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Occurrence of phthalate diesters (phthalates), *p*-hydroxybenzoic acid esters (parabens), bisphenol A diglycidyl ether (BADGE) and their derivatives in indoor dust from Vietnam: Implications for exposure

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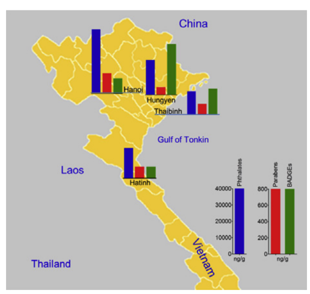
HIGHLIGHTS

- Phthalates, parabens, and BADGEs were found widely in indoor dust from Vietnam.
- Median concentration of phthalates and parabens was 22,600 and 123 ng/g, respectively.
- Median concentration of BADGEs in dust was 184 ng/g.
- Median exposure to phthalates through dust ingestion by toddlers was 90 ng/kg-bw/d.

GRAPHICAL ABSTRACT



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ABSTRACT

Phthalate diesters (phthalates), esters of *p*-hydroxybenzoic acid (parabens), and bisphenol A diglycidyl ether (BADGE) are used in personal care products, food packages, household products, or pharmaceuticals. These compounds possess endocrine-disrupting potentials and have been reported to occur in the environment. Nevertheless, no previous studies have reported the occurrence of these compounds in indoor dust from Vietnam. In this study, nine phthalates, six parabens, and four BADGEs were determined in indoor dust samples collected from Hanoi, Hatinh, Hungyen, and Thaibinh, in Vietnam. Total concentrations of phthalates, parabens, and BADGEs in indoor dust ranged from 3440 to 106,000 ng/g (median: 22,600 ng/g), 40–840 ng/g (median: 123 ng/g), and 23 to 1750 ng/g (median: 184 ng/g), respectively. Based on the measured median concentration of phthalates, parabens, and BADGEs in indoor dust, we estimated human exposure doses to these compounds through indoor dust ingestion for various age groups. The exposure doses to phthalates, parabens, and BADGEs decreased with age and ranged from 19.4 to 90.4 ng/kg-bw/d, 0.113–0.528 ng/kg-bw/d, and 0.158–0.736 ng/kg-bw/d, respectively. This is the first study on the occurrence and human exposure of phthalates, parabens, and BADGEs in indoor dust from Vietnam.

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1. Introduction

Owing to the widespread use of phthalates, parabens, and BADGEs as additives or preservatives in various consumer products, contamination by these compounds in the indoor environment is inevitable. The ubiquitous occurrence of phthalates, parabens, and BADGEs in the indoor environment in many countries has been reported in recent studies (Wang et al., 2012a; Guo and Kannan, 2013; Guo et al., 2014; Ma et al., 2014). These compounds possess endocrine-disrupting potentials, and, therefore, human exposure to these chemicals is a public health concern.

Phthalates are used in various consumer, household, and industrial products as plasticizers. In cosmetics and personal care products, diethyl phthalate (DEP) and di-*n*-butyl phthalate (DBP) were found at concentrations as high as 25,500 and 24,300 μg/g, respectively (Koniecki et al., 2011; Guo and Kannan, 2013; Guo et al., 2014). Phthalates were reported to occur in indoor air at hundreds to thousands of ng/m^3 and in indoor dust at hundreds to thousands of ng/g (Fromme et al., 2004, 2005; Bergh et al., 2011; Guo and Kannan, 2011; Bergh et al., 2012; Kubwabo et al., 2013; Blanchard et al., 2014; Tran and Kannan, 2015). Phthalates elicit reproductive and developmental toxicities in laboratory animals (Gray et al., 2000, 2006; Boberg et al., 2008). Gaspar et al. (2014) reported that 82–89% of children in California had DBP exposures exceeding the reproductive health benchmarks. Therefore, to develop strategies to mitigate exposures, a comprehensive assessment of sources of human exposure to phthalates is necessary.

Parabens are widely used in foods, cosmetics, and pharmaceuticals (Soni et al., 2005; Fei et al., 2011; SCCS, 2011). The concentrations of five parabens in foods from the USA were as high as 409 ng/g (Liao et al., 2013b) and ranged from 0.839 ng/g in beverages to 100 ng/g in vegetables collected from China (Liao et al., 2013a). Wang et al. (2012a) reported median concentration of six parabens in indoor dust samples from the USA (1560 ng/g), China (573 ng/g), South Korea (2180 ng/g), and Japan (1850 ng/g). Several earlier studies have reported the occurrence of parabens in human blood, urine, and milk (Darbre et al., 2004; Schlumpf et al., 2010; Frederiksen et al., 2011). The concentrations of the sum of four parabens in urine samples collected from Chinese young adults ranged from 0.82 to 728 ng/mL (Ma et al., 2013), and concentrations as high as 10,000 ng/mL were found in urine from Chinese adult females (Wang et al., 2013). Studies have reported toxic effects of parabens in laboratory animals (Miller et al., 2001; Okubo et al., 2001; Byford et al., 2002; Oishi, 2002a, 2002b; Golden et al., 2005; Darbre and Harvey, 2008; Kim et al., 2011). In an effort to reduce the risk of exposure to parabens, Denmark announced a ban on the use of two parabens, propyl- (PrP) and butyl-paraben (BuP), their isoforms, and salts in children's cosmetic products (SCCS, 2011).

