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Sorption of Semi-Volatile Organic Compounds to Dust and Other Surfaces in Indoor Environments

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Dedication

To my wife and parents.

Acknowledgements

Five years ago, as a fresh-faced master student who decided to pursue a Ph.D. degree at UT Austin. I thought the process of completing research work for obtaining a doctoral degree should be like walking on a treacherous road at dark night. I have to rely heavily on myself and struggle to find a way towards the light. Looking back, I realized I had it all wrong. It is not the challenges and hurdles that make me stronger and unbreakable, but the support and companionship of the inspiring people around me encourage me to achieve the goals.

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Abstract

Sorption of Semi-Volatile Organic Compounds to Dust and Other Surfaces in Indoor Environments

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Semi-volatile organic compounds (SVOCs) are ubiquitous in indoor environments. Because they partition strongly to dust other surfaces in the indoor environments, most SVOCs persist for years after the source is removed. Biomonitoring data based on blood and urine testing suggested the universal and significant human exposure to SVOCs, which may result in serious adverse health effects. However, because of the poor understanding of their transfer process from sources to indoor surfaces, significant uncertainties exist for the estimation of exposure to SVOCs through various pathways and effective strategies to limit such exposure remains hamstrung. The goal of this dissertation is to explicitly elucidate the sorption of important and emerging SVOCs to dust and other surfaces in the indoor environments. The specific research objectives are to 1) investigate the emission, sorption, and fate of phthalates in a residential test house; 2) characterize the direct transfer of SVOCs from sources to settled dust through systematic chamber study; and 3) measure SVOC levels in heating, ventilation, and air conditioning (HVAC) filter dust of U.S. low-income homes and investigate their association with concentrations in settled dust, seasons, building characteristics, and childhood asthma. Strong sorption of phthalates was observed on interior surfaces, including dust, dish plates, windows, mirrors, fabric cloth,

and wood, in a residential test house. In addition, when dust is in contact with the PVC floorings, equilibrium dust concentrations of phthalates are orders of magnitude higher than typical dust concentrations reported in the literature. And we found that the equilibrium concentrations of phthalates in dust can be predicted with the concentrations of phthalates within the gas layer in adjacent with the flooring materials. Finally, the results suggest that HVAC filter dust is a useful sampling media to monitor indoor SVOC concentrations with high sensitivity. When using settled dust, in addition to considering seasonal influences, it is very important to know the sampling location because the types and levels of SVOCs might be related to the local materials.

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Chapter 1: Introduction

Building materials and consumer products contain SVOCs, which can be slowly emitted to the indoor environments (Weschler, 2009). Emissions from these sources produce indoor concentrations that are substantially higher than those found outdoors (Rudel and Perovich, 2009). Since the 1950s, increased levels of some volatile indoor pollutants (e.g., formaldehyde, aromatic and chlorinated solvents, chlorinated pesticides and polychlorinated biphenyls (PCBs)) raised the concerns of researchers and experiments have been conducted to reduce the levels of those pollutants. In contrast, levels of phthalates and organophosphates have increased and remained high (Klepeis et al., 2001; Rudel and Perovich, 2009). Previous studies have shown that phthalates and organophosphates are ubiquitous and among the most abundant indoor contaminants in residential homes (Bennett et al., 2014; Blanchard et al., 2014b; Takeuchi et al., 2014; Mandin et al., 2016). Those SVOCs are extensively used in building materials and consumer products such as vinyl flooring, carpeting, wall coverings, electronics, and furniture, and they are often present in the product at percent to tens-of-percent levels (Weschler and Nazaroff, 2008). Because these additives are not chemically bounded to the polymer matric, they slowly migrate from the sources to indoor air, settled dust, suspended particles, and other indoor surfaces.

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). However, following the restrictions on using certain phthalates in toys and child care products (CPSC, 2008), phthalates used in PVC products are changing rapidly, with a trend toward using phthalates of higher molecular weight and lower volatility (Weschler, 2009; Schossler et al., 2011). The use of alternative plasticizers, such as diisononyl cyclohexane-1,2-dicarboxylate (DINCH) and di(2-ethylhexyl) adipate (DEHA), has also occurred very recently. Organophosphates are estimated to account for about 11.5% of world consumption of flame retardants (200 000 tones/year, 800 million USD by value) (Xu et al., 2016). Phosphate additives are commonly used as flame retardants, stabilizers, and/or plasticizers in a variety of consumer goods such as furniture, textile, electronics, and baby products (Saito et al., 2007; Stapleton et al., 2009, 2011, 2012; Gallen et al., 2014a; Cooper et al., 2016). Restrictions on the use of polybrominated diphenyl ether (PBDE) flame retardants have led to an increase in the consumption of organophosphates as replacements (Stapleton et al., 2011; Bergman et al., 2012a; van der Veen and de Boer, 2012; Araki et al., 2014; Castorina et al., 2017). The global consumption of organophosphate flame retardants was 300,000 tons in 2004, increasing to 680,000 tons in 2015 (Giulivo et al., 2017), and was projected to grow at a rate of 5.2% per year until 2021 (Lucintel, 2016).

Exposure to phthalates and organophosphates may result in adverse health effects, which are primarily associated with carcinogenic, neurological, reproductive and developmental toxicity. For example, exposure to certain phthalates, such as dibutyl phthalate (DnBP), butyl benzyl phthalate (BBzP), and di-(2-ethylhexyl) phthalate (DEHP), has shown profound and irreversible changes in the development of the reproductive tract, especially in males (Latini et al., 2006; Matsumoto et al., 2008; Martino-Andrade and Chahoud, 2010; Kay et al., 2013, 2014; Zarean et al., 2016). Two widely used organophosphates, tri(2-chloroethyl) phosphate (TCEP) and tris(1,3,-dichloroisopropyl) phosphate (TDCPP), have been designated as carcinogens by the state of California (State of California, 2016). Adverse health effects such as decreased fetal growth, autism spectrum disorders, disruption of hormones, altered behavior at early life stages, decreased quality of semen, and alteration of thyroid function have also been associated with exposure to phthalates and organophosphates (Meeker and Stapleton, 2010; van der Veen and de Boer, 2012; Meeker et al., 2013; Patisaul et al., 2013; Araki et al., 2014; Dishaw et al., 2014; Miodovnik et al., 2014; Ferguson et al., 2016; Zhao et al., 2017; Hoffman et al., 2017; Preston et al., 2017; Schang et al., 2018; Ospina et al., 2018). In contrast, the association between exposure to phthalates and organophosphates and allergic diseases has not received substantial attention. Several epidemiological studies have reported links between certain phthalates present in dust or PVC materials in homes and asthma and allergic diseases in children

(Bornehag et al., 2004; Jaakkola and Knight, 2008; Kolarik et al., 2008a; Larsson et al., 2010; Hsu et al., 2012a; Ait Bamai et al., 2014b, 2016). One study reported an association between organophosphates and the prevalence of asthma and allergies (Araki et al., 2014). However, other studies conducted with similar approaches seemed to contradict these findings (Callesen et al., 2014b; Beko et al., 2015).

SVOCs such as phthalates and organophosphates are known to sorb strongly to indoor surfaces including airborne particles, settled dust, stationery room surfaces, clothing, and even human (and animal) occupants due to their low vapor pressure. Especially for some high molecular weight SVOCs, more than 90 percent of their total mass indoors are ad/absorbed on indoor surfaces (Weschler and Nazaroff, 2008). Several studies have found that most impenetrable indoor surfaces contain a very thin organic film at their interface with room air (Diamond et al., 2000; Liu et al., 2003; Butt et al., 2004; Weschler and Nazaroff, 2008, 2017). Gas-phase SVOCs in indoor air can sorb to these surfaces by partitioning to the film layer. Therefore, these surfaces may affect surface-air exchange of SVOCs and their residence time in indoor environments. For porous material surfaces such as gypsum wallboard, carpet, clothing and furniture foam, SVOCs can diffuse into the material and sorb there (Petrick et al., 2010; Liang and Xu, 2015). Over time, such sorption processes may establish significant reservoirs that may act as secondary sources in indoor environments. Although the sorption of volatile organic compounds (VOCs) to building materials and other common indoor materials has been well studied (Won et al., 2000, 2001), few studies have investigated the sorption of phthalates and organophosphates (Clausen and Hansen, 2004; Xu et al., 2012; Liang and Xu, 2014a, 2015; Wu et al., 2017).

Environmental factors such as indoor temperature and humidity may significantly impact the sorption of SVOCs to surfaces in indoor environment. Temperature variations inside buildings are very common and are mainly associated with outdoor temperature changes (Coley and Kershaw, 2010; Mavrogianni et al., 2012; Nazaroff, 2013; Nguyen et al., 2014). Temperature may have a strong influence on the fate of phthalates in indoor environments because phthalate emission and sorption properties depend strongly on temperature. Clausen et al. (2012) observed an order of magnitude increase in di-2-ethylhexyl phthalate (DEHP) emissions in experimental chambers when the temperature was increased by 10 ºC. Similar results were obtained by Liang and Xu (2014a) when measuring the emission rate of phthalates from a range of vinyl flooring and crib mattress covers at different temperatures. In contrast, with increasing temperature, decreasing surface/air partition coefficients were observed for SVOCs (i.e., polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and DEHP between air and surfaces such as particles (Naumova et al., 2002, 2003; Arp et al., 2008), plant leaves (Kömp and McLachlan, 1997), and stainless steel (Clausen et al., 2012). Despite that the emission of phthalates were found to be sensitive to temperature through chamber measurements, limited work has investigated the influence of temperature on the fate and transport of phthalates in a real residential environment under well-controlled temperature program. Dampness (represented by relative humidity) in buildings has been found to be a strong risk indicator for health effects such as asthma (Jaakkola and Knight, 2008). Several field studies suggested that increased levels of phthalates were associated with signs of higher dampness in residential homes (Hsu et al., 2012b; Zhang et al., 2013; Ait Bamai et al., 2014a). However, little is known on the association between humidity and the concentrations of phthalates in the indoor environments. No study has investigated the influence of humidity on the sorption of phthalates between source materials and indoor settled dust.

Particles have a great influence on the fate and transport of indoor SVOCs. Because the sorption of SVOCs to airborne particles, which have large surface to volume ratio, are significantly strong and sufficiently rapid (Weschler and Nazaroff, 2008; Benning et al., 2013; Liu et al., 2013; Xie et al., 2013), particles become important carriers that accelerate the transport of SVOCs from their original source to other indoor surfaces through deposition, resuspension, and advection of air. Settled dust is deposited particle and may serve as strong sinks for SVOCs due to its wide occurrence in indoor environments. In particular for dust, which is in contact with the sources of SVOCs, the SVOC concentrations were possibly orders of magnitudes higher than those in dust settled on non-source surfaces due to the potential direct transfer of SVOCs from source to dust. Clausen and Hansen (2004) observed a seven-fold increase of DEHP concentration in dust settled on PVC flooring containing DEHP. Other chamber studies also suggested that PVC materials which contained phthalates can greatly enhance the phthalate concentration in dust which were in contact with those source materials (Schripp et al., 2010; Jeon et al., 2016). Besides, several field studies provided evidence that there might be a direct transfer of phthalates from PVC floorings into dust in contact with the floorings (Sukiene et al., 2016, 2017). However, very few of those studies have investigated the mass transfer process of SVOCs from source to dust in a systematic way.

The highly contaminated dust may result in significant exposure when inhaled and ingested by indoor occupants. The settled dust sorbed with high levels of SVOCs may transfer to hand and other skin surfaces when occupants have physical contacts with indoor surfaces and consequently be ingested through hand-to mouth activities. Particularly for infants and toddlers who frequently play on the ground, pick up dust through touching indoor surfaces, and constantly put their hands in the mouth, the exposure through ingestion of dust-phase SVOCs can be considerably greater than those for adults. Inhalation of the resuspended dust is also important for human exposure to SVOCs. Previous study showed that crawling-induced resuspension of settled floor dust may significantly enhance the exposure to inhalable particles for infants (Boor et al., 2016). Particles-bound phthalates may desorb from the particle after entering the airways and may create high localized concentrations at the particle deposition site and further cause bronchial obstruction (Oie et al., 1997; Jaakkola and Knight, 2008; Liu et al., 2017). Therefore, to accurately determine the exposure to SVOCs for indoor occupants, it is necessary to have a comprehensive understanding on the sorption of SVOCs to particles and dust.

To fill the knowledge gap, there is a need to explicitly elucidate the sorption of SVOCs to dust and other surfaces in indoor environments. The work in this dissertation is grouped to fulfill three objectives:

1. Investigate the Emission, Sorption, and Fate of phthalates in a Residential Test House.
2. Characterize the Direct Transfer of SVOCs from Sources to Settled Dust Through Systematic Chamber Study.
3. Measure SVOC Levels in HVAC Filter Dust of U.S. Low-Income Homes and Investigate Their Association with Concentrations in Settled Dust, Seasons, Building Characteristics, and Childhood Asthma.

The dissertation is organized in two major sections: (1) the Summary of Methods and Research Findings (Chapter 3), and (2) Appendices A-C, which include three full-length manuscripts corresponding to the three investigations. The three paper topics are as follows:

Paper 1. Fate and Transport of Phthalates in Indoor Environments and the Influence of Temperature: A Case Study in a Test House (Objective 1; Appendix A).

Paper 2. Transfer of Phthalates and Their Alternatives from Polyvinyl Chloride Flooring and Crib Mattress Cover into Settled Dust (Objective 2; Appendix B).

Paper 3. Phthalates and Organophosphates in Settled Dust and HVAC Filter Dust of U.S. Low-Income Homes: Association with Season, Building Characteristics, and Childhood Asthma (Objective 3; Appendix C).

Chapter 2: Literature Review

2.1 DUST AS A METRIC FOR STUDYING THE OCCURRENCE OF SVOCS

Settled dust (i.e., deposited particles) that is sampled from floors, mattresses, moldings, and shelves has been widely used as an indicator for residential contamination (Butte and Heinzow, 2002; Lioy et al., 2002; Mercier et al., 2011), especially for SVOCs. The house dust serves as an indoor sink, collecting and accumulating SVOC pollutants in a manner similar to a passive sampler. Field studies of phthalates were measured in settled dust samples in field campaigns (Table 1), with DEHP as the most abundant phthalate compound in indoor settled dusts. A CTEPP study measured the total exposure of 257 preschool children (ages 2 to 5 years) and their primary adult caregivers to more than 50 target compounds in homes and daycare centers (US EPA, 2005). The two phthalates targeted in the CTEPP study (BBzP and DEHP) were detected in residential air, house dust, and dermal wipe samples. The measured phthalate concentrations were the highest among the selected pollutants (including pesticides, PAHs, and polychlorinated biphenyls). Although the concentrations of organophosphates are substantially less compared with those of phthalates, they are also wildly present in indoor environments. As shown in Table 2, TCPP and TBEP are the most abundant organophosphates in indoor dust samples because of their use as flame retardants in polyurethane foams as building insulation materials. Collectively, these field studies reveal the presence of phthalates and organophosphates in indoor environments is in large amounts and in a wide range of products.

However, the age of settled dust is usually unknown, which may result in potential uncertainty for exposure assessments. For chemicals with high K_{oa} values, if the dust does not have sufficient residence time indoors to equilibrate with the gas phase, the SVOC compounds tend to be underrepresented in dust relative to their abundance in air (Weschler and Nazaroff, 2010). Furthermore, settled dust samples are localized and generally only representative for the location where they are collected. Samples collected in the same house but at multiple locations may vary significantly in chemical composition, due to existence of different sources and different origins of dust (Bergh et al., 2011b; Sukiene et al., 2017). In addition, to understand the potential respiratory health effects of exposure to SVOCs, it is crucial to measure their concentrations in inhalable particles (with aerodynamic diameter <10 μm) and respirable particles (with aerodynamic diameter <2.5 μm). Settled dust may not be suitable for this purpose, because it only contains a small fraction of particles in these size ranges (Butte and Heinzow, 2002; Gustafsson et al., 2018). Although traditional air sampling has also been applied to determine SVOC levels in suspended particles (e.g., PM_{10}) (Wang et al., 2014; Zhang et al., 2014; Mandin et al., 2016), it may only provide a snapshot of the concentrations, which can vary significantly with time and within a building.

A potential alternative to settled dust is the dust collected by filters of building heating, ventilation, and air conditioning (HVAC) systems. More than 70% of residential homes in the U.S. have a central forced-air HVAC system (US HUD, 2011), almost all with built-in filters that can be collected with minimal effort. The HVAC filters are in place for long, potentially known periods of time, collect particles from a wide spatial area with large volumes of air, and thus provide a spatially and temporally integrated way for evaluations of indoor contamination. HVAC filter dust has been successfully used to characterize indoor microbial communities (Stanley et al., 2008a; Noris et al., 2009, 2011) and heavy metals (Noris et al., 2009) but to date, there is very limited research addressing SVOC contaminants (Batterman et al., 2010; Xu et al., 2014b; He et al., 2016). To our knowledge, very limited studies has measured phthalate and organophosphate concentrations in HVAC filter dust in residential homes. Their relationship with SVOC concentrations in settled dust and the influence of seasonal variations and building characteristics remain unknown. Such information is needed not only for better understanding the indoor sources and exposures to SVOCs, but also for investigating the feasibility of the novel filter forensics methodology comparing to conventional sampling approaches in indoor environments.

Table 1: Phthalate concentrations (µg/g) in indoor dust as reported in relevant studies. All values are medians with range (minimum – maximum) unless stated otherwise.

Country	**Reference**	**N**	**Building type**	**DMP**	**DEP**	**DnBP**	**BBzP**	**DEHP**	**DnOP**
North America									
U.S.	Wilson et al. (2001)	29	Day care	n.a.	n.a.	18.4[a] (1.58-46.3)	67.7[a] (15.1-175)	n.a.	n.a.
U.S.	Rudel et a l. (2003)	120	Residence	n.a.	4.98 (<d.l.-111)	20.1 (<d.l.-352)	45.4 (3.87-1310)	340 (16.7-7700)	n.a.
U.S.	Wilson et al. (2003)	4	Day care	n.a.	n.a.	1.87[a] (0.06-5.85)	3.72[a] (0.02-7.43)	n.a.	n.a.
U.S.	Wilson et al. (2003)	9	Residence	n.a.	n.a.	1.21[a] (0.38-3.03)	5.86[a] (0.50-15.6)	n.a.	n.a.
U.S.	Hwang et al. (2008)	10	Residence	n.a.	n.a.	n.a.	n.a.	386 (104-2050)	n.a.
U.S.	Guo and Kannan (2011)	33	Residence	0.08 (>d.l.-3.3)	2 (0.7-11.8)	13.1 (4.5-94.5)	21.1 (3.6-393)	304 (37.2-9650)	0.4 (<d.l.-14.1)
U.S.	Xu et al. (2014)	24	Retail store	<d.l. <d.l.	72 (6-591)	258 (8-961)	148.5 (14-3262)	192 (12-4187)	117 (16-343)
U.S.	Dodson et al. (2015)	49	Residence	n.a.	2.1 (<d.l.-85)	11 (<d.l.-56)	19 (<d.l.-330)	140 (50-800)	1.6 (0.42-9.4)
Europe									
Norway	Oie et al. (1997)	372	Residence	n.a.	10[a] (<d.l.-110)	100[a] 10-1030	110[a] (<d.l.-440)	640[a] (100-1610)	n.a.
Germany	Butte et al. (2001)	286	Residence	n.a.	n.a.	49 n.a.	49 n.a.	740 n.a.	n.a.
Germany	Becker et al. (2002)	199	Residence	n.a.	3.3 n.a.	42 n.a.	15 n.a.	416 n.a.	n.a.
Demark	Clausen et al. (2003)	15	School	n.a.	n.a.	n.a.	n.a.	3214[a] (400-8500)	n.a.
Germany	Kersten and Reich (2003)	65	Residence	n.a.	5 n.a.	47 n.a.	19 n.a.	600 n.a.	n.a.
France	Santillo et al. (2003)	31	Residence	<d.l. n.a.	6.87 (<d.l.-49.4)	55.3 (11.6-624)	28.2 (<d.l.-3551)	504.6 (14.9-3289)	n.a.
Germany	Santillo et al. (2003)	5	Residence	1.42 (<d.l.-2.83)	12.9 (1.86-368)	44.1 (22.3-1511)	82.2 (4.4-218)	996 (547-1586)	n.a.
Italy	Santillo et al. (2003)	5	Residence	<d.l. (<d.l.-1.5)	6.78 (1.92-23.6)	42.8 (22.8-46.8)	23.6 (9.0-308)	434 (314-933)	n.a.
Spain	Santillo et al. (2003)	22	Residence	<d.l. (<d.l.-0.92)	5.33 (1.09-64.6)	79.4 (48.6-201)	4.54 (0.81-153)	317.2 (113-2151)	n.a.
U.K.	Santillo et al. (2003)	29	Residence	<d.l. (<d.l.-1.1)	3.5 (0.6-114.8)	52.8 (0.1-106.4)	24.5 (<d.l.-238.9)	195.4 (0.5-416.4)	n.a.
Germany	Becker et al. (2004)	252	Residence	n.a.	n.a.	n.a.	n.a.	515 (22-5330)	n.a.
Germany	Fromme et al. (2004)	59	Residence	1.5 n.a.	6.1 n.a.	47 n.a.	29.7 n.a.	703.4 n.a.	n.a.
Sweden	Bornehag et al. (2005)	346	Residence	n.a.	<d.l. (<d.l.-2425)	150 (<d.l.-5446)	135 (<d.l.-45549)	770 (<d.l.-40459)	n.a.
Germany	Nagorka et al. (2005)	278	Residence	n.a.	n.a.	29 n.a.	13 n.a.	480 n.a.	n.a.

Table 1: continued.

Germany	Butte et al. (2008)	29	Residence	n.a.	n.a.	28 n.a.	51 n.a.	970 n.a.	n.a.
Bulgaria	Kolarik et al. (2008)	177	Residence	260[b] (<d.l.-4300)	350[b] (<d.l.-9070)	7860[b] (<d.l.-58070)	320[b] (<d.l.-2730)	960[b] (<d.l.-29440)	250[b] (<d.l.-2510)
Germany	Abb et al. (2009)	30	Residence	n.a.	n.a.	87.4 n.a.	15.2 n.a.	604 n.a.	n.a.
Denmark	Langer et al. (2010)	497	Residence	n.a.	1.7 (8.8)[c]	15 (7.2)[c]	3.7 (4.0)[c]	210 (2.4)[c]	n.a.
Denmark	Langer et al. (2010)	151	Day care	n.a.	2.2 (2.6)[c]	38 (4.6)[c]	17 (3.4)[c]	500 (1.9)[c]	n.a.
Sweden	Bergh et al. (2011)	10	Residence	0.04 (0.03-0.1)	3.7 (1.3-63)	130 (17-260)	17 (3.1-110)	680 (130-3200)	n.a.
Sweden	Bergh et al. (2011)	10	Office	0.2 (0.05-1.2)	20 (3.7-180)	100 (20-450)	8.8 (1.4-110)	1100 (57-3700)	n.a.
Sweden	Bergh et al. (2011)	10	Day care	0.1 (0.01-1.5)	4.2 (1.0-23)	150 (38-560)	31 (9.0-120)	1600 (260-5800)	n.a.
France	Blanchard et al. (2014)	30	Residence	0.2 (<d.l.-2.3)	2.8 (<d.l.-93.6)	11.9 (<d.l.-59.7)	8.5 (0.17-79.5)	289 (13.7-1520)	n.a.
Sweden	Luongo el al. (2016)	62	Residence	0.47 (<d.l.-2.3)	14 (<d.l.-323)	103 (10-5945)	16 (3.4-397)	449 (33-4843)	n.a.
Asia									
Japan	Kanazawa et al. (2010)	41	Residence	<d.l. (<d.l.-1.01)	0.35 (<d.l.-6.3)	22.3 (5.1-549)	2.4 (<d.l.-35.8)	1200 (220-10200)	n.a.
China	Guo and Kannan (2011)	75	Residence	0.2 (>d.l.-8.2)	0.4 (0.7-45.5)	20.1 (1.5-1160)	0.2 (<d.l.-12)	228 (9.9-8400)	0.2 (<d.l.-45.7)
Taiwan	Hsu et al. (2012)	101	Residence	0.1 n.a.	1 n.a.	20.2 n.a.	1 n.a.	753 n.a.	n.a.
Kuwait	Gevao et al. (2013)	21	Residence	0.03 (>d.l.-0.1)	1.8 (0.1-16)	45 (8.3-160)	8.6 (<d.l.-160)	2256 (380-7800)	14 (<d.l.-1300)
China	Zhang et al. (2013)	215	Residence	0.1 (<d.l.-24)	0.2 (<d.l.-33.9)	23.7 (<d.l.-2150)	1.6 (<d.l.-38.7)	183 (<d.l.-9950)	0.1 (<d.l.-39.5)
China	Wang el al. (2014)	28	Residence	<d.l. (<d.l.-68.84)	n.a.	134 (3.64-4357)	n.a.	581 (67.1-3475)	n.a.
Japan	Ait Bamai et al. (2014)	128	Residence	<d.l. (<d.l.-4.6)	<d.l. (<d.l.-58.7)	16.6 (<d.l.-1670)	2.0 (<d.l.-139)	1110 (213-7090)	n.a.
China	Bu et al. (2016)	30	Residence	3.4 (<d.l.-25.3)	7.6 (0.7-91.3)	165 (24.7-1087)	<d.l. (<d.l.-2.8)	1543 (279-7424)	n.a.
China	Bu et al. (2016)	30	Residence	1.9 (<d.l.-32.0)	6.4 (0.2-99.4)	139 (33.6-493)	21.2 (<d.l.-10.9)	1450 (121.8-7958)	n.a.
Kuwait	Albar et al. (2017)	15	Residence	0.1 (<d.l.-2.5)	2.7 (0.004-32.4)	1.6 (0.49-34.4)	0.8 (0.12-4.8)	240 (49.4-450)	2.8 (0.21-4.4)
Saudi Arabia	Albar et al. (2017)	15	Residence	0.6 (0.17-5.2)	1.4 (0.29-23.4)	33.3 (5.2-375)	0.8 (0.37-3.8)	1020 (71.5-2401)	26.8 (3.28-827)

a. Mean value
b. Geometric mean value
c. Geometric standard deviation

Table 2: Organophosphate concentrations (µg/g) in indoor dust as reported in relevant studies. All values are medians with range (minimum – maximum) unless stated otherwise.

Country	**Reference**	**N**	**Building type**	**TnBP**	**TCPP**	**TBEP**	**TPhP**	**TDCPP**
North America								
U.S.	Stapleton et al. (2009)	50	Residence	n.a.	0.572 [a] (<d.l.-5.49)	n.a.	7.36 [a] (<d.l.-1798)	1.89 [a] (<d.l.-56.1)
U.S.	Meeker and Stapleton (2010)	50	Residence	n.a.	n.a.	n.a.	7.4 [a] (<d.l.-1798)	1.88 [a] (<d.l.-56.1)
U.S.	Dodson et al. (2012)	16	Residence	0.032 (<d.l.-1.8)	2.1 (0.34-120)	12 (2.3-68)	<d.l. (<d.l.- <d.l.)	2.8 (0.73-24)
U.S.	Dodson et al. (2012)	16	Residence	<d.l. (<d.l.-1.8)	2.2 (0.49-140)	11 (0.79-170)	<d.l. (<d.l.-<d.l.)	2.1 (0.92-44)
U.S.	Stapleton et al. (2014)	30	Residence	n.a.	3.44 [a] (0.217-67.8)	n.a.	n.a.	2.73 [a] (0.62-13.1)
U.S.	Shin et al. (2014)	30	Residence	n.a.	n.a.	n.a.	2.0 (n.a.-11.7)	3.6 (n.a.-72.5)
Canada	Fan et al. (2014)	134	Residence	0.25 (<d.l.-7.1)	1.4 (<d.l.-56)	31.9 (3.6-143)	1.7 (0.26-63)	2.7 (0.12-77)
Canada	Fan et al. (2014)	134	Residence	0.26 (<d.l.-4.4)	1.1 (<d.l.-49)	22.8 (2.4-236)	1.6 (<d.l.-95)	2.0 (0.11-101)
U.S.	Schreder and Guardia (2015)	20	Residence	n.a.	4.82 (n.a.-82.7)	n.a.	n.a.	1.62 (n.a.)
U.S.	Hoffman et al. (2015)	49	Residence	n.a.	n.a.	n.a.	1.02 (0.099-40.3)	1.39 (0.197-39.5)
Europe								
Norway	Cequier et al. (2014)	48	Residence	0.055 (n.a.)	2.68 (n.a.)	13.4 (n.a.)	0.98 (n.a.)	0.50 (n.a.)
Norway	Cequier et al (2014)	6	School	0.043 (n.a.)	2.04 (n.a.)	87.2 (n.a.)	1.54 (n.a.)	1.49 (n.a.)
UK	Brommer and Harrad (2003)	32	Residence	<d.l. (<d.l.-0.09)	21 (3.7-100)	n.a.	3.3 (0.49-110)	0.71 (0.06-14)
Denmark	Langer et al. (2016)	497	Residence	<d.l. (<d.l.-4.0)	<d.l. (<d.l.-100)	<d.l. (<d.l.-1300)	<d.l. (<d.l.-91)	<d.l. (<d.l.-860)
Denmark	Langer et al. (2016)	151	Daycare	<d.l. (<d.l.-4.3)	5.6 (n.a.-350)	26 (<d.l.-11000)	2.0 (<d.l.-210)	7.1 (<d.l.-320)
Germany	Zhou et al. (2017)	15	Residence	0.25 (<d.l.-1.4)	4.3 (2.1-99)	4.2 (1.7-10)	1.2 (0.48-23)	<d.l. (<d.l.-4.3)
Germany	Zhou et al. (2017)	5	Daycare	0.42 (<d.l.-0.79)	60 (12-1000)	4.0 (3.4-16)	1.2 (0.34-2.5)	<d.l. (<d.l.-4.4)
Germany	Zhou et al. (2017)	11	Office	0.28 (<d.l.-1.2)	8.6 (4.8-150)	8.5 (1.8-470)	2.9 (0.77-9.2)	3.2 (0.80-39)
Germany	Zhou et al. (2017)	7	School	0.66 (0.24-1.7)	110 (10-2700)	5.7 (3.4-22_	1.5 (1.2-9.5)	7.4 (1.2-67)
Asia								
Japan	Tajima et al. (2014)	48	School	<d.l. (<d.l.-2.46)	0.74 (<d.l.-392)	30.9 (n.a.-936)	0.87 (<d.l.-23.4)	<d.l. (<d.l.-9745)
Japan	Tajima et al. (2014)	128	School	0.74 (<d.l.-60.6)	2.23 (n.a.-621)	26.5 (n.a.-1933)	3.13 (n.a.-27.5)	<d.l.- (<d.l.-73.1)
Philippines	Kim et al. (2013)	17	Residence	0.019 (<d.l.-0.079)	n.a.	n.a.	0.089 (0.008-2.1)	n.a.
Philippines	Kim et al. (2013)	20	Residence	0.02 (<d.l.-0.28)	n.a.	n.a.	0.071 (0.013-0.44)	n.a.

a. Geometric mean value

2.2 AD/ABSORPTION OF SVOCS ON OTHER INDOOR SURFACES

Because of their low vapor pressure, SVOCs emitted in the gas-phase tend to partition strongly to indoor surfaces. For porous material surfaces such as gypsum wallboard, carpet, clothing and furniture foam, SVOCs can diffuse into the material and sorb there (Petrick et al., 2010; Liang and Xu, 2015). Over time, such sorption processes may establish significant reservoirs that may act as secondary sources in indoor environments. Furthermore, SVOCs sorbed to interior surfaces may represent an important source of potential contact and exposure through dermal absorption and nondietary ingestion pathways (Little et al., 2012; Weschler and Nazaroff, 2012), particularly for young children who may frequently contact those surfaces and have enhanced hand-to-mouth activities. Therefore, assessing the levels of SVOCs on indoor surfaces is one of the top priorities to determine human exposure. However, due to substantial analytical challenge of SVOCs (Melymuk et al., 2014, 2015; Wang et al., 2015), and extensively long equilibrium time scale of their partitioning (Liu et al., 2016; Wu et al., 2017), few studies have investigated the partition mechanisms of SVOCs between the gas-phase and indoor surfaces.

Impervious Surfaces

Ad/absorption of SVOCs between the gas-phase and impervious surfaces generally assumes that the surfaces are impermeable materials and the transport of SVOCs solely considers the external mass-transfer between the gas-phase and surfaces. The internal mass-transfer, which is the diffusion of SVOCs inside the material, is considered negligible for impervious surfaces. Langmuir and Freundlich adsorption isotherms are commonly used to describe the partition between the gas-phase and the impervious surfaces. Particularly, since the concentrations of SVOCs are relatively low (Rudel et al., 2003, 2010a), a linear partition relationship based on a surface/air partition coefficient, K, was also frequently used in the literature (Guo, 2013). Liang and Xu, (2014) measured the partition coefficient of DEHP between the gas-phase and polished stainless-steel by pass a gas stream of air containing DEHP in a specifically designed chamber. Liu et al., (2014) measured the surface/air partition coefficients of polychlorinated biphenyl congeners (PCBs) for glass and stainless-steel by applying a degree of sorption saturation model. A more recent study argued that the adsorption of SVOCs to impervious surfaces is significantly affected by surfaces morphology, more specifically, the true surface area of an impermeable material (Wu et al., 2017). They measured the surface roughness of pre-cleaned aluminum, stainless-steel, glass, and acrylic and then obtained the surface/air partition coefficients of DEHP, which are the mass adsorbed per unit true surface area divided by the concentration in the air.

However, those measurements of surface/air partition coefficients were usually conducted in experimental chambers while the interior surfaces in real indoor environments might be more complicated. Several studies found that the impervious surfaces of indoor environments are mostly covered by a thin layer of greasy films (Liu et al., 2003; Butt et al., 2004). Those films typically have a thickness ranging from 5−20 nm and organic content from 15−30% (Liu et al., 2003), grow at a rate of 0.11−0.31 nm/day (Wu et al., 2008), and are conceptualized as a combination of accumulated organic constituents, sorbed water, inorganic molecules and ions, and deposited particles (Diamond et al., 2000). More recently, Wu et al. (2017) showed that although the surface/air partition coefficients of DEHP on pre-cleaned aluminum, stainless-steel, glass, and acrylic differed by several orders of magnitudes, the partition coefficients are similar when the surfaces are soiled. As described in Weschler and Nazaroff (2017), such findings in Wu et al. (2017) supported the idea that organic films are expected to produce a degree of commonality among surfaces in a given indoor environment. Although the chemical properties of indoor surfaces differ significantly, the films would have similar similar chemical-physical properties as the compounds constituting the films are usually derived from human occupants themselves and the SVOCs used in indoor materials. Those studies suggest that modeling the partition relationship between a variety of different surfaces and air can be simplified by modeling the absorption between the organic films and air.

Porous Surfaces

Ad/absorption of SVOCs on porous surfaces can be considered as a diffusive transport process. In the model, molecules can diffuse into the pore space of the materials and ad/absorbed there. Therefore, the material-phase diffusion coefficient and the surface/air partition coefficient are two key parameters to characterize the ad/absorption of SVOCs on porous surfaces. Liu et al., (2016) estimated the surface/air partition coefficients and material-phase diffusion coefficients between gas-phase organophosphorus flame retardants and materials such as carpet, gypsum wallboard, tile, clothing, flooring, and mattress pad using model described in (Deng et al., 2010). Another study developed a C_m-history model to estimate the gas-phase concentration in adjacent with the material, y_0, and cloth/air partition coefficients simultaneously. However, both studies require fitting two parameters from a single time-series data set. The inability to obtain unique parameter estimates from the data may result in uncertainties in the results.

Recently, several studies started investigating the sorption of SVOCs on clothing, considering that dermal uptake of SVOCs might be a significant exposure pathway (Weschler and Nazaroff, 2012). Morrison et al. (2015) calculated the partition coefficients of diethyl phthalate (DEP) and DnBP between cotton clothing and air through chamber measurements. They further measured the partitioning of PCBs from air to clothing and found that clothing may act as a reservoir for PCBs that extends exposure.

2.3 ASSOCIATION WITH CHILDHOOD ASTHMA

Several studies in literature examined the associations between phthalates in dust or PVC materials in homes and allergic diseases in children. In most of the studies, non-floor settled dust were sampled. Among Swedish children, Bornehag et al. (2004) showed an association between higher concentrations of DEHP in house dust with presence of asthma. This association was also observed with house dust concentrations of BBzP and the presence of allergic rhinitis and atopic dermatitis. Similar results were found in Bulgaria and Taiwan, where positive associations between house dust DEHP or BBzP concentrations and childhood asthma (Kolarik et al., 2008a) and rhinitis and eczema (Hsu et al., 2012a), were reported. However, the results were not always consistent. Two studies conducted in Denmark and Japan (Kanazawa et al., 2010; Callesen et al., 2014a) did not find a significant association between phthalates in dust and allergies in children. In another cross-sectional study (Ait Bamai et al., 2014a), levels of several phthalates in floor dust had linear associations with the prevalence of allergic rhinitis, conjunctivitis, and atopic dermatitis in children. But the associations were not found for non-floor dust. A recent study in Denmark (Beko et al., 2015) involving children with asthma, allergic rhinitis, or eczema did not show a direct association between the presence of allergic disease and exposure to phthalates. However, among kids with allergic disease, there was association between indoor allergen sensitization and exposure to BBzP, DBP, and DEHP. In another controlled study (Deutschie et al., 2008), 16 healthy and 16 house-dust-mite allergic human subjects were exposed to house dust containing low and high concentrations of DEHP. Healthy subjects were not affected by the dust exposure, but the allergic group showed a silent inflammation in nasal mucosa in response to the dust containing low DEHP doses.

Phthalate metabolites in urine were also used to examine their relations with allergic diseases, but conflicting data were produced. Positive associations were found between urinary metabolites of BBzP, DEHP, diisononyl phthalate (DINP), or diisodecyl phthalate (DIDP) and children's asthma in Belgium (Franken et al., 2017), Norway (Bertelsen et al., 2013), and Taiwan (Hsu et al., 2012a). But, no associations were reported between urinary concentrations of any phthalate metabolites and either asthma or rhinitis in two studies of Danish children (Callesen et al., 2014b). A recent study among U.S. participants found that BBzP metabolite was associated with allergic sensitization in adults, but this was not observed in children (Hoppin et al., 2013). Callesen et al., (2014b) argued that urinary metabolite concentrations, which represent the total intake of phthalate ester, could fail to reveal an association between a given exposure pathway (e.g., inhalation or dermal) and an allergic disease, if another exposure pathway (e.g., diet) overwhelmingly dominates total exposure. Additionally, the presence of asthma and asthma severity may also be influenced by exposures to microbes in indoor environments. Studies assessing increased exposure to indoor fungi before the development of asthma symptoms suggested that certain fungal species pose a respiratory health risk in susceptible populations (Sharpe et al., 2015). Several other studies found that exposure to bacterial endotoxin and mold also affected asthma severity in children and additional respiratory outcomes in subjects with and without allergy (Michel et al., 1996; Zambelli-Weiner et al., 2004; Mendell et al., 2011; Dannemiller et al., 2016).

