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Phthalates and polybrominated diphenyl ethers in retail stores

Ying Xu [a,*], Yirui Liang [a], Jorge R. Urquidi [a], Jeffrey A. Siegel [b]

[a] Department of Civil, Architectural and Environmental Engineering, The University of Texas at Austin, 301 E. Dean Keeton Street, Austin, TX 78712, USA
[b] Department of Civil Engineering, University of Toronto, 35 St. George Street, Toronto, ON M5S 1A4, Canada

HIGHLIGHTS

- We measured 6 phthalates and 14 PBDEs in 12 retail stores in TX and PA, U.S.
- DEP, DnBP, and DEHP were the most abundant compounds among phthalates.
- PBDEs were dominated with BDE-28, -99, and -209 in indoor air samples.
- Results are comparable to previous studies in other types of indoor environments.
- Air concentration of pollutants was not a significant function of air change rate.

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ABSTRACT

Retail stores contain a wide range of products that can emit a variety of indoor pollutants. Among these chemicals, phthalate esters and polybrominated diphenyl ethers (PBDEs) are two important categories, because they are extensively used as additives in consumer products and associated with serious health concerns. This study measured six phthalate and 14 PBDE compounds inside of 12 retail stores in Texas and Pennsylvania, U.S. Phthalates and PBDEs were widely found in the retail environment, indicating that they are ubiquitous indoor air pollutants. DEP, DnBP, and DEHP were the most abundant phthalates, with DnBP showing the highest concentration (0.23 ± 0.36 μg m^{-3}). PBDEs were dominated by BDE-28, -99, and -209, having concentrations as high as 0.85 ± 1.99 ng m^{-3} (BDE-99). The levels of phthalates and PBDEs measured in this study are comparable to concentrations found in previous investigations of residential buildings, with phthalates showing lower concentrations and PBDEs exhibiting higher concentrations in retail stores. The potential co-occurrence of phthalates was not as strong as that of PBDEs, suggesting that phthalates might have more diverse sources. Whole building emission rates were calculated and showed similar patterns of variations as indoor air concentrations, suggestion the diversity of indoor sources of phthalates and PBDEs in retail environments.

1. Introduction

Many building materials and consumer products used in indoor environments emit harmful contaminants (Weschler, 2009). Emissions from these sources produce indoor concentrations that are substantially higher than those found outdoors (Rudel and Perovich, 2009). Since the 1950s, levels of some indoor pollutants (formaldehyde, aromatic and chlorinated solvents, chlorinated pesticides, and polychlorinated biphenyls [PCBs]) have increased and then decreased (Weschler, 2009). In contrast, levels of phthalates and polybrominated diphenyl ethers (PBDEs) have increased and remain high (Rudel and Perovich, 2009; Weschler, 2009). Phthalates and PBDEs are used extensively in building materials and consumer products such as vinyl flooring, carpeting, wall coverings, electronics, and furniture, as plasticizers and flame retardants, respectively. Members of both the chemical families are classified as semi-volatile organic compounds (SVOCs) due to their low vapor pressures at room temperature, and they are often present in the finished product at percent to tens-of-percent levels (Darnerud et al., 2001; Hites, 2004; Weschler and Nazaroff, 2008). Because these additives are not chemically bound to polymers, they slowly migrate into the surrounding environment. Phthalates and PBDEs are ubiquitous indoors, redistributing from their original source to indoor air and all interior surfaces, including airborne particles, dust, and skin (Weschler and Nazaroff, 2008, 2012; Xu et al., 2009, 2010; Xu and Little, 2006). Recent field measurements suggest that indoor environments play an important role in phthalates and PBDE exposures (Allen et al., 2007; Bornehag et al.,

* Corresponding author. The University of Texas at Austin, Civil, Architectural and Environmental Engineering Department-ARE, 301 E. Dean Keeton St., Stop C1752, Austin, TX 78712-1098, USA. Tel.: +1 512 471 6507; fax: +1 512 475 3191.
E-mail address: xuying@mail.utexas.edu (Y. Xu).

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2005; Frederiksen et al., 2009; Harrad et al., 2006; Jaakkola et al., 2004; Meeker et al., 2009; Sjodin et al., 2008).

The serious adverse effects associated with phthalates and PBDEs have been detailed in several recent reviews (Alcock et al., 2011; Birnbaum et al., 2003; Darnerud, 2003; Heudorf et al., 2007; Jaakkola and Knight, 2008; Latini et al., 2006; Legler and Brouwer, 2003; Matsumoto et al., 2008; McKee et al., 2004; Meerts et al., 2000; Ritter and Arbucklet, 2007; Vonderheide et al., 2008). Collectively, these reviews show that exposure to phthalates results in profound and irreversible changes in the development of the reproductive tract, especially in males. Effects such as increases in prenatal mortality, reduced growth and birth weight, and skeletal, visceral, and external malformations, are also associated with exposure to phthalates. In addition, epidemiologic studies in children show associations between phthalates and the risk of asthma and allergies. Exposure to PBDEs may cause changes in the development of brain and nerve tissues. Effects such as permanent learning and memory impairment, behavioral changes, delayed puberty onset, decreased sperm count, sex hormone alteration, fetal malformations, thyroid hormone disruption, and possibly cancer are also associated with exposure to PBDEs.