Bisphenol A diglycidyl ether (BADGE) is a building block of epoxy resins and is used to make the inner surface coating of food cans (Wang et al., 2012a, 2012b). BADGE is a reactive molecule, and, following contact with aqueous and acidic media, chlorinated and hydrated derivatives such as bisphenol A (2,3-dihydroxypropyl) glycidyl ether (BADGE·H_2O), bisphenol A (2,3-dihydroxypropyl) ether (BADGE·$2H_2O$), and bisphenol A (3-chloro-2-hydroxypropyl) (2,3-dihydroxypropyl) ether (BADGE·HCl·H_2O) (collectively referred to in this study as BADGEs) can be formed. The toxic effects of BADGEs have been reported in several laboratory studies (Nakazawa et al., 2002; Satoh et al., 2004; Ramilo et al., 2006; Hyoung et al., 2007; Yang et al., 2010). Studies also have reported the occurrence of BADGEs in foodstuffs (Lintschinger and Rauter, 2000; Petersen et al., 2003; Coulier et al., 2010) and in human urine from the USA and China (Wang et al., 2012b), Japan (Hanaoka et al., 2002), Greece (Asimakopoulos et al., 2014), and India (Xue et al., 2015). An earlier study reported median concentrations of BADGEs in dust samples collect from the USA, China, South Korea, and Japan at 1350, 1410, 2380, and 2020 ng/g, respectively (Wang et al., 2012a). Prior to this study, no study reported the occurrence of phthalates, parabens, or BADGEs in dust from Vietnam. In this study, nine phthalates, six parabens, and four BAGDEs were determined in 46 indoor dust samples collected from four cities in Vietnam. Based on the concentrations measured in dust, human exposure to these compounds via dust ingestion was estimated.

2. Materials and methods

2.1. Standards and chemicals

Analytical grade acetone was purchased from Macron Chemical (Nashville, TN, USA), and formic acid (98.2%) was purchased from Sigma–Aldrich (St. Louis, MO, USA). Hexane, dichloromethane, and methanol (HPLC grade) were purchased from J. T. Baker (Phillipsburg, NJ, USA). Milli-Q water was prepared using an ultrapure water system (Barnstead International, Dubuque, IA, USA). Nine phthalate diesters, *viz*., dimethyl phthalate (DMP), diethyl phthalate (DEP), diisobutyl phthalate (DIBP), di-*n*-butyl phthalate (DBP), di-*n*-hexyl phthalate (DNHP), benzyl butyl phthalate (BzBP), dicyclohexyl phthalate (DCHP), di(2-ethylhexyl) phthalate (DEHP), and di-*n*-octyl phthalate (DOP), along with their corresponding d_4 (deuterated) internal standards, with a purity of >99%, were purchased from AccuStandard Inc. (New Haven, CT, USA). Methyl paraben (MeP), ethyl paraben (EtP), propyl paraben (PrP), butyl paraben (BuP), benzyl paraben (BzP), and heptyl paraben (HepP) were purchased from AccuStandard, Inc. Bisphenol A diglycidyl ether (BADGE, ≥95%), bisphenol A (2,3-dihydroxypropyl) glycidyl ether (BADGE·H_2O, ≥95%), bisphenol A (3-chloro-2-hydroxypropyl) (2,3-dihydroxypropyl) ether (BADGE·HCl·H_2O, ≥95%), and bisphenol A (2,3-dihydroxypropyl) ether (BADGE·$2H_2O$, ≥97%) were purchased from Sigma–Aldrich (Table S1; Supporting Information). ^{13}C-labeled 2-hydroxy-4-methoxybenzophenone ($^{13}C_{12}$-BP-3) (99%), $^{13}C_6$-MeP (99%), $^{13}C_6$-BuP (99%), and $^{13}C_{12}$-BADGE (99%) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA).

2.2. Sample collection

Indoor dust samples were collected during April and May 2014 in Hanoi ($n = 18$), Hatinh ($n = 12$), Hungyen ($n = 8$), and Thaibinh ($n = 8$), Vietnam. The sampling locations were grouped into four categories: homes (living rooms and kitchens, $n = 16$), supermarkets/grocery shops, electronic stores and pharmacies; ($n = 18$), laboratories (Hanoi only, $n = 7$), and offices ($n = 5$); all of the sites were built after 2000. Floor dust samples were collected using a vacuum cleaner or by sweeping the floor directly with a broom. Samples were stored in polyethylene bags and then placed in glass jars at 4 °C until analysis.

2.3. Sample preparation

Prior to analysis, dust samples were sieved and homogenized by passage through a 150 μm sieve. Fifty nanograms of d_4-phthalates (except for d_4-DEHP, for which 250 ng was spiked) were spiked onto 50–60 mg of dust samples, as internal standards. The spiked dust samples were equilibrated for 30 min at room temperature. The dust samples were extracted three times by shaking in an orbital shaker (Eberbach Corp., Ann Arbor, MI, USA) with a 4 mL mixture of dichloromethane (DCM) and hexane (3:1, v:v) for 10 min each time. After shaking, samples were centrifuged at 2000 g for 5 min (Eppendorf Centrifuge 5804, Hamburg, Germany), and the supernatant was transferred into a 15 mL glass tube. The extracts