The mechanism responsible for the association between phthalate exposure and doctor-diagnosed asthma remains unclear. Some in vitro and in vivo animal studies showed that phthalates or their metabolites may have different modes of action (e.g., direct pharmacological effect on receptors involved in allergenic pathology or indirect effect as adjuvants for different causative agents) to influence immune and inflammatory parameters that play a role in respiratory diseases (Oie et al., 1997; Larsen et al., 2002, 2003; Larsen and Nielsen, 2008; Bornehag and Nanberg, 2010; Kimber and Dearman, 2010; Beko et al., 2015; Franken et al., 2017). Additionally, discovery by recent in vivo studies that viable fungi can induce asthma-like disease in experimental animals supports the argument that fungi are possible etiologic agents of asthma (Porter et al., 2011a, 2011b).

Compared with phthalates, organophosphates were paid significantly limited attentions on the associations with respiratory diseases. Canbaz et al., (2017) demonstrated that phosphorus flame retardants including TDCPP and TPHP can be immunotoxic for dentritic cells and indicated that those phosphorus flame retardants can impact the immunological health in the respiratory tract. A cross-sectional study in Japan reported increased odds ratios for increased concentrations of TNBP and TDCPP in house dust with allergic diseases such as asthma, rhinitis (Araki et al., 2014). In contrast, another case-control study conducted in Stockholm did not observe any association between flame retardants, including seven organophosphates in mattress dust and the development of childhood asthma (Canbaz et al., 2015). The controversial results make it hard to draw any conclusions for the relation between flame retardants and childhood asthma.

Chapter 3: Summary of Methods and Research Findings

This chapter describes the investigations conducted in support of the three research objectives outlined in Chapter 1. Full manuscripts describing each investigation appear in Appendices A-C with complete introduction, methods, results, discussion, and supporting information.

3.1 INVESTIGATE THE EMISSION, SORPTION, AND FATE OF PHTHALATES IN A RESIDENTIAL TEST HOUSE (OBJECTIVE 1)[1]

3.1.1 Field Measurements

As shown in Figure 1, field measurements were conducted in the test house to investigate concentrations of phthalates in air and on different indoor surfaces with the change of room temperature. The test house, which is a three-bedroom, two-bathroom 110 m^2 manufactured home located at the University of Texas at Austin, has two independent heating ventilation, and air conditioning systems and is fully instrumented with air exchange measurement system, weather stations, and several particle, temperature, relative humidity (RH), and air velocity measurement stations. The temperature of the house was strictly controlled at 21±0.8 ºC for about six months and then increased to 30±0.5 ºC. This temperature range was selected to represent typical temperature fluctuations of buildings. After three months, the temperature was dropped back to 21 ºC to allow comparison with previous results at the same temperature and to examine the reproducibility of the data. Finally, the temperature was increased to 25±1.0 ºC to further verify the sensitivity of phthalate levels to temperature. At each experimental stage, total airborne concentrations of phthalates were monitored periodically. Once steady state conditions at 21 and 30 ºC were achieved, concentrations of sorbed phthalates were measured on different surfaces, including dust, windows, stoneware dish plates, mirrors, fabric

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[1] Bi, C., Liang, Y., & Xu, Y. (2015). Fate and Transport of Phthalates in Indoor Environments and the Influence of Temperature: A Case Study in a Test House. *Environmental Science & Technology, 49*(16), 9674–9681. The first author did all the experimental work and consequent data analysis with some suggestions from the second and third authors.

cloth, and wood. In addition, at 21 ºC, the kinetic aspect of phthalate sorption was investigated at selected, pre-cleaned surface locations by measuring the accumulation of phthalates on these surfaces. Finally, temperature, RH, air change rate, and total suspended particle (TSP) concentrations were monitored periodically for the entire study period. Test house conditions are listed in Table 3.

Air Sampling Surface Sampling Dust Sampling Interior View

Wood Cloth Dish Plate Mirror

Figure 1. Photos of sampling method and materials.

Table 3. Test conditions and model parameters.

Parameter	21°C	25°C	30°C
Relative humidity (%)	30.4±13.8	40.4±4.0	24.9±5.9
Total suspended particle concentration (μg/m^3)	4.0±3.8		
Air exchange rate (hr^{-1})	0.50±0.06	-	0.71±0.10
Area of flooring material (m^2)	77		
Area of wall, ceiling, and furniture (m^2)	220		
Area of windows (m^2)	7		
Volume of the test house (m^3)	250		
Surface/gas partition coefficient for BBzP (K_{surf}, m)	wall, ceiling, and furniture: 100 [a] windows: 108 [b]		
Mass transfer coefficient (h_m, m/hr)	0.106 [c]		
y_0 for BBzP in the flooring material [d] (μg/m^3)	2.7	5.9	12.4
Particle/gas partition coefficient for BBzP [e] (K_p, m^3/mg)	52.7	32.5	18.1
Particle/gas partition coefficient for DEHP [e] (K_p, m^3/mg)	350	207	110

a. Directly read from Figure S6 in Ref. (Xu and Hubal, 2009). We ignored the effect of temperature, because this parameter does not have a significant influence on model results.

b. Measured in the current study. We ignored the effect of temperature, because this parameter does not have a significant influence on model results.

c. Calculated using empirical correlations in Ref. (Axley, 1991), which are based on Reynolds and Schmidt numbers. We measured near-surface air velocity in the test house (0.01 m/s) and used this value to obtain the Reynolds number, and finally, the mass transfer coefficient. Similar low velocity was also reported in other field studies (Huang et al., 2004).

d. Measured in the current study using a specially designed chamber.

e. Calculated using Equation 15 in Ref. (Naumova et al., 2003) with the vapour pressure estimated using Equation 9 in Ref. (Gobble et al., 2014).

3.1.2 Indoor Air

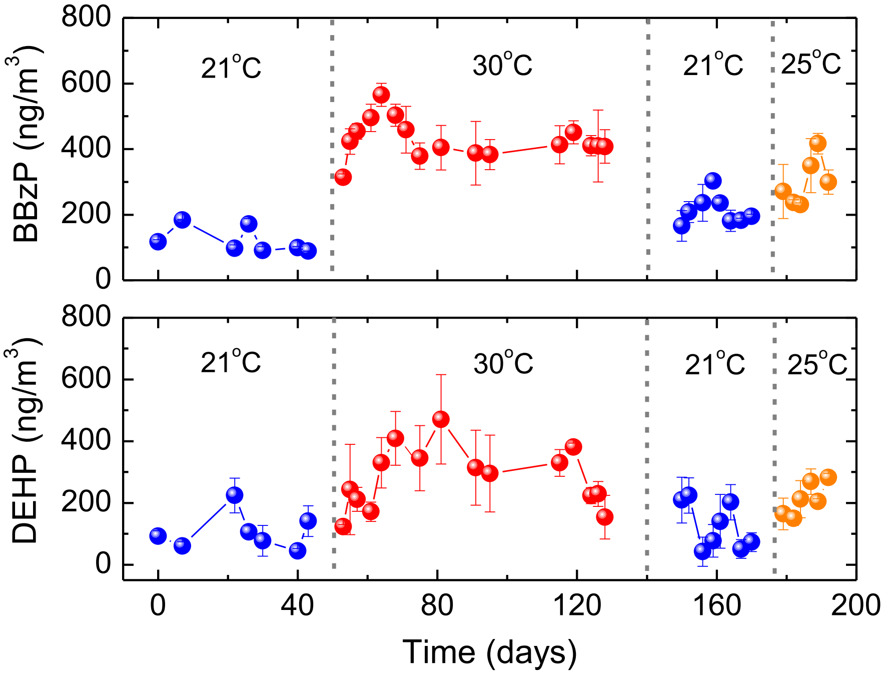


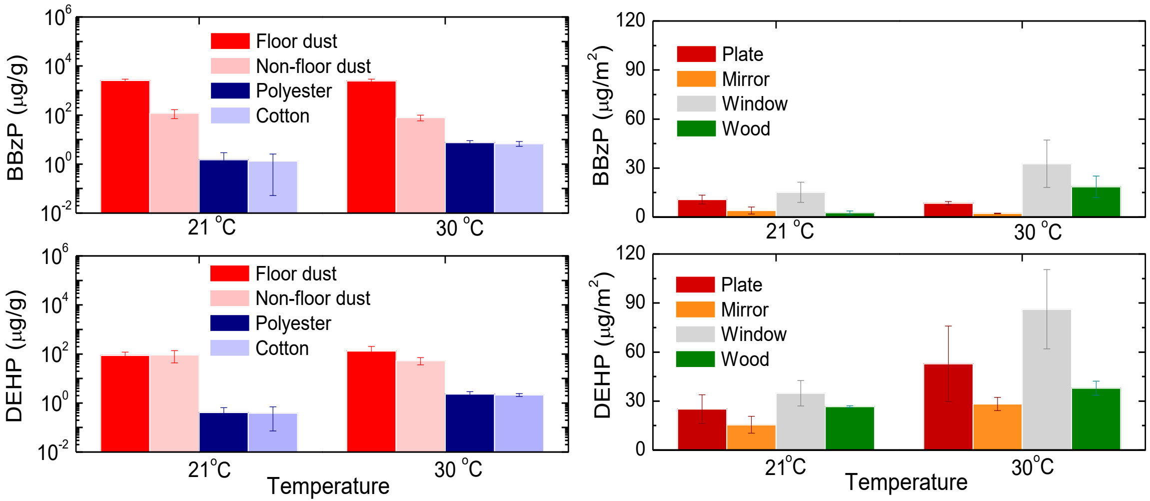
Figure 2. Total airborne concentration of BBzP and DEHP in the test house.

Phthalate levels in indoor air is highly sensitive to temperature. As shown in Figure 2, the mean steady-state concentrations at 21 °C were 149 and 110 ng/m^3 for BBzP and DEHP, respectively. When the temperature was increased to 30 °C, about four weeks were required for the indoor air concentration of phthalates to reach steady state. The steady-state concentrations at 30 °C were 419 and 300 ng/m^3 for BBzP and DEHP, respectively, which were about three times higher than the steady-state levels at 21 °C. The measured indoor air concentrations of phthalates in the test house are comparable to those measured in previous field studies in residential homes (Wilson et al., 2001; Fromme et al., 2004; Kanazawa et al., 2010; Rudel et al., 2010a; Bergh et al., 2011b; Xu et al., 2014a), which were in a range of 20–50 ng/m^3 for BBzP and 120–600 ng/m^3 for DEHP (although most of the studies did not provide temperature information at sampling sites). The measured BBzP concentrations in the test house were higher than levels reported in these studies, possibly because the test house was completely covered with vinyl flooring, which is a significant source of BBzP. After the temperature was dropped back to 21 ºC, the concentrations of phthalates reduced significantly. The results generally verified the reproducibility of the data, although the levels at 21 ºC were slightly higher than previous data. This is possibly due to the "baking effect" (i.e., enhanced emission and desorption) at 30 ºC for about three months, and thus, additional time may be needed for the equilibrium of surface partitioning to be re-established. Finally, when the temperature was increased to 25 ºC, the concentrations of BBzP and DEHP both increased instantly and to levels between the steady-state concentrations at 21 and 30 ºC. Considering that climate change may result in increased indoor temperatures as outdoor temperatures increase and heat waves become more frequent, indoor air concentrations of phthalates and human inhalation exposure to the contaminants could significantly increase as global warming becomes more serious.

3.1.3 Interior Surfaces

Dust

Figure 3a shows the concentration of phthalates in non-floor dust (i.e., dust collected from molding, ceilings, and walls) and floor dust (i.e., dust accumulated on vinyl flooring) at 21 and 30 ºC. At each temperature, the concentration of BBzP in floor dust was about 20–30 times higher than in non-floor dust, while little difference was observed for DEHP. The results indicated that dust captured a great amount of BBzP from vinyl flooring through direct contact with this BBzP source. The results also suggested that the sampling location of settled dust should be carefully selected in future indoor field studies, and building occupants may be exposed to a significantly high level of phthalates via ingestion of dust that deposited on source surfaces. At 21 ºC, the measured mean (± s.d.) concentrations in floor dust were 2560±360 and 88±30 µg/g for BBzP and DEHP, respectively, and in non-floor dust were 119±48 and 91±47 µg/g for BBzP and DEHP, respectively. Temperature did not significantly influence phthalate concentrations. One possible reason might be the offsetting effect of increased gas-phase concentration and decreased dust/gas sorption partitioning with increasing temperature. Generally, the results are comparable to previous field studies (Oie et al., 1997; Butte et al., 2001; Wilson et al., 2001; Rudel et al., 2003; Santillo et al., 2003; Bornehag et al., 2005; Langer et al., 2010; Bergh et al., 2011a), where mean concentrations in settled dust were reported in a range of 5 to 135 µg/g for BBzP and 200 to 1600 µg/g for DEHP. The slightly lower concentrations of DEHP in the current study might be due to the limited sources of DEHP presented in the test house, where not as many consumer products were used as in real residential homes.



(a) (b)

Figure 3. BBzP and DEHP concentrations in (a) dust and fabric cloth and on (b) plate, mirror, window, and wood.

We assumed that sorption equilibrium had been reached at 21 ºC, considering that the settled dust had been accumulating for six years in the test house, which was conditioned for half a year at 21 ºC before measurements started. The equilibrium coefficients (K_{dust}, m^3/g) that describe the partitioning between non-floor dust and gas phase were determined to be: 962 and 1970 m^3/g for BBzP and DEHP, respectively (Table 4). Weschler and Nazaroff (2010) proposed a method to estimate K_{dust} using the octanol-air partition coefficient of a chemical (K_{oa}). Because there are great deviations for the K_{oa} values of BBzP and DEHP reported in the literature (Cousins and Mackay, 2000; Xiao and Wania, 2003; Weschler and Nazaroff, 2008), we estimated the possible values of K_{dust} using the available ranges of K_{oa} and compared these values with our measurement results. The measured K_{dust} of BBzP (962 m^3/g) falls into the estimated range of $10^{2.2}$–$10^{4.6}$ m^3/g, while the K_{dust} of DEHP was an order of magnitude lower than the estimated range of 10^4–10^6 m^3/g. One possible reason might be the limited sources of DEHP presented in the test house, and another reason might be the significantly low organic content in the dust (e.g., skin flakes and organic particles generated during cooking) that resulted in weak dust/gas partitioning, considering that the test house was not occupied. In addition, the values of K_{dust} decreased greatly with increasing temperature (Table 4). However, caution should be taken when comparing results because it was possible that the dust/gas partitioning had not reached steady state within the three months at 30 ºC, considering the slow rate for SVOCs to approach equilibrium partitioning (Weschler and Nazaroff, 2010).

Interestingly, for floor dust, we found that a simple linear partitioning relationship was sufficient to explain its high BBzP level. Multiplying the partition coefficient (K_{dust}, Table 4) with the gas-phase concentration of BBzP in equilibrium with vinyl flooring (y_0, Table 3) (e.g., 962 m^3/g ×2.7 $\mu g/m^3$=2597 $\mu g/g$ at 21 ºC), the difference between the calculated and measured BBzP concentrations in floor dust (e.g., 2560 $\mu g/g$ at 21 ºC) was less than 10% at each temperature. The results suggested that the high BBzP concentration in floor dust are possibly due to the partitioning between dust and the gas layer adjacent to the flooring rather than direct solid-to-solid mass transfer. Although further verification is needed, this finding allows us to estimate phthalate concentrations in settled dust accumulated above source surfaces by only rapidly measuring y_0 for the source (Liang and Xu, 2014b).

Table 4. Measured indoor air concentration and surface/gas partition coefficients

		BBzP		DEHP	
Indoor air		21 ºC	30 ºC	21 ºC	30 ºC
Steady-state airborne concentration (ng/m^3)		149	419	110	300
Calculated gas-phase concentration [a] (ng/m^3)		123	391	46.0	209
Surfaces					
Mirror	K_{surf} (m)	33.5	5.78	288	135
	Log K_{film} [b]	10.2	9.46	11.2	10.8
Plate	K_{surf} (m)	86.9	21.8	582	253
	Log K_{film} [b]	10.6	10.0	11.5	11.1
Window	K_{surf} (m)	123	83.6	758	413
	Log K_{film} [b]	10.8	10.6	11.6	11.3
Dust	K_{dust} [c] (m^3/g)	962	202	1970	258

a. The measured concentrations in indoor air are actually the total airborne concentrations; that is, the sum of the gas-phase (C_g) and particle-phase (F) concentrations, C_g+F. Based on the definition of the particle/gas partition coefficient [K_p= (F/TSP)/C_g], the gas-phase concentration can be calculated using the equation C_g= (C_g+F)/(1+K_pTSP). Because C_g+F and TSP were measured and the values of K_p were estimated in Table 3, we calculated the gas-phase phthalate concentration (C_g) and used it to further obtain the surface/gas partition coefficients (K_{surf}).

b. Assuming the thickness of organic film is 10 nm and the film contains 20% organics, we calculated the dimensionless film/gas partition coefficients using equation 3.12 in Ref. (Weschler and Nazaroff, 2008).

c. Non-floor settled dust. Caution should be taken when comparing results, because the dust/gas partitioning possibly has not reached steady state at 30 ºC.

Plate, mirror, window, and wood

Because the time scale to achieve equilibrium sorption on impervious surfaces was fast (i.e., hours and days; see next section), we assumed that sorption equilibrium had been reached when measuring surface concentrations. Figure 3b shows the equilibrium concentrations of phthalates on impervious surfaces, including plate, mirror, and window. Generally, the concentration of DEHP on these surfaces was higher than that of BBzP because DEHP's lower vapor pressure resulted in stronger surface sorption (Xu and Hubal, 2009). We calculated the equilibrium surface/gas partition coefficient (K_{surf}) at different temperatures in Table 4. As expected, K_{surf} decreased by about a factor of five for BBzP and a factor of two for DEHP when temperature increased from 21 to 30 ºC. The K_{surf} values for windows did not reduce as much as other surfaces because its surface temperature was actually lower than 30 ºC due to the low outdoor ambient temperature (about 0–20 ºC). In addition, the effect of temperature on the surface concentration of phthalates was not consistent (Figure 3b). For example, with an increasing temperature, BBzP concentration on the plate decreased, while DEHP concentration increased. The change of surface concentration with temperature mainly depended on competition between the extent of the K_{surf} decrease and the increase in gas-phase concentration. Finally, the surface concentration on wood increased with increasing temperature. However, because phthalates may diffuse into the porous material, the method used (surface wipe) may not be sufficient to fully characterize wood's sorption property.

Organic films have been found to develop rapidly on impervious indoor surfaces through partitioning of gas-phase compounds from air to the film and enhanced dry deposition of airborne particles (Liu et al., 2003; Butt et al., 2004). Such films typically have a thickness ranging from 5–20 nm and organic content from 15%–30% (Liu et al., 2003), grow at a rate of 0.11–0.31 nm/day (Wu et al., 2008), and are conceptualized as a combination of accumulated organic constituents, sorbed water, inorganic molecules and ions, and deposited particles (Diamond et al., 2000; Weschler and Nazaroff, 2008). In the current study, the windows of the test house had not been cleaned since it was built, and we observed a "greasy" coating on the surface. This may explain the higher concentration of phthalates on window glass than on plate and mirror (Figure 3b). Assuming such films exist on all impervious surfaces measured in this study with a thickness of 10 nm and organic content of 20%, we converted the measured equilibrium surface/gas partition coefficients (K_{surf}) to dimensionless film-to-gas partitioning ratios (K_{film}) (Table 4). Interestingly, the values of Log K_{film} are comparable to those of Log K_{oa} reported in the literature (Cousins and Mackay, 2000; Xiao and Wania, 2003), which further verified the SVOC equilibrium model between air and indoor surfaces (Weschler and Nazaroff, 2008). The results also indicated that the gas-phase phthalate concentrations in indoor air may be used to estimate their composition on impervious indoor surfaces and vice versa.

Fabric cloth

With temperature increasing from 21 to 30 °C, the concentrations of phthalates in both cotton and polyester increased in approximately three times (Figure 3a), mainly due to the increase in their gas-phase concentration in indoor air. When chemical concentrations were converted from µg/g of fabric cloth to µg/m^2 of surface area with the known cloth density (about 0.005 m^2/g of fabric cloth), the calculated concentrations, compared with the levels on plate, mirror and window surfaces (Figure 3b), were orders of magnitude higher. This result indicated that phthalates preferably sorbed to the fabric material and less so to the smooth impervious surfaces, which is similar to other SVOCs such as nicotine (Petrick et al., 2010). However, we did not observe any phthalate sorption differences between cotton and polyester, possibly due to the relatively low gas-phase concentration of phthalates in the test house compared with other chamber studies of SVOCs sorption that typically involved high gas-phase concentrations (Piade et al., 1999; Petrick et al., 2010). In addition, we observed higher BBzP concentrations than those of DEHP at both temperatures and in both fabric cloths. We believed this might be related to the kinetic aspect of phthalate partitioning to fabric cloth. The time scale required for sorption equilibrium to be reached in cloth could be significantly longer for DEHP than for BBzP due to its larger K_{oa} value (Weschler and Nazaroff, 2010). Other possible reasons might be associated with chemical polarity (Buchert et al., 2001; Cao, 2010; Laing and Kean, 2011), RH variations in the test house (Piade et al., 1999), and fabric structure (Chien et al., 2011).

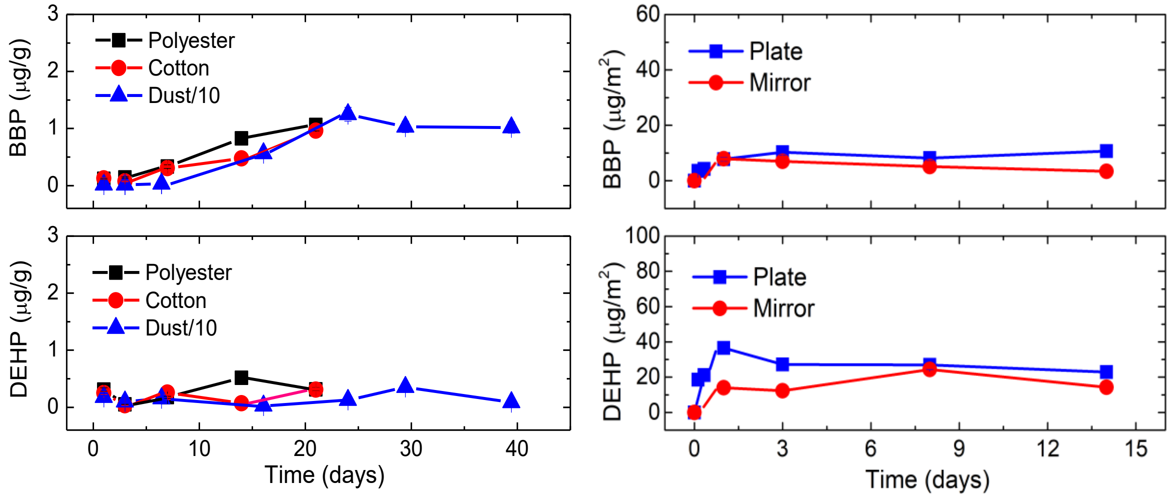
3.1.4 Sorption Kinetics

When indoor air concentrations of phthalates reach steady state, phthalate levels on interior surfaces will evolve over time and finally achieve sorption equilibrium. The preceding measurements of surface concentration were based on the assumption of equilibrium conditions.

In this section, we experimentally assessed the time scale required for sorption equilibrium to be reached between interior surfaces and gas-phase phthalates at 21 ºC.

Figure 4a shows that equilibration time scale can be fast (i.e., less than 24 hours) for plate and mirror surfaces. The steady-state concentrations of BBzP and DEHP (Figure 4) were similar to those in the measurements of surface concentrations (Figure 4b), indicating that the assumption that sorption equilibrium had been reached was reasonable. The variation of surface concentration at steady state might be related to the fluctuation of gas-phase concentration in the test house. Weschler and Nazaroff (2008) proposed a method to estimate the equilibrium time for SVOC sorption reservoirs modeled as organic thin films. Based on their method and the K_{oa} values of phthalates (Cousins and Mackay, 2000), the estimated time scales (e.g., ~ 12 hours for BBzP and ~ 20 days for DEHP assuming a film thickness of 10 nm, and ~ 1 hour for BBzP and ~ 2 days for DEHP assuming a film thickness of 1 nm) were comparable to our observations. Considering clean dishware and cups are typically air dried, the fast sorption from air to those surfaces may result in meaningful daily exposure to phthalates when eating and drinking, especially for compounds with high K_{surf}.

In contrast to the impervious surfaces, Figure 4b shows that about 20 days were required for BBzP to reach its equilibrium concentration in fabric cloth, while for DEHP the equilibration time can be even longer. However, given sufficient time, cloths, furnishings, and textile toys exposed to indoor air could accumulate a great amount of phthalates (Figure 4a), resulting in dermal exposure when touching the surfaces or wearing clothes. Previous research has shown that skin oil could even significantly enhance the sorption (Morrison et al., 2014). For children, phthalate sorption may pose a serious ingestion risk because children frequently put objects in their mouths. Dust concentration of phthalates (Figure 4b) was lower than those in pervious measurements (Figure 3a) because the dust used for the kinetic experiments was pre-cleaned and may have lost some of its organic components, thereby influencing its sorption properties.



(a) (b)

Figure 4. (a) Sorption kinetics for cloth and dust. (b) Sorption kinetics for plates and mirrors.

2.1.5 Indoor Fate Model of Phthalates

Because the flooring material was the major source of BBzP in the test house, we measured its emissions using a specially designed experimental chamber (Liang and Xu, 2014b). The obtained gas-phase concentration of BBzP in equilibrium with the material phase (y_0) was

2.7, 5.9, and 12.4 μg/m^3 at 21, 25, and 30 ºC, respectively (Figure 5). The values of y_0 allowed us to use a fate and transport model (Xu and Hubal, 2009; Xu et al., 2010) to interpret the field measurement results of BBzP. Other model parameters used are summarized in Table 3. Figure 5 shows good agreement between model predictions and observed air concentrations of BBzP in the test house at different temperatures. The difference between predictions and measurements was possibly due to the change in infiltration rate in the test house. During the second experimental stage when the indoor temperature was maintained at 30°C, we observed significant daily fluctuations in ambient air temperature (0–20 °C), which may have caused great variations in the amount of outdoor air penetrating into the house, although we were unable to monitor the infiltration rate in real time. However, the airborne concentration of BBzP was very sensitive to the infiltration rate, as shown in Figure 5 (the two dashed lines indicate an infiltration rate of 0.8 and 0.6 /h, respectively). Another possible reason might be the ignorance of some indoor sink reservoirs. For example, there are various interior surfaces that can serve as sorption surfaces, but only major surfaces (i.e., walls, ceilings, furniture, and windows) were accounted in the model. Considering that the K_{oa} value of BBzP is relatively large compared with other large molecular weight phthalates such as DEHP, ignorance of some sorption surfaces may not have great influences on model predictions. Although there were other limitations on use of the model (e.g., only the major source was considered), the results suggested that data obtained from chamber studies on source emissions can be directly applied to the model and provide a reasonably well interpretation of the fate and transport of phthalates in actual indoor environments. With knowledge of their indoor air and surface concentrations, this approach can help health professionals estimate screening-level indoor exposures to phthalates as well as other similar SVOC compounds of concern and develop effective strategies to limit the exposures.

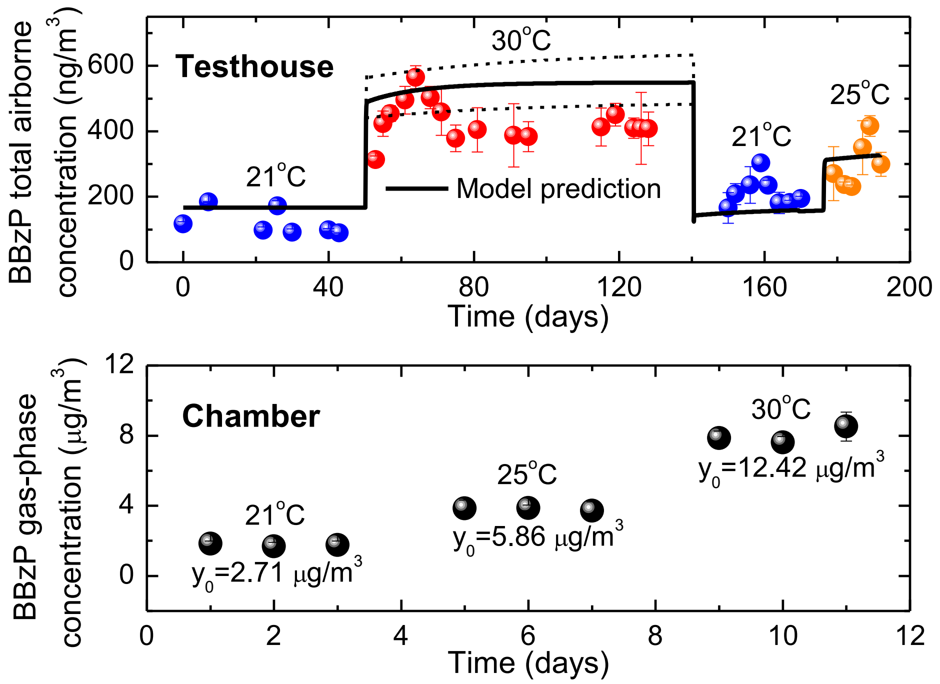


Figure 5. Total airborne concentrations of BBzP in the test house and gas-phase chamber concentrations of BBzP emitted from the vinyl flooring sample used in the test house.

3.2 Transfer of Phthalates and Their Alternatives from vinyl floorings and crib mattress covers to Settled house Dust (Objective 2)

3.2.1 Tested Materials

As shown in Table 5, three PVC floorings and two crib mattress covers were used in this study. The test materials were pre-cleaned with wipes dipped with methanol before the experiments. The plasticizers in the tested materials were DnBP, BBzP, DEHP, DEHA, and DINCH, collectively, with mass fractions from 4% to 23%. The y_0, which is the gas-phase concentration of SVOCs immediately adjacent to the material phase, were directly measured using the method developed by Liang and Xu (2014) for these materials.

Dust was collected from a residential house with a vacuum cleaner. The dust was then sieved with a 150 μm laboratory sieve and stored in an amber glass container with PTFE lid at -18 °C. Background concentrations of target compounds in the dust were measured at 302 ± 23.4

μg/g and 257 ± 17 μg/g for DnBP and DEHP, respectively. The concentrations of BBzP, DEHA, and DINCH in dust were below detection limit. The mass fractions of organics in the dust were determined by the thermal/optical carbon analyzers (Atmoslytic Inc., Calabasas, CA) using reflectance and transmittance analysis and pyrolysis correction, and the value is 32.6 ± 2.80 %.

Table 5. Content, y_0 values, and dust loadings for tested source materials

ID	Content	Mass fraction (m/m)	y_0 (μg/m^3)	Dust loading (g/m^2)	
				Low	High
1 (vinyl flooring)	DEHP	23±3%	2.37	0.512±0.054	15.7±1.02
2 (vinyl flooring)	BBzP	15±2%	8.47	2.46±0.234	16.2±0.845
3 (vinyl flooring)	DnBP	4±1%	24.7	2.97±0.286	19.4±1.24
	DEHP	7±1%	1.54		
4 (mattress cover)	DEHA	4±0.3%	1.05	2.35±0.147	N.A.
5 (mattress cover)	DINCH	11±1%	0.70	2.88±0.165	N.A.

3.2.2 Experimental Setup

As shown in Figure 6, the 10 L stainless steel test canisters (model No. CTH-24, Eagle Stainless Container Inc.) were customized with an inlet and an outlet on the cap using two 0.25-

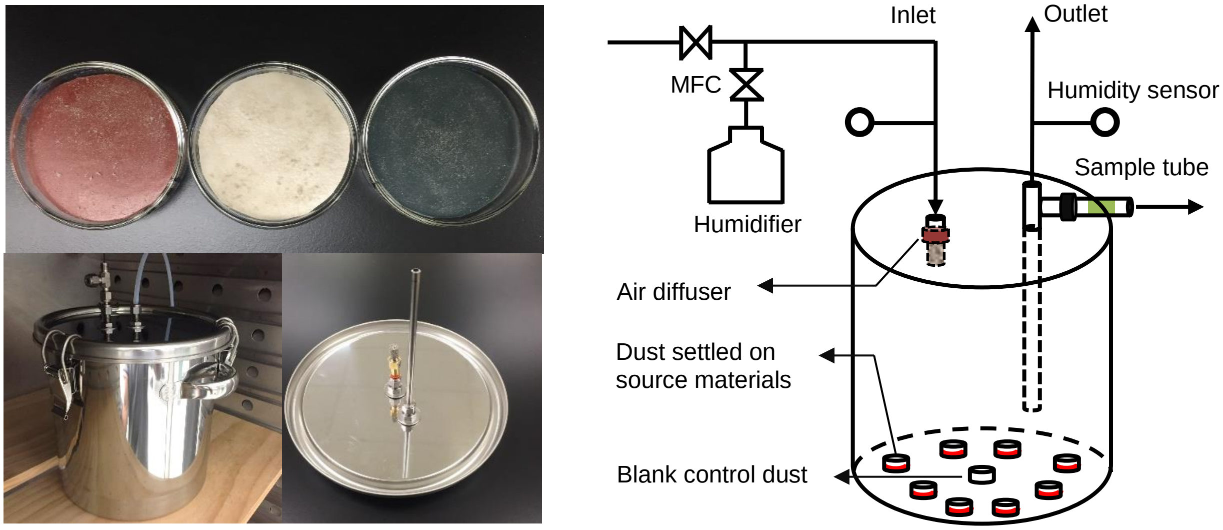


Figure 6. Configuration of test canister.

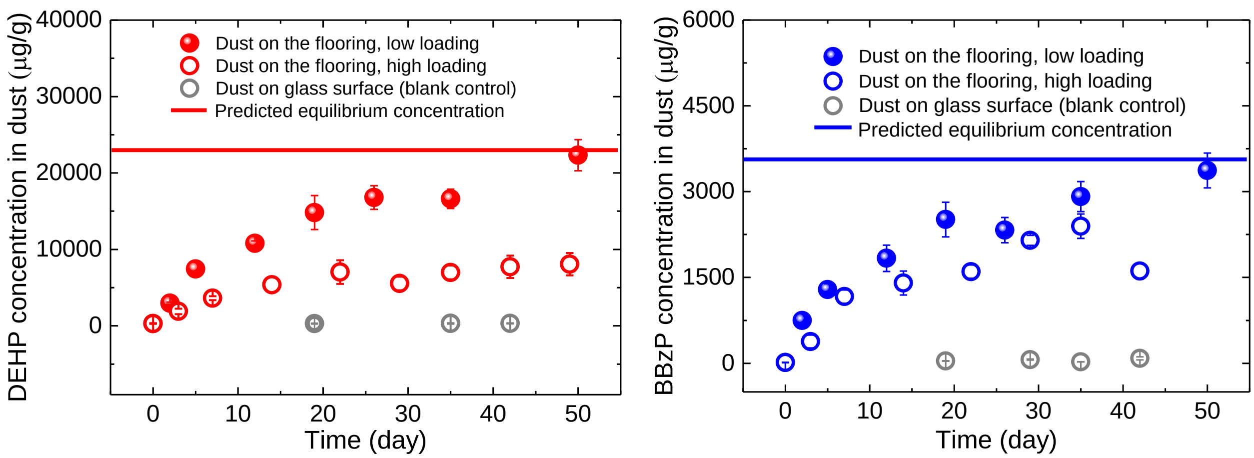
inch bulkhead unions (SS-400-61, Swagelok). The air inlet was connected to a brass gas diffuser inside the canister to avoid a direct blow of gas streams onto the samples. The air outlet was connected to a stainless-steel tube which extends into the chamber to enhance the mixing of air inside the chamber. The air leakage rate was less than 2% of the total flow rate.

Source materials were cut into 60 mm diameter pieces and were placed into 60 mm diameter glass petri dish. Dust was evenly sprayed onto the flooring. Eight dishes containing the same material with dust were then placed at the bottom of a 10 L stainless-steel canister with one inlet and one outlet for airflow. The stainless-steel canister was operated at an air exchange rate of 0.5 hr^{-1}, ~0% relative humidity (RH), and 25 ± 0.5 °C. In addition, a blank control sample, which is a petri dish containing dust but without source materials, was also placed in each canister. During sampling, one of the eight petri dish samples was taken out of the canister for further extraction and analysis of the dust. In this way, dust samples were periodically collected from the chamber to monitor the increase of phthalate and their alternative concentration in dust until steady state was reached. Concentrations of the target compounds in blank control samples were also analyzed by taking a portion of dust from the blank control petri dish at each sampling time. In addition, , to measure the gas-phase concentration of target compounds in the chamber, air samples were collected from the canister using Tenax TA tubes with a pump (SKC 224-PCXR4) calibrated to a nominal flow rate of 50 mL/min for approximately 2 hours. Background concentrations of target compounds in the canister were examined for two weeks prior to the experiments and no contamination was found.

To study the influence of dust loading on the sorption kinetics, Sample 1, 2, and 3, were tested with low and high dust loadings, which were 0.512 - 2.97 and 15.7 - 19.4 g/m^2, respectively (Table 5). Sample 3, which contains DnBP and DEHP, was used to investigate the influence of humidity on the direct transfer of phthalates to dust. The experiment was conducted in three canisters with identical procedure described above except different relative humidity in the supplied air. The three canisters were supplied with air with relative humidity (RH) at 0%, 50% and 80%, respectively. Humidified air was prepared by supply air flow through a bubble humidifier. The relative humidity was monitored with a HOBO data logger (Model U12-012, HOBOware Pro, Onset Computer Co.) at the inlet and outlet of each canister. The experiments started after the relative humidity at the inlet had less than 3% difference with the one at the outlet of the canister. The differences of average relative humidity between the inlet and outlet of the canisters were less than 7% during the experimental period.

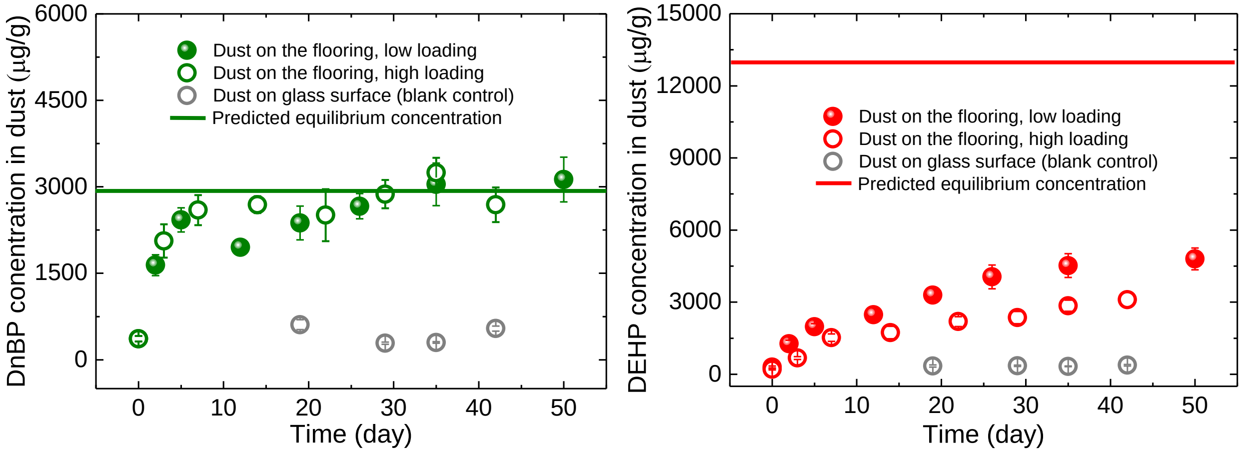
3.2.3 SVOC Levels in Dust

Figure 7 showed the accumulation of phthalates and their alternatives in dust settled on three floorings and two crib mattress covers. The results suggested the concentration of phthalates and their alternatives in dust settled on the source materials increased significantly within the experimental period. In the case of low dust mass loading, it took shorter time for the low-molecular-weight compounds (e.g., DnBP and BBzP) to reach steady state (possibly sorption equilibrium) than the high-molecular-weight ones (e.g., DEHP and DINCH). Figure 7a) shows that the steady-state concentration of DEHP in dust settled on surface of Sample 1 reached 29890 ± 2679 μg/g, which was two orders of magnitudes higher than the background levels of DEHP (302 ± 23.4 μg/g) in the dust measured prior to the experiments. The level was the highest among all target compounds in dust settled on their source materials. For DnBP, BBzP, DEHA, and DINCH, their steady-state concentrations in dust ranged from 1500 to 3500 μg/g, which were also 1 – 2 orders of magnitudes higher than their measured background levels in the dust. In contrast, the levels of phthalates and their alternatives in the blank control samples (i.e., dust settled on clean glass surfaces) at 50 days increased less than 50% for DEHP and fluctuated within the range of 20% for other target compounds compared with their background levels in the dust. The results suggest that there is significant direct transfer of phthalates and their alternatives from the source material to dust settled on the material surface. Such transfer is particularly important for high molecular weight phthalates, such as DEHP, due to their low vapor pressure and strong sorption to dust. The significantly enhanced concentrations of phthalates and their alternatives in dust settled on their source surfaces supported the findings in several recent field studies in the literature (Bi et al., 2015; Sukiene et al., 2016, 2017).