Despite the negative health impacts, phthalates are still dominating the plasticizer market (Schossler et al., 2011). The global production rate of phthalate plasticizers has increased from 2.5 million tons/year to 6 million tons/year within a decade (Cadogan and Howick, 1996; Rudel and Perovich, 2009). In 2009, the U.S. Consumer Product Safety Improvement Act (CPSIA) was enacted to restrict the use of phthalates in toys and child care articles (GovTrack, 2007). As a result, phthalates used in polymeric products are changing rapidly, with a shift from low to high molecular phthalates (Weschler, 2009). PBDEs have 209 possible congeners and are manufactured in three commercial products that are used in building and consumer products: Penta-, Octa-, and Deca-BDE mixtures. In the United States, Penta- and Octa-BDE mixtures were phased out in 2004. Deca-BDE is banned in electrical and electronic applications in Europe and is committed to end production, import and sales by the end of 2013 by U.S. producers and importers (Dodson et al., 2012). Alternative plasticizers, such as di(2-ethylhexyl) terephthalate (DEHT) and diisononyl cyclohexane-1,2-dicarboxylate (DINCH), and alternate flame retardants, such as tetrabromobisphenol-A, hexabromocyclododecane, and bis(2,4,6,-tribromphenoxy) ethane, have emerged very recently, but toxicological information on these compounds is lacking. Given that these alternatives share properties similar to those of phthalates and PBDEs, respectively, similar levels of emission, environmental fate and transport, and human exposure are expected (Stapleton et al., 2008). Furthermore, because typical lifetimes for some building materials and consumer products may exceed ten years (Stapleton et al., 2006), and these chemicals may continue to be produced in developing countries and exported to the U.S. as additives in finished products, the levels of phthalate and PBDE in indoor environments are not likely to drop for a long time (Park et al., 2011).

Retail stores are an understudied indoor environment considering occupant risk and economic implications associated with poor indoor air quality. Retail buildings account for approximately one-third of U.S. commercial buildings (Diamond, 2001). The retail sector employs about 15 million workers, approximately 10% of the U.S. workforce (NRF, 2010). Additionally, the average percentage of people engaged in a shopping activity per day is estimated to be around 43% of the U.S. population. These shoppers spend 1.7 h on average of their daily time shopping (ATUS, 2012). However, retail stores contain a wide range of new products that may emit a variety of indoor pollutants, leading to higher levels of these compounds indoors (Wu et al., 2011). Thus, the indoor environmental quality of retail buildings has a potential to significantly affect occupants' health, their perception of the retail environments, as well as the occupational health exposures for retail workers. Although several articles have examined indoor pollutant concentrations in retail spaces (e.g., Caselli et al., 2009; Eklund et al., 2008; Loh et al., 2006; Wu et al., 2011), these studies tend to focus on volatile organic compounds (VOCs) and particle mass concentrations and there have been few investigations into SVOCs. Hartmann et al. (2004) measured concentrations of eight organophosphates in car, retail and office environments in Switzerland. The retail environments included electronics and furniture stores. Tributyl phosphate (TBP), tris (2-chloroethyl) phosphate (TCEP), and triphenyl phosphate (TPP) were found in all the retail stores. Tris (2-chloro-isopropyl)phosphate (TCPP) was found in furniture stores, but not in electronics stores, and tris (1,3-dichloroisopropyl) phosphate (TDCP) was not found in any of the locations studied. Wu et al. (2011) measured diethyl phthalate (DEP), as well as several VOC concentrations in 37 small- and medium-sized commercial buildings distributed across different sizes, ages, uses, and regions of California, and DEP was found in all seven of the retail sites. Phthalates and PBDEs have been measured in indoor air and dust in studies of residential and educational environments in North America, Europe and Asia (Bergh et al., 2011b; Harrad et al., 2006; Kanazawa et al., 2010; Rudel et al., 2010; Thuresson et al., 2012; Vorkamp et al., 2011), but their data for other types of buildings, such as retail spaces, are rare.

This study is part of a research project focusing on relationships between ventilation rate and indoor air quality and occupant satisfaction in retail environments. The objectives of the present paper are: (i) to report the concentrations of targeted phthalates and PBDEs measured in air samples collected from retail stores ($n = 12$) in Texas (TX) and Pennsylvania (PA); (ii) to compare the results with concentrations reported in other studies to provide insights into the variations over location and building types; (iii) to examine potential correlations among concentrations of different SVOC pollutants; (iv) to investigate relationships between SVOC concentrations and ventilation; and (v) to report whole building emission rates for phthalates and PBDEs in retail environments.

2. Materials and methods

2.1. Buildings sampled

The retail buildings investigated for this paper were part of a large project, Ventilation and Indoor Air Quality in Retail Stores (ASHRAE RP-1596). Twelve retail buildings located in Austin, TX and central Pennsylvania were visited. Table 1 describes the sample in more detail. The first column of Table 1 is a unique site identifier where the first letter is the store type (H = home improvement, M = general merchandise, F = furniture, E = electronics, G = mid-sized grocery, and S = small grocery), the second letter is a unique brand identifier, and the third character indicates whether the site was located in Pennsylvania (P) or Texas (T). In later parts of this paper, for those stores that were visited more than once, a number is added to the site identifier to differentiate between visits to the same site. Additionally, one site, a general merchandise store (MiT), was an intervention site and was tested for two consecutive weeks. During the first week, the ventilation rate was increased to its maximum capacity and the second week the store was returned to its typical operation. Indoor concentrations of the target compounds were measured at the same location during each sampling event. The test at the higher ventilation rate is generally excluded from summary data. Details on each building site and other measurements can be found in Siegel et al. (2013).