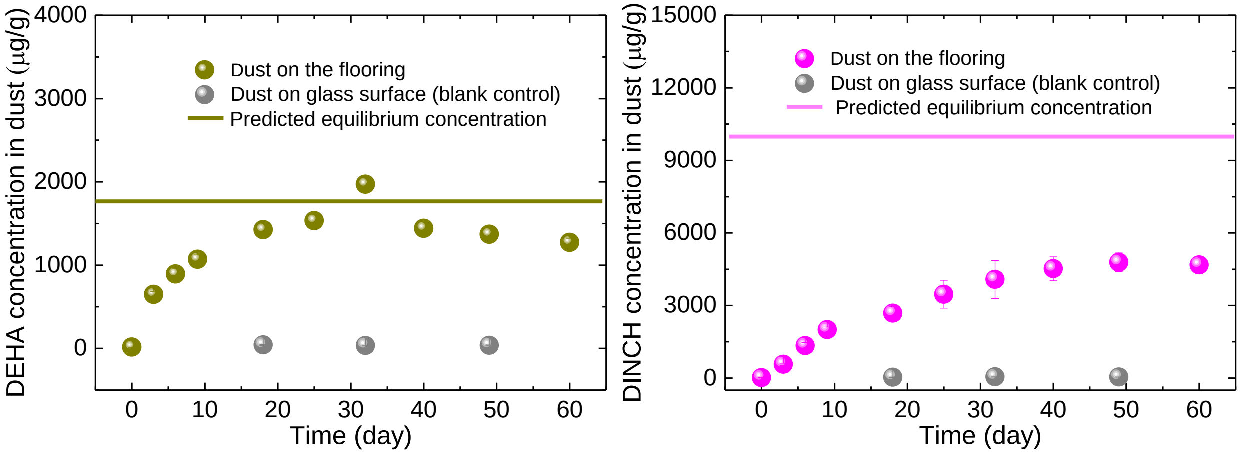


a) Sample 1

b) Sample 2



c) Sample 3



d) Sample 4

e) Sample 5

Figure 7. Concentrations of phthalates and phthalate alternatives in dust in contact with the source materials and in blank controls.

Figure 8 compared the measured steady-state concentrations in dust with the concentrations of target SVOCs in settled dust in residential homes reported in the literature (Rudel et al., 2003; Wilson et al., 2003; Hwang et al., 2008; Kanazawa et al., 2010; Bergh et al., 2011a; Guo and Kannan, 2011; Nagorka et al., 2011; Blanchard et al., 2014a; Fromme et al., 2016; Luongo and Ostman, 2016; Subedi et al., 2017; Larsson et al., 2017; Giovanoulis et al., 2018). The concentrations in dust measured in this study are 2 – 3 orders of magnitudes higher than the reported median concentration in the literature and are even higher than most of the reported maximum concentrations in these studies. Because the settled dust in the field campaigns might be collected on a variety of indoor surfaces which are not necessarily the source of the target compounds, the concentrations of phthalates and their alternatives in the dust could be lower than those measured in this study. Another possible reason is that the residence time of settled dust collected in these field studies is unknown. If it is not sufficient for SVOCs in the dust to equilibrium with the gas phase, the SVOC concentrations could be underrepresented in the dust. Finally, dust larger than 150 μm was screened out in this study while some of those large particles may be kept for analysis in field studies. Larger particles may have lower concentration of SVOCs due to their smaller specific surface area and lower mass transfer coefficients (Liu et al., 2014a). The results of current study indicate that when using settled dust as a sampling medium of SVOCs in indoor environments, it is important to know the sampling location and types of material surfaces because the types and levels of SVOCs may be strongly related to local materials used in indoor environments.

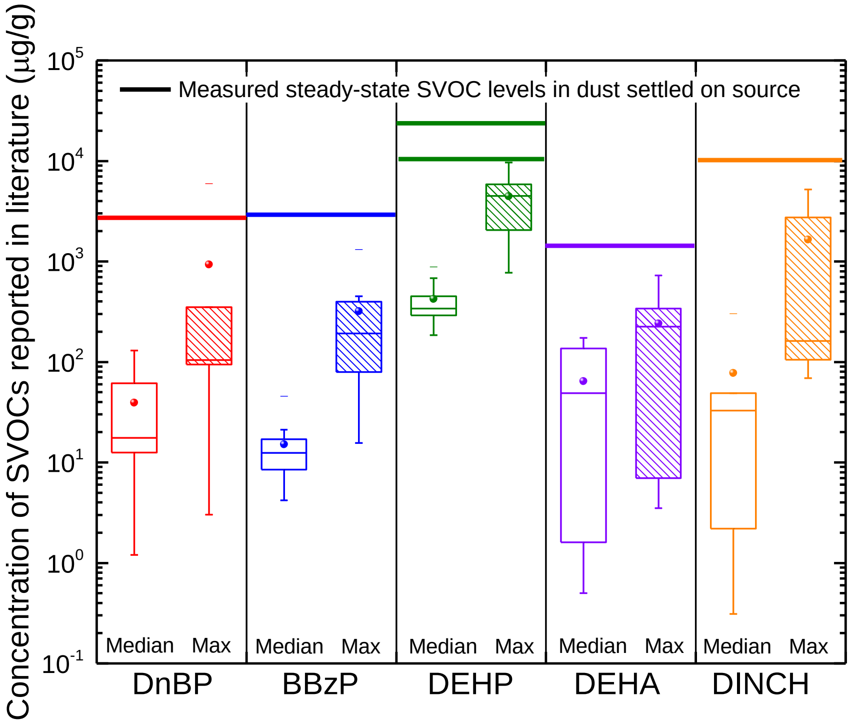


Figure 8. Comparisons between measured SVOC levels in dust and median and maximum concentrations of SVOCs reported in the literature.

3.2.4 Influence of Dust Loading

The influence of dust loading was further examined by increasing the mass loading from 0.5 - 3 g/m^2 to 15 - 20 g/m^2 on the surfaces of Sample 1, 2, and 3, as shown in Figure 7. The steady-state dust concentrations of BBzP and DEHP in the case of low dust loading were about twice as high as those in the case of high dust loading. Under low dust loading condition, it took about 22 and 40 days for the dust to reach sorption equilibrium for BBzP and DEHP, respectively; while the time increased to 40 days for BBzP and longer than 50 days for DEHP, when dust loadings were elevated. In contrast, no significant differences were observed for DnBP between the low and high dust loading cases either on the steady-state dust concentration or sorption kinetics, possibly due to its high volatility and weak sorption to particles. The results suggest that high dust loading may reduce the equilibrium concentration of phthalates in dust settled on the source surfaces and slow down the sorption kinetics. In residential homes, the dust loading on flooring materials ranges from 0.002 to 4 g/m^2 (Lioy et al., 2002; Johnson et al., 2009; Roberts et al., 2009; Patisaul et al., 2013; Rasmussen et al., 2013), which is even lower than the minimum loading used in the current experiments. Therefore, faster transfer of SVOCs from source surfaces to the dust deposited on the surface and higher SVOC levels in the dust are expected in actual indoor environments, compared to the experimental measurements.

3.2.5 Influence of Humidity

Dampness in buildings has been found to be a strong risk indicator for health effects such as asthma (Jaakkola and Knight, 2008). Several field studies suggested that increased levels of phthalates were associated with signs of higher dampness in residential homes (Hsu et al., 2012b; Zhang et al., 2013; Ait Bamai et al., 2014a). However, it is still unknown that whether there is any association between dampness (represented by RH) and the concentrations of phthalates in the indoor environments. In this study, DnBP and DEHP concentrations in dust settled on Flooring 3 were monitored under the RH at 0%, 50% and 80%, respectively. Phthalates accumulated in the dust at close rates and reached to similar steady-state levels under the different RH conditions (Figure 9). Previous studies showed that humidity has little or no influences on the emission of phthalates from PVC products (Clausen et al. 2007; Ekelund et al., 2010; Manoukian et al., 2016). Therefore, the amount of phthalates emitted from the vinyl flooring sample is expected to be similar at the different RHs in the current study. Recent studies suggested that when absorption was the dominant sorption mechanism, RH may have no influence on the soil/air partition of SVOCs; while when adsorption dominated the sorption process, the increased moisture in the air could competitively displacing SVOC molecules adsorbed on the soil surfaces (Hippelein and McLachlan, 2000; Goss et al., 2004; Davie-Martin et al., 2015). The results of current study suggest that RH may have little impact on the transfer of phthalates to the dust settled on the source surfaces and absorption may dominate the sorption to the dust.

3.2.6 Equilibrium model

A linear partition relationship was assumed between the concentration of target compounds in the dust and in the gas phase (Weschler and Nazaroff 2010), or

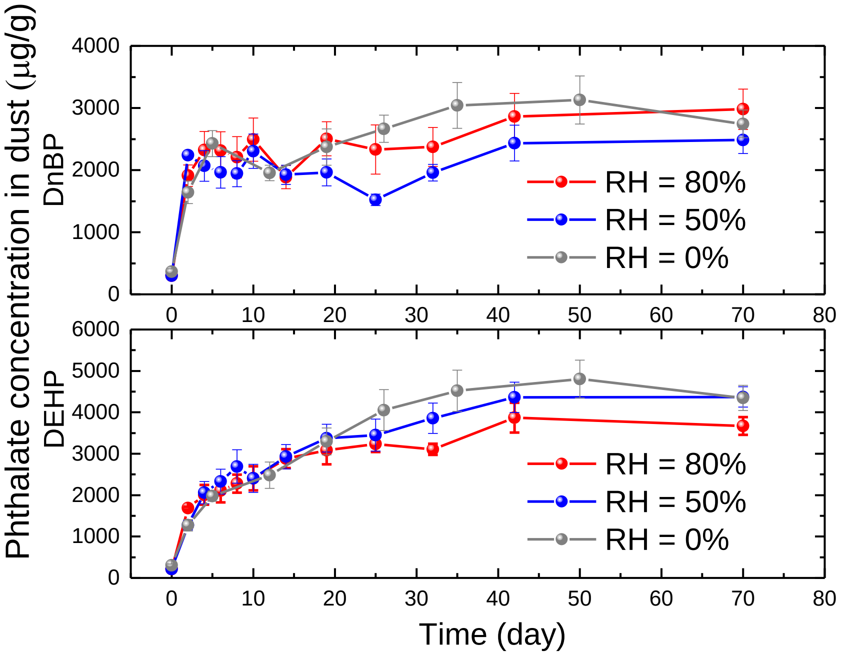


Figure 9. Influence of humidity on dust concentration

$$X_{dust} = \frac{f_{om\_dust} \times K_{oa} \times C_g}{\rho_{dust}} \tag{1}$$

Where X_{dust} is the dust-phase concentration of SVOCs, f_{om_dust} is the volume fraction of organics, ρ_{dust} is the density of dust, K_{oa} is the octanol-air partition coefficients, and C_g is the gas-phase concentration of SVOCs. f_{om_dust} was measured as 0.32 in this study and ρ_{dust} is the density of dust, which was assumed as 2.0×10^6 g/m^3 (Weschler and Nazaroff, 2010). Median values of K_{oa} for the target compounds reported in the literature was used (Xiao and Wania, 2003; Weschler and Nazaroff, 2008). y_0, which is the gas-phase concentration immediately adjacent to the material, was used for C_g. It is relatively constant and its values for the vinyl flooring and crib mattress cover samples were measured using a previous developed chamber method, and the results were listed in Table 5. The predicted equilibrium concentrations of target compounds in dust were shown as solid lines in Figure 7. The predicted equilibrium concentrations agreed well with most of the measured steady-state levels of target compounds, indicating sorption saturation in dust had been reached. In addition, the results suggest that when dust is in contact with the source of certain SVOCs, it is the gas-phase SVOCs immediately adjacent to the material partitioning with the dust phase rather than the gas phase in the bulk room air. Because y_0 values for a range of building materials and consumer products, such as vinyl floorings, crib mattress covers, backpacks, toys, table cloth, and wall coverings, have been reported in previous studies (Liang and Xu, 2014a; Xie et al., 2015; Cao et al., 2017), Equation 1 can be used to rapidly determine the equilibrium concentrations of SVOCs in dust settled on these sources.

For DEHP, the measured steady-state concentration in dust settled on Sample 1 agreed with the predicted concentrations, but the level is significantly higher than the concentrations measured in dust settled on Sample 3. In addition, the concentration of DINCH measured in this study also did not agree with the prediction. The discrepancy between measured and predicted equilibrium concentrations of DEHP and DINCH might be due to the uncertainties of K_{oa} values in the literature and differences of dust loadings used in the experiments. The K_{oa} values of DEHP reported in the literature range over three orders of magnitudes, which may significantly increase the uncertainty in the prediction. Figure B6 shows the potential uncertainties of the predicted equilibrium concentrations of DEHP (dashed line), estimated using the minimum and maximum K_{oa} values reported in literature. The measured steady-state DEHP concentrations fell within the estimated range. DINCH is an emerging phthalate alternative recently used in the market, few studies have reported its K_{oa} value or vapor pressure that can be used to calculate its K_{oa}. In addition, dust loading may also have a significant impact on the equilibrium concentrations of phthalates in dust (discussed in the next section). As shown in Table 5, even under low dust loading condition, the loading on Sample 3 is six times higher than those on Sample 1. The increase of dust loading may result in accumulation of multiple-layer dust on the source materials. Due to the existence of concentration gradient within the boundary layer, the gas-phase concentrations near the upper layer of the dust may be lower than that immediately adjacent to the materials (y_0). Therefore, the lower overall gas-phase concentration near the dust due to higher dust loading may result in a reduced equilibrium concentration in the dust.

3.2.7 Exposure for Children through Dust Ingestion

Children spend considerable amount of time crawling and playing on the floor. These activities may result in the transfer of dust from flooring materials to their hands. Because children frequently put their hands in their mouth, the hand-to-mouth transfer may result in significant amount ingestion of the contaminated dust. Although the total daily ingestion of soil and dust was estimated for children (Moya and Phillip 2014), there are limited studies focusing specifically on the ingestion of dust through hand-to-mouth transfer. Wilson et al. (2013) developed a mechanistic model to estimate the soil and dust ingestion rates through hand to mouth transfer and found that the mean dust ingestion rate for toddlers who play on hard surfaces was 53 mg/day. Another study used U.S. Environmental Protection Agency (EPA)'s Stochastic Human Exposure and Dose Simulation (SHEDS) model to differentiate among various activities of children and reported mean hand-to-mouth dust ingestion to be 20 mg/day. They also reported that hand-to-mouth dust ingestion typically consisted of about 29 percent of the amount of the total daily soil and dust ingestion. Therefore, we used the hand-to-mouth dust ingestion rate reported in the literature to estimate the potential exposure to phthalates and phthalate alternatives due to ingestion of the settled dust measured in the current study. For studies only reported the total daily dust ingestion rate, we applied the ratio of 29 percent to obtain the estimated hand-to-mouth ingestion rates. The exposure through ingestion of dust was calculated for a three-year-old toddler with the following equation:

$$Exposure\ (\mu g/kg/d) = \frac{IngR_{HtoM} \times X_{dust}}{BW} \tag{1}$$

Where $IngR_{HtoM}$ (mg/day) is the hand-to-mouth dust ingestion rate; X_{dust} (µg/g) is the equilibrium concentration of target compounds in dust measured in this study; and BW (kg) is the body weight. Parameters used in the calculation were listed in Table 6.

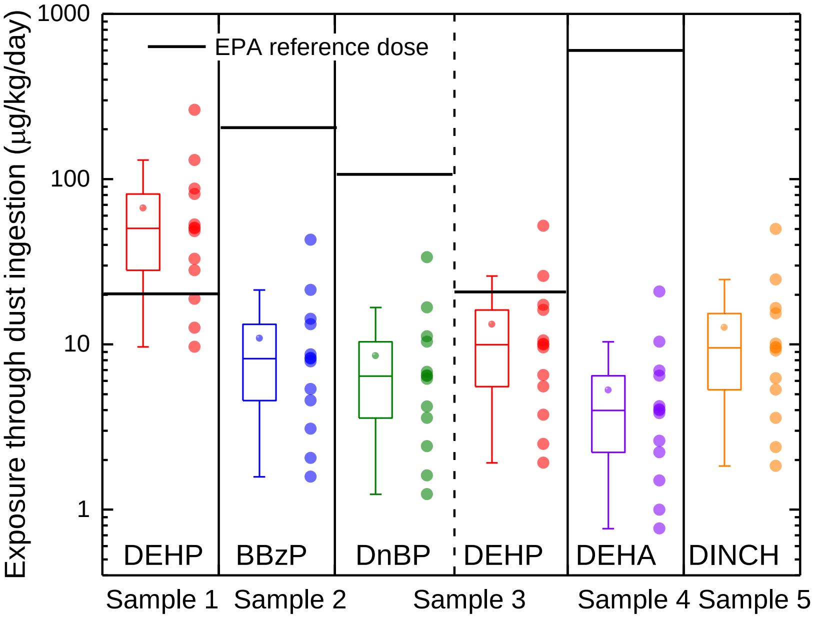


Figure 10. Toddlers' exposure to phthalates and phthalate alternatives via ingestion of dust settled on source surfaces due to hand-mouth activities.

Figure 10 showed the estimated exposure of toddlers to phthalates and phthalate alternatives via ingestion of dust settled on source surfaces due to their hand-to-mouth activities. The results suggest that the potential exposure could be significantly high. The maximum estimated exposure to DEHP was 263 µg/kg/day, which was more than 20 times higher than the oral reference dose reported by U.S. EPA. The median exposure to DEHP was 50 and 10 µg/kg/day for dust settled on flooring Sample 1 and Sample 3, respectively, which were also over two times higher than and half of the reference dose. The estimated median values of exposure to DnBP, BBzP, and DEHA were 6.43, 8.20, and 9.53 µg/kg/day, respectively. Although they were significantly lower than the reference doses (100, 200, and 600 µg/kg/day, respectively), the exposure levels via only ingestion of hand-to-mouth dust settled on source surfaces remains comparable to the daily overall intake of DnBP, BBzP, DEHA, or DINCH in typical residential homes reported in the literature (Oomen et al., 2008; Shi and Zhao, 2014; Zhang et al., 2014). The results indicate that ingestion of dust settled on the sources of SVOCs may play an important role on children's exposure to these compounds.

Table 6. Parameters used in the model prediction

ID	Sample 1	Sample 2	Sample 3		Reference
Content	DEHP	BBzP	DnBP	DEHP	
Dust-phase concentration, X_{dust} (ug/g)	22700	3700	2900	4500	Measured in this study
3-year-old toddler body weight, BW (kg)	13.8				(US EPA, 2011)
Hand-to-mouth dust ingestion rate (mg/day)	20-160				(Lepow et al., 1975; Duggan and Williams, 1977; Binder et al., 1986; Clausing et al., 1987; Davis et al., 1990; Stanek and Calabrese, 1995; Calabrese et al., 1997b, 1997a; Hogan et al., 1998; Özkaynak et al., 2011; Stanek et al., 2012)

3.3 PHTHALATES AND ORGANOPHOSPHATES IN SETTLED DUST AND HVAC FILTER DUST OF U.S. LOW-INCOME HOMES: ASSOCIATION WITH SEASON, BUILDING CHARACTERISTICS, AND CHILDHOOD ASTHMA (OBJECTIVE 3)

3.3.1 Building Investigations

A total of 54 homes from rural/semi-rural areas of central Texas enrolled in this study. Environmental sampling was conducted at each home during both winter (December 2014 – February 2015) and summer (June 2014 – September 2014) seasons. Questionnaires including personal and dwelling characteristics were distributed and collected during home visits. Through the questionnaires and onsite visual inspections, building characteristics such as the age and type of the house, floor area, number of rooms, frequency of cleaning, signs of mold and dampness, and floor materials were recorded. There were five categories of floor materials: PVC flooring, wall-to-wall carpet, wood floor, concrete, and ceramic tile. Additionally, the HVAC run-times and indoor temperatures in each household were monitored using Onset U12-012/013 monitors installed in each home; the airflow rate through the HVAC system was determined for each site; and the microbial concentrations and community were also investigated with details described in Maestre et al. (2018) and Givehchi et al. (2018).

The study enrolled 54 children (60% boys and 40% girls) with a mean age of 10.3 years (SD = 2.8 years). There were 37 children who had physician-diagnosed asthma and experienced asthma symptoms in the previous 12 months, leaving 17 children in the healthy control group. Asthma severity was measured with the validated Severity of Chronic Asthma scale with three questions derived from national clinical guidelines that parents answered about the frequency of daytime asthma symptoms, night-time wakening for asthma, and activity limitations due to asthma (Horner et al., 2006).

3.3.2 Sample Collection and analysis

HVAC filter dust and settled dust samples were collected in both summer and winter seasons. Brand-new synthetic HVAC filters, with minimum efficiency reporting value (MERV) ratings of 7-8, were installed at each house for one month. These filters have an efficiency of 50-85% for 3-10 μm particles when tested according to the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) Standard 52.2. The filters were placed in return grilles located on the ceiling or the wall in each home. After sampling, filters were removed, sealed in static shielding bags, and transported back to Austin, Texas inside an ice box and stored in the laboratory at -18°C until further processing. Settled dust samples were collected immediately before removal of the installed HVAC filter at each sampling site, using a previously reported protocol (Bi et al., 2015). Briefly, a modified vacuum cleaner was used, which was connected to a special aluminum nozzle holder to avoid contact between dust and plastic parts and limit potential contamination. Settled dust (approximately 100 mg) was collected onto a pre-weighed cellulose thimble filter (26 × 60 mm, Whatman Inc.) inserted into the nozzle holder. Dust sampling was conducted mainly in children's rooms. Dust was collected from the floor surface and from objects within 30 cm above the floor. After sampling, cellulose thimbles were placed in pre-cleaned amber glass jars and transported at 4°C to the laboratory for storage at -18°C.

Six phthalates, including dimethyl phthalate (DMP), DEP, DnBP, BBzP, DEHP, and di-n-octyl phthalate (DnOP), and five organophosphates, including tri-n-butyl phosphate (TnBP), tris (1-chloro-2-propyl) phosphate (TCPP), tri (2-butoxyethanol) phosphate (TBEP), triphenyl phosphate (TPhP), and TDCPP, were targeted in this study. Two small pieces of filter samples (10 × 10 cm) were cut out of each HVAC filter obtained at randomly chosen positions, followed by determination of area and weight of the filter pieces. The remaining filter section was vacuumed, and a piece of cleaned filter (100 cm^2) was obtained and weighed using a five-digit microbalance. The mass of dust on HVAC filters was determined from the difference in mass between the filter samples and the cleaned filter piece. The dust samples were then ultrasonically extracted for 30 minutes with hexane, which was repeated three times per sample. Extracts were concentrated using a rotary evaporator (IKA RV-10) and a high purity N_2 stream. After concentrating to approximately 5 mL, the samples were filtered (Whatman AutovialTM syringless PTFE filters with 0.2 μm pore). Next, a nitrogen purge was used to further concentrate the samples to approximately 500 μL. Analysis of the extracted samples was conducted by gas chromatography (GC) coupled with a flame ionization detector (Agilent 7890A GC-FID). Details of the method are provided in the Appendix.

For data analysis, dust concentrations below the limit of detection were assigned a value corresponding to half of the respective detection limit. A Shapiro-Wilk test of normality indicated that the dust concentrations of phthalates and organophosphates were not normally distributed. Thus, non-parametric Spearman's rank correlation analyses were performed to study the co-occurrence of compounds. Wilcoxon signed-rank test was used to compare the levels of target compounds between HVAC filter and settled dust during different seasons. Potential associations between concentrations of target SVOCs in dust and questionnaire data (i.e., building and personal characteristics) were analyzed with Mann-Whitney *U* test. Linear regression analyses were performed to examine the association between the severity of childhood asthma and SVOC concentrations in dust. For certain analyses, the total number of test samples was lower than 54, due to unavailability of participating homes or inadequate dust mass. DMP, DEP, TnBP and TBEP were not included in the statistical analysis due to the relatively low detection frequency. Statistical analysis was carried out using R v3.4.4, an open-source statistical software environment; and a 5% significance level was applied.

3.3.3 Concentrations of Phthalates and Organophosphates in Dust

Phthalates and organophosphates were detected in all HVAC filter dust and settled dust samples. Their concentrations and detection rates are summarized in Table 7, in which the data for winter and summer seasons were combined (the data for each season are reported in Table C4 and discussed in the following sections). The mean concentration of each compound was found to be higher than the median value, suggesting a skewed concentration distribution as shown in the cumulative frequency distribution (Figure C5). In general, the measured concentrations varied considerably across all sampling sites, and the concentrations of phthalates were much higher than those of organophosphates. Little to no correlation for most of the phthalates and organophosphates were observed (Tables C5 and C6), indicating their indoor sources are diverse. However, concentrations of BBzP and DEHP in both HVAC filter and settled dust correlated significantly, suggesting these two compounds may have originated from the same sources.

For phthalates, the total concentrations of target compounds in HVAC filter dust ranged from 2.91 to 6370 μg/g, with a median value of 286 μg/g, while in settled dust, total phthalate concentration ranged from 26.4 to 5420 μg/g, with a median value of 288 μg/g. For the two-season combined data (Table 7), no significant concentration differences were observed between HVAC filter dust and settled dust samples for all phthalate compounds ($p > 0.05$, Mann-Whitney U test). The most frequently detected phthalate was DEHP for both HVAC filter and settled dust, followed by BBzP, DnOP, and DnBP. These are the same phthalates observed to be most abundant in urine samples of the U.S. population (Meeker and Ferguson, 2014; Zota et al., 2014).

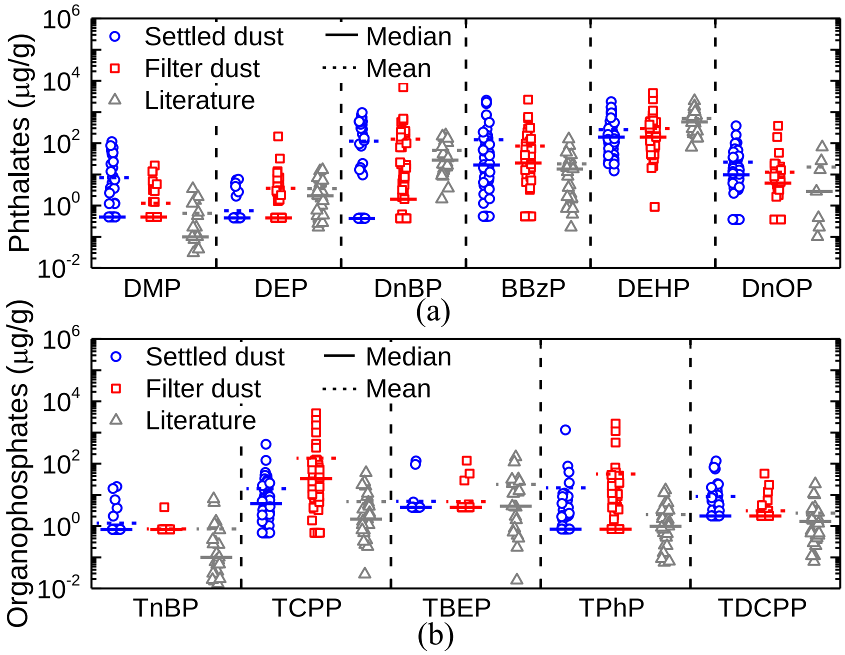
The compounds detected at the highest concentration in both filter and settled dust were DEHP and BBzP (HVAC filter dust: 3980 and 2440 μg/g, respectively; settled dust: 2120 and 2380 μg/g, respectively). These compounds are widely used as plasticizers in PVC products and building materials (e.g., flooring and wall covering) with material-phase concentrations of approximately 10% – 30% by weight (Liang and Xu, 2014b, 2014a). Health concerns surrounding phthalate plasticizers, especially DEHP and BBzP, have caused a trend toward using phthalates of lower volatility or alternative plasticizers (Schossler et al., 2011); however, the findings of the present study demonstrated that DEHP and BBzP are still the most abundant phthalates in indoor dust from low-income homes. The low detection rates of DMP and DEP are most likely due to its high volatility, with their majority present in gas phase rather than dust phase. Since very limited published data on phthalate levels in HVAC filter dust in residential homes are available, Figure 11a shows a comparison of measured phthalate concentrations with median values reported in the literature for settled dust in residential homes (Ait Bamai et al., 2014a; Albar et al., 2017; Bergh et al., 2011; Olivier Blanchard et al., 2014; Bornehag et al., 2005; Bu et al., 2016; Dodson et al., 2015; Fromme et al., 2004; Gevao et al., 2013; Guo and Kannan, 2011; He et al., 2016; Hsu et al., 2012; Kanazawa et al., 2010; Kubwabo et al., 2013; Langer et al., 2010; Luongo and Ostman, 2016; Rudel et al., 2003; Santillo et al., 2003; Shin et al., 2014; Subedi et al., 2017; Wang et al., 2014; Zhang et al., 2013). The median concentrations of DEHP (155 μg/g) and BBzP (20.1 μg/g) in settled dust from low-income houses determined in the present study were comparable to concentrations measured in common U.S. homes at other locations in the United States (Rudel et al., 2003; Guo and Kannan, 2011; Shin et al., 2014; Dodson et al., 2015), such as Richmond and Bolinas, CA (DEHP: 140 μg/g and BBzP: 19 μg/g), Albany, NY (DEHP: 304 μg/g and BBzP: 21.1 μg/g), Cape Cod, MA (DEHP: 340 μg/g and BBzP: 45.4 μg/g), and across the United States (DEHP: 144 μg/g and BBzP: 14.9 μg/g).

Table 7. Detection rates and the levels of phthalates and organophosphates in dust

	HVAC filter dust (μg/g), N = 91						**Settled dust (μg/g),** N = 92					
	>MDL[a]	Min	Median	Max	Mean	SD[b]	>MDL[a]	Min	Median	Max	Mean	SD[b]
Phthalates												
DMP	13%	<MDL	<MDL	19.0	1.19	2.73	28%	<MDL	<MDL	111	7.85	19.8
DEP	31%	<MDL	<MDL	163	3.54	17.3	7%	<MDL	<MDL	6.93	0.68	1.14
DnBP	52%	<MDL	1.60	6200	133	654	30%	<MDL	<MDL	950	115	228
BBzP	81%	<MDL	23.1	2440	80.8	270	80%	<MDL	20.1	2380	128	383
DEHP	98%	<MDL	155	3980	293	502	100%	12.8	155	2120	271	347
DnOP	59%	<MDL	5.27	358	11.7	40.3	74%	<MDL	9.65	355	24.6	45.4
Total		2.91	286	6370	524	894		26.4	288	5420	547	749
Organophosphates												
TnBP	1%	<MDL	<MDL	4.01	0.81	0.34	5%	<MDL	<MDL	18.3	1.24	2.46
TCPP	91%	<MDL	33.3	4090	150	529	77%	<MDL	5.22	418	15.8	45.5
TBEP	4%	<MDL	<MDL	124	6.04	13.5	3%	<MDL	<MDL	121	6.23	15.2
TPhP	43%	<MDL	<MDL	1930	46.4	233	36%	<MDL	<MDL	1190	16.8	124
TDCPP	11%	<MDL	<MDL	47.7	3.10	5.23	37%	<MDL	<MDL	122	8.89	18.2
Total		8.24	56.9	5190	206	738		8.24	19.3	1220	48.9	133

[a] The method detection limits (MDLs) for dust samples were reported in the Supplementary Material. Detection rates are reported as % above MDL.

[b] Standard deviation.



(a)

(b)

Figure 11. Comparison of measured concentrations of (a) phthalates and (b) organophosphates in dust with median values reported in literature for residential homes (over 4000 homes in 42 studies).

For organophosphates, the total concentrations of target compounds in HVAC filter dust ranged from 8.24 to 5190 µg/g with a median value of 56.9 µg/g, while in settled dust, concentrations ranged from 8.24 to 1220 µg/g, with a median value of 19.3 µg/g. Similar to phthalates, no significant concentration differences were observed between HVAC filter dust and settled dust samples for each compound in the two-season combined data ($p > 0.05$, Mann-Whitney *U* test), except for TCPP. In HVAC filter dust, TCPP was detected in the majority of the samples (91%) and at the highest concentrations (mean: 150 µg/g; range: <MDL to 4090 µg/g). TPhP and TDCPP were detected in a smaller percentage of samples (43% and 11%, respectively) and at lower concentrations (mean: 46.4 µg/g; range <MDL to 1930 µg/g and mean: 3.10 µg/g; range <MDL to 47.7 µg/g, respectively). In settled dust, the most abundant compounds were similar to those detected in HVAC filter dust, but their levels and detection rates were lower, except for TDCPP and TnBP. TCPP is majorly applied in rigid polyurethane foams (PUFs) for building insulation (BAUA 2008a). TPhP is used as a flame retardant and/or a plasticizer in polymeric materials and products such as wallpaper, LCD-TV, computers, curtains, and insulation boards (Kajiwara et al., 2011). TDCPP and TCPP have been detected in flexible PUFs furnishings including couches, chairs, mattress pads, and pillows (Stapleton et al., 2009, 2011). The widespread use of these compounds in building materials and consumer products may contribute to their high levels in both the HVAC filter dust and settled dust samples. These results are in agreement with previous research assessing exposure to organophosphates, in which the metabolites of TDCPP, TPhP, and TCPP were the most frequently detected biomarkers in humans (Stapleton et al., 2009; Cooper et al., 2011; Meeker et al., 2013; Butt et al., 2014; Dodson et al., 2014; Jayatilaka et al., 2017; Ospina et al., 2018). Figure 11b shows a comparison of measured organophosphate concentrations to median values reported in the literature for settled dust in residential homes (Abdallah and Covaci, 2014; Ali et al., 2016, 2013, 2012; Araki et al., 2014; Bergh et al., 2011; Brommer and Harrad, 2015; Castorina et al., 2017; Cequier et al., 2014; Dodson and Perovich, 2012; Fan et al., 2014; He et al., 2016; Hoffman et al., 2015; Kanazawa et al., 2010; Kim et al., 2013; Langer et al., 2016; Luongo and Ostman, 2016; Meeker and Stapleton, 2010;

Schreder and La Guardia, 2014; Shin et al., 2014; Stapleton et al., 2014, 2009; Tajima et al., 2014; Van den Eede et al., 2011; Wu et al., 2016; Zhou et al., 2017). The median concentrations measured in settled dust in the present study were comparable to those reported in the literature, except for TCPP. The median level of TCPP in the present study was 3- to 180-times higher than reported levels in Belgium, Canada, China, Denmark, Egypt, Japan, New Zealand, Kingdom of Saudi Arabia, and Sweden, (Abdallah and Covaci, 2014; Ali et al., 2012, 2016; Bergh et al., 2011; Fan et al., 2014; He et al., 2016; Tajima et al., 2014; Van den Eede et al., 2011; Wu et al., 2016), and was slightly higher than levels reported in U.S., Germany, and Norway (Stapleton et al., 2009, 2014; Dodson and Perovich, 2012; Cequier et al., 2014; Schreder and La Guardia, 2014; Zhou et al., 2017), but was relatively lower than levels reported in two studies in Japan (Kanazawa et al., 2010; Araki et al., 2014). The high level of TCPP in dust samples in the present study could be attributed to the use of rigid PUFs as roof and wall insulation for low-cost housing, especially considering that over 80% of produced TCPP is used in rigid PUFs, mainly for construction applications (BAUA, 2008).

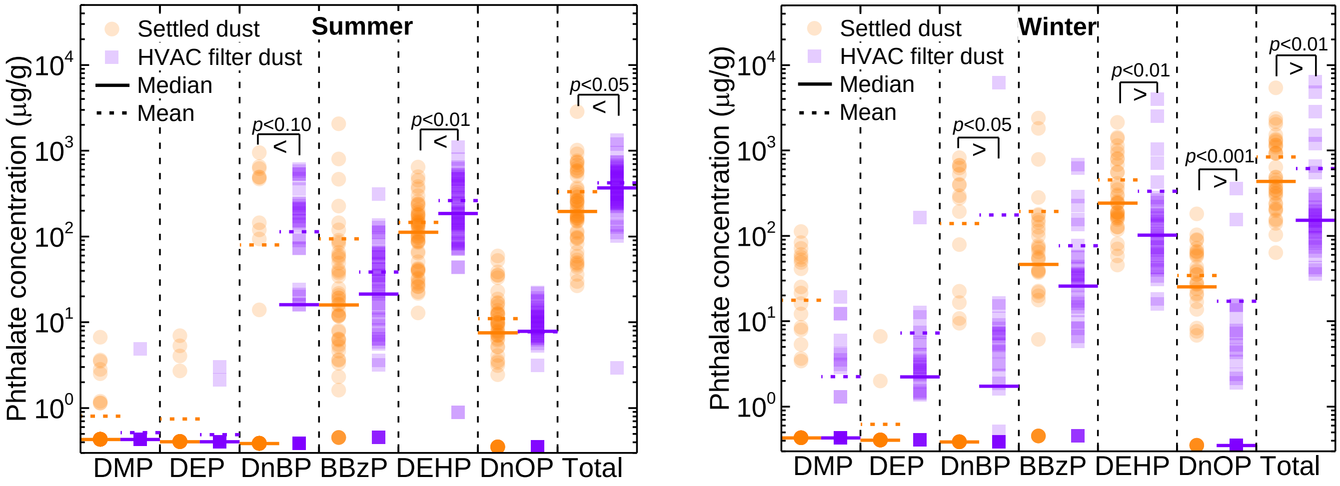
3.3.4 Comparison Between HVAC Filter Dust and Settled Dust

Spearman's rank correlation analysis was applied to explore the possible correlations between HVAC filter dust and settled dust. Significant positive correlation for TCPP (r_s = 0.41, p = 0.0001) was observed, suggesting their common sources inside the house. Currently, the vast majority of TCPP is used in rigid PUFs as construction materials and a small fraction is used in flexible PUFs for furniture and upholstery (WHO, 2004; BAUA, 2008). Therefore, it is possible that a large amount of TCPP was emitted from walls, roofs, and/or furniture into the air and then captured by suspended particles and floor dust via the same gas/particle absorptive partitioning process. Since the equilibration time of TCPP is short due to its relatively small K_{oa} (Weschler and Nazaroff, 2008, 2010), sorption equilibrium between the gas and particle phases may occur at a fast rate. TCPP has been detected in indoor air at concentrations ranging from 2 ng/m^3 to 1300 ng/m^3 in residential homes (Bergh et al., 2011; Marklund et al., 2005; Saito et al.,

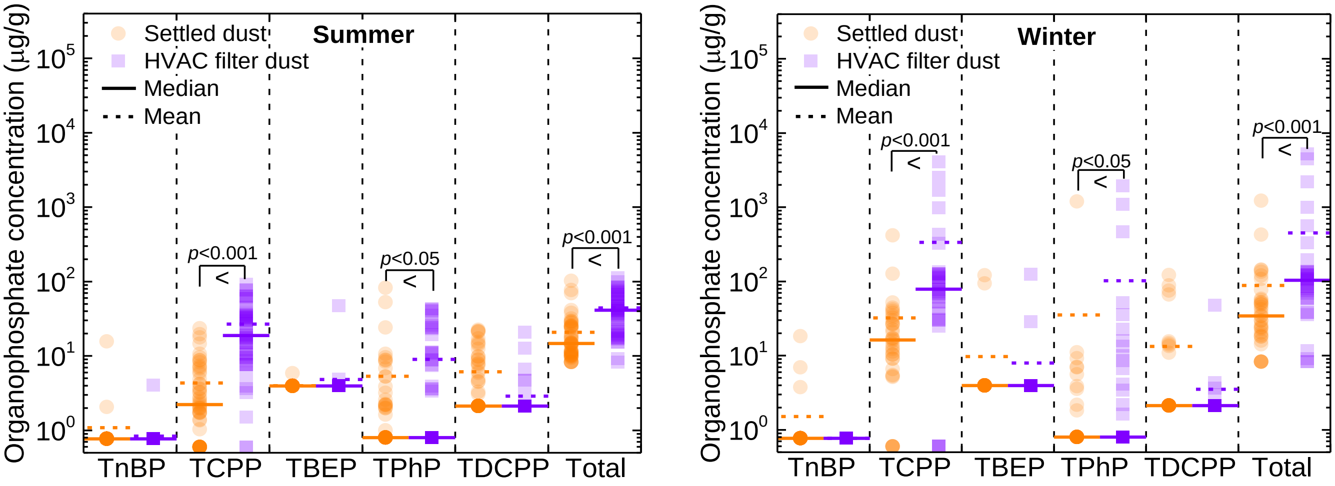
2007) and was recently classified by the U.S. EPA as having a high hazard for reproductive and developmental effects (US EPA, 2015). No such correlation between HVAC filter dust and settled dust was observed for other target compounds, suggesting different sources and/or different environmental fates for these compounds in an indoor environment.

HVAC filter dust and settled dust samples were then paired for each single house and compared by season using paired Wilcoxon signed rank tests. As shown in Figure 12, for most of the frequently detected compounds, statistically significant higher concentrations were observed in HVAC filter dust than in settled dust, except for the case of phthalates in winter. The median concentrations of DnBP, DEHP, and total phthalates in HVAC filter dust in summer were 16, 185, and 368 μg/g, respectively, which were higher than those in settled dust (0.39, 112, and 195 μg/g, respectively). Similarly, for organophosphates, the concentrations of TCPP, TPhP, and total organophosphates in HVAC filter dust were significantly higher than in settled dust in both summer and winter (Figure 12). In summer, the median levels of TCPP and total organophosphates in HVAC filter dust were 18.7 and 41.3 μg/g, respectively; while in settled dust TCPP and total organophosphate concentrations were 2.22 and 14.7 μg/g, respectively. In winter, the median levels of TCPP and total organophosphates in HVAC filter dust were 78.6 and 104 μg/g, respectively; while in settled dust were 16.2 and 34.3 μg/g, respectively.

One possible reason for the higher SVOC concentration in HVAC filter dust may be related to the smaller particle size in HVAC filter dust as compared to settled dust. HVAC filter dust predominately consists of particles suspended in indoor air, with a small portion of coarse particles originating from indoor surfaces due to particle resuspension caused by airflow and occupant activities such as walking, cleaning, and vacuuming (Mukai et al., 2009). In this study, particle size distribution was determined for filter dust samples with details described in Givehchi et al. (2018). The mean value of particle volume median diameters is 32.3 μm (SD = 4.78 μm). In contrast, settled dust consists of particles deposited from indoor aerosols, soil particles tracked inside by foot, and biologically derived material such as skin flakes. Settled dust is generally assumed to be a mixture of large particles that have high settling rates. Although its particle size distribution was not determined in this study, the volume or mass median diameters of settled dust particles were reported in other indoor studies, with a range of 86-685 μm (Lewis et al., 1999; Morawska and Salthammer, 2003; Cao et al., 2014b; Gustafsson et al., 2018). Previous studies (Weschler and Nazaroff, 2008, 2010) have demonstrated that the equilibration of an SVOC in dust is largely controlled by its absorption to the organic matter of dust. Considering that the fraction of organic matter in dust (f_{om}) increases as particle size decreases (Morawska and Salthammer, 2003), higher equilibrium concentration of SVOCs are expected in smaller-sized particles such as HVAC filter dust. Additionally, the time scale for SVOCs to achieve the equilibrium concentration in dust is also affected by particle size. Smaller particles show faster mass transfer rates due to their larger specific surface area and higher mass transfer coefficients (Liu et al., 2014a). The high-velocity airflow pulled by a fan in the HVAC system further enhances the mass transfer coefficient between gas and particle phases (EI-Amin, 2011; Clack, 2017). As a result, the levels of SVOCs in HVAC filter dust may be close to their equilibrium concentrations, if the filters were installed for a long enough period of time. In contrast, measured SVOC concentrations in settled dust are observed to be systematically less than their predicted equilibrium levels in typical indoor conditions, particularly for compounds with high K_{oa} values, due to slow sorption kinetics (Weschler and Nazaroff, 2010). Finally, the large volume of air that passed through HVAC filters increased the sensitivity of sampling and analysis. In this study, since the filters were typically installed at the return air grille indoors, the environmental conditions (e.g., temperature or humidity) were probably similar between HVAC filter dust and settled dust; therefore, if the presence of SVOCs in dust is dominated by the process of sorption from the gas-phase in indoor air, SVOC concentrations in HVAC filter dust should be no less than their concentrations in settled dust.



(a) Phthalates



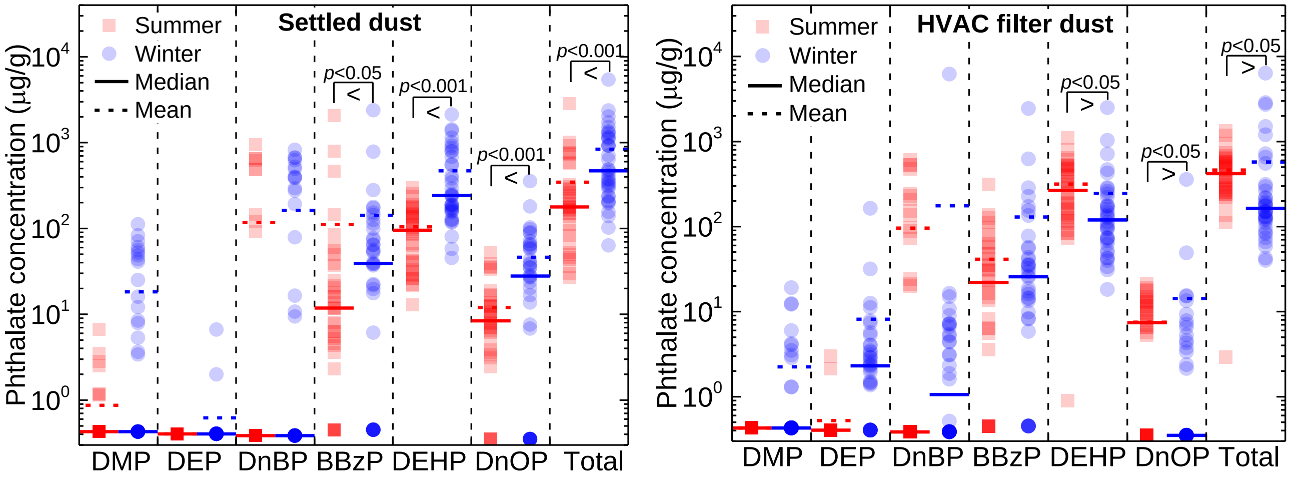
(b) Organophosphates

Figure 12. Comparison of SVOC concentrations between HVAC filter dust and settled dust in summer (N = 51) and winter (N = 36)

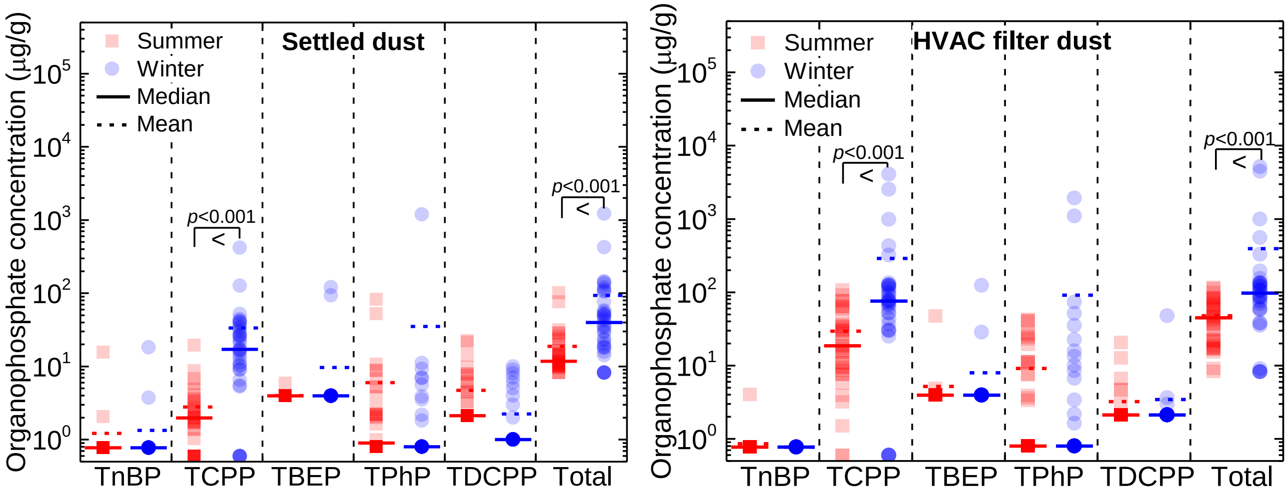
In winter, higher phthalate levels were observed in settled dust as compared to HVAC filter dust (Figure 12a). The median concentrations of DEHP, DnOP, and total phthalates in settled dust were 242, 25.3, and 434 μg/g, respectively, which were significantly higher than those in HVAC filter dust (101, 0.35, and 151 μg/g, respectively). Although no statistically significant difference was observed for BBzP, its median and mean levels in settled dust (46.1 and184 μg/g, respectively) was also higher than those in HVAC filter dust (26.2 and 138 μg/g, respectively). The high phthalate concentrations in settled dust could be attributed to the combination effect of local phthalate sources and use of space heaters in winter. Phthalates have been widely used in vinyl flooring as plasticizers. Liang and Xu (2014a, 2014b) demonstrated that 15 out of 16 vinyl flooring samples purchased in the U.S. in their study contained high concentrations of phthalates in the material phase. In the present study, approximately 40% of the low-income homes had vinyl flooring covering the entire floor area in the children's room. Phthalate concentrations in settled dust, which was in direct contact with vinyl flooring, were likely to be increased due to direct transfer from the source into dust (Bi et al., 2015; Bi and Xu, 2016; Sukiene et al., 2017). Furthermore, use of space heaters may substantially enhance the transfer of phthalates to dust. Space heaters were observed in children's rooms as well as other bedrooms in the sampling sites, possibly as a way to keep thermostat settings low and save on energy bills. Although the number of space heaters was not recorded, the runtime of the HVAC system was monitored at each site. The average runtime for each HVAC system in the winter was only 10%, which is much shorter than in the summer (31%). For a number of homes, the HVAC runtime during the winter was even less than 5%, but room temperatures were maintained approximately 10°C above the outdoor ambient temperature during cold weather. Use of space heaters can generate high temperatures on the immediate floor surface, and thus significantly increase the emission of phthalates from the source. Although dust/gas partition coefficient also decreases with increasing temperature, it may not offset the increase of emission, resulting in higher phthalate concentrations in settled dust. For example, when the surface temperature increased from 21 to 40°C, DEHP gas-phase concentration immediately adjacent to the surface of vinyl flooring increased by approximately 2-fold (Liang and Xu, 2014a), while the K_{oa} value of DEHP, which is proportional to the dust/gas partition coefficient, decreased from $10^{11.5}$ to $10^{10.4}$ (Xiao and Wania, 2003; Weschler and Nazaroff, 2010; Gobble et al., 2014; Bi et al., 2015). As a result, under equilibrium conditions, DEHP concentration in settled dust in contact with vinyl flooring increased by a factor of two. In addition, smaller particle size and lower dust loading on surfaces due to seasonal variation may further shorten the time to reach equilibrium and contribute to the high phthalate concentrations in settled dust in winter, as discussed in the next section.

3.3.5 Seasonal Variation

Seasonal variation of SVOC contamination in indoor dust may significantly influence the reliability of human exposure assessments that are based on single-point sampling (Cao et al., 2014a). However, only a few studies of seasonal effects have been performed (Allen et al., 2008; Harrad et al., 2008; Batterman et al., 2009; Vorkamp et al., 2011; Yu et al., 2012; Cao et al., 2014a, 2015), and most of these were based on limited sampling sizes. The average daily outdoor air temperature in central Texas changed from 14.6±0.3°C in winter to 28.1±1.9°C in summer, while the average daily indoor air temperature changed slightly from 21.9±1.9°C in winter to 24.1±1.6°C in summer. Figure 13 shows the seasonal variation of phthalate and organophosphate concentrations in settled dust and HVAC filter dust. Significant seasonal differences were observed in settled dust, especially for phthalates. The median concentrations of BBzP, DEHP, DnOP, and total phthalates in settled dust in the winter were 39.1, 242, 27.9, and 468 μg/g, respectively, which were approximately three times higher than those in the summer (11.8, 95, 8.39, and 177 μg/g, respectively). For organophosphates, higher median concentrations of TCPP and total organophosphates were also observed in settled dust in winter (17.1 and 39.9 μg/g, respectively) than in summer (1.97 and 11.7 μg/g, respectively). The seasonal trend in the present work is similar to the few previous studies available (Yu et al., 2012; Cao et al., 2014a, 2015), which found higher winter concentrations in indoor settled dust for SVOCs including total organophosphates, hexabromocyclododecanes, and PBDEs. For HVAC filter dust, the seasonal trends were inconsistent: DEHP, DnOP, and total phthalates showed higher median concentrations in summer (266, 7.41, and 420 μg/g, respectively) than in winter (119, 0.35, 163 μg/g, respectively), while TCPP and total organophosphates showed higher median concentrations in winter (76.1 and 97.8 μg/g, respectively) than in summer (18.5 and 44.7 μg/g, respectively). To our knowledge, no studies have examined the seasonal influences on SVOC concentrations in HVAC filter dust, but several groups measured their levels in indoor suspended particles. Zhang et al. (2014) found significantly higher concentrations of phthalates on PM_{10} and $PM_{2.5}$ in summer as compared to winter, while Mandin et al. (2016) observed opposite results.



(a) Phthalates



(b) Organophosphates

Figure 13. Seasonal concentration variations of (a) phthalates and (b) organophosphates in HVAC filter and settled dust (N = 36).

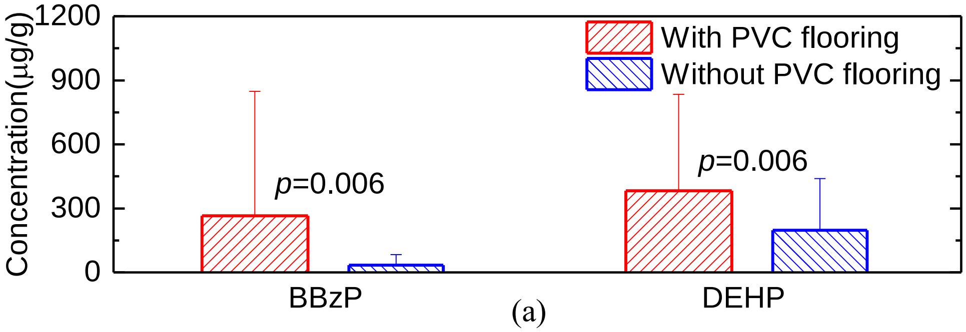
There are many factors that may contribute to the seasonal variations in the concentrations of phthalates and organophosphates in dust. Temperature is one of the most important factors, which could have a significant influence on the SVOC equilibrium concentration in dust. For equilibrium conditions, an SVOC's concentration in dust (C_{dust}) is directly proportional to its gaseous concentration (C_g), to the K_{oa} value, and to the fraction of organic matter in dust (f_{om}), or $C_{dust} = C_g \times K_{oa} \times f_{om}/\rho_{dust}$ (Weschler and Nazaroff, 2010). The change of C_{dust} with an increasing temperature mainly depends on competition between the extent of the K_{oa} decrease and the increase in C_g. For example, based on the correlation developed by Liang and Xu (2014a), when indoor temperature increases from 21.9 to 24.1 °C, emission of DEHP from different vinyl flooring samples can be enhanced 1.5- to 3.4-times, while the K_{oa} value of DEHP decreased only by 30%, from 3×10^{11} to 2.2×10^{11} (Xiao and Wania, 2003). Therefore, if there are plenty of DEHP sources present in the indoor environment, such as the large surface area of vinyl flooring, it is possible to have a higher equilibrium concentration of DEHP in dust in the summer than in winter. In contrast, for other SVOCs, especially those with higher vapor pressures whose emissions are less sensitive to temperature changes (Kemmlein et al., 2003; Liang and Xu, 2014a), the decrease of K_{oa} may dominate the process, resulting in a lower equilibrium dust concentration in the summer. In addition to temperature, ventilation is another important factor. Better ventilation conditions may have a dilution effect of indoor contaminants and thus result in a lower gas-phase concentration (C_g) of SVOCs in air (Xu et al., 2009). However, ventilation inside residential buildings depends mainly on infiltration, weather conditions, and occupant behaviors (e.g., window opening). The air-tightness of the building envelope and living habits of the residents may not be consistent among different sampling sites. Finally, introduction or removal of source materials/products may have an immediate impact on the C_g of SVOCs, as previously demonstrated (Harrad et al., 2008; Zhang et al., 2009). These factors may complicate analysis of seasonal influences.

For HVAC filter dust, the concentration of phthalates and organophosphates might be close to their equilibrium concentration in dust due to small particle size, large airflow rates through filters, and long sampling durations (over one month). We observed higher concentrations of compounds with very low vapor pressure, such as DEHP and DnOP, in summer, but similar trends were not detected for other compounds. For settled dust, the unknown indoor residence time may have not been sufficient for the dust phase SVOC to equilibrate with its gas phase. Therefore, sorption kinetics directly impacted SVOC concentrations in dust. Some studies found that dust mass deposited on household surfaces during winter was less than during summer, and with smaller particle sizes (Edwards et al., 1998; Butte and Heinzow, 2002). The seasonal difference in dust properties may allow equilibrium partitioning to occur at a faster rate in winter than in summer. In the current study, higher levels of TCPP, total organophosphates, BBzP, DEHP, DnOP, and total phthalates in settled dust were observed in winter than in summer, which likely resulted from a combination of faster sorption kinetics, higher K_{oa} value, and poorer ventilation in winter. In addition, for phthalates, significantly enhanced emission from local sources (i.e., vinyl flooring) due to use of space heaters could also be an important factor, as discussed previously.

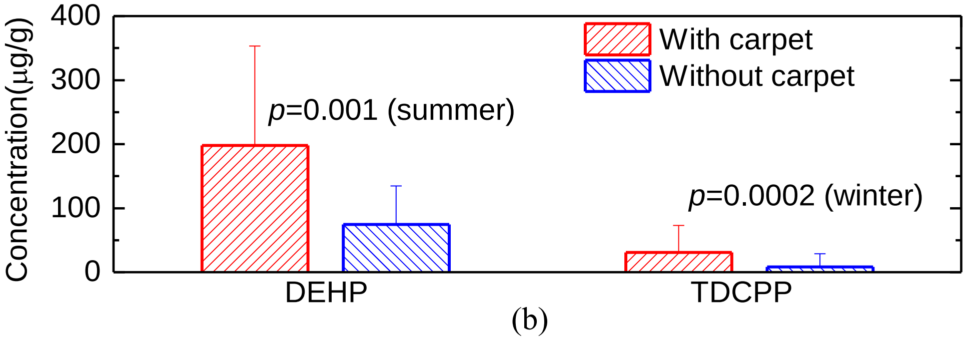
3.3.6 Influence of building characteristics

The association between the type of floor materials and concentration of target compounds in dust was investigated. Of all studied houses, 40% used PVC flooring, 30% had wall-to-wall carpet, and the rest had compressed wood, ceramic tile, or concrete flooring. As shown in Figure 14a, significantly higher median concentrations of BBzP and DEHP in settled dust were observed in homes with PVC flooring (BBzP: 39.8 and 13.6 µg/g in homes with and without PVC flooring, respectively, $p = 0.006$; DEHP: 189 and 127 µg/g in homes with and without PVC flooring, respectively, $p = 0.006$). After adjusting by season, statistical significance remained. The median concentrations of BBzP and DEHP in settled dust were observed higher in homes with PVC flooring in winter (BBzP: 75.3 and 6.09 µg/g in homes with and without PVC flooring, respectively, $p = 0.03$; DEHP: 553 and 187 µg/g in homes with and without PVC flooring, respectively, $p = 0.009$) and in summer (BBzP: 18.9 and 5.2 µg/g in homes with and without PVC flooring, respectively, $p = 0.009$; DEHP: 138 and 50.6 µg/g in homes with and without PVC flooring, respectively, $p = 0.007$). However, no such relationship between concentration of HVAC filter dust and floor type was found for any target compound. These results indicate that the content of BBzP and DEHP in settled dust in homes with PVC flooring may partially originate from the floor which the dust is in contact with. Recent studies (Bi et al., 2015; Sukiene et al., 2017) have observed direct transfer of DBiP and BBzP from vinyl flooring to dust on the floor surface, with an increased concentration greater than one order of magnitude relative to dust on other surfaces. Bornehag et al. (2005) also reported that concentrations of BBzP and DEHP in house dust were associated with the amount of PVC flooring in the house, which could further increase uptake of phthalates in infants (Carlstedt et al., 2013). Results in this study showing a correlation between the levels of BBzP and DEHP and use of PVC flooring in homes supports previous reports (Bornehag et al., 2005; Ait Bamai et al., 2014a; Bi et al., 2015; Sukiene et al., 2017).

Figure 14b shows that TDCPP levels in settled dust were significantly associated with the presence of carpets (median values of 14 and 2.1 µg/g in carpeted and uncarpeted homes, respectively, in winter; $p = 0.002$). When houses with vinyl flooring were excluded, significantly higher concentrations of DEHP were found in dust collected from carpeted homes (median values of 131 and 50.6 µg/g in carpeted and uncarpeted homes, respectively; in summer, $p = 0.001$). These results suggest that carpets may serve as a significant reservoir of phthalates and organophosphates, especially for those with relatively low vapor pressures. Carpets are one type



(a)



(b)

Figure 14. Phthalate and organophosphate concentrations in settled dust in relation to the presence of PVC flooring (a) and carpet (b). The *p*-values were calculated using the Mann–Whitney *U*-test and $p<0.05$ indicates a statically significant difference.

of fleecy indoor material where particles containing absorbed SVOCs could accumulate, due to high particle deposition velocity on rough surfaces (Abadie et al., 2001) and low efficiency of removing dust from carpet (Yiin et al., 2007; Causer et al., 2008). Additionally, carpet has a large capacity to adsorb gas-phase SVOCs, due to its large specific surface area. Abb et al. (2009) found that the concentration of total phthalates in house dust increased as carpet area increased in a study conducted in Germany. Several studies in the U.S. and Australia (Castorina et al., 2017; He et al., 2018) found significantly higher dust loadings of TCEP, TCPP, TPHP, and TDCPP in houses with the presence of carpets, although a similar association was not found in Japan (Araki et al., 2014; Tajima et al., 2014). In addition, carpets could also serve as a source of organophosphates due to their use as flame retardants in carpet padding and fibers (Yadav et al., 2018). However, a recent study (Ionas et al., 2015) reported that organophosphates detected in carpet were typically at low levels (less than 0.01% by weight in the material phase). Generally, it is difficult to precisely identify the sources of SVOCs in indoor environments due to the complexity of the environment, existence of multiple sources, and limited sample size. Although some associations between the type of floor materials and concentration of organophosphates and phthalates were found in this study, further research is needed to confirm such findings.

The results of other associated building characteristics are listed in Tables C.S6 and C.S7. The average (± SD) floor area and house age were 160 (± 55) m^2 and 18 (± 13) years, respectively. Higher concentrations of DnBP, BBzP, DEHP, TCPP, and TPhP in indoor dust in different seasons were found in smaller homes. Since lower income populations commonly have smaller homes (Adamkiewicz et al., 2011), they are likely exposed to elevated concentrations of phthalates and organophosphates due to smaller house size, elevated source strength, and possibly inadequate ventilation. Newer buildings were found to have higher concentrations of TCPP in both HVAC filter dust and settled dust in the summer. Such a finding may be attributed to the increasing use of organophosphates in consumer products (as replacements of PBDEs), but there was no correlation between the concentration of other organophosphates in dust and the age of homes. Significant relationships were found between dust concentrations of target SVOCs and participants' house-cleaning activities. For vacuuming of carpets, the concentration of TDCPP in settled dust was lower in the high-frequency vacuuming group than in the low-frequency vacuuming group. In contrast, interestingly, for activities of cleaning furniture and hard floors, the concentrations of TCPP, TDCPP, and total organophosphates in HVAC filter dust and BBzP and TPhP in settled dust were significantly higher in the high-frequency cleaning group. This finding suggests that furniture and floor cleaning activities may actually cause contaminated dust to resuspend and become airborne, thereby increasing residents' inhalation exposure to some SVOCs. However, frequent vacuuming of carpets may help to decrease dust concentrations of certain SVOCs and thus reduce exposure.

3.3.7 Relationship with childhood asthma

For HVAC filter dust samples collected in the summer, a statistically significant linear correlation was found between DEHP concentration and asthma severity in children who had physician-diagnosed asthma ($R^2 = 0.36$, $N = 37$, $p < 0.001$), indicating slightly elevated DEHP levels are present in homes of children with greater asthma severity compared to those with lower severity. However, there was no statistically significant difference in DEHP concentration in HVAC filter dust between the asthma and the control groups. Also, no associations were found for winter sampling events (perhaps due to short winter season in the central Texas and/or short HVAC runtime in winter), for other phthalates or organophosphates, or in settled dust. The results suggest that it is only the severity of asthma in children with physician-diagnosed asthma that is associated with DEHP concentration found in home HVAC filters in the summer.

Several previous studies examined the association between phthalates in dust or PVC materials in homes and allergic diseases in children. In most of these studies, non-floor settled dust was sampled. Among Swedish children, Bornehag et al. (2004) showed an association between higher concentrations of DEHP in house dust and the occurrence of asthma. This association was also observed with house dust concentrations of BBzP and the presence of allergic rhinitis and atopic dermatitis. Similar results were found in Bulgaria and Taiwan, where positive associations between house dust DEHP or BBzP concentrations and childhood asthma, rhinitis, and eczema were reported (Kolarik et al., 2008a; Hsu et al., 2012a). However, results were not always consistent. Two studies conducted in Denmark and Japan (Callesen et al., 2014; Kanazawa et al., 2010) did not find a significant association between phthalates in dust and allergies in children. In another cross-sectional study (Ait Bamai et al., 2014a), levels of phthalates in floor dust had a linear association with the prevalence of allergic rhinitis, conjunctivitis, and atopic dermatitis in children; however, this association was not found for non-floor dust. A recent study in Denmark (Beko et al., 2015) involving children with asthma, allergic rhinitis, or eczema did not show a direct association between the occurrence of allergic disease and exposure to phthalates. However, among children with allergic disease, there was an association between indoor allergen sensitization and exposure to BBzP, DnBP, and DEHP. In another controlled study (Deutschie et al., 2008), 16 healthy human subjects and 16 human allergic to dust mites were exposed to house dust containing low and high concentrations of DEHP. Healthy subjects were not affected by the dust exposure, but the allergic group showed silent inflammation in nasal mucosa in response to the dust containing low DEHP doses. To some extent, our current finding that only the severity of asthma in children with asthma disease correlates with DEHP concentration in HVAC filter dust in the summer supported the results of Beko et al. (2015) and Deutschie et al. (2008).

The mechanism responsible for the association between phthalate exposure and doctor-diagnosed asthma remains unclear. Some *in vitro* and *in vivo* animal studies showed that phthalates or their metabolites may have different modes of action (e.g., direct pharmacological effect on receptors involved in allergenic pathology or indirect effect as adjuvants for different causative agents) to influence immune and inflammatory parameters that play a role in respiratory diseases (Oie et al., 1997; Larsen et al., 2002, 2003; Larsen and Nielsen, 2008; Bornehag and Nanberg, 2010; Kimber and Dearman, 2010; Beko et al., 2015; Franken et al., 2017). Additionally, indoor microbes may also influence the presence of asthma and asthma severity. Studies assessing increased exposure to indoor fungi prior to the development of asthma symptoms suggested that certain fungal species pose a respiratory health risk in susceptible populations (Sharpe et al., 2015). *In vivo* studies revealed that viable fungi can induce asthma-like disease in experimental animals supports the argument that fungi are possible etiologic agents of asthma (Porter et al., 2011; Porter et al., 2011). Several other studies found that exposure to bacterial endotoxin and mold also affect asthma severity in children and additional respiratory outcomes in subjects with and without allergies (Michel et al., 1996; Zambelli-Weiner et al., 2004; Mendell et al., 2011; Dannemiller et al., 2016).

Chemical and biological contaminants concurrently present in indoor environments may interact with each other. Biodegradation of phthalates, particularly DEHP, by a variety of bacteria and fungi present in household dust, PVC materials, and soil have been reported in previous studies (Webb et al., 2000; Nakamiya et al., 2005; Nalli et al., 2006b, 2006a; Liang et al., 2008; Pradeep et al., 2013). In the current project, significant associations between the concentrations of fungi and bacteria and the levels of DEHP in summertime HVAC filter dust were observed (reported in details in Maestre et al., 2018). Therefore, it is possible that DEHP concentrations and concentrations of fungi and/or bacteria are intimately related. Warm summer air, which contains more moisture than winter air, may strengthen this effect. However, we did not collect enough data to determine if the HVAC filters themselves were a source of air contamination and thus exacerbated asthma symptoms in children. Furthermore, certain toxic volatile organic compounds could be produced during the process of microbial degradation of plasticizers. Previous studies reported that microbial degradation of DEHP and di-2-ethylhexyl adipate (DEHA) was the most likely source of 2-ethylhexanol in indoor air (Nalli et al., 2006b, 2006a), which has been associated with nasal symptoms (Wieslander et al., 1999) or asthma (Norbäck et al., 2000; Kanazawa et al., 2010) in building occupants. Future confirmatory studies are needed to examine whether the presence of phthalates alters the indoor microbial communities and whether the interaction between indoor chemicals and the environmental microbiome results in adverse health effects.

3.3.8 Implications for exposure assessments

Phthalates and organophosphates measured in HVAC filter dust could be considered to represent what mostly went into the airway via inhalation, while those measured in settled dust was assumed to be digested through oral intake of contaminated particles adhered to food and objects. We compared the estimated daily intake of target compounds through these two exposure pathways. The results and the equations and parameters used in the estimation are listed in Tables C.S8 and C.S9. Median values of inhalation rates, dust ingestion levels, and body weights were used for 3-year-old children and adults, based on the EPA exposure factor handbook (US EPA, 2011). We assumed the average time spent at home to be 64% (Harrad et al., 2006). For all target SVOC compounds, the intake through ingestion of settled dust was significantly higher than via inhalation of suspended particles, especially for children. However, the association between target compounds in dust and asthma severity in children was found only for HVAC filter dust, but not settled dust. The results suggest that the impact of an SVOC compound on a particular health endpoint is related not only to the amount of intake but also to the route of exposure. As air travels through the respiratory tract via inhalation, contaminated particles may deposit in the respiratory tract and release (i.e., desorb) a fraction of SVOCs which then interact with the tissue. The subsequent effect depends on the type of the compound, the size of particles, and the region of the respiratory tract (Liu et al., 2017). Oie et al. (1997) found that inhalation exposure to particles containing DEHP was important due to its low clearance and extensive penetration into the pulmonary region. They reported that high local concentrations of DEHP can be created in the airways at the particle deposition site and the primary hydrolysis product of DEHP increased the risk of inducing inflammation in the airways, which is a characteristic of asthma. Therefore, to evaluate the health effects in respiratory tract such as asthma, it may be more direct, representative, and meaningful to use airborne particles than settled floor dust.

HVAC filter dust may become a useful sampling medium to measure SVOC concentrations in airborne particles. HVAC filters can be installed with minimal effort for a particular period of time and capture particles from large volumes of air, resulting in the collection of spatially and temporally integrated concentrations. The large volume of air that passes through the filters simply increases the sensitivity of sampling and analysis. This approach overcomes the limitation of traditional air sampling methods that only provides a “snapshot” of compound concentrations, which can vary significantly with time and within a building. Although filters may affect SVOC distribution in gas-, dust-, and filter-phases in a short term, because of sorption of gaseous species on the HVAC filter or filter cake or desorption of SVOCs from particles collected on the filter or the filter itself, it is expected that any impact will be minimal after the sorption equilibrium is established. When combined with HVAC system characterization such as system run-times, volumetric flow rates, and mass of dust collected, HVAC filter dust sampling provides a novel filter forensics methodology to detect and quantify particulate SVOC concentrations in indoor air. In contrast, settled floor dust can be used to assess SVOC exposures via inadvertent ingestion of dust, especially for young children who spend considerable time on the floor and frequently put their hands and other objects in their mouths. However, our findings show that when using house settled dust, it is important to note where the samples were collected from, because the type and level of SVOCs in floor dust may be directly related to local materials used in the house and could be different at other indoor locations. Additionally, since the age of settled dust is usually unknown, for SVOCs with high K_{oa} values, the mass fraction in settled dust may not have sufficient time to equilibrate with the gas phase concentration and thus may be underrepresented in settled dust. Seasonal variations were observed for both HVAC filter dust and settled dust, resulting from the combined effect of several factors discussed previously. The concentrations of some phthalates and organophosphates in settled dust in winter were generally higher than in summer, but the seasonal pattern was not well-defined for HVAC filter dust. More integrated and consecutive studies involving seasonal variations of SVOCs in HVAC filter dust and settled dust should be conducted in the future.

Appendix A

Paper 1. Fate and Transport of Phthalates in Indoor Environments and the Influence of Temperature: A Case Study in a Test House[2]

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ABSTRACT

A case study in a test house was conducted to investigate the fate and transport of benzyl butyl phthalate (BBzP) and di-2-ethylhexyl phthalate (DEHP) in residential indoor environments and the influence of temperature. Total airborne concentrations of phthalates were sensitive to indoor temperatures, and their steady-state concentration levels increased by a factor of three with an increase in temperature from 21 to 30 ºC. Strong sorption of phthalates was observed on interior surfaces, including dust, dish plates, windows, mirrors, fabric cloth, and wood. Equilibrium partitioning coefficients for phthalates adsorbed to these surfaces were determined, and their values decreased with increasing temperature. For impervious surfaces, dimensionless partitioning coefficients were calculated and found to be comparable to reported values of the octanol-air partition coefficients of phthalates, K_{oa}, suggesting that an organic film may develop on these surfaces. In addition, sorption kinetics was studied experimentally, and the equilibration time scale for impervious surfaces was found to be faster than that of fabric cloth. Finally, using an indoor fate model to interpret the measurement results, there was good agreement between model predictions and the observed indoor air concentrations of BBzP in the test house.

INTRODUCTION

Phthalates have been widely used as plasticizers to enhance the flexibility of polyvinylchloride (PVC) products. These semi-volatile organic compounds (SVOCs) are found in a wide range of building materials and consumer products such as vinyl flooring, carpet padding,

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[2] Bi, C., Liang, Y., & Xu, Y. (2015). Fate and Transport of Phthalates in Indoor Environments and the Influence of Temperature: A Case Study in a Test House. *Environmental Science & Technology*, *49*(16), 9674–9681. The first author did all the experimental work and consequent data analysis with some suggestions from the second and third authors.

wall coverings, floor tiles, furniture, and electronics (Bornehag et al., 2005). Phthalates are often present in products at single-percent to tens-of-percent levels (Weschler and Nazaroff, 2008). For example, PVC flooring typically contains 10%–30% by mass phthalate plasticizers (Bornehag et al., 2005; Liang and Xu, 2014a, 2014b). Because phthalates are not chemically bound to the polymer matrix, these SVOCs usually are emitted from the products into the air and other media (Clausen and Hansen, 2004; Xu and Little, 2006; Xu et al., 2012; Liang and Xu, 2014b). Previous studies have shown that phthalates are ubiquitous and among the most abundant indoor contaminants (Rudel et al., 2003, 2010a; Blanchard et al., 2014b). Exposure to phthalates may cause irreversible changes in the development of the human reproductive system (Martino-Andrade and Chahoud, 2010; Witorsch and Thomas, 2010; Kay et al., 2013, 2014); elevate the risk of asthma and allergies (Kolarik et al., 2008b; Bornehag and Nanberg, 2010; Hsu et al., 2012a; Callesen et al., 2014a; Shu et al., 2014); increase the risk of obesity (Hatch et al., 2010); and result in adverse pregnancy outcomes such as preterm birth, low birth-weight, and pregnancy loss (Toft et al., 2012; Sathyanarayana et al., 2014).

Interior surfaces play an important role in the fate and transport of phthalates in indoor environments. Because phthalates partition strongly to surfaces such as particles (Benning et al., 2013; Liu et al., 2013), dust (Weschler and Nazaroff, 2010; Xu et al., 2014b, 2014a), windows (Diamond et al., 2000; Bennett et al., 2014), and skin (Gong et al., 2014), the sorption process may affect the dynamic behavior of phthalates and their indoor fate. Recent studies suggest that airborne particles can enhance emissions of SVOCs from source materials by reducing the mass transfer resistance at the air/material interface (Benning et al., 2013; Liu et al., 2013). Several studies have found that most impenetrable indoor surfaces contain a very thin organic film at their interface with room air (Diamond et al., 2000; Liu et al., 2003; Butt et al., 2004; Weschler and Nazaroff, 2008). Gas-phase phthalates in indoor air can sorb to these surfaces by partitioning to the film layer. Therefore, these surfaces may affect surface-air exchange of phthalates and their residence time in indoor environments. For porous material surfaces such as gypsum wallboard, carpet, clothing, and furniture foam, phthalates can also diffuse into the material and sorb there (Petrick et al., 2010; Liang and Xu, 2015). Over time, such processes may establish significant reservoirs that may act as secondary sources, thereby extending the long-term persistence of phthalates in indoor environments. Furthermore, phthalates sorbed to interior surfaces may represent an important source of potential contact and exposure through dermal absorption and non-dietary ingestion pathways (Weschler and Nazaroff, 2008), particularly for young children who may frequently contact those surfaces and have enhanced hand-to-mouth activities. Therefore, it is important to determine the extent to which phthalates sorb to different indoor surfaces. Although the sorption of volatile organic compounds (VOCs) to building materials and other common indoor materials has been well studied (Won et al., 2000, 2001), few studies have investigated the sorption of phthalates (Clausen and Hansen, 2004; Xu et al., 2012; Liang and Xu, 2014a, 2015).

Temperature variations inside buildings are very common (Table A3) and are mainly associated with outdoor temperature changes (Coley and Kershaw, 2010; Mavrogianni et al., 2012; Nazaroff, 2013; Nguyen et al., 2014). Temperature may have a strong influence on the fate of phthalates in indoor environments because phthalate emission and sorption properties depend strongly on temperature. Clausen et al. (2012) observed an order of magnitude increase in di-2-ethylhexyl phthalate (DEHP) emissions in experimental chambers when the temperature was increased by 10 ºC. Similar results were obtained by Liang and Xu (2014a) when measuring the emission rate of phthalates from a range of vinyl flooring and crib mattress covers at different temperatures. In contrast, with increasing temperature, decreasing surface/air partition coefficients were observed for SVOCs (i.e., polychlorinated biphenyls [PCBs], polycyclic aromatic hydrocarbons[PAHs], and DEHP) between air and surfaces such as particles (Naumova et al., 2002, 2003; Arp et al., 2008), plant leaves (Kömp and McLachlan, 1997), and stainless steel (Clausen et al., 2012).

Field measurements to investigate the influence of temperature on the fate of indoor phthalates are limited. Several studies reported no association between room temperature and phthalate concentrations in indoor air (Fromme et al., 2004; Bergh et al., 2011a, 2011b), while another study found significant correlations for higher vapor pressure phthalates (i.e., diethyl phthalate [DEP], di-n-butyl phthalate [DBP], and di-isobutyl phthalate [DiBP]) (Gaspar et al., 2014). Although no studies have investigated seasonal variations in indoor phthalates, higher indoor concentrations in the summer than in the winter have been noted for some SVOCs, including PCBs (Balfanz et al., 1993; Kohler et al., 2005) and polybrominated diphenyl ethers (PBDEs) (Hazrati and Harrad, 2006). However, all the above field studies were not designed to investigate the influence of temperature in a systematic way. In those studies, indoor air and dust samples were typically collected from a number of sampling sites (e.g., public buildings and private houses), which may have significant differences from site to site in pollutant sources, sink surfaces, and other environmental conditions. As a result, it is difficult to clarify the actual influence of temperature on the emission and distribution of indoor SVOCs.

Any strategies to limit exposure to phthalates require an understanding of their sources and fate in the environment. Thus, the goal of this research was to conduct a case study in a test house to investigate the fate and transport of phthalates in residential indoor environments and the influence of temperature. The specific objectives were to: (1) measure the dynamic changes in indoor air concentrations of phthalates with changes in indoor temperatures; (2) determine the equilibrium concentrations of phthalates at different temperatures on interior surfaces, including dust, windows, dish plates, mirrors, fabric cloth, and wood; (3) investigate the sorption kinetics of phthalates for some of these surfaces; and (4) interpret the measurement data using a previous developed multimedia indoor fate model for phthalates.

MATERIALS AND METHODS

Test house. The test house (Figure A4) is a three-bedroom, two-bathroom 110 m^2 manufactured home located at the University of Texas at Austin. The house has two independent heating ventilation, and air conditioning systems and is fully instrumented in and around the house, including CO_2 tracer gas systems for air exchange measurements, weather stations, and several particle, temperature, relative humidity (RH), and air velocity measurement stations. Furniture and appliances used in typical residential homes were also installed in the test house (Figure A5). This provided an intermediate environment between full-scale chambers and actual occupied houses while removing interferences from human activity and allowing field measurements under controlled conditions. The test house was built in 2008 and the floor of the house was completely covered with vinyl flooring, which was found to contain approximately 10% (w/w) of benzyl butyl phthalate (BBzP).

Field measurements. Field measurements were conducted in the living room and two bedrooms of the test house. The temperature of the house was strictly controlled at 21±0.8 ºC for about six months and then increased to 30±0.5 ºC. This temperature range was selected to represent typical temperature fluctuations of buildings (Table A3). After three months, the temperature was dropped back to 21 ºC to allow comparison with previous results at the same temperature and to examine the reproducibility of the data. Finally, the temperature was increased to 25±1.0 ºC to further verify the sensitivity of phthalate levels to temperature. At each experimental stage, total airborne concentrations of phthalates were monitored periodically. Once steady state conditions at 21 and 30 ºC were achieved (i.e., change of indoor air concentrations less than 20% for three consecutive data points) concentrations of sorbed phthalates were measured on different surfaces, including dust, windows, stoneware dish plates, mirrors, fabric cloth, and wood. In addition, at 21 ºC, the kinetic aspect of phthalate sorption was investigated at selected, pre-cleaned surface locations by measuring the accumulation of phthalates on these surfaces. Finally, temperature, RH, air change rate, and total suspended particle (TSP) concentrations were monitored periodically for the entire study period. Test house conditions are listed in Table A1.

Table A1. Test conditions and model parameters.

Parameter	21°C	25°C	30°C
Relative humidity (%)	30.4±13.8	40.4±4.0	24.9±5.9
Total suspended particle concentration ($\mu g/m^3$)	4.0±3.8		
Air exchange rate (hr^{-1})	0.50±0.06	-[e]	0.71±0.10
Area of flooring material (m^2)	77		
Area of wall, ceiling, and furniture (m^2)	220		
Area of windows (m^2)	7		
Volume of the test house (m^3)	250		
Surface/gas partition coefficient for BBzP (K_{surf}, m) K_{surf} for wall, ceiling, and furniture	100 [a]		
K_{surf} for windows	123	- [e]	83.6
Mass transfer coefficient (h_m, m/hr)	0.106 [b]		
y_0 for BBzP in the flooring material [c] ($\mu g/m^3$)	2.7	5.9	12.4
Particle/gas partition coefficient for BBzP [d] (K_p, m^3/mg)	52.7	32.5	18.1
Particle/gas partition coefficient for DEHP [d] (K_p, m^3/mg)	350	207	110

f. Directly read from Figure S6 in Ref. (Xu and Hubal, 2009). We ignored the effect of temperature, because this parameter does not have a significant influence on model results.

g. Calculated using empirical correlations in Ref. (Axley, 1991), which are based on Reynolds and Schmidt numbers. We measured near-surface air velocity in the test house (0.01 m/s) and used this value to obtain the Reynolds number, and finally, the mass transfer coefficient. Similar low velocity was also reported in other field studies.(Huang et al., 2004)

h. Measured in the current study using a specially designed chamber.

i. Calculated using Equation 15 in Ref. (Naumova et al., 2003) with the vapour pressure estimated using Equation 9 in Ref. (Gobble et al., 2014).

j. Values were not measured in this study and were assumed to be the same as those at 21 °C in the model.

Air sampling. To collect air samples, a low volume pump (A.P. Buck Manufacturing Inc.) was connected with a container holding polyurethane foam (PUF) (22 × 100 mm, 1-section, 76 mm sorbent) and a glass fiber filter. Prior to use, the PUF and container were ultrasonically cleaned three times for 30 min each using hexane, after which they were dried, wrapped in aluminum foil, and sealed in ziplock bags. Duplicate air samples were collected at a flow rate of 3 $L \cdot min^{-1}$ for 48 hours with a field blank at each sampling location. After sampling, the sample tubes were stored in a protective casing wrapped in aluminum foil at a temperature below 4 °C, and analyzed within 1 hour of sampling. Before analysis, the PUFs were spiked with three surrogate chemicals (i.e., dibenzyl phthalate, diphenyl isophthalate, and diphenyl phthalate) to determine analytical recovery efficiencies. The average recovery for the surrogates in all samples was 85.3%± 7.3%.

Dust sampling. Dust samples from floors (a source of BBzP) and other non-source surfaces, such as molding and shelves, were collected separately to examine potential differences in phthalate concentrations. The settled dust (about 0.1 g) was collected onto cellulose thimble filters (26 × 60 mm, Whatman Inc.) using a special aluminum nozzle holder connected to a vacuum cleaner (Eureka model 71B) to avoid contamination due to contact between dust and plastic parts of the vacuum cleaner. Prior to use, the filters were ultrasonically cleaned with hexane, after which they were dried in an oven for 1 h. The filter was weighed before and after sampling to obtain the mass of settled dust collected. In the sorption kinetics study for dust, dust collected from a residential home was evenly sprayed on an aluminum foil surface with a layer thickness of approximate 1 mm. Dust samples were periodically collected from the layer with a brush to monitor the accumulation of phthalates from air to the surface dust. After sampling, dust samples were placed in a beaker covered with aluminum foil, sealed in a glass jar with a polytetrafluoroethylene (PTFE) lid, stored below 4 °C, and then analyzed within 1 hour of sampling. Similar to the air samples, spiked surrogate chemicals were applied before analysis. The average recovery for the surrogates in all dust samples was 91.3% ± 4.4%.

Surface sampling. Surface-wipe samples were collected to determine the sorption of phthalates to hard surfaces. All surfaces were selected or placed in vertical positions to avoid significant particle deposition (Figure A5). Gauze pads (10 × 10 cm), which were pre-cleaned by extracting with hexane, air-dried, and stored in glass jars, were saturated with 10 mL of hexane and wiped twice in opposite directions across the designated area of surfaces (i.e., windows and mirrors: 0.2 m^2; plates: 0.05 m^2; and wood: 0.14 m^2). This process was repeated two times with new gauze pads used each time, resulting in three consecutive wipe samples collected at each location. Field blank preparation was identical to sample preparation, except that the field blank gauze pads did not contact hard surfaces. All sampled gauze pads were stored below 4 °C and analyzed within 30 min of sampling. Preliminary tests comparing wipe times (three, five, and ten consecutive wipes) as well as solvents (hexane and dichloromethane) showed no difference in removal efficiency. The recovery efficiency of the method for wiping the surfaces was also tested. A volume of 10 µL of methanol containing 2 µg of DBP, BBzP, and DEHP was applied onto a pre-cleaned glass surface (30 × 30 cm) in small droplets. After allowing the surface to completely dry, the surface was wiped using the method described above. The recovery check was conducted three times, and the mean recovery ratios averaged 87.9%± 8.6% for all of the phthalate compounds.

Sorption of phthalates to soft surfaces (i.e., fabric cloth) was examined. Two materials that are commonly encountered in indoor furnishings or clothing but with different surface characteristics were selected: cotton and polyester. Both fabrics, which were white and purchased at a local store, were cut into 15 × 15 cm pieces. The cloth pieces were then ultrasonically pre-cleaned three times for 30 min each with hexane, completely dried, and hung in a bedroom wardrobe. Fabric samples were periodically collected and placed in a beaker covered with aluminum foil, sealed in a glass jar with a PTFE lid, and stored below 4 °C prior to extraction and analysis.

Sample extraction and chemical analysis. The collected air, dust and surface samples were ultrasonically extracted three times for 30 min each with hexane. The volume of the extract was concentrated to approximately 100 µL, following rotary evaporation, cleanup, and nitrogen purge, except that the dust samples were concentrated to 500 µL due to a higher chemical loading. The extract was then analyzed for dimethyl phthalate (DMP), DEP, DBP, di-n-octyl phthalate (DnOP), BBzP, and DEHP using a GC-FID system (Agilent 7890A) equipped with a DB-5ht column (30 m, 0.25 mm, 0.1 µm). However, the former four phthalates were not detected in any samples. The GC-FID system was operated using a 4:1 split injection at 275°C. The temperature program was set at 120°C and held for 2 min, ramped up 12°C/min for 15 min and held 3 min, and then ramped up 20°C/min for 2 min and held 2 min. The detector was set at 320°C. Helium was used as the carrier gas with a constant column flow set at 1.2 ml/min. A calibration standard was run prior to each GC injection, and the variance was always below 10% for all injections. In addition, because laboratory equipment such as glassware and solvents may be contaminated with phthalates, we pre-cleaned all glassware and rigorously collected and analyzed solvents, extracts, and blanks.

Indoor fate model of phthalates. Xu et al.(Xu and Hubal, 2009; Xu et al., 2010) extended a chamber-based SVOC emission model (Xu and Little, 2006) to predict human exposures to phthalates emitted from vinyl flooring in a realistic residential environment. This model was employed in the present study to investigate the fate of indoor phthalates. In Xu et al.'s model, the most important emission parameter is y_0, the gas-phase concentration of phthalates in equilibrium with the material phase. Using a specially designed experimental chamber, Liang and Xu (Liang and Xu, 2014b, 2014a) recently developed a novel, rapid method to measure y_0 for a range of phthalate compounds released from building materials. Similar emission chamber tests were conducted in the current study for the vinyl flooring, which completely covered the floor of the test house and contained approximately 10% (w/w) of BBzP. The obtained y_0 values at 21, 25, and 30 °C allowed us to use Xu et al.'s model to interpret the field measurement results of BBzP and understand the fundamental mechanisms governing the fate and transport of phthalates in indoor environments.

RESULTS AND DISCUSSIONS

Indoor air. Figure A1 shows the total airborne concentrations of BBzP and DEHP at different temperatures. At 21 °C, we assumed that steady state had been reached because the test house had been conditioned at this temperature for about half a year before measurements started and the change of measured air concentrations were within 20%. The concentration fluctuation (Figure A1) may be related to changes in humidity and infiltration rate due to changing outdoor weather conditions, unexpected human activities in the test house, and uncertainties associated with sampling and analysis. The mean steady-state concentrations at 21 °C were 149 and 110 ng/m^3 for BBzP and DEHP, respectively. When the temperature was increased to 30 °C, about four weeks were required for the indoor air concentration of phthalates to reach steady state. The steady-state concentrations at 30 °C were 419 and 300 ng/m^3 for BBzP and DEHP, respectively, which were about three times higher than the steady-state levels at 21°C. The measured indoor air concentrations of phthalates in the test house are comparable to those measured in previous field studies in residential homes (Wilson et al., 2001; Fromme et al., 2004; Kanazawa et al., 2010; Rudel et al., 2010a; Bergh et al., 2011b; Xu et al., 2014a), which were in a range of 20–50 ng/m^3 for BBzP and 120–600 ng/m^3 for DEHP (although most of the studies did not provide temperature information at sampling sites). The measured BBzP concentrations in the test house were higher than levels reported in these studies, possibly because the test house was completely covered with vinyl flooring, which is a significant source of BBzP.

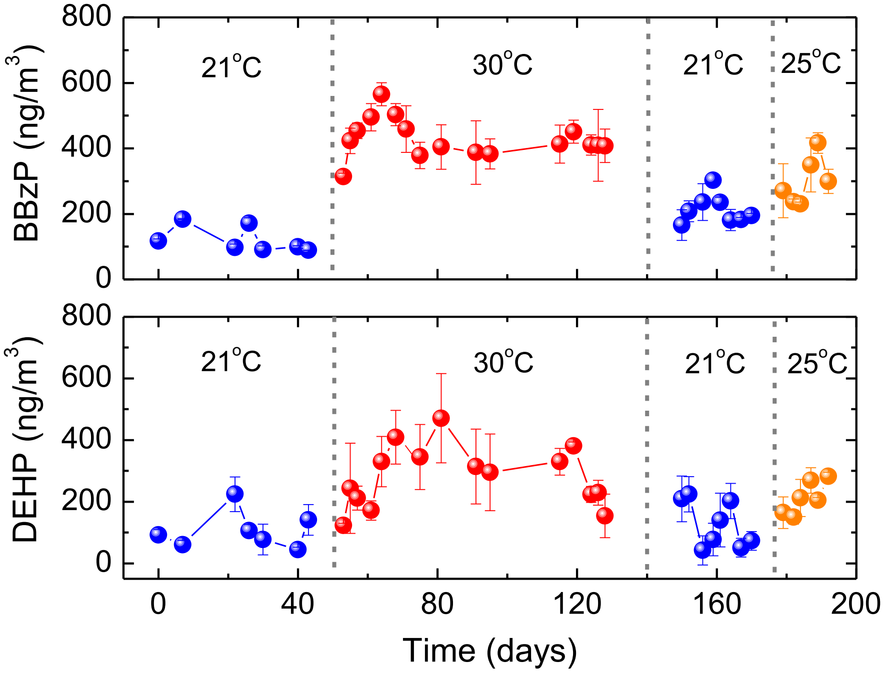
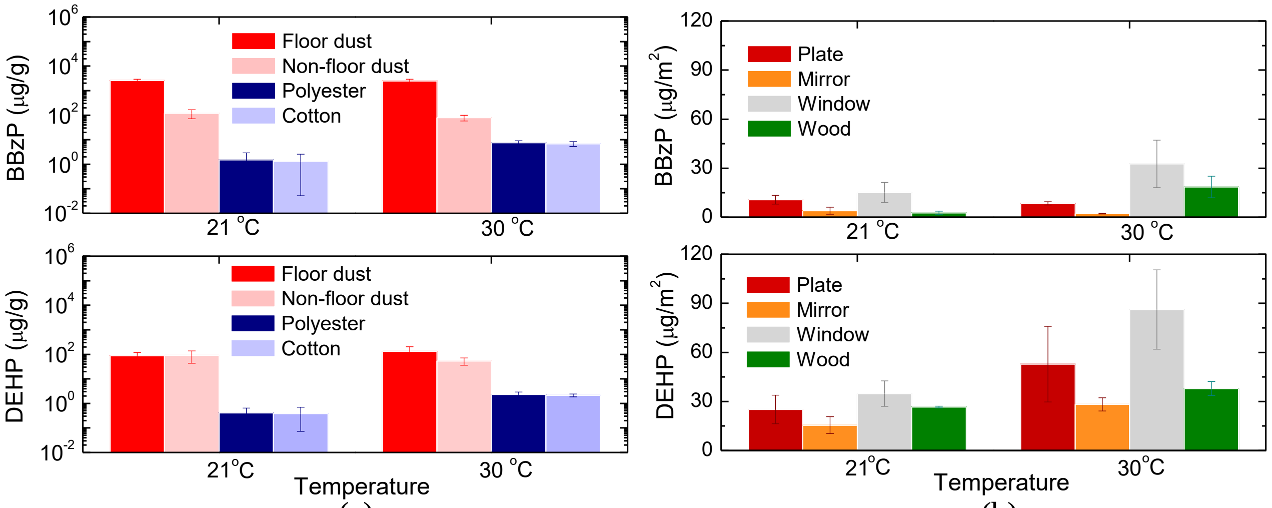


Figure A1. Total airborne concentration of BBzP and DEHP in the test house.

(Note: dashed lines indicate the time at which temperature was changed)

After the temperature was dropped back to 21 ºC, the concentrations of phthalates reduced significantly. The results generally verified the reproducibility of the data, although the levels at 21 ºC were slightly higher than previous data. This is possibly due to the "baking effect" (i.e., enhanced emission and desorption) at 30 ºC for about three months, and thus, additional time may be needed for the equilibrium of surface partitioning to be re-established. Finally, when the temperature was increased to 25 ºC, the concentrations of BBzP and DEHP both increased rapidly and to levels between the steady-state concentrations at 21 and 30 ºC. These results further verified the sensitivity of indoor phthalate levels to temperature. Considering that climate change may result in increased indoor temperatures as outdoor temperatures increase and heat waves become more frequent, indoor air concentrations of phthalates and human inhalation exposure to the contaminants could significantly increase as global warming becomes more serious.

Interior surfaces. *Dust.* Figure A2a shows the concentration of phthalates in non-floor dust (i.e., dust collected from molding, ceilings, and walls) and floor dust (i.e., dust accumulated on vinyl flooring) at 21 and 30 ºC. At each temperature, the concentration of BBzP in floor dust was about 20–30 times higher than in non-floor dust, while little difference was observed for DEHP. The results indicated that dust captured a great amount of BBzP from vinyl flooring through direct contact with this BBzP source. The results also suggested that the sampling location of settled dust should be carefully selected in future indoor field studies, and building occupants may be exposed to a significantly high level of phthalates via ingestion of dust that deposited on source surfaces. At 21 ºC, the measured mean (± s.d.) concentrations in floor dust were 2560±360 and 88±30 μg/g for BBzP and DEHP, respectively, and in non-floor dust were 119±48 and 91±47 μg/g for BBzP and DEHP, respectively. Temperature did not significantly influence phthalate mass fractions in the dust. One possible reason might be the offsetting effect of increased gas-phase concentration and decreased dust/gas sorption partitioning with increasing temperature. Generally, the results are comparable to previous field studies (Oie et al., 1997; Butte et al., 2001; Wilson et al., 2001; Rudel et al., 2003; Santillo et al., 2003; Bornehag et al., 2005; Langer et al., 2010; Bergh et al., 2011a), where mean concentrations in settled dust were reported in a range of 5 to 135 µg/g for BBzP and 200 to 1600 µg/g for DEHP. The slightly lower concentrations of DEHP in the current study might be due to the limited sources of DEHP present in the test house, where not as many consumer products were used as in real residential homes.



(a) (b)

Figure A2. BBzP and DEHP concentrations in (a) dust and fabric cloth and on (b) plate, mirror, window, and wood.

We assumed that sorption equilibrium had been reached at 21 ºC, considering that the settled dust had been accumulating for six years in the test house, which was conditioned for half a year at 21 ºC before measurements started. The equilibrium coefficients (K_{dust}, m^3/g) that describe the partitioning between non-floor dust and gas phase were determined to be: 962 and 1970 m^3/g for BBzP and DEHP, respectively (Table A2). Weschler and Nazaroff (2010) proposed a method to estimate K_{dust} using the octanol-air partition coefficient of a chemical (K_{oa}). Because there are great deviations for the K_{oa} values of BBzP and DEHP reported in the literature (Table A5), we estimated the possible values of K_{dust} using the available ranges of K_{oa} and compared these values with our measurement results. The measured K_{dust} of BBzP (962 m^3/g) falls into the estimated range of $10^{2.2}$–$10^{4.6}$ m^3/g, while the K_{dust} of DEHP was an order of magnitude lower than the estimated range of 10^4–10^6 m^3/g. One possible reason might be the significantly low organic content in the dust (e.g., skin flakes and organic particles generated during cooking) that resulted in weak dust/gas partitioning, considering that the test house was not occupied. In addition, the values of K_{dust} decreased greatly with increasing temperature (Table A2). However, caution should be taken when comparing results because it was possible that the dust/gas partitioning had not reached steady state within the three months at 30 ºC, considering the slow rate for SVOCs to approach equilibrium partitioning (Weschler and Nazaroff, 2010).

Interestingly, for floor dust, we found that a simple linear partitioning relationship was sufficient to explain its high BBzP level. Multiplying the partition coefficient (K_{dust}, Table A2) with the gas-phase concentration of BBzP in equilibrium with vinyl flooring (y_0, Table A1) (e.g., 962 m^3/g ×2.7 μg/m^3=2597 μg/g at 21 ºC), the difference between the calculated and measured BBzP concentrations in floor dust (e.g., 2560 μg/g at 21 ºC) was less than 10% at each temperature. The results suggested that the high BBzP concentration in floor dust are possibly due to the partitioning between dust and the gas layer adjacent to the flooring rather than direct solid-to-solid mass transfer. Although further verification is needed, this finding allows us to estimate phthalate concentrations in settled dust accumulated above source surfaces by only rapidly measuring y_0 for the source (Liang and Xu, 2014b).

Table A2. Measured indoor air concentration and surface/gas partition coefficients.

		BBzP		DEHP	
Indoor air		21 ºC	30 ºC	21 ºC	30 ºC
Steady-state airborne concentration (ng/m^3)		149	419	110	300
Calculated gas-phase concentration [a] (ng/m^3)		123	391	46.0	209
Surfaces					
Mirror	K_{surf} (m)	33.5	5.78	288	135
	Log K_{film} [b]	10.2	9.46	11.2	10.8
Plate	K_{surf} (m)	86.9	21.8	582	253
	Log K_{film} [b]	10.6	10.0	11.5	11.1
Window [c]	K_{surf} (m)	123	83.6	758	413
	Log K_{film} [b]	10.8	10.6	11.6	11.3
Dust	K_{dust} [d] (m^3/g)	962	202	1970	258

d. The measured concentrations in indoor air are actually the total airborne concentrations; that is, the sum of the gas-phase (C_g) and particle-phase (F) concentrations, C_g+F. Based on the definition of the particle/gas partition coefficient [K_p= (F/TSP)/C_g], the gas-phase concentration can be calculated using the equation C_g=

$(C_g+F)/(1+K_pTSP)$. Because C_g+F and TSP were measured and the values of K_p were estimated in Table A1, we calculated the gas-phase phthalate concentration (C_g) and used it to further obtain the surface/gas partition coefficients (K_{surf}).

e. Assuming the thickness of organic film is 10 nm and the film contains 20% organics, we calculated the dimensionless film/gas partition coefficients using equation 3.12 in Ref. (Weschler and Nazaroff, 2008).

f. The temperature of windows at 30 °C was actually less than 30 °C because of cool outdoor conditions.

g. Non-floor settled dust. Caution should be taken when comparing results, because the dust/gas partitioning possibly had not reached equilibrium at 30 °C.

Plate, mirror, window, and wood. Because the time scale to achieve equilibrium sorption on impervious surfaces was fast (i.e., hours and days; see next section), we assumed that sorption equilibrium had been reached when measuring surface concentrations. Figure A2b shows the equilibrium concentrations of phthalates on impervious surfaces, including plate, mirror, and window. Generally, the concentration of DEHP on these surfaces was higher than that of BBzP because DEHP's lower vapor pressure resulted in stronger surface sorption (Xu and Hubal, 2009). We calculated the equilibrium surface/gas partition coefficient (K_{surf}) at different temperatures in Table A2. K_{surf} decreased by about a factor of five for BBzP and a factor of two for DEHP when temperature increased from 21 to 30 °C. The K_{surf} values for windows did not reduce as much as other surfaces because its surface temperature was actually lower than 30 °C due to the low outdoor ambient temperature (about 0–20 °C). In addition, the effect of temperature on the surface concentration of phthalates was not consistent (Figure A2b). For example, with an increasing temperature, BBzP concentration on the plate decreased, while DEHP concentration increased. The change of surface concentration with temperature mainly depended on competition between the extent of the K_{surf} decrease and the increase in gas-phase concentration. Finally, the surface concentration on wood increased with increasing temperature. However, because phthalates may diffuse into the porous material, the method used (surface wipe) may not be sufficient to fully characterize wood's sorption property.

Organic films have been found to develop rapidly on impervious indoor surfaces through partitioning of gas-phase compounds from air to the film and enhanced dry deposition of airborne particles (Liu et al., 2003; Butt et al., 2004). Such films typically have a thickness ranging from 5–20 nm and organic content from 15%–30% (Liu et al., 2003), grow at a rate of 0.11–0.31 nm/day

(Wu et al., 2008), and are conceptualized as a combination of accumulated organic constituents, sorbed water, inorganic molecules and ions, and deposited particles (Diamond et al., 2000; Weschler and Nazaroff, 2008). In the current study, the windows of the test house had not been cleaned since it was built, and we observed a “greasy” coating on the surface. This may explain the higher surface concentration of phthalates on window glass than on plate and mirror (Figure A2b). Assuming such films exist on all impervious surfaces measured in this study with a thickness of 10 nm and organic content of 20%, we converted the measured equilibrium surface/gas partition coefficients (K_{surf}) to dimensionless film-to-gas partitioning ratios (K_{film}) (Table A2). Interestingly, the values of Log K_{film} are comparable to those of Log K_{oa} reported in the literature (Table A5), which further verified the SVOC equilibrium model between air and indoor surfaces (Weschler and Nazaroff, 2008). The results also indicated that the gas-phase phthalate concentrations in indoor air may be used to estimate their composition on impervious indoor surfaces and vice versa.

Fabric cloth. With temperature increasing from 21 to 30 °C, the concentrations of phthalates in both cotton and polyester increased by approximately three times (Figure A2a), mainly due to the increase in their gas-phase concentration in indoor air. When the concentration in cloth was divided by the gas-phase concentration and then multiplied by the density of fabric cloth (~0.18 g/cm^3), we obtained the dimensionless partition coefficient, K_{cloth} (Table A6), which were comparable to those measured in a recent study of airborne phthalate partitioning to cotton clothing (Morrison et al., 2015). The values of K_{cloth} were orders of magnitude lower than those of K_{flim} on impervious surfaces (Table A2). However, because the density of fabric cloth was calculated by dividing the mass of the fabric piece by its volume, it may not represent the actual fiber density. Therefore, it is not possible to conclude that phthalates either preferably sorbed to the fabric material or to the smooth impervious surfaces. We did not observe any phthalate sorption differences between cotton and polyester, possibly due to the relatively low gas-phase concentration of phthalates in the test house compared with other chamber studies of SVOCs sorption that typically involved high gas-phase concentrations (Piade et al., 1999; Petrick et al., 2010). In addition, we observed higher BBzP concentrations than those of DEHP at both temperatures and in both fabric cloths. We believed this might be related to the kinetic aspect of phthalate partitioning to fabric cloth. The time scale required for sorption equilibrium to be reached in cloth could be significantly longer for DEHP than for BBzP due to its larger K_{oa} value (Weschler and Nazaroff, 2010). Other possible reasons might be associated with chemical polarity (Buchert et al., 2001; Cao, 2010; Laing and Kean, 2011), RH variations in the test house (Piade et al., 1999), and fabric structure (Chien et al., 2011).

Sorption kinetics. When indoor air concentrations of phthalates reach steady state, phthalate levels on interior surfaces will evolve over time and finally achieve sorption equilibrium. The preceding measurements of surface concentration were based on the assumption of equilibrium conditions. In this section, we experimentally assessed the time scale required for sorption equilibrium to be reached between interior surfaces and gas-phase phthalates at 21 ºC.

Figure A6b shows that equilibration time scale can be fast (i.e., less than 24 hours) for plate and mirror surfaces. The steady-state concentrations of BBzP and DEHP (Figure A6) were similar to those in the measurements of surface concentrations (Figure A2b), indicating that the assumption that sorption equilibrium had been reached was reasonable. The variation of surface concentration at steady state might be related to the fluctuation of gas-phase concentration in the test house. Weschler and Nazaroff (2008) proposed a method to estimate the equilibrium time for SVOC sorption reservoirs modeled as organic thin films. Based on their method and the K_{oa} values of phthalates (Cousins and Mackay, 2000), the estimated time scales (e.g., ~ 12 hours for BBzP and ~ 20 days for DEHP assuming a film thickness of 10 nm, and ~ 1 hour for BBzP and ~ 2 days for DEHP assuming a film thickness of 1 nm) were comparable to our observations. Considering clean dishware and cups are typically air dried, the fast sorption from air to those surfaces may result in meaningful daily exposure to phthalates when eating and drinking, especially for compounds with high K_{surf}.

In contrast to the impervious surfaces, Figure A6a shows that sorption equilibrium was not reached in fabric cloth after about 20 days for BBzP and DEHP. However, given sufficient time, cloths, furnishings, and textile toys exposed to indoor air could accumulate a great amount of phthalates (Figure A2a), resulting in dermal exposure when touching the surfaces or wearing clothes. Previous research has shown that skin oil could even significantly enhance the sorption (Morrison et al., 2014). For children, phthalate sorption may pose a serious ingestion risk because children frequently put objects in their mouths. Dust concentration of phthalates (Figure A6b) was lower than those in pervious measurements (Figure A2a) because the dust used for the kinetic experiments was pre-cleaned and may have lost some of its organic components, thereby influencing its sorption properties.

Indoor fate model of phthalates. Because the flooring material was the major source of BBzP in the test house, we measured its emissions using a specially designed experimental chamber (Liang and Xu, 2014b). The obtained gas-phase concentration of BBzP in equilibrium with the material phase (y_0) was 2.7, 5.9, and 12.4 µg/m^3 at 21, 25, and 30 ºC, respectively (Figure A3). The values of y_0 allowed us to use a fate and transport model (Xu and Hubal, 2009; Xu et al., 2010) to interpret the field measurement results of BBzP. Other model parameters used are summarized in Table A1. Figure A3 shows good agreement between model predictions and observed air concentrations of BBzP in the test house at different temperatures. The difference between predictions and measurements was possibly due to the change in infiltration rate in the test house or the fluctuation of TSP concentration (Liu et al., 2015). During the second experimental stage when the indoor temperature was maintained at 30°C, we observed significant daily fluctuations in ambient air temperature (0–20 °C), which may have caused great variations in the amount of outdoor air penetrating into the house, although we were unable to monitor the infiltration rate in real time. However, the airborne concentration of BBzP was very sensitive to the infiltration rate, as shown in Figure A3 (the two dashed lines indicate an infiltration rate of 0.8 and 0.6 /h, respectively). Another possible reason might be the ignorance of some indoor sink reservoirs. For example, there are various interior surfaces that can serve as sorption surfaces, but only major surfaces (i.e., walls, ceilings, furniture, and windows) were accounted in the model. Considering that the K_{oa} value of BBzP is relatively small compared with other large molecular weight phthalates such as DEHP, ignorance of some sorption surfaces may not have great influences on model predictions. Although there were other limitations on use of the model (e.g., only the major source was considered and indoor air was assumed well mixed), the results suggested that data obtained from chamber studies on source emissions can be directly applied to the model and provide a reasonably well interpretation of the fate and transport of phthalates in actual indoor environments. With knowledge of their indoor air and surface concentrations, this approach can help health professionals estimate screening-level indoor exposures to phthalates as well as other similar SVOC compounds of concern and develop effective strategies to limit the exposures.

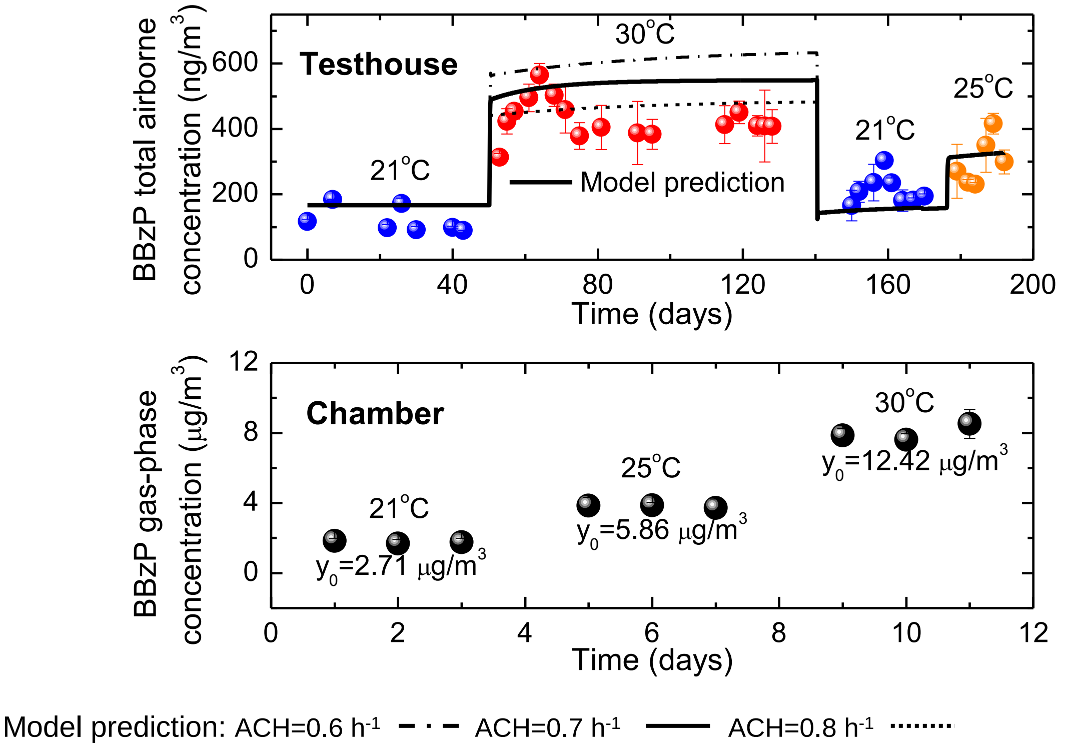


Figure A3. Total airborne concentrations of BBzP in the test house and gas-phase chamber concentrations of BBzP emitted from the vinyl flooring sample used in the test house. (Note: there were no airborne particles in the chamber.)

ACKNOWLEDGEMENTS

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SUPPORTING INFORMATION

Table A3. Typical indoor temperature range

Country	Reference	Number of buildings	Building type	Temperature range (ºC)
Korea	[1]	713	Multi-use facilities	21.8-24.6 (season)
Japan	[1]	213	Multi-use facilities	20.1-26 (season)
U.S.	[1]	807	Multi-use facilities	20.9-23.2 (season)
Turkey	[2]	119	Education	22-30 (year)
Nigeria	[3]	528	Residence	29-33 (day)
U.S.	[4]	356	Residence	18.2-27.2 (year)
U.K.	[5]	909	Office building	16-33 (season)
India	[6]	200	Residence	28-32 (season)

Table A4. Measured indoor air concentration and surface/gas partition coefficients[a].

		BBzP		DEHP	
Indoor air		21 °C	30 °C	21 °C	30 °C
Steady-state airborne concentration (ng/m^3)		149±63.7	419±64.8	110±77.1	300±116
Calculated gas-phase concentration [b] (ng/m^3)		123±52.6	391±60.5	46.0±32.1	209±80.9
Surfaces					
Mirror	K_{surf} (m)	33.5±22.0	5.78±1.04	288±227	135±55.9
	Log K_{film} [c]	10.2	9.46	11.2	10.8
Plate	K_{surf} (m)	86.9±42.9	21.8±4.24	582±433	253±148
	Log K_{film} [c]	10.6	10.0	11.5	11.1
Window	K_{surf} (m)	123±73.0	83.6±39.2	758±556	413±198
	Log K_{film} [c]	10.8	10.6	11.6	11.3

Dust	K_{dust} [d] (m^3/g)	962±567	202±61.8	1970±1720	258±132

a. The calculation of the standard deviations of the partition coefficients were based on the uncertainties of measurements in airborne concentrations and surface concentrations. For $K=\frac{A}{B}$, $\sigma_K = \frac{\bar{A}}{\bar{B}}\sqrt{\left(\frac{\sigma_A}{\bar{A}}\right)^2 + \left(\frac{\sigma_B}{\bar{B}}\right)^2}$

b. The measured concentrations in indoor air are actually the total airborne concentrations; that is, the sum of the gas-phase (C_g) and particle-phase concentration (F), C_g+F. Based on the definition of particle/gas partition coefficient (K_p= (F/TSP)/C_g), the gas-phase concentration can be calculated using equation: C_g= (C_g+F)/(1+K_pTSP). Since C_g+F and TSP were measured and the values of K_p were estimated in Table 1, we calculated gas-phase phthalate concentration (C_g) and used it to further obtain surface/gas partition coefficients (K_{surf}).

c. Assuming the thickness of organic film is 10 nm and the film contains 20% organics, we calculated the dimensionless film/gas partition coefficients using equation 3.12 in ref (Weschler and Nazaroff, 2008).

d. Non-floor settled dust. Caution should be take when comparing results, because the dust/gas partitioning may have not reached steady state at 30 °C due to the short waiting time (~several weeks).

Table A5. The values of Log K_{oa} in literature.

	Log K_{oa}					
	BBzP			DEHP		
	21 °C	25 °C	30 °C	21 °C	25 °C	30 °C
Cousins and Mackay (2000)(Cousins and Mackay, 2000)	9.2[a]	8.8	8.2[a]	11.0[a]	10.5	9.9[a]
Weschler and Nazaroff (2008)(Weschler and Nazaroff, 2008)	11.6[a]	11.2	10.7[a]	13.1[a]	12.7	12.2[a]
Xiao and Wania (2003)(Xiao and Wania, 2003)	10.6[b]	10.4[b]	10.1[b]	11.5[b]	11.3[b]	11.0[b]

a. Based on the K_{oa} value of BBzP and DEHP reported in 25 °C, the values at 21 °C and 30 °C were extrapolated with their corresponding vapor pressures at different temperatures.

b. Calculated using Equation 6 in ref (Xiao and Wania, 2003) with vapor pressure estimated using Equation 9 in ref (Gobble et al., 2014).

Table A6. The values of dimensionless partition coefficients for fabric cloth (K_{cloth})

	Cotton		Polyester	
	21 °C	30 °C	21 °C	30 °C
BBzP	2.93×10^6	3.14×10^6	3.35×10^6	3.44×10^6
DEHP	2.00×10^6	1.83×10^6	2.17×10^6	1.99×10^6

Note: The cloth/gas partitioning possibly had not reached equilibrium for DEHP

Figure A4. Exterior, interior, and floor plan of the UTest House

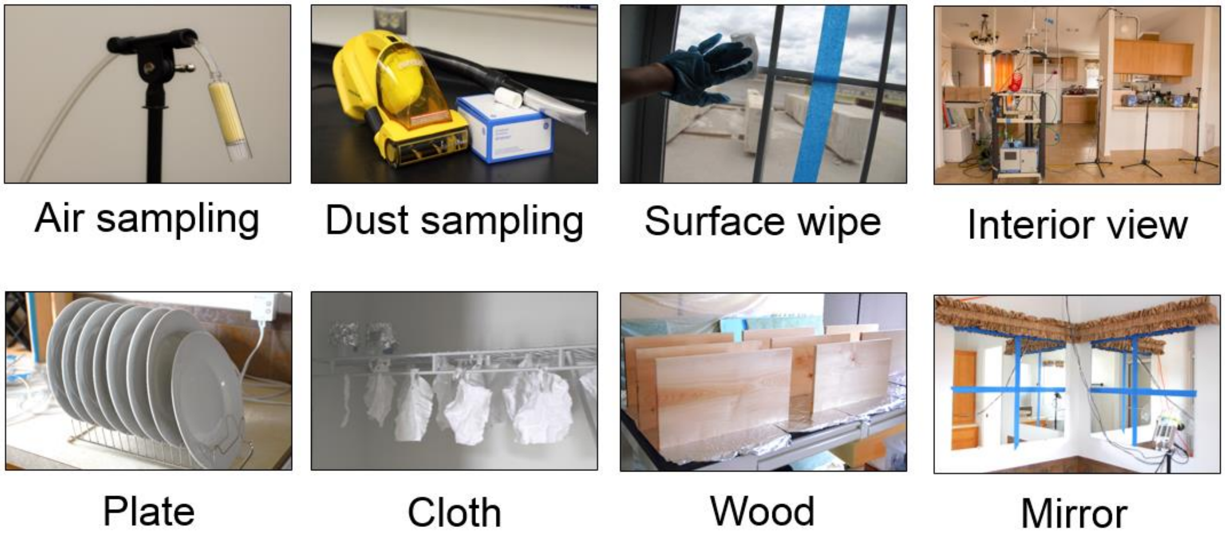
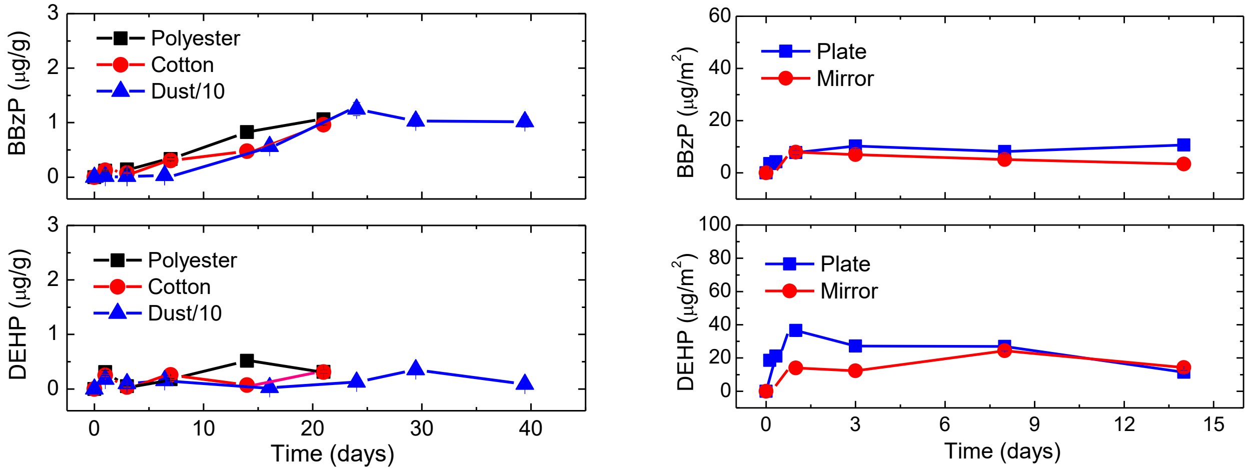


Figure A5. Photos of sampling method and materials



(a) (b)

Figure A6. (a) Sorption kinetics for cloth and dust. (b) Sorption kinetics for plates and mirrors.

Appendix B

Paper 2. Transfer of Phthalates and Their Alternatives from Polyvinyl Chloride Flooring and Crib Mattress Cover into Settled Dust

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(In preparation for *Environmental Science & Technology*)

ABSTRACT

Phthalates have been widely used as plasticizers in building materials and consumer products where they may be present at percent to tens-of-percentage levels. Previous field studies showed that settled dust, which was in contact with the source materials, have significantly higher phthalate concentrations than dust deposited on other surfaces. Therefore, it is important to investigate the transfer of phthalates and their alternatives from sources to settled dust through systematic chamber study. In this work, vinyl floorings and crib mattress covers with settled dust on the top was placed in a well-mixed chamber at different relative humidity and dust loadings. The dust was periodically collected for chemical analysis to monitor the increase of phthalate concentration in dust until steady state was reached. The results showed that the phthalates and their alternatives accumulated in the dust settled on the vinyl flooring and crib mattress covers, and reached to significantly high equilibrium concentrations in the dust. Model predictions indicate that when dust is in contact with the source materials, equilibrium concentrations of phthalates and their alternatives in dust are linearly correlated with y_0, the gas-phase concentrations of phthalates immediately adjacent to the surface of those materials, and the chemical K_{oa} values. We found that higher dust loading on source materials may slow down the transfer of phthalates from source to settled dust and reduce the equilibrium concentrations of phthalates in dust. The change of humidity in air did not influence the sorption of phthalates on dust. The results suggest that ingestion of dust settled on the source materials may significantly increase the exposure to phthalates and their alternatives, especially for young children, and the level could exceed the reference dose for certain compounds.

INTRODUCTION

Phthalates have been widely used as plasticizers to enhance the flexibility of polyvinyl chloride (PVC) products. These semi-volatile organic compounds (SVOCs) are found in a wide range of building materials and consumer products such as vinyl flooring, carpet padding, wall coverings, floor tiles, furniture, and electronics (Bornehag et al., 2005). Phthalates are often present in products at single-percent to tens-of-percent levels (Weschler and Nazaroff, 2008). For example, PVC flooring typically contains 10%-30% by mass phthalate plasticizers (Liang and Xu, 2014a). Because phthalates are not chemically bound to the polymer matrix, these SVOCs usually are emitted from the products into the air and other media. Previous studies have shown that phthalates are ubiquitous and among the most abundant indoor contaminants (Blanchard et al., 2014b).

Exposure to phthalates may result in severe adverse health effects, which have been detailed in several reviews (Jaakkola and Knight, 2008; Matsumoto et al., 2008). For example, exposure to certain phthalates, such as di-n-butyl phthalate (DnBP), butyl benzyl phthalate (BBzP), and di-(2-ethylhexyl) phthalate (DEHP), has been lined to profound and irreversible changes in the development of the reproductive tract, especially in males (Matsumoto et al., 2008; Martino-Andrade and Chahoud, 2010; Kay et al., 2013, 2014; Høyer et al., 2018). Phthalate exposure may also increase the chances of prenatal mortality, reduced growth and birth weight, and skeletal, visceral, and external malformations (Latini et al., 2006; Heudorf et al., 2007; Zarean et al., 2016). Besides, several epidemiologic studies reported the association between phthalates and childhood asthma (Oie et al., 1997; Kolarik et al., 2008a; Bornehag and Nanberg, 2010). In addition, alternative plasticizers, such as diisononyl cyclohexane-1,2-dicarboxylate (DINCH) and diethylhexyl adipate (DEHA) have emerged very recently, but currently there is a lack of toxicological information for these compounds (Schossler et al., 2011). Given that these alternatives have properties that are similar to those of phthalates, similar levels of emissions and environmental fates may be expected.

Phthalates and their alternatives partition strongly to particles due to their low vapor pressures and high K_{oa} values. Previous studies have suggested that high molecular weight phthalates such as BBzP and DEHP are mostly sorbed in the particle phase (>75%) rather than in the gas phase in indoor environments (Liu et al., 2013; Xie et al., 2013). Settled dust is deposited particles, which has large surface to volume ratio. Overtime, it may establish significant reservoirs of phthalates in indoor environments. When deposited particles are in contact with the source material of phthalates, their concentrations of phthalates could be orders of magnitudes higher than those in dust settled on non-source surfaces. Clausen et al. (2004) investigated phthalate uptakes in dust which was in contact with the PVC flooring containing 17% (w/w) DEHP in a controlled chamber. They found that the DEHP concentration in dust increased about seven-fold over the two-month experimental period. Other chamber studies also showed that PVC materials which contained phthalates can greatly enhance the concentration of phthalates in dust which were in contact with those materials (Schripp et al., 2010; Jeon et al., 2016). In addition to chamber studies, several field studies also provided evidence that there is direct transfer of phthalates between PVC floorings and dust in contact with the floorings. Bi et al., (2015) compared the BBzP concentrations in dust accumulated on source surfaces (flooring) and other non-source surfaces in a residential test house, and found that BBzP concentrations in dust settled on source surfaces were one order of magnitude higher than those on non-source surfaces. Sukiene et al., (2016, 2017) placed artificial consumer products doped with deuterium-labeled plasticizers such as phthalates and adipates in residential apartments. They monitored the concentration of SVOCs in dust settled on those surfaces and found that direct transfer from source into dust substantially increases the final SVOC concentration in dust in contact with the source, regardless of the vapor pressure of investigated SVOCs. However, none of those studies investigated the mass transfer mechanism from source to dust in a systematic way. Therefore, it is important to understand the transfer process of phthalates from their source materials to settled dust.

Ingestion and inhalation of the highly contaminated settled dust may result in significant exposure for indoor occupants. Little et al., (2012) estimated that ingestion of dust might be the most important pathway of phthalate exposure. Occupants can be exposed to dust-phase phthalates through dietary ingestion when dust is settled on food surfaces, surface-to-hand transfer when touching indoor surfaces, and inhalation when the dust is re-suspended from the flooring by human activities. Particularly for infants and toddlers who frequently play on the ground, pick up dust through touching indoor surfaces, and put their hands in the mouth, the exposure through ingestion of dust-phase phthalate can be considerably greater than those for adults. In addition, previous study showed that crawling-induced resuspension of settled floor dust may significantly enhance the exposure to inhalable particles for infants (Boor et al., 2016). Particles-bound phthalates may desorb from the particle after entering the airways and may create high localized concentrations at the particle deposition site and further cause bronchial obstruction (Oie et al., 1997; Jaakkola and Knight, 2008; Liu et al., 2017).

The goal of this study is to investigate the mass transport of phthalates and their alternatives from PVC floorings and crib mattress cover to settled dust through systematic chamber studies. The specific objectives of these studies are to 1) measure the direct transfer of phthalates and their alternatives from source to dust through systematic chamber studies; 2) study the mechanism controlling the transfer process; 3) investigate the influence of dust loading and humidity on the sorption kinetics and partition relationship; and 4) estimate the potential exposure for young children due to ingestion of highly contaminated dust through hand to mouth activities.

MATERIALS AND METHODS

Source Materials

As shown in Table B1, three PVC floorings and two crib mattress covers were used in this study. The test materials were pre-cleaned with wipes dipped with methanol before the experiments. The plasticizers in the tested materials were DnBP, BBzP, DEHP, DEHA, and DINCH, collectively, with mass fractions from 4% to 23%. The y_0, which is the gas-phase concentration of SVOCs immediately adjacent to the material phase, were directly measured using the method developed by Liang and Xu (2014) for these materials.

Dust Characteristics

Dust was collected from a residential house with a vacuum cleaner. The dust was then sieved with a 150 μm laboratory sieve and stored in an amber glass container with PTFE lid at -18 ºC. Background concentrations of target compounds in the dust were measured at 302 ± 23.4 μg/g and 257 ± 17 μg/g for DnBP and DEHP, respectively. The concentrations of BBzP, DEHA, and DINCH in dust were below detection limit. The mass fractions of organics in the dust were determined by the thermal/optical carbon analyzers (Atmoslytic Inc., Calabasas, CA) using reflectance and transmittance analysis and pyrolysis correction, and the value is 32.6 ± 2.80 %.

Table B1. Content, y_0 values, and dust loadings for tested source materials

ID	Content	Mass fraction (m/m)	y_0 (μg/m^3)	Dust loading (g/m^2)	
				Low	High
1 (vinyl flooring)	DEHP	23±3%	2.37	0.512±0.054	15.7±1.02
2 (vinyl flooring)	BBzP	15±2%	8.47	2.46±0.234	16.2±0.845
3 (vinyl flooring)	DnBP	4±1%	24.7	2.97±0.286	19.4±1.24
	DEHP	7±1%	1.54		
4 (mattress cover)	DEHA	4±0.3%	1.05	2.35±0.147	N.A.
5 (mattress cover)	DINCH	11±1%	0.70	2.88±0.165	N.A.

Experimental Setup

As shown in Figure 6B1, the 10 L stainless steel test canisters (model No. CTH-24, Eagle Stainless Container Inc.) were customized with an inlet and an outlet on the cap using two 0.25-inch bulkhead unions (SS-400-61, Swagelok). The air inlet was connected to a brass gas diffuser inside the canister to avoid a direct blow of gas streams onto the samples. The air outlet was connected to a stainless-steel tube which extends into the chamber to enhance the mixing of air inside the chamber. The air leakage rate was less than 2% of the total flow rate.

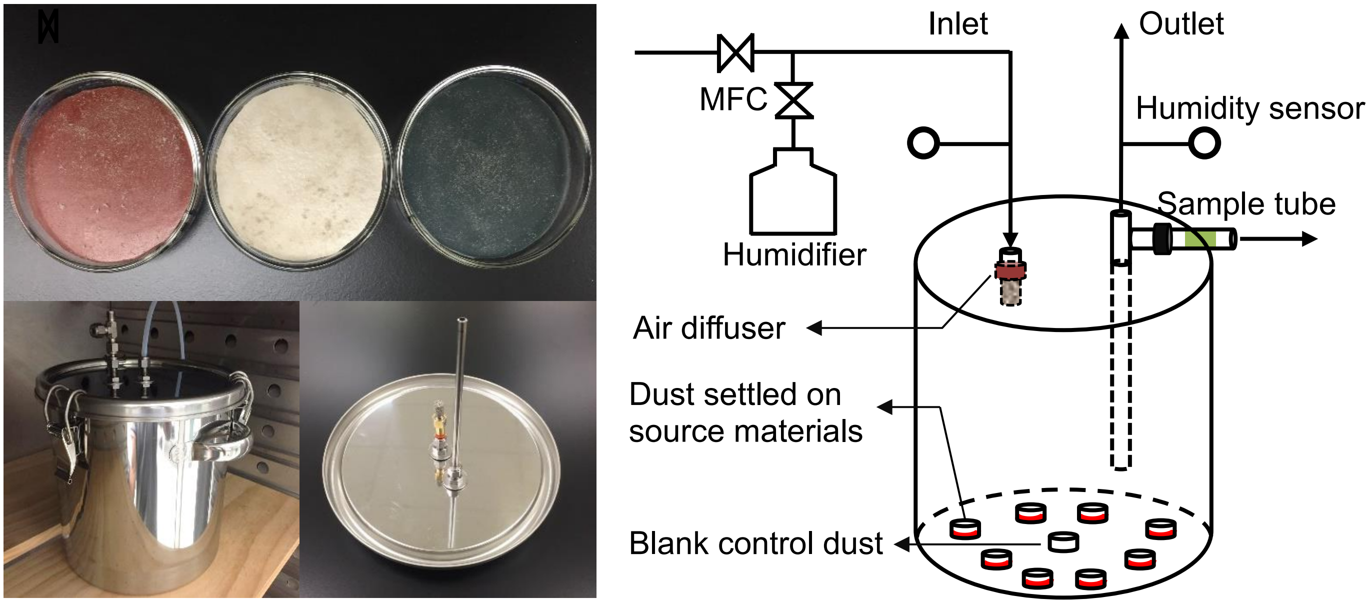


Figure B1. Configuration of test canister.

Source materials were cut into 60 mm diameter pieces and were placed into 60 mm diameter glass petri dish. Dust was evenly sprayed onto the flooring. Eight dishes containing the same material with dust were then placed at the bottom of a 10 L stainless-steel canister with one inlet and one outlet for airflow. The stainless-steel canister was operated at an air exchange rate of 0.5 hr^{-1}, ~0% relative humidity (RH), and 25 ± 0.5 °C. In addition, a blank control sample, which is a petri dish containing dust but without source materials, was also placed in each canister. During sampling, one of the eight petri dish samples was taken out of the canister for further extraction and analysis of the dust. In this way, dust samples were periodically collected from the chamber to monitor the increase of phthalate and their alternative concentration in dust until steady state was reached. Concentrations of the target compounds in blank control samples were also analyzed by taking a portion of dust from the blank control petri dish at each sampling time. In addition, , to measure the gas-phase concentration of target compounds in the chamber, air samples were collected from the canister using Tenax TA tubes with a pump (SKC 224-PCXR4) calibrated to a nominal flow rate of 50 mL/min for approximately 2 hours. Background concentrations of target compounds in the canister were examined for two weeks prior to the experiments and no contamination was found.

To study the influence of dust loading on the sorption kinetics, Sample 1, 2, and 3, were tested with low and high dust loadings, which were 0.512 - 2.97 and 15.7 - 19.4 g/m^2, respectively (Table B1). Sample 3, which contains DnBP and DEHP, was used to investigate the influence of humidity on the direct transfer of phthalates to dust. The experiment was conducted in three canisters with identical procedure described above except different relative humidity in the supplied air. The three canisters were supplied with air with relative humidity (RH) at 0%, 50% and 80%, respectively. Humidified air was prepared by supply air flow through a bubble humidifier. The relative humidity was monitored with a HOBO data logger (Model U12-012, HOBOware Pro, Onset Computer Co.) at the inlet and outlet of each canister. The experiments started after the relative humidity at the inlet had less than 3% difference with the one at the outlet of the canister. The differences of average relative humidity between the inlet and outlet of the canisters were less than 7% during the experimental period.

Dust Extraction and Chemical Analysis

The dust settled on the source material within the petri dish was removed gently with a quantitative filter and transferred to an 8-ml glass vial. The vial was weighted before and after placing the dust to determine the mass of dust collected in each petri dish. The collected dust samples were ultrasonically extracted three times for 30 min each with hexane. The volume of the extract was concentrated to approximately 100 μL by gently bubbling the samples with ultrapure nitrogen. The extract was then analyzed for DnBP, BBzP, DEHP, and DEHA using a GC-FID system (Agilent 7890A) equipped with a DB-5ht column (30 m, 0.25 mm, 0.1 μm). The GC-FID system was operated using a splitless injection at 275°C. The temperature program was set at 120°C and held for 2 min, ramped up 12°C/min for 15 min and held 3 min, and then ramped up 20°C/min for 2 min and held 2 min. The detector was set at 320°C. Helium was used as the carrier gas with a constant column flow set at 1.2 ml/min. A calibration standard was run prior to each GC injection, and the variance was always below 10% for all injections.

5 µL of the dust extract was injected to Tenax TA sorbent tubes for the analysis of DINCH. Tenax TA sorbet tubes collecting gas-phase SVOCs in the chamber as well as tubes with the dust extract injected were desorbed by a thermal desorber (TD) (Turbomatrix 650 ATD) and analyzed by a GC-MS system (Agilent 7890A GC-5975MS). Before analysis, 5 µL solution of d4-DEHP (2 µg/mL) in methanol was injected into the Tenax tubes as internal standard. The sorbent tubes were then desorbed for 30 min at 300 °C, with a He flow of 50 ml/min, and a cold trap temperature of minus 25 °C. Flash heating of the cold trap to 350 °C transferred the analyte through the valves at 250 °C and the transfer line at 250 °C to the GC. The GC-MS had a constant pressure resulting in a flow rate of 1.6 ml/min at 80 °C, and was equipped with a 30 m × 0.25 mm DB-5ht column and operated at a 5:1 split injection. The temperature program was 80 °C, hold for 0.5 min, ramp 20 °C/min for 8.5 min, then ramp 30 °C/min for 2 min, and finally hold for 9 min. All analyses were performed in full scan mode. All tubes were analyzed in two successive desorption to ensure complete desorption of both the tube and the TD-GC-MS system.

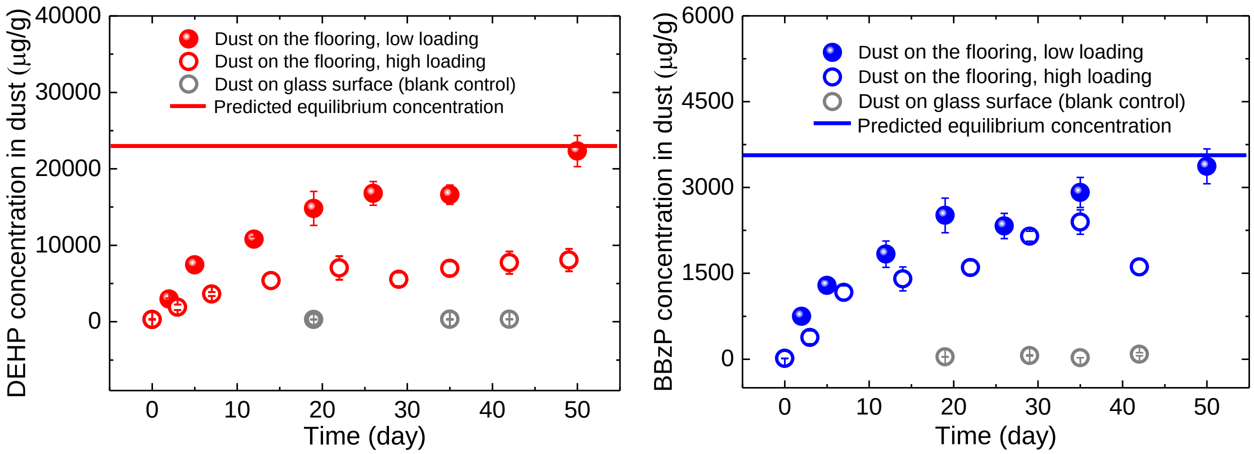
Quality assurance and control

Petri dishes were cleaned three times with hexane through ultrasonication and baked at 80 °C for 12 hours before experiments. Prior to sample measurements, matrix spiking experiments were conducted for all the target compounds; and the average recovery efficiencies were between 72% and 102%. Additionally, all samples were spiked with chemical standards prior to extraction to determine analytical extraction efficiencies, as described above. Each sample was weighed three times using a five-digit microbalance and mean weight was calculated. To check drift of the GC system, calibration standards were run after every six samples, and results were accepted only when the standards varied less than 10%. A calibration curve was constructed using six different concentrations in the range of 1-500 ng/µL for each of the five target compounds. Instrumental detection limits (IDLs) were estimated from calibration results based on evaluation guidelines for chromatographic analysis (OSHA, 1999). Method detection limits

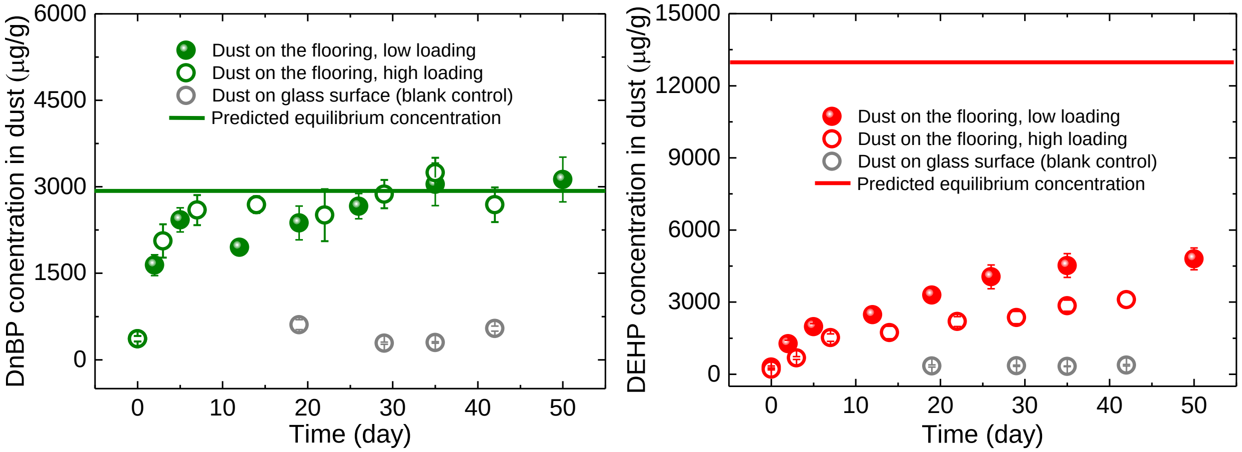
(MDLs) under the chromatographic conditions were calculated based on IDLs, median volume of extracts, and median weights of the dust samples. The IDLs and MDLs for phthalates and organophosphates are listed in Table B3. Lab blanks (i.e., empty petri dish) were collected and treated identically to the samples (except that no dust was collected). None of the target compounds were detected in these laboratory blanks. To avoid contamination, all glassware and metal ware used for sample collection and analysis were ultrasonically cleaned in methanol for 20 minutes, rinsed with methanol three times, and then baked at 80 °C in an oven prior to use.

RESULTS AND DISCUSSION

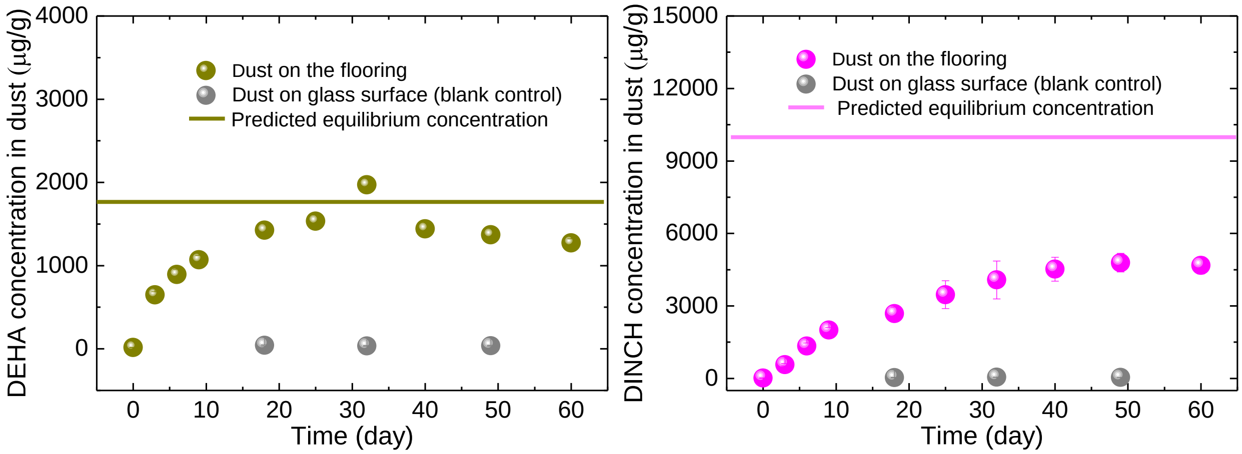
Figure B2 showed the accumulation of phthalates and their alternatives in dust settled on three floorings and two crib mattress covers. The results suggested the concentration of phthalates and their alternatives in dust settled on the source materials increased significantly within the experimental period. In the case of low dust mass loading, it took shorter time for the low-molecular-weight compounds (e.g., DnBP and BBzP) to reach steady state (possibly sorption equilibrium) than the high-molecular-weight ones (e.g., DEHP and DINCH). Figure B2a) shows that the steady-state concentration of DEHP in dust settled on surface of Sample 1 reached 29890 ± 2679 μg/g, which was two orders of magnitudes higher than the background levels of DEHP (302 ± 23.4 μg/g) in the dust measured prior to the experiments. The level was the highest among all target compounds in dust settled on their source materials. For DnBP, BBzP, DEHA, and DINCH, their steady-state concentrations in dust ranged from 1500 to 3500 μg/g, which were also 1 – 2 orders of magnitudes higher than their measured background levels in the dust. In contrast, the levels of phthalates and their alternatives in the blank control samples (i.e., dust settled on clean glass surfaces) at 50 days increased less than 50% for DEHP and fluctuated within the range of 20% for other target compounds compared with their background levels in the dust. The results suggest that there is significant direct transfer of phthalates and their alternatives from the source material to dust settled on the material surface. Such transfer is particularly important for high molecular weight phthalates, such as DEHP, due to their low vapor pressure and strong sorption to dust. The significantly enhanced concentrations of phthalates and their alternatives in dust settled on their source surfaces supported the findings in several recent field studies in the literature (Bi et al., 2015; Sukiene et al., 2016, 2017).



a) Sample 1 b) Sample 2



c) Sample 3



d) Sample 4 e) Sample 5

Figure B2. Concentrations of phthalates and phthalate alternatives in dust in contact with the source materials and in blank controls.

Figure B3 compared the measured steady-state concentrations in dust with the concentrations of target SVOCs in settled dust in residential homes reported in the literature (Rudel et al., 2003; Wilson et al., 2003; Hwang et al., 2008; Kanazawa et al., 2010; Bergh et al., 2011a; Guo and Kannan, 2011; Nagorka et al., 2011; Blanchard et al., 2014a; Fromme et al., 2016; Luongo and Ostman, 2016; Subedi et al., 2017; Larsson et al., 2017; Giovanoulis et al., 2018). The concentrations in dust measured in this study are 2 – 3 orders of magnitudes higher than the reported median concentration in the literature and are even higher than most of the reported maximum concentrations in these studies. Because the settled dust in the field campaigns might be collected on a variety of indoor surfaces which are not necessarily the source of the target compounds, the concentrations of phthalates and their alternatives in the dust could be lower than those measured in this study. Another possible reason is that the residence time of settled dust collected in these field studies is unknown. If it is not sufficient for SVOCs in the dust to equilibrium with the gas phase, the SVOC concentrations could be underrepresented in the dust. Finally, dust larger than 150 μm was screened out in this study while some of those large particles may be kept for analysis in field studies. Larger particles may have lower concentration of SVOCs due to their smaller specific surface area and lower mass transfer coefficients (Liu et al., 2014a). The results of current study indicate that when using settled dust as a sampling medium of SVOCs in indoor environments, it is important to know the sampling location and types of material surfaces because the types and levels of SVOCs may be strongly related to local materials used in indoor environments.

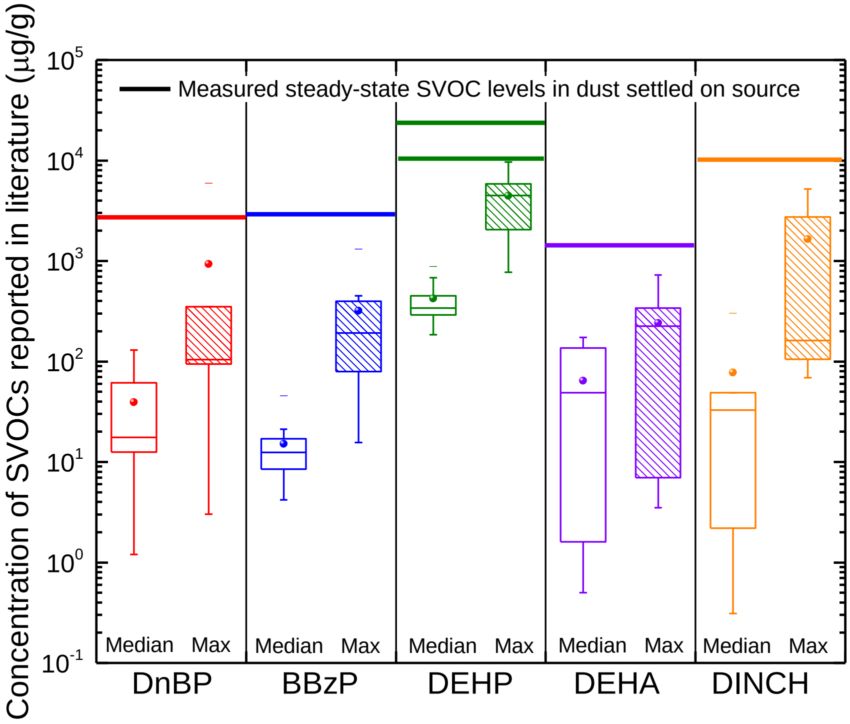


Figure B3. Comparisons between measured SVOC levels in dust and median and maximum concentrations of SVOCs reported in the literature.

Influence of dust loading

The influence of dust loading was further examined by increasing the mass loading from 0.5 - 3 g/m^2 to 15 - 20 g/m^2 on the surfaces of Sample 1, 2, and 3, as shown in Figure 7B2. The steady-state dust concentrations of BBzP and DEHP in the case of low dust loading were about twice as high as those in the case of high dust loading. Under low dust loading condition, it took about 22 and 40 days for the dust to reach sorption equilibrium for BBzP and DEHP, respectively; while the time increased to 40 days for BBzP and longer than 50 days for DEHP, when dust loadings were elevated. In contrast, no significant differences were observed for DnBP between the low and high dust loading cases either on the steady-state dust concentration or sorption kinetics, possibly due to its high volatility and weak sorption to particles. The results suggest that high dust loading may reduce the equilibrium concentration of phthalates in dust settled on the source surfaces and slow down the sorption kinetics. In residential homes, the dust loading on flooring materials ranges from 0.002 to 4 g/m^2 (Lioy et al., 2002; Johnson et al., 2009; Roberts et al., 2009; Patisaul et al., 2013; Rasmussen et al., 2013), which is even lower than the minimum loading used in the current experiments. Therefore, faster transfer of SVOCs from source surfaces to the dust deposited on the surface and higher SVOC levels in the dust are expected in actual indoor environments, compared to the experimental measurements.

Influence of Humidity

Dampness in buildings has been found to be a strong risk indicator for health effects such as asthma (Jaakkola and Knight, 2008). Several field studies suggested that increased levels of phthalates were associated with signs of higher dampness in residential homes (Hsu et al., 2012b; Zhang et al., 2013; Ait Bamai et al., 2014a). However, it is still unknown that whether there is any association between dampness (represented by RH) and the concentrations of phthalates in the indoor environments. In this study, DnBP and DEHP concentrations in dust settled on Flooring 3 were monitored under the RH at 0%, 50% and 80%, respectively. Phthalates accumulated in the dust at close rates and reached to similar steady-state levels under the different RH conditions (Figure B4). Previous studies showed that humidity has little or no influences on the emission of phthalates from PVC products (Clausen et al. 2007; Ekelund et al., 2010; Manoukian et al., 2016). Therefore, the amount of phthalates emitted from the vinyl flooring sample is expected to be similar at the different RHs in the current study. Recent studies suggested that when absorption was the dominant sorption mechanism, RH may have no influence on the soil/air partition of SVOCs; while when adsorption dominated the sorption process, the increased moisture in the air could competitively displacing SVOC molecules adsorbed on the soil surfaces (Hippelein and McLachlan, 2000; Goss et al., 2004; Davie-Martin et al., 2015). The results of current study suggest that RH may have little impact on the transfer of phthalates to the dust settled on the source surfaces and absorption may dominate the sorption to the dust.

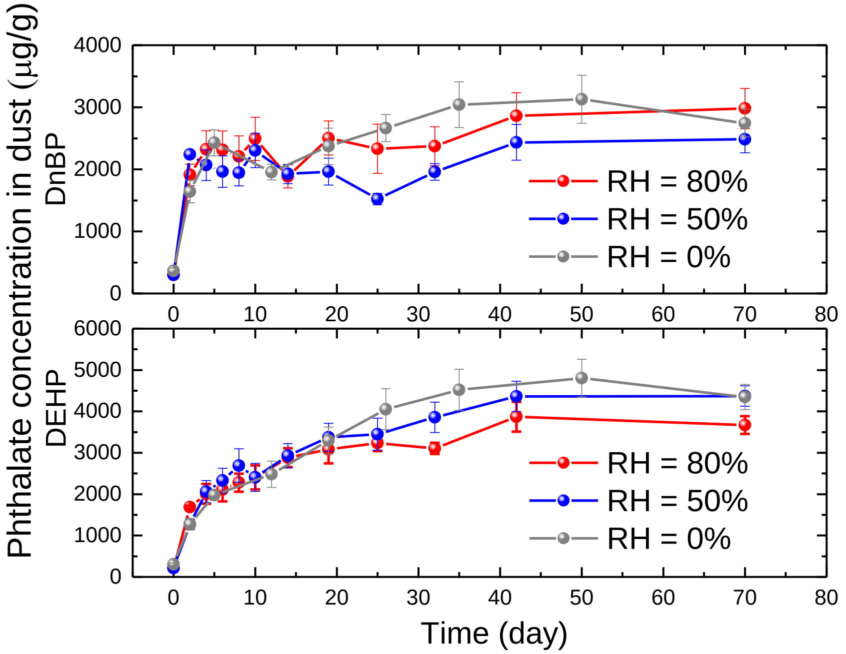


Figure B4. Influence of humidity on dust concentration

Equilibrium model

A linear partition relationship was assumed between the concentration of target compounds in the dust and in the gas phase (Weschler and Nazaroff 2010), or

$$X_{dust} = \frac{f_{om\_dust} \times K_{oa} \times C_g}{\rho_{dust}} \quad (1)$$

where X_{dust} is the dust-phase concentration of SVOCs, f_{om_dust} is the volume fraction of organics, ρ_{dust} is the density of dust, K_{oa} is the octanol-air partition coefficients, and C_g is the gas-phase concentration of SVOCs. f_{om_dust} was measured as 0.32 in this study and ρ_{dust} is the density of dust, which was assumed as 2.0×10^6 g/m^3 (Weschler and Nazaroff, 2010). Median values of K_{oa} for the target compounds reported in the literature was used (Table B4). y_0, which is the gas-phase concentration immediately adjacent to the material, was used for C_g. It is relatively constant and its values for the vinyl flooring and crib mattress cover samples were measured using a previous developed chamber method, and the results were listed in Table B1. The predicted equilibrium concentrations of target compounds in dust were shown as solid lines in Figure B2. The predicted equilibrium concentrations agreed well with most of the measured steady-state levels of target compounds, indicating sorption saturation in dust had been reached. In addition, the results suggest that when dust is in contact with the source of certain SVOCs, it is the gas-phase SVOCs immediately adjacent to the material partitioning with the dust phase rather than the gas phase in the bulk room air. Because y_0 values for a range of building materials and consumer products, such as vinyl floorings, crib mattress covers, backpacks, toys, table cloth, and wall coverings, have been reported in previous studies (Liang and Xu, 2014a; Xie et al., 2015; Cao et al., 2017), Equation 1 can be used to rapidly determine the equilibrium concentrations of SVOCs in dust settled on these sources.

For DEHP, the measured steady-state concentration in dust settled on Sample 1 agreed with the predicted concentrations, but the level is significantly higher than the concentrations measured in dust settled on Sample 3. In addition, the concentration of DINCH measured in this study also did not agree with the prediction. The discrepancy between measured and predicted equilibrium concentrations of DEHP and DINCH might be due to the uncertainties of K_{oa} values in the literature and differences of dust loadings used in the experiments. The K_{oa} values of DEHP reported in the literature range over three orders of magnitudes, which may significantly increase the uncertainty in the prediction. Figure B6 shows the potential uncertainties of the predicted equilibrium concentrations of DEHP (dashed line), estimated using the minimum and maximum K_{oa} values reported in literature. The measured steady-state DEHP concentrations fell within the estimated range. DINCH is an emerging phthalate alternative recently used in the market, few studies have reported its K_{oa} value or vapor pressure that can be used to calculate its K_{oa}. In addition, dust loading may also have a significant impact on the equilibrium concentrations of phthalates in dust (discussed in the next section). As shown in Table B1, even under low dust loading condition, the loading on Sample 3 is six times higher than those on Sample 1. The increase of dust loading may result in accumulation of multiple-layer dust on the source materials. Due to the existence of concentration gradient within the boundary layer, the gas-phase concentrations near the upper layer of the dust may be lower than that immediately adjacent to the materials (y_0). Therefore, the lower overall gas-phase concentration near the dust due to higher dust loading may result in a reduced equilibrium concentration in the dust.

Exposure for children through dust ingestion

Children spend considerable amount of time crawling and playing on the floor. These activities may result in the transfer of dust from flooring materials to their hands. Because children frequently put their hands in their mouth, the hand-to-mouth transfer may result in significant amount ingestion of the contaminated dust. Although the total daily ingestion of soil and dust was estimated for children (Moya and Phillip 2014), there are limited studies focusing specifically on the ingestion of dust through hand-to-mouth transfer. Wilson et al. (2013) developed a mechanistic model to estimate the soil and dust ingestion rates through hand to mouth transfer and found that the mean dust ingestion rate for toddlers who play on hard surfaces was 53 mg/day. Another study used U.S. Environmental Protection Agency (EPA)'s Stochastic Human Exposure and Dose Simulation (SHEDS) model to differentiate among various activities of children and reported mean hand-to-mouth dust ingestion to be 20 mg/day. They also reported that hand-to-mouth dust ingestion typically consisted of about 29 percent of the amount of the total daily soil and dust ingestion. Therefore, we used the hand-to-mouth dust ingestion rate reported in the literature to estimate the potential exposure to phthalates and phthalate alternatives due to ingestion of the settled dust measured in the current study. For studies only reported the total daily dust ingestion rate, we applied the ratio of 29 percent to obtain the estimated hand-to-mouth ingestion rates. The exposure through ingestion of dust was calculated for a three-year-old toddler with the following equation:

$$Exposure\ (\mu g/kg/d) = \frac{IngR_{HtoM} \times X_{dust}}{BW} \tag{2}$$

Where $IngR_{HtoM}$ (mg/day) is the hand-to-mouth dust ingestion rate; X_{dust} (μg/g) is the equilibrium concentration of target compounds in dust measured in this study; and BW (kg) is the body weight. Parameters used in the calculation were listed in Table B2.

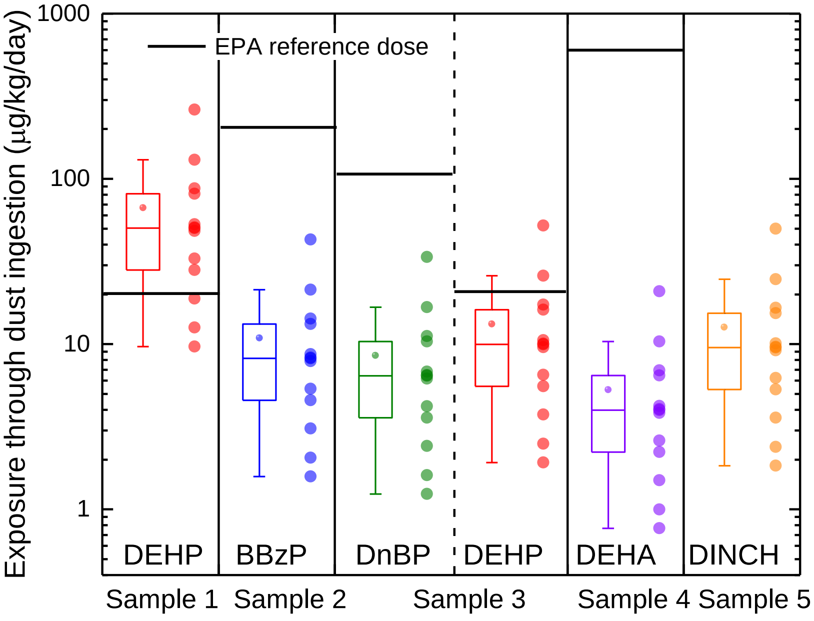


Figure B5. Toddlers' exposure to phthalates and phthalate alternatives via ingestion of dust settled on source surfaces due to hand-mouth activities.

Figure B5 showed the estimated exposure of toddlers to phthalates and phthalate alternatives via ingestion of dust settled on source surfaces due to their hand-to-mouth activities. The results suggest that the potential exposure could be significantly high. The maximum estimated exposure to DEHP was 263 μg/kg/day, which was more than 20 times higher than the oral reference dose reported by U.S. EPA. The median exposure to DEHP was 50 and 10 μg/kg/day for dust settled on flooring Sample 1 and Sample 3, respectively, which were also over two times higher than and half of the reference dose. The estimated median values of exposure to DnBP, BBzP, and DEHA were 6.43, 8.20, and 9.53 μg/kg/day, respectively. Although they were significantly lower than the reference doses (100, 200, and 600 μg/kg/day, respectively), the exposure levels via only ingestion of hand-to-mouth dust settled on source surfaces remains comparable to the daily overall intake of DnBP, BBzP, DEHA, or DINCH in typical residential homes reported in the literature (Oomen et al., 2008; Shi and Zhao, 2014; Zhang et al., 2014). The results indicate that ingestion of dust settled on the sources of SVOCs may an play important role on children's exposure to these compounds.

Table B2. Parameters used in the model prediction

ID	Sample 1	Sample 2	Sample 3		Reference
Content	DEHP	BBzP	DnBP	DEHP	
Dust-phase concentration, X_{dust} (ug/g)	22700	3700	2900	4500	Measured in this study
3-year-old toddler body weight, BW (kg)	13.8				(US EPA, 2011)
Hand-to-mouth dust ingestion rate (mg/day)	20-160				(Lepow et al., 1975; Duggan and Williams, 1977; Binder et al., 1986; Clausing et al., 1987; Davis et al., 1990; Stanek and Calabrese, 1995; Calabrese et al., 1997b, 1997a; Hogan et al., 1998; Özkaynak et al., 2011; Stanek et al., 2012)

CONCLUSIONS

This study investigated the transfer of phthalates and their alternatives from source surfaces to dust settled on the surface through systematic chamber study. The results showed that the phthalates and their alternatives accumulated in the dust settled on the vinyl flooring and crib mattress covers, and reached to significantly high equilibrium concentrations in the dust. Model predictions indicate that when dust is in contact with the source materials, equilibrium concentrations of phthalates and their alternatives in dust are linearly correlated with y_0, the gas-phase concentrations of phthalates immediately adjacent to the surface of those materials, and the chemical Koa values. We found that higher dust loading on source materials may slow down the transfer of phthalates from source to settled dust and reduce the equilibrium concentrations of phthalates in dust. The change of humidity in air did not influence the sorption of phthalates on dust. The results suggest that ingestion of dust settled on the source materials may significantly increase the exposure to phthalates and their alternatives, especially for young children, and the level could exceed the reference dose for certain compounds.

ACKNOWLEDGEMENT

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SUPPORTING INFORMATION

Table B3. Instrumental detection limits (IDLs) and method detection limits (MDLs) of target compounds.

Abbreviation	Name	IDLs (ng)	MDLs (µg/g)
DnBP	Di-n-butyl phthalate	1.28	25.6
BBzP	Benzyl butyl phthalate	1.50	30.0
DEHP	Bis (2-Ethylhexyl) phthalate	2.97	59.4
DEHA	Di ethylhexyl adipate	1.37	27.4
DINCH	Diisononyl cyclohexane-1,2-dicarboxylate	1.06	21.2

Table B4. Calculated K_{oa} values based on the vapor pressures reported in literature

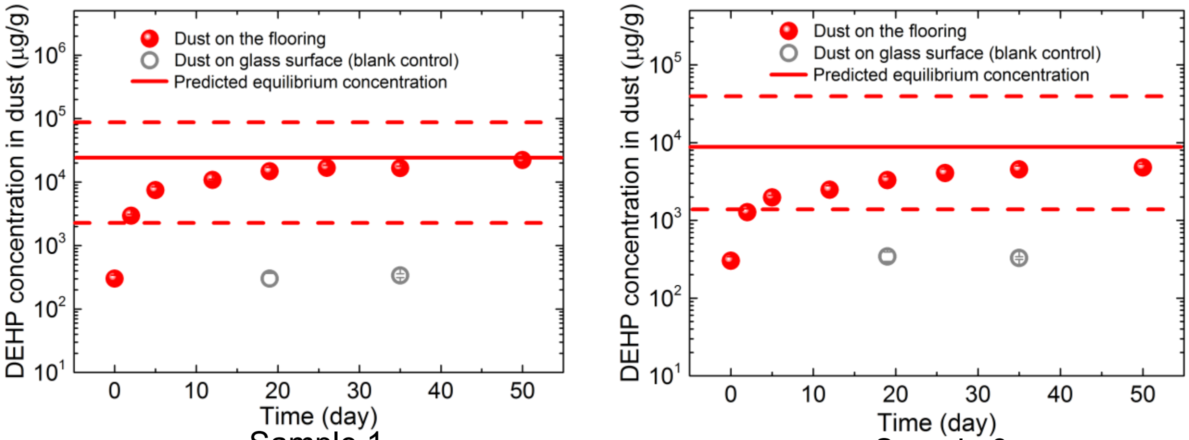
Compound	Vapor pressure (pa)	Calculated logK_{oa} values
DnBP	2.70×10^{-3}[a], 4.70×10^{-3}[b], 9.70×10^{-3}[b], and 3.60×10^{-3}[b]	9.23[a], 8.99[b], 8.68[b], 9.11[b], and 8.54[d]
BBzP	1.10×10^{-3}[a], 2.50×10^{-3}[b], 1.20×10^{-3}[b], and 6.70×10^{-4}[b],	9.61[a], 9.26[b], 9.58[b], 9.83[b], and 8.78[d]
DEHP	1.90×10^{-5}[a], 1.70×10^{-4}[a], 5.10×10^{-5}[a], 2.10×10^{-5}[b], 2.50×10^{-5}[b], and 8.60×10^{-4}[b]	11.4[a], 10.4[a], 10.9[a], 11.3[b], 11.2[b], 9.72[b], and 10.53[d]
DEHA	4.27×10^{-4}[a]	10.0[a]
DINCH	2.20×10^{-5}[c]	11.3[c]

[a] calculated in EPI suite

[b] Weschler and Nazaroff, (2008)

[c] NICNAS, (2012)

[d] Cousins and Mackay, (2000)



Sample 1

Sample 3

Figure B6. Predicted equilibrium concentrations of DEHP using the minimum and maximum K_{oa} values reported in the literature

Appendix C

Paper 3. Phthalates and organophosphates in settled dust and HVAC filter dust of U.S. low-income homes: association with season, building characteristics, and childhood asthma

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ABSTRACT

Phthalates and organophosphates are ubiquitous indoor semi-volatile organic contaminants (SVOCs) that have been widely used as plasticizers and flame retardants in consumer products. Although many studies have assessed their levels in house dust, only a few used dust samples captured by filters of building heating, ventilation, and air conditioning (HVAC) systems. HVAC filters collect particles from large volumes of air over a long period of time (potentially known) and thus provide a spatially and temporally integrated concentration. This study measured concentrations of phthalates and organophosphates in HVAC filter dust and settled floor dust collected from low-income homes in Texas, United States, in both the summer and winter seasons. The most frequently detected compounds were benzyl butyl phthalate (BBzP), di-(2-ethylhexyl) phthalate (DEHP), di-n-octyl phthalate (DnOP), tris (1-chloro-2-propyl) phosphate (TCPP), triphenyl phosphate (TPhP), and tris (1,3-dichloroisopropyl) phosphate (TDCPP). The median level of TCPP in settled dust was 3- to 180-times higher than levels reported in other studies of residential homes, which was likely due to the use of TCPP in roof and wall insulation installed in low-cost housing units. No correlation between HVAC filter dust and settled dust was observed for the target compounds other than TCPP. Significantly higher concentrations were observed in HVAC filter dust as compared to settled dust for most of the frequently detected compounds in both seasons, except for several phthalates in the winter. SVOC concentrations in settled dust in winter were generally higher than in summer, while different seasonality patterns were found for HVAC filter dust. Settled dust samples from homes with vinyl flooring contained significantly higher levels of BBzP and DEHP as compared to homes with other types of floor

material. The concentration of DEHP and TDCPP in settled dust also significantly associated with the presence of carpet in homes. Cleaning activities to remove dust from furniture actually increased the levels of certain compounds in HVAC filter dust, while frequent vacuuming of carpet helped to decrease the concentrations of some compounds in settled dust. Additionally, the size and age of a given house also correlated with the levels of some pollutants in dust. A statistically significant association between DEHP concentration in HVAC filter dust in summer and the severity of asthma in children was observed. These results suggest that HVAC filter dust represents a useful sampling medium to monitor indoor SVOC concentrations with high sensitivity; in contrast, when using settled dust, in addition to consideration of seasonal influences, it is critical to know the sampling location because the type and level of SVOCs may be related to local materials used there.

INTRODUCTION

Phthalates and organophosphates are widely used as chemical additives to enhance performance of polymeric materials and products. Phthalate additives are typically used as plasticizers to improve the flexibility of polyvinyl chloride (PVC) products such as flooring, wall covering, and electrical cables (Liang and Xu, 2014b). The global production rate of phthalates has increased to 6 million tons/year within a decade; currently, phthalates account for more than 70% of the worldwide plasticizer market (Rudel and Perovich, 2009; Erickson, 2015). Phosphate additives are commonly used as flame retardants, stabilizers, and/or plasticizers in a variety of consumer goods such as furniture, textiles, electronics, and baby products (Saito et al., 2007; Stapleton et al., 2009, 2011, 2012; Gallen et al., 2014b; Cooper et al., 2016). Restrictions on the use of polybrominated diphenyl ether (PBDE) flame retardants have led to increased use of organophosphates as a replacement (Stapleton et al., 2011; Bergman et al., 2012b; van der Veen and de Boer, 2012; Araki et al., 2014; Castorina et al., 2017). The global consumption of organophosphate flame retardants was 300,000 tons in 2004, increased to 680,000 tons in 2015 (Giulivo et al., 2017), and was projected to grow at a rate of 5.2% per year until 2021 (Lucintel, 2016). Phthalates and organophosphates are not chemically bonded to the given polymer matrix and, as a result, these compounds slowly migrate into the surrounding environment (Kemmlein et al., 2003; Xu et al., 2010; Xu et al., 2009; Xu and Little, 2006) and are now ubiquitous indoor pollutants (Bergh et al., 2011; Luongo and Ostman, 2016; Weschler and Nazaroff, 2008). The indoor air concentration of phthalates and organophosphates is several orders of magnitude higher than their outdoor concentration, indicating that these compounds originate from indoor environmental sources (Staaf and Östman, 2005; Rudel et al., 2010b).

Health concerns related to phthalates and organophosphates have primarily focused on carcinogenic, neurological, reproductive and developmental toxicity. For example, exposure to certain phthalates, such as di-n-butyl phthalate (DnBP), butyl benzyl phthalate (BBzP), and di-(2-ethylhexyl) phthalate (DEHP), has been lined to profound and irreversible changes in the development of the reproductive tract, especially in males (Latini et al., 2006; Matsumoto et al., 2008; Martino-Andrade and Chahoud, 2010; Kay et al., 2013, 2014; Zarean et al., 2016; Høyer et al., 2018). Two widely used organophosphates, tri(2-chloroethyl) phosphate (TCEP) and tris(1,3,-dichloroisopropyl) phosphate (TDCPP), have been designated as carcinogens by the state of California (State of California, 2016). Adverse health effects, such as decreased fetal growth, autism spectrum disorders, disruption of hormones, altered behavior in early stages of life, decreased quality of semen, and alteration of thyroid function have also been associated with exposure to phthalates and organophosphates (Meeker and Stapleton, 2010; Dishaw et al., 2011, 2014; van der Veen and de Boer, 2012; Meeker et al., 2013; Patisaul et al., 2013; Miodovnik et al., 2014; Jeddi et al., 2015; Oliveri et al., 2015; Ferguson et al., 2016; Preston et al., 2017; Zhao et al., 2017; Hoffman et al., 2017; Schang et al., 2018). In contrast, the association between exposure to phthalates and organophosphates and allergic diseases has not received substantial attention. Several epidemiological studies have reported links between certain phthalates present in dust or PVC materials in homes and asthma and allergic diseases in children (Bornehag et al., 2004; Jaakkola and Knight, 2008; Kolarik et al., 2008a; Larsson et al., 2010; Hsu et al., 2012a; Ait Bamai et al., 2014b, 2016). One study reported an association between organophosphates and the prevalence of asthma and allergies (Araki et al., 2014). However, other studies conducted with similar approaches seemed to contradict these findings (Callesen et al., 2014b; Beko et al., 2015).

Most phthalates and organophosphates are classified as semi-volatile organic compounds (SVOCs) due to their low vapor pressures at room temperature. SVOCs are known to sorb strongly to particles, and the fraction of SVOCs in the particle phase increases with increasing octanol-air partition coefficients (K_{oa}) (Weschler and Nazaroff, 2008, 2010). For most high-molecular-weight SVOCs, the particle-phase fractions are greater than 75% as compared to the gas phase (Liu et al., 2013; Xie et al., 2013). Therefore, indoor particles play an important role in human exposure to phthalates and organophosphates. Recent studies have shown that particle-bound SVOCs, following inhalation, may create high localized concentrations in the airway at the deposition site of particles and cause bronchial obstruction (Oie et al., 1997; Jaakkola and Knight, 2008). Additionally, humans can be exposed to particle-phase SVOCs through ingestion of deposited particles (i.e., settled dust) adhered to food, objects, and the skin. Ingestion of particle-phase SVOCs is particularly relevant to children, who spend considerable time on floors where contaminated particles accumulate and frequently put their hands and other objects into their mouths. Ingestion of house dust and inhalation of airborne particles have been identified as the major exposure pathways for certain phthalates (Bekö et al., 2013; Little et al., 2012; Xu et al., 2010; Xu et al., 2009) and organophosphates (Marklund et al., 2005; Mäkinen, 2009; Schreder et al., 2016), respectively.

Settled dust (i.e., deposited particles) that is sampled from floors, mattresses, moldings, and shelves has been widely used as an indicator for residential contamination (Butte and Heinzow, 2002; Lioy et al., 2002; Mercier et al., 2011), especially for SVOCs. House dust serves as an “indoor sink”, collecting and accumulating SVOC pollutants in a manner similar to a passive sampler. Phthalates and organophosphates have been frequently detected in settled dust at high concentrations in various countries (Abdallah and Covaci, 2014; Ait Bamai et al., 2014a; Albar et al., 2017; Ali et al., 2016, 2013, 2012; Bergh et al., 2011; Blanchard et al., 2014; Bornehag and

Nanberg, 2010; Castorina et al., 2017; Dodson et al., 2015; Gevao et al., 2013; Guo and Kannan, 2011; He et al., 2016; Hsu et al., 2012; Kanazawa et al., 2010; Kubwabo et al., 2013; Luongo and Ostman, 2016; Rudel et al., 2003; Santillo et al., 2003; Stapleton et al., 2009, 2008; Van den Eede et al., 2011; Wu et al., 2016). However, the age of settled dust is usually unknown, which can impact the accuracy of exposure assessments. For SVOCs with high K_{oa} values, if house dust does not have sufficient residence time indoors to equilibrate with the gas phase, the SVOC compounds tend to be underrepresented in dust relative to their abundance in air (Weschler and Nazaroff, 2010). Furthermore, settled dust samples are localized and typically only represent the immediate collection area. Samples collected in the same house but at multiple locations may vary significantly in chemical composition, due to existence of different sources and different origins of dust (Bergh et al., 2011; Morawska and He, 2018; Sukiene et al., 2016). To fully understand the potential respiratory health effects of exposure to SVOCs, it is crucial to measure their concentrations in inhalable particles (with aerodynamic diameter < 15 μm) and respirable particles (with aerodynamic diameter < 2.5 μm) (Miller et al., 1979). Settled dust may not be suitable for this purpose, because only a small fraction of particles in settled dust falls in these size ranges (Butte and Heinzow, 2002; Gustafsson et al., 2018). Although traditional air sampling techniques have been used to determine SVOC levels in suspended particles (e.g., PM_{10}) (Wang et al., 2014; Zhang et al., 2014; Mandin et al., 2016), these may only provide a brief "snapshot" of the concentration, since SVOC levels can vary significantly over time and within the same building.

An alternative to conventional approaches to collect particle-phase SVOCs is use of filters in building heating, ventilation, and air conditioning (HVAC) systems. More than 70% of residential homes in the U.S. have a central forced-air HVAC system (US HUD, 2011), almost all with built-in filters that can be collected with minimal effort. HVAC filters remain in place for relatively long periods of time (which can be measured), collect particles from a wide spatial area with large volumes of air, and thus provide a spatially and temporally integrated concentration. HVAC filter dust has been successfully used to characterize indoor microbial communities

(Stanley et al., 2008b; Noris et al., 2009, 2011; Maestre et al., 2018a) and heavy metals (Noris et al., 2009) but has been applied only in a limited number of studies on phthalates and organophosphates (Xu et al., 2014b; He et al., 2016). To our knowledge, no study has measured their concentrations in HVAC filter dust in residential homes, and the correlation of SVOC levels between HVAC filter dust and settled dust has not been investigated. Additionally, the influence of seasonal variations and building characteristics on SVOC accumulation in HVAC filter dust is unknown. Such information is needed not only for better understanding the indoor sources of and human exposures to SVOCs, but also for evaluating the feasibility of this novel filter forensics method compared to conventional sampling approaches in indoor environments.

The present study focused on phthalates and organophosphates (the two major groups of indoor SVOC contaminants) in low-income homes, where building characteristics (e.g., smaller home size and older age) and occupant behavior (e.g., cleaning and smoking) linked with socioeconomic status (SES) may result in higher exposure to indoor pollutants (Adamkiewicz et al., 2011; Dodson et al., 2017). The objectives of this study are as follows: (1) determine the concentrations of target phthalates and organophosphates in settled dust and HVAC filter dust samples collected from low-income homes (N = 54) in Texas, United States; (2) compare the composition of selected compounds in settled dust and HVAC filter dust; (3) investigate the influence of seasonal variation on concentrations of these compounds in dust; (4) examine the associations between selected building characteristics and the concentration of target phthalates and organophosphates in dust; and (5) investigate the relationship between the concentrations of these compounds in dust and asthma and asthma severity in children.

MATERIALS AND METHODS

Sampling sites

The research is part of a large project to investigate asthma triggers for children in low-income homes (HUD: TXHHU0023-13). A total of 54 homes from rural/semi-rural areas of central Texas enrolled in this study. The mean SES score of the participating homes was 23.7 (sample range: 8 – 47), with most belonging to the two lowest social classes defined by the Hollingshead Index (i.e., <29). The enrolled occupied homes all had the typical appliances, furniture, electronics, and household goods present. Environmental sampling was conducted at each home during both winter (December 2014 – February 2015) and summer (June 2014 – September 2014) seasons. Questionnaires including personal and dwelling characteristics were distributed and collected during home visits. Through the questionnaires and onsite visual inspections, building characteristics such as the age and type of the house, floor area, number of rooms, frequency of cleaning, signs of mold and dampness, and floor materials were recorded. There were five categories of floor materials: PVC flooring, wall-to-wall carpet, wood floor, concrete, and ceramic tile. Additionally, the HVAC run-times and indoor temperatures in each household were monitored using Onset U12-012/013 monitors installed in each home; the airflow rate through the HVAC system was determined for each site; and the microbial concentrations and community were also investigated with details described in Maestre et al. (2018) and Givehchi et al. (2018).

Study population

Participants were recruited from the Children’s Wellness Center – a safety net clinic for low-income Medicaid-eligible children in central Texas. The study enrolled 54 children (60% boys and 40% girls) with a mean age of 10.3 years (SD = 2.8 years). Approval was given by all participants, in accordance with the Institutional Review Board Protocol by the University of Texas at Austin. Data collected from the children included pulmonary function testing results, questionnaire information, and inflammatory markers contained in exhaled breath samples. There were 37 children who had physician-diagnosed asthma and experienced asthma symptoms in the previous 12 months, leaving 17 children in the healthy control group. Asthma severity was measured with the validated Severity of Chronic Asthma scale with three questions derived from national clinical guidelines that parents answered about the frequency of daytime asthma symptoms, night-time wakening for asthma, and activity limitations due to asthma (Horner et al., 2006). A score of 1 was recorded for the "does not have asthma" response, followed by 2 for "has asthma symptoms 0-2 times each week," to 5 for "has asthma symptoms all the time." The responses to the 3 questions were summed. Consistent with the national clinical guidelines, families that indicated their child had asthma symptoms "0-2" times a week (score of 6) were classified as having intermittent asthma, and children who had asthma symptoms more frequently than two times per week were classified as having persistent asthma (scores of 7 or higher).

Questionnaires

Questionnaires were distributed and collected from each home during site visits. The questionnaire consisted of two sections concerning the dwelling and personal characteristics. The section of dwelling and living conditions included questions such as building age, floor area, type of building, number of occupants, floor materials, renovations, frequency of room cleanings, presence of pets indoors, presence of smokers in the home, signs of dampness, water leakage, usage of mechanical ventilation equipment, and the frequency of window opening/closing. Parents of participating children answered questions concerning personal characteristics and health conditions of their children. The questionnaire gathered information on gender, age, school grade, parental education and occupation, time typically spent in house, history of allergies, parental history of allergies, food intake, health status, and medication. Questions on asthma and asthma symptoms were based on the core questionnaire of the International Study of Asthma and Allergies in Childhood (ISAAC) (ISAAC, 1993).

Sample collection and analysis

HVAC filter dust and settled dust samples were collected in both summer and winter seasons. Brand-new synthetic HVAC filters, with minimum efficiency reporting value (MERV) ratings of 7-8, were installed at each house for one month. These filters have an efficiency of 50-85% for 3-10 μm particles when tested according to the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) Standard 52.2. The filters were placed in return grilles located on the ceiling or the wall in each home. After sampling, filters were removed, sealed in static shielding bags, and transported back to Austin, Texas inside an ice box and stored in the laboratory at -18°C until further processing. Settled dust samples were collected immediately before removal of the installed HVAC filter at each sampling site, using a previously reported protocol (Bi et al., 2015). Briefly, a modified vacuum cleaner was used, which was connected to a special aluminum nozzle holder to avoid contact between dust and plastic parts and limit potential contamination. Settled dust (approximately 100 mg) was collected onto a pre-weighed cellulose thimble filter (26 × 60 mm, Whatman Inc.) inserted into the nozzle holder. Dust sampling was conducted mainly in children's rooms. Dust was collected from the floor surface and from objects within 30 cm above the floor. After sampling, cellulose thimbles were placed in pre-cleaned amber glass jars and transported at 4°C to the laboratory for storage at -18°C.

Six phthalates and five organophosphates were targeted in this study, and their names, abbreviations, and CAS numbers are listed in Table C1. Two small pieces of filter samples (10 × 10 cm) were cut out of each HVAC filter obtained at randomly chosen positions, followed by determination of area and weight of the filter pieces. The remaining filter section was vacuumed, and a piece of cleaned filter (100 cm^2) was obtained and weighed using a five-digit microbalance. The mass of dust on HVAC filters was determined from the difference in mass between the filter samples and the cleaned filter piece. Prior to analysis, the HVAC filter and settled dust samples were fortified with known amounts of surrogate standards (i.e., dibenzyl phthalate, diphenyl isophthalate, and diphenyl phthalate) to explore extraction efficiency. The dust samples were then ultrasonically extracted for 30 minutes with hexane, which was repeated three times per sample. Extracts were concentrated using a rotary evaporator (IKA RV-10) and a high purity N_2 stream. After concentrating to approximately 5 mL, the samples were filtered (Whatman AutovialTM syringless PTFE filters with 0.2 μm pore). Next, a nitrogen purge was used to further concentrate the samples to approximately 500 μL. Analysis of the extracted samples was conducted by gas chromatography (GC) coupled with a flame ionization detector (Agilent 7890A GC-FID). Details of the method are provided in the Supplementary Materials.

Table C1. Abbreviations, names, and CAS numbers of the target compounds.

Abbreviation	**Name**	**CAS no.**
Phthalates		
DMP	Dimethyl phthalate	131-11-3
DEP	Diethyl phthalate	84-66-2
DnBP	Di-n-butyl phthalate	84-74-2
BBzP	Benzyl butyl phthalate	85-68-7
DEHP	Di-(2-ethylhexyl) phthalate	117-81-7
DnOP	Di-n-octyl phthalate	117-84-0
Organophosphates		
TnBP	Tri-n-butyl phosphate	126-73-8
TCPP	Tris (1-chloro-2-propyl) phosphate	13674-84-5
TBEP	Tri (2-butoxyethanol) phosphate	78-51-3
TPhP	Triphenyl phosphate	115-86-6
TDCPP	Tris (1,3-dichloroisopropyl) phosphate	13674-87-8

Quality assurance and control

Unused HVAC filters and cellulose thimbles were examined to determine background levels, which were below detection limits for all the target phthalates and organophosphates. Prior to sample measurements, matrix spiking experiments were conducted for all the target compounds; and the average recovery efficiencies were between 72% and 102%. Additionally, all samples were spiked with chemical standards prior to extraction to determine analytical extraction efficiencies, as described above. Each sample was weighed three times using a five-digit microbalance and mean weight was calculated. To check drift of the GC system, calibration standards were run after every six samples, and results were accepted only when the standards varied less than 10%. A calibration curve was constructed using six different concentrations in the range of 10-200 ng/μL for each of the 11 target compounds. Instrumental detection limits (IDLs) were estimated from calibration results based on evaluation guidelines for chromatographic analysis (OSHA, 1999). Method detection limits (MDLs) under the chromatographic conditions were calculated based on IDLs, median volume of extracts, and median weights of the dust samples. The IDLs and MDLs for phthalates and organophosphates are listed in Table C3. Field blanks (i.e., cellulose thimble filter) were collected and treated identically to the samples (except that no dust was vacuumed through the filter). These, along with laboratory blanks, were analyzed with the field samples. None of the target compounds were detected in these field and laboratory blanks. To avoid contamination, all glassware and metal ware used for sample collection and analysis were ultrasonically cleaned in methanol for 20 minutes, rinsed with methanol three times, and then baked in an oven prior to use.

Statistical analysis

For data analysis, dust concentrations below the limit of detection were assigned a value corresponding to half of the respective detection limit. A Shapiro-Wilk test of normality indicated that the dust concentrations of phthalates and organophosphates were not normally distributed. Thus, non-parametric Spearman's rank correlation analyses were performed to study the co-occurrence of compounds. Wilcoxon signed-rank test was used to compare the levels of target compounds between HVAC filter and settled dust during different seasons. Potential associations between concentrations of target SVOCs in dust and questionnaire data (i.e., building and personal characteristics) were analyzed with Mann-Whitney *U* test. Linear regression analyses were performed to examine the association between the severity of childhood asthma and SVOC concentrations in dust. For certain analyses, the total number of test samples was lower than 54, due to unavailability of participating homes or inadequate dust mass. Dimethyl phthalate (DMP), diethyl phthalate (DEP), tri-n-butyl phosphate (TnBP) and tri (2-butoxyethanol) phosphate (TBEP) were not included in the statistical analysis due to the relatively low detection frequency. Statistical analysis was carried out using R v3.4.4, an open-source statistical software environment; and a 5% significance level was applied.

RESULTS AND DISCUSSION

Concentrations of phthalates and organophosphates in dust

Phthalates and organophosphates were detected in all HVAC filter dust and settled dust samples. Their concentrations and detection rates are summarized in Table C2, in which the data for winter and summer seasons were combined (the data for each season are reported in Table C4 and discussed in the following sections). The mean concentration of each compound was found to be higher than the median value, suggesting a skewed concentration distribution as shown in the cumulative frequency distribution (Figure C5). In general, the measured concentrations varied considerably across all sampling sites, and the concentrations of phthalates were much higher than those of organophosphates. Little to no correlation for most of the phthalates and organophosphates were observed (Tables C5 and C6), indicating their indoor sources are diverse. However, concentrations of BBzP and DEHP in both HVAC filter and settled dust correlated significantly, suggesting these two compounds may have originated from the same sources.

For phthalates, the total concentrations of target compounds in HVAC filter dust ranged from 2.91 to 6370 µg/g, with a median value of 286 µg/g, while in settled dust, total phthalate concentration ranged from 26.4 to 5420 µg/g, with a median value of 288 µg/g. For the two-season combined data (Table C2), no significant concentration differences were observed between HVAC filter dust and settled dust samples for all phthalate compounds ($p > 0.05$, Mann-Whitney U test). The most frequently detected phthalate was DEHP for both HVAC filter and settled dust, followed by BBzP, di-n-octyl phthalate (DnOP), and DnBP. These are the same phthalates observed to be most abundant in urine samples of the U.S. population (Meeker and Ferguson, 2014; Zota et al., 2014). The compounds detected at the highest concentration in both filter and settled dust were DEHP and BBzP (HVAC filter dust: 3980 and 2440 µg/g, respectively; settled dust: 2120 and 2380 µg/g, respectively). These compounds are widely used as plasticizers in PVC products and building materials (e.g., flooring and wall covering) with material-phase concentrations of approximately 10% – 30% by weight (Liang and Xu, 2014b, 2014a). Health concerns surrounding phthalate plasticizers, especially DEHP and BBzP, have caused a trend toward using phthalates of lower volatility or alternative plasticizers (Schossler et al., 2011); however, the findings of the present study demonstrated that DEHP and BBzP are still the most abundant phthalates in indoor dust from low-income homes. The low detection rates of DMP and DEP are most likely due to its high volatility, with their majority present in gas phase rather than dust phase. Since very limited published data on phthalate levels in HVAC filter dust in residential homes are available, Figure C1a shows a comparison of measured phthalate concentrations with median values reported in the literature for settled dust in residential homes (Ait Bamai et al., 2014a; Albar et al., 2017; Bergh et al., 2011; Olivier Blanchard et al., 2014; Bornehag et al., 2005; Bu et al., 2016; Dodson et al., 2015; Fromme et al., 2004; Gevao et al., 2013; Guo and Kannan, 2011; He et al., 2016; Hsu et al., 2012; Kanazawa et al., 2010; Kubwabo et al., 2013; Langer et al., 2010; Luongo and Ostman, 2016; Rudel et al., 2003; Santillo et al., 2003; Shin et al., 2014; Subedi et al., 2017; Wang et al., 2014; Zhang et al., 2013). The median concentrations of DEHP (155 μg/g) and BBzP (20.1 μg/g) in settled dust from low-income houses determined in the present study were comparable to concentrations measured in common U.S. homes at other locations in the United States (Rudel et al., 2003; Guo and Kannan, 2011; Shin et al., 2014; Dodson et al., 2015), such as Richmond and Bolinas, CA (DEHP: 140 μg/g and BBzP: 19 μg/g), Albany, NY (DEHP: 304 μg/g and BBzP: 21.1 μg/g), Cape Cod, MA (DEHP: 340 μg/g and BBzP: 45.4 μg/g), and across the United States (DEHP: 144 μg/g and BBzP: 14.9 μg/g).

Table C2. Detection rates and the concentrations of phthalates and organophosphates in indoor dust.

	HVAC filter dust (μg/g), N = 91						**Settled dust (μg/g),** N = 92					
	>MDL[a]	Min	Median	Max	Mean	SD[b]	>MDL[a]	Min	Median	Max	Mean	SD[b]
Phthalates												
DMP	13%	<MDL	<MDL	19.0	1.19	2.73	28%	<MDL	<MDL	111	7.85	19.8
DEP	31%	<MDL	<MDL	163	3.54	17.3	7%	<MDL	<MDL	6.93	0.68	1.14
DnBP	52%	<MDL	1.60	6200	133	654	30%	<MDL	<MDL	950	115	228
BBzP	81%	<MDL	23.1	2440	80.8	270	80%	<MDL	20.1	2380	128	383
DEHP	98%	<MDL	155	3980	293	502	100%	12.8	155	2120	271	347
DnOP	59%	<MDL	5.27	358	11.7	40.3	74%	<MDL	9.65	355	24.6	45.4
Total		2.91	286	6370	524	894		26.4	288	5420	547	749
Organophosphates												
TnBP	1%	<MDL	<MDL	4.01	0.81	0.34	5%	<MDL	<MDL	18.3	1.24	2.46
TCPP	91%	<MDL	33.3	4090	150	529	77%	<MDL	5.22	418	15.8	45.5
TBEP	4%	<MDL	<MDL	124	6.04	13.5	3%	<MDL	<MDL	121	6.23	15.2
TPhP	43%	<MDL	<MDL	1930	46.4	233	36%	<MDL	<MDL	1190	16.8	124
TDCPP	11%	<MDL	<MDL	47.7	3.10	5.23	37%	<MDL	<MDL	122	8.89	18.2
Total		8.24	56.9	5190	206	738		8.24	19.3	1220	48.9	133

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[a] The method detection limits (MDLs) for dust samples were reported in the Supplementary Material. Detection rates are reported as % above MDL.

[b] Standard deviation.

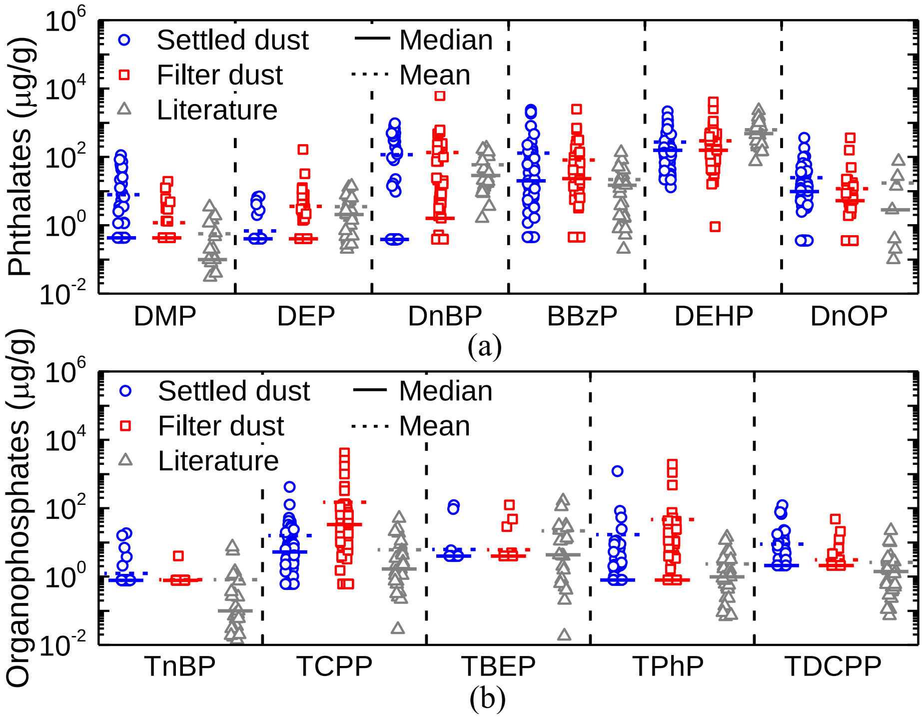


Figure C1. Comparison of measured concentrations of (a) phthalates and (b) organophosphates in dust with median values reported in literature for residential homes (over 4000 homes in 42 studies).

For organophosphates, the total concentrations of target compounds in HVAC filter dust ranged from 8.24 to 5190 μg/g with a median value of 56.9 μg/g, while in settled dust, concentrations ranged from 8.24 to 1220 μg/g, with a median value of 19.3 μg/g. Similar to phthalates, no significant concentration differences were observed between HVAC filter dust and settled dust samples for each compound in the two-season combined data ($p > 0.05$, Mann-Whitney *U* test), except for tris (1-chloro-2-propyl) phosphate (TCPP). In HVAC filter dust, TCPP was detected in the majority of the samples (91%) and at the highest concentrations (mean: 150 μg/g; range: <MDL to 4090 μg/g). Triphenyl phosphate (TPhP) and TDCPP were detected in a smaller percentage of samples (43% and 11%, respectively) and at lower concentrations (mean: 46.4 μg/g; range <MDL to 1930 μg/g and mean: 3.10 μg/g; range <MDL to 47.7 μg/g, respectively). In settled dust, the most abundant compounds were similar to those detected in HVAC filter dust, but

their levels and detection rates were lower, except for TDCPP and TnBP. TCPP is majorly applied in rigid polyurethane foams (PUFs) for building insulation (BAUA 2008a). TPhP is used as a flame retardant and/or a plasticizer in polymeric materials and products such as wallpaper, LCD-TV, computers, curtains, and insulation boards (Kajiwara et al., 2011). TDCPP and TCPP have been detected in flexible PUFs furnishings including couches, chairs, mattress pads, and pillows (Stapleton et al., 2009, 2011). The widespread use of these compounds in building materials and consumer products may contribute to their high levels in both the HVAC filter dust and settled dust samples. These results are in agreement with previous research assessing exposure to organophosphates, in which the metabolites of TDCPP, TPhP, and TCPP were the most frequently detected biomarkers in humans (Stapleton et al., 2009; Cooper et al., 2011; Meeker et al., 2013; Butt et al., 2014; Dodson et al., 2014; Jayatilaka et al., 2017; Ospina et al., 2018). Figure C1b shows a comparison of measured organophosphate concentrations to median values reported in the literature for settled dust in residential homes (Abdallah and Covaci, 2014; Ali et al., 2016, 2013, 2012; Araki et al., 2014; Bergh et al., 2011; Brommer and Harrad, 2015; Castorina et al., 2017; Cequier et al., 2014; Dodson and Perovich, 2012; Fan et al., 2014; He et al., 2016; Hoffman et al., 2015; Kanazawa et al., 2010; Kim et al., 2013; Langer et al., 2016; Luongo and Ostman, 2016; Meeker and Stapleton, 2010; Schreder and La Guardia, 2014; Shin et al., 2014; Stapleton et al., 2014, 2009; Tajima et al., 2014; Van den Eede et al., 2011; Wu et al., 2016; Zhou et al., 2017). The median concentrations measured in settled dust in the present study were comparable to those reported in the literature, except for TCPP. The median level of TCPP in the present study was 3- to 180-times higher than reported levels in Belgium, Canada, China, Denmark, Egypt, Japan, New Zealand, Kingdom of Saudi Arabia, and Sweden, (Abdallah and Covaci, 2014; Ali et al., 2012, 2016; Bergh et al., 2011; Fan et al., 2014; He et al., 2016; Tajima et al., 2014; Van den Eede et al., 2011; Wu et al., 2016), and was slightly higher than levels reported in U.S., Germany, and Norway (Stapleton et al., 2009, 2014; Dodson and Perovich, 2012; Cequier et al., 2014; Schreder and La Guardia, 2014; Zhou et al., 2017), but was relatively lower than levels reported in two studies in Japan (Kanazawa et al., 2010; Araki et al., 2014). The high level of TCPP in dust samples in the present study could be attributed to the use of rigid PUFs as roof and wall insulation for low-cost housing, especially considering that over 80% of produced TCPP is used in rigid PUFs, mainly for construction applications (BAUA, 2008).

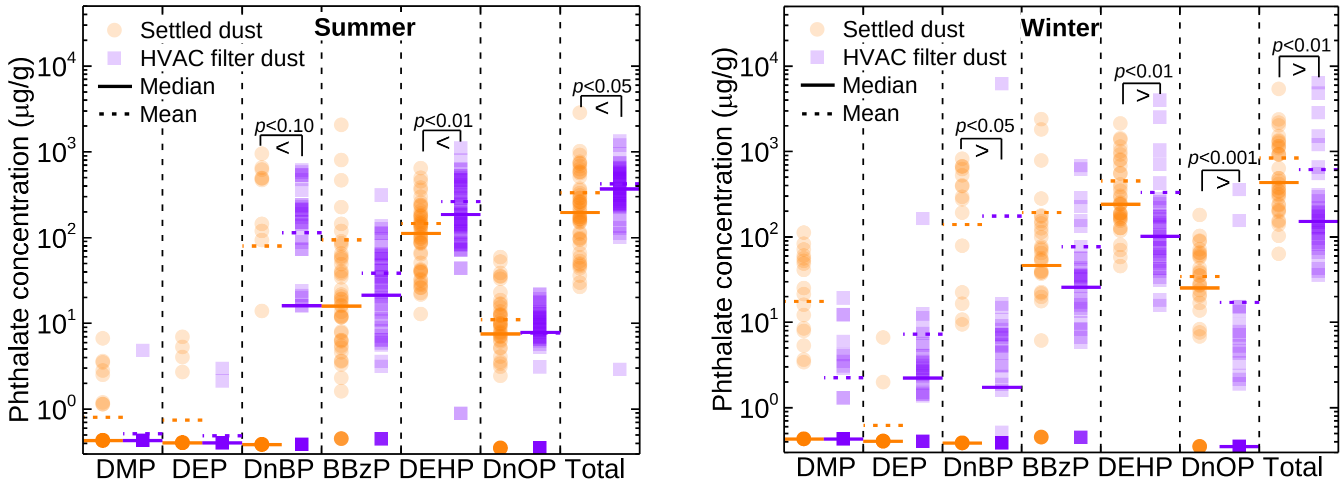
Comparison between HVAC filter dust and settled dust

Spearman's rank correlation analysis was applied to explore the possible correlations between HVAC filter dust and settled dust. Significant positive correlation for TCPP (r_s = 0.41, p = 0.0001) was observed, suggesting their common sources inside the house. Currently, the vast majority of TCPP is used in rigid PUFs as construction materials and a small fraction is used in flexible PUFs for furniture and upholstery (WHO, 2004; BAUA, 2008). Therefore, it is possible that a large amount of TCPP was emitted from walls, roofs, and/or furniture into the air and then captured by suspended particles and floor dust via the same gas/particle absorptive partitioning process. Since the equilibration time of TCPP is short due to its relatively small K_{oa} (Weschler and Nazaroff, 2008, 2010), sorption equilibrium between the gas and particle phases may occur at a fast rate. TCPP has been detected in indoor air at concentrations ranging from 2 ng/m^3 to 1300 ng/m^3 in residential homes (Bergh et al., 2011; Marklund et al., 2005; Saito et al., 2007) and was recently classified by the U.S. EPA as having a high hazard for reproductive and developmental effects (US EPA, 2015). No such correlation between HVAC filter dust and settled dust was observed for other target compounds, suggesting different sources and/or different environmental fates for these compounds in an indoor environment.

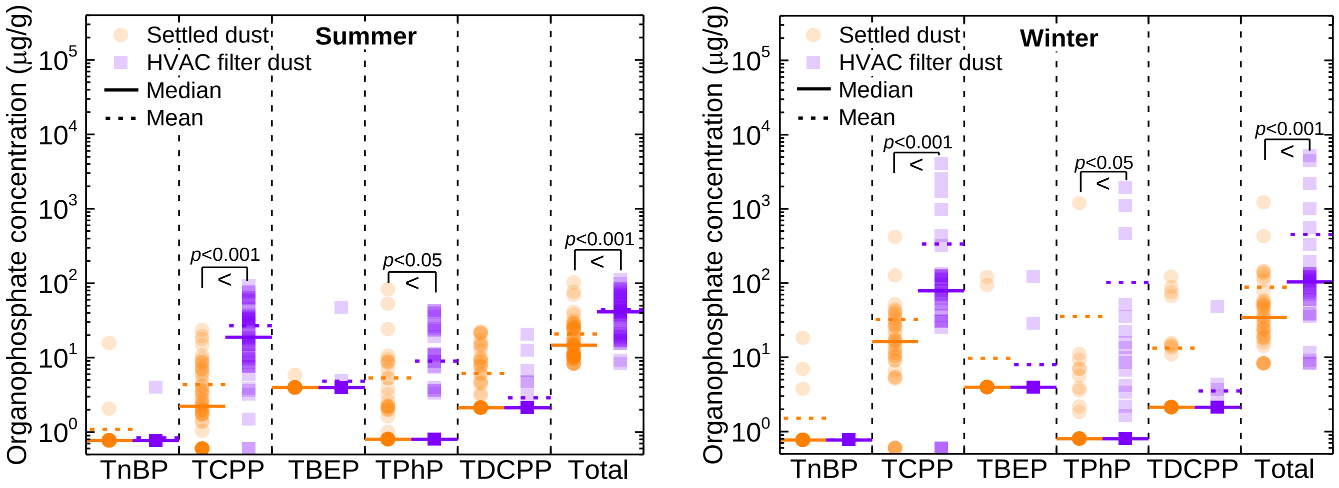
HVAC filter dust and settled dust samples were then paired for each single house and compared by season using paired Wilcoxon signed rank tests. As shown in Figure C2, for most of the frequently detected compounds, statistically significant higher concentrations were observed in HVAC filter dust than in settled dust, except for the case of phthalates in winter. The median concentrations of DnBP, DEHP, and total phthalates in HVAC filter dust in summer were 16, 185, and 368 μg/g, respectively, which were higher than those in settled dust (0.39, 112, and 195 μg/g, respectively). Similarly, for organophosphates, the concentrations of TCPP, TPhP, and total organophosphates in HVAC filter dust were significantly higher than in settled dust in both summer and winter (Figure C2). In summer, the median levels of TCPP and total organophosphates in HVAC filter dust were 18.7 and 41.3 μg/g, respectively; while in settled dust TCPP and total organophosphate concentrations were 2.22 and 14.7 μg/g, respectively. In winter, the median levels of TCPP and total organophosphates in HVAC filter dust were 78.6 and 104 μg/g, respectively; while in settled dust were 16.2 and 34.3 μg/g, respectively.

One possible reason for the higher SVOC concentration in HVAC filter dust may be related to the smaller particle size in HVAC filter dust as compared to settled dust. HVAC filter dust predominately consists of particles suspended in indoor air, with a small portion of coarse particles originating from indoor surfaces due to particle resuspension caused by airflow and occupant activities such as walking, cleaning, and vacuuming (Mukai et al., 2009). In this study, particle size distribution was determined for filter dust samples with details described in Givehchi et al. (2018). The mean value of particle volume median diameters is 32.3 μm (SD = 4.78 μm). In contrast, settled dust consists of particles deposited from indoor aerosols, soil particles tracked inside by foot, and biologically derived material such as skin flakes. Settled dust is generally assumed to be a mixture of large particles that have high settling rates. Although its particle size distribution was not determined in this study, the volume or mass median diameters of settled dust particles were reported in other indoor studies, with a range of 86-685 μm (Lewis et al., 1999; Morawska and Salthammer, 2003; Cao et al., 2014b; Gustafsson et al., 2018). Previous studies (Weschler and Nazaroff, 2008, 2010) have demonstrated that the equilibration of an SVOC in dust is largely controlled by its absorption to the organic matter of dust. Considering that the fraction of organic matter in dust (f_{om}) increases as particle size decreases (Morawska and Salthammer, 2003), higher equilibrium concentration of SVOCs are expected in smaller-sized particles such as HVAC filter dust. Additionally, the time scale for SVOCs to achieve the equilibrium concentration in dust is also affected by particle size. Smaller particles show faster mass transfer rates due to their larger specific surface area and higher mass transfer coefficients (Liu et al., 2014a). The high-velocity airflow pulled by a fan in the HVAC system further enhances the mass transfer coefficient between gas and particle phases (EI-Amin, 2011; Clack, 2017). As a result, the levels of SVOCs in HVAC filter dust may be close to their equilibrium concentrations, if the filters were installed for a long enough period of time. In contrast, measured SVOC concentrations in settled dust are observed to be systematically less than their predicted equilibrium levels in typical indoor conditions, particularly for compounds with high K_{oa} values, due to slow sorption kinetics (Weschler and Nazaroff, 2010). Finally, the large volume of air that passed through HVAC filters increased the sensitivity of sampling and analysis. In this study, since the filters were typically installed at the return air grille indoors, the environmental conditions (e.g., temperature or humidity) were probably similar between HVAC filter dust and settled dust; therefore, if the presence of SVOCs in dust is dominated by the process of sorption from the gas-phase in indoor air, SVOC concentrations in HVAC filter dust should be no less than their concentrations in settled dust.

In winter, higher phthalate levels were observed in settled dust as compared to HVAC filter dust (Figure C2a). The median concentrations of DEHP, DnOP, and total phthalates in settled dust were 242, 25.3, and 434 µg/g, respectively, which were significantly higher than those in HVAC filter dust (101, 0.35, and 151 µg/g, respectively). Although no statistically significant



(a) Phthalates

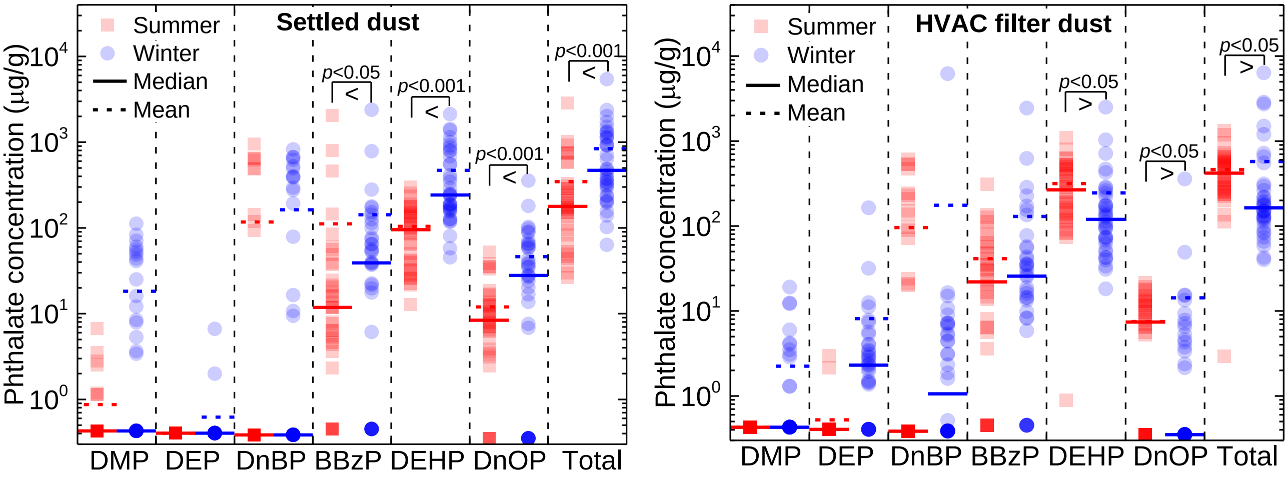


(b) Organophosphates

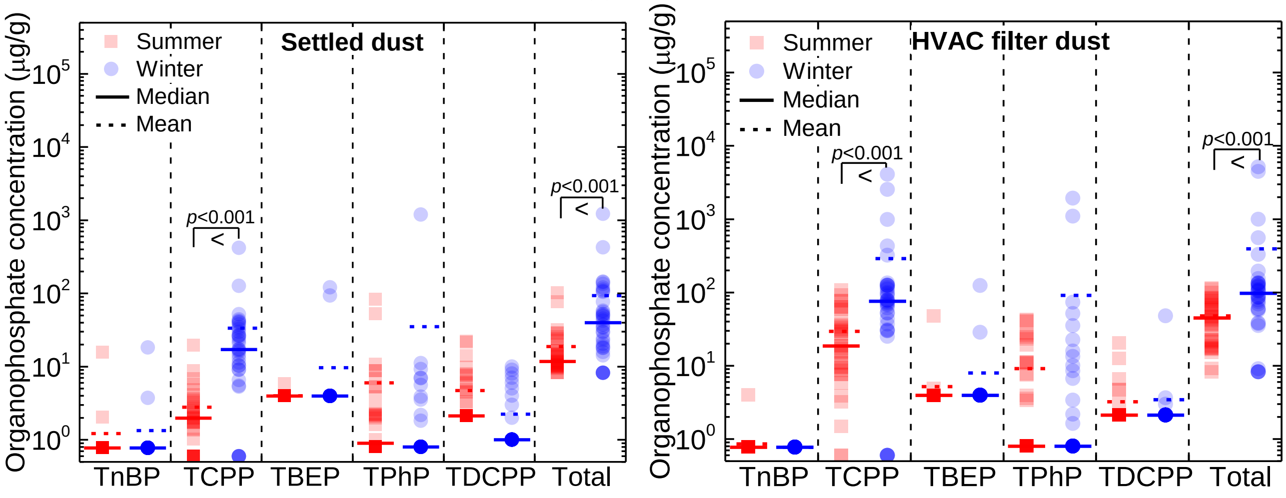
Figure C2. Comparison of SVOC concentrations between HVAC filter dust and settled dust in summer (N = 51) and winter (N = 36) difference was observed for BBzP, its median and mean levels in settled dust (46.1 and184 µg/g, respectively) was also higher than those in HVAC filter dust (26.2 and 138 µg/g, respectively). The high phthalate concentrations in settled dust could be attributed to the combination effect of local phthalate sources and use of space heaters in winter. Phthalates have been widely used in vinyl flooring as plasticizers. Liang and Xu (2014a, 2014b) demonstrated that 15 out of 16 vinyl flooring samples purchased in the U.S. in their study contained high concentrations of phthalates in the material phase. In the present study, approximately 40% of the low-income homes had vinyl flooring covering the entire floor area in the children's room. Phthalate concentrations in settled dust, which was in direct contact with vinyl flooring, were likely to be increased due to direct transfer from the source into dust (Bi et al., 2015; Bi and Xu, 2016; Sukiene et al., 2017). Furthermore, use of space heaters may substantially enhance the transfer of phthalates to dust. Space heaters were observed in children's rooms as well as other bedrooms in the sampling sites, possibly as a way to keep thermostat settings low and save on energy bills. Although the number of space heaters was not recorded, the runtime of the HVAC system was monitored at each site. The average runtime for each HVAC system in the winter was only 10%, which is much shorter than in the summer (31%). For a number of homes, the HVAC runtime during the winter was even less than 5%, but room temperatures were maintained approximately 10°C above the outdoor ambient temperature during cold weather. Use of space heaters can generate high temperatures on the immediate floor surface, and thus significantly increase the emission of phthalates from the source. Although dust/gas partition coefficient also decreases with increasing temperature, it may not offset the increase of emission, resulting in higher phthalate concentrations in settled dust. For example, when the surface temperature increased from 21 to 40°C, DEHP gas-phase concentration immediately adjacent to the surface of vinyl flooring increased by approximately 2-fold (Liang and Xu, 2014a), while the K_{oa} value of DEHP, which is proportional to the dust/gas partition coefficient, decreased from $10^{11.5}$ to $10^{10.4}$ (Xiao and Wania, 2003; Weschler and Nazaroff, 2010; Gobble et al., 2014; Bi et al., 2015). As a result, under equilibrium conditions, DEHP concentration in settled dust in contact with vinyl flooring increased by a factor of two. In addition, smaller particle size and lower dust loading on surfaces due to seasonal variation may further shorten the time to reach equilibrium and contribute to the high phthalate concentrations in settled dust in winter, as discussed in the next section.

Seasonal variation

Seasonal variation of SVOC contamination in indoor dust may significantly influence the reliability of human exposure assessments that are based on single-point sampling (Cao et al., 2014a). However, only a few studies of seasonal effects have been performed (Allen et al., 2008; Harrad et al., 2008; Batterman et al., 2009; Vorkamp et al., 2011; Yu et al., 2012; Cao et al., 2014a, 2015), and most of these were based on limited sampling sizes. The average daily outdoor air temperature in central Texas changed from 14.6±0.3°C in winter to 28.1±1.9°C in summer, while the average daily indoor air temperature changed slightly from 21.9±1.9°C in winter to 24.1±1.6°C in summer. Figure C3 shows the seasonal variation of phthalate and organophosphate concentrations in settled dust and HVAC filter dust. Significant seasonal differences were observed in settled dust, especially for phthalates. The median concentrations of BBzP, DEHP, DnOP, and total phthalates in settled dust in the winter were 39.1, 242, 27.9, and 468 μg/g, respectively, which were approximately three times higher than those in the summer (11.8, 95, 8.39, and 177 μg/g, respectively). For organophosphates, higher median concentrations of TCPP and total organophosphates were also observed in settled dust in winter (17.1 and 39.9 μg/g, respectively) than in summer (1.97 and 11.7 μg/g, respectively). The seasonal trend in the present work is similar to the few previous studies available (Yu et al., 2012; Cao et al., 2014a, 2015), which found higher winter concentrations in indoor settled dust for SVOCs including total organophosphates, hexabromocyclododecanes, and PBDEs. For HVAC filter dust, the seasonal trends were inconsistent: DEHP, DnOP, and total phthalates showed higher median concentrations in summer (266, 7.41, and 420 μg/g, respectively) than in winter (119, 0.35, 163 μg/g, respectively), while TCPP and total organophosphates showed higher median concentrations in winter (76.1 and 97.8 μg/g, respectively) than in summer (18.5 and 44.7 μg/g, respectively). To our knowledge, no studies have examined the seasonal influences on SVOC concentrations in HVAC filter dust, but several groups measured their levels in indoor suspended particles. Zhang et al. (2014) found significantly higher concentrations of phthalates on PM_{10} and $PM_{2.5}$ in summer as compared to winter, while Mandin et al. (2016) observed opposite results.



(a) Phthalates



(b) Organophosphates

Figure C3. Seasonal concentration variations of (a) phthalates and (b) organophosphates in HVAC filter and settled dust (N = 36).

There are many factors that may contribute to the seasonal variations in the concentrations of phthalates and organophosphates in dust. Temperature is one of the most important factors, which could have a significant influence on the SVOC equilibrium concentration in dust. For equilibrium conditions, an SVOC's concentration in dust (C_{dust}) is directly proportional to its gaseous concentration (C_g), to the K_{oa} value, and to the fraction of organic matter in dust (f_{om}), or $C_{dust} = C_g \times K_{oa} \times f_{om}/\rho_{dust}$ (Weschler and Nazaroff, 2010). The change of C_{dust} with an increasing temperature mainly depends on competition between the extent of the K_{oa} decrease and the increase in C_g. For example, based on the correlation developed by Liang and Xu (2014a), when indoor temperature increases from 21.9 to 24.1 °C, emission of DEHP from different vinyl flooring samples can be enhanced 1.5- to 3.4-times, while the K_{oa} value of DEHP decreased only by 30%, from 3×10^{11} to 2.2×10^{11} (Xiao and Wania, 2003). Therefore, if there are plenty of DEHP sources present in the indoor environment, such as the large surface area of vinyl flooring, it is possible to have a higher equilibrium concentration of DEHP in dust in the summer than in winter. In contrast, for other SVOCs, especially those with higher vapor pressures whose emissions are less sensitive to temperature changes (Kemmlein et al., 2003; Liang and Xu, 2014a), the decrease of K_{oa} may dominate the process, resulting in a lower equilibrium dust concentration in the summer. In addition to temperature, ventilation is another important factor. Better ventilation conditions may have a dilution effect of indoor contaminants and thus result in a lower gas-phase concentration (C_g) of SVOCs in air (Xu et al., 2009). However, ventilation inside residential buildings depends mainly on infiltration, weather conditions, and occupant behaviors (e.g., window opening). The air-tightness of the building envelope and living habits of the residents may not be consistent among different sampling sites. Finally, introduction or removal of source materials/products may have an immediate impact on the C_g of SVOCs, as previously demonstrated (Harrad et al., 2008; Zhang et al., 2009). These factors may complicate analysis of seasonal influences.

For HVAC filter dust, the concentration of phthalates and organophosphates might be close to their equilibrium concentration in dust due to small particle size, large airflow rates through filters, and long sampling durations (over one month). We observed higher concentrations of compounds with very low vapor pressure, such as DEHP and DnOP, in summer, but similar trends were not detected for other compounds. For settled dust, the unknown indoor residence time may have not been sufficient for the dust phase SVOC to equilibrate with its gas phase. Therefore, sorption kinetics directly impacted SVOC concentrations in dust. Some studies found that dust mass deposited on household surfaces during winter was less than during summer, and with smaller particle sizes (Edwards et al., 1998; Butte and Heinzow, 2002). The seasonal difference in dust properties may allow equilibrium partitioning to occur at a faster rate in winter than in summer. In the current study, higher levels of TCPP, total organophosphates, BBzP, DEHP, DnOP, and total phthalates in settled dust were observed in winter than in summer, which likely resulted from a combination of faster sorption kinetics, higher K_{oa} value, and poorer ventilation in winter. In addition, for phthalates, significantly enhanced emission from local sources (i.e., vinyl flooring) due to use of space heaters could also be an important factor, as discussed previously.

Influence of building characteristics

The association between the type of floor materials and concentration of target compounds in dust was investigated. Of all studied houses, 40% used PVC flooring, 30% had wall-to-wall carpet, and the rest had compressed wood, ceramic tile, or concrete flooring. As shown in Figure C4a, significantly higher median concentrations of BBzP and DEHP in settled dust were observed in homes with PVC flooring (BBzP: 39.8 and 13.6 μg/g in homes with and without PVC flooring, respectively, $p = 0.006$; DEHP: 189 and 127 μg/g in homes with and without PVC flooring, respectively, $p = 0.006$). After adjusting by season, statistical significance remained. The median concentrations of BBzP and DEHP in settled dust were observed higher in homes with PVC flooring in winter (BBzP: 75.3 and 6.09 μg/g in homes with and without PVC flooring, respectively, $p = 0.03$; DEHP: 553 and 187 μg/g in homes with and without PVC flooring, respectively, $p = 0.009$) and in summer (BBzP: 18.9 and 5.2 μg/g in homes with and without PVC flooring, respectively, $p = 0.009$; DEHP: 138 and 50.6 μg/g in homes with and without PVC flooring, respectively, $p = 0.007$). However, no such relationship between concentration of HVAC filter dust and floor type was found for any target compound. These results indicate that the content of BBzP and DEHP in settled dust in homes with PVC flooring may partially originate from the floor which the dust is in contact with. Recent studies (Bi et al., 2015; Sukiene et al., 2017) have observed direct transfer of DBiP and BBzP from vinyl flooring to dust on the floor surface, with an increased concentration greater than one order of magnitude relative to dust on other surfaces. Bornehag et al. (2005) also reported that concentrations of BBzP and DEHP in house dust were associated with the amount of PVC flooring in the house, which could further increase uptake of phthalates in infants (Carlstedt et al., 2013). Results in this study showing a correlation between the levels of BBzP and DEHP and use of PVC flooring in homes supports previous reports (Bornehag et al., 2005; Ait Bamai et al., 2014a; Bi et al., 2015; Sukiene et al., 2017).

Figure C4b shows that TDCPP levels in settled dust were significantly associated with the presence of carpets (median values of 14 and 2.1 µg/g in carpeted and uncarpeted homes, respectively, in winter; $p = 0.002$). When houses with vinyl flooring were excluded, significantly

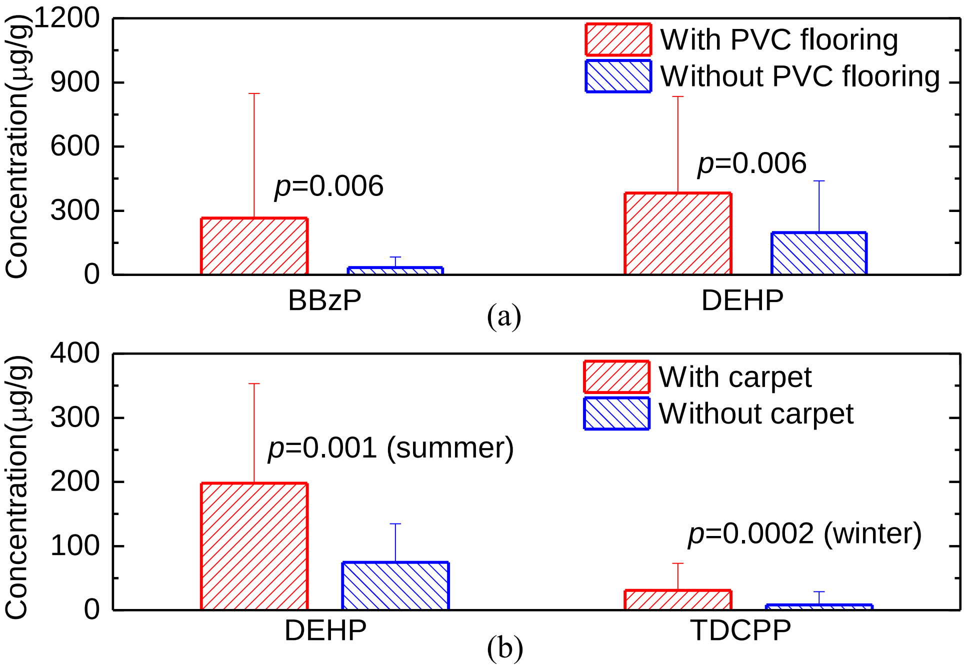


Figure C4. Phthalate and organophosphate concentrations in settled dust in relation to the presence of PVC flooring (a) and carpet (b). The *p*-values were calculated using the Mann–Whitney *U*-test and $p<0.05$ indicates a statically significant difference.

higher concentrations of DEHP were found in dust collected from carpeted homes (median values of 131 and 50.6 μg/g in carpeted and uncarpeted homes, respectively; in summer, $p = 0.001$). These results suggest that carpets may serve as a significant reservoir of phthalates and organophosphates, especially for those with relatively low vapor pressures. Carpets are one type of fleecy indoor material where particles containing absorbed SVOCs could accumulate, due to high particle deposition velocity on rough surfaces (Abadie et al., 2001) and low efficiency of removing dust from carpet (Yiin et al., 2007; Causer et al., 2008). Additionally, carpet has a large capacity to adsorb gas-phase SVOCs, due to its large specific surface area. Abb et al. (2009) found that the concentration of total phthalates in house dust increased as carpet area increased in a study conducted in Germany. Several studies in the U.S. and Australia (Castorina et al., 2017; He et al., 2018) found significantly higher dust loadings of TCEP, TCPP, TPHP, and TDCPP in houses with the presence of carpets, although a similar association was not found in Japan (Araki et al., 2014; Tajima et al., 2014). In addition, carpets could also serve as a source of organophosphates due to their use as flame retardants in carpet padding and fibers (Yadav et al., 2018). However, a recent study (Ionas et al., 2015) reported that organophosphates detected in carpet were typically at low levels (less than 0.01% by weight in the material phase). Generally, it is difficult to precisely identify the sources of SVOCs in indoor environments due to the complexity of the environment, existence of multiple sources, and limited sample size. Although some associations between the type of floor materials and concentration of organophosphates and phthalates were found in this study, further research is needed to confirm such findings.

The results of other associated building characteristics are listed in Tables C8 and C9. The average (± SD) floor area and house age were 160 (± 55) m^2 and 18 (± 13) years, respectively. Higher concentrations of DnBP, BBzP, DEHP, TCPP, and TPhP in indoor dust in different seasons were found in smaller homes. Since lower income populations commonly have smaller homes (Adamkiewicz et al., 2011), they are likely exposed to elevated concentrations of phthalates and organophosphates due to smaller house size, elevated source strength, and possibly inadequate ventilation. Newer buildings were found to have higher concentrations of TCPP in both HVAC filter dust and settled dust in the summer. Such a finding may be attributed to the increasing use of organophosphates in consumer products (as replacements of PBDEs), but there was no correlation between the concentration of other organophosphates in dust and the age of homes. Significant relationships were found between dust concentrations of target SVOCs and participants' house-cleaning activities. For vacuuming of carpets, the concentration of TDCPP in settled dust was lower in the high-frequency vacuuming group than in the low-frequency vacuuming group. In contrast, interestingly, for activities of cleaning furniture and hard floors, the concentrations of TCPP, TDCPP, and total organophosphates in HVAC filter dust and BBzP and TPhP in settled dust were significantly higher in the high-frequency cleaning group. This finding suggests that furniture and floor cleaning activities may actually cause contaminated dust to resuspend and become airborne, thereby increasing residents' inhalation exposure to some SVOCs. However, frequent vacuuming of carpets may help to decrease dust concentrations of certain SVOCs and thus reduce exposure.

Relationship with childhood asthma

For HVAC filter dust samples collected in the summer, a statistically significant linear correlation was found between DEHP concentration and asthma severity in children who had physician-diagnosed asthma ($R^2 = 0.36$, $N = 37$, $p < 0.001$), indicating slightly elevated DEHP levels are present in homes of children with greater asthma severity compared to those with lower severity. However, there was no statistically significant difference in DEHP concentration in HVAC filter dust between the asthma and the control groups. Also, no associations were found for winter sampling events (perhaps due to short winter season in the central Texas and/or short HVAC runtime in winter), for other phthalates or organophosphates, or in settled dust. The results suggest that it is only the severity of asthma in children with physician-diagnosed asthma that is associated with DEHP concentration found in home HVAC filters in the summer.

Several previous studies examined the association between phthalates in dust or PVC materials in homes and allergic diseases in children. In most of these studies, non-floor settled dust was sampled. Among Swedish children, Bornehag et al. (2004) showed an association between higher concentrations of DEHP in house dust and the occurrence of asthma. This association was also observed with house dust concentrations of BBzP and the presence of allergic rhinitis and atopic dermatitis. Similar results were found in Bulgaria and Taiwan, where positive associations between house dust DEHP or BBzP concentrations and childhood asthma, rhinitis, and eczema were reported (Kolarik et al., 2008a; Hsu et al., 2012a). However, results were not always consistent. Two studies conducted in Denmark and Japan (Callesen et al., 2014; Kanazawa et al., 2010) did not find a significant association between phthalates in dust and allergies in children. In another cross-sectional study (Ait Bamai et al., 2014a), levels of phthalates in floor dust had a linear association with the prevalence of allergic rhinitis, conjunctivitis, and atopic dermatitis in children; however, this association was not found for non-floor dust. A recent study in Denmark (Beko et al., 2015) involving children with asthma, allergic rhinitis, or eczema did not show a direct association between the occurrence of allergic disease and exposure to phthalates. However, among children with allergic disease, there was an association between indoor allergen sensitization and exposure to BBzP, DnBP, and DEHP. In another controlled study (Deutschie et al., 2008), 16 healthy human subjects and 16 human allergic to dust mites were exposed to house dust containing low and high concentrations of DEHP. Healthy subjects were not affected by the dust exposure, but the allergic group showed silent inflammation in nasal mucosa in response to the dust containing low DEHP doses. To some extent, our current finding that only the severity of asthma in children with asthma disease correlates with DEHP concentration in HVAC filter dust in the summer supported the results of Beko et al. (2015) and Deutschie et al. (2008).

The mechanism responsible for the association between phthalate exposure and doctor-diagnosed asthma remains unclear. Some *in vitro* and *in vivo* animal studies showed that phthalates or their metabolites may have different modes of action (e.g., direct pharmacological effect on receptors involved in allergenic pathology or indirect effect as adjuvants for different causative agents) to influence immune and inflammatory parameters that play a role in respiratory diseases (Oie et al., 1997; Larsen et al., 2002, 2003; Larsen and Nielsen, 2008; Bornehag and Nanberg, 2010; Kimber and Dearman, 2010; Beko et al., 2015; Franken et al., 2017). Additionally, indoor microbes may also influence the presence of asthma and asthma severity. Studies assessing increased exposure to indoor fungi prior to the development of asthma symptoms suggested that certain fungal species pose a respiratory health risk in susceptible populations (Sharpe et al., 2015). *In vivo* studies revealed that viable fungi can induce asthma-like disease in experimental animals supports the argument that fungi are possible etiologic agents of asthma (Porter et al., 2011; Porter et al., 2011). Several other studies found that exposure to bacterial endotoxin and mold also affect asthma severity in children and additional respiratory outcomes in subjects with and without allergies (Michel et al., 1996; Zambelli-Weiner et al., 2004; Mendell et al., 2011; Dannemiller et al., 2016).

Chemical and biological contaminants concurrently present in indoor environments may interact with each other. Biodegradation of phthalates, particularly DEHP, by a variety of bacteria and fungi present in household dust, PVC materials, and soil have been reported in previous studies (Webb et al., 2000; Nakamiya et al., 2005; Nalli et al., 2006b, 2006a; Liang et al., 2008; Pradeep et al., 2013). In the current project, significant associations between the concentrations of fungi and bacteria and the levels of DEHP in summertime HVAC filter dust were observed (reported in details in Maestre et al., 2018). Therefore, it is possible that DEHP concentrations and concentrations of fungi and/or bacteria are intimately related. Warm summer air, which contains more moisture than winter air, may strengthen this effect. However, we did not collect enough data to determine if the HVAC filters themselves were a source of air contamination and thus exacerbated asthma symptoms in children. Furthermore, certain toxic volatile organic compounds could be produced during the process of microbial degradation of plasticizers. Previous studies reported that microbial degradation of DEHP and di-2-ethylhexyl adipate (DEHA) was the most likely source of 2-ethylhexanol in indoor air (Nalli et al., 2006b, 2006a), which has been associated with nasal symptoms (Wieslander et al., 1999) or asthma (Norbäck et al., 2000; Kanazawa et al., 2010) in building occupants. Future confirmatory studies are needed to examine whether the presence of phthalates alters the indoor microbial communities and whether the interaction between indoor chemicals and the environmental microbiome results in adverse health effects.

Implications for exposure assessments

Phthalates and organophosphates measured in HVAC filter dust could be considered to represent what mostly went into the airway via inhalation, while those measured in settled dust was assumed to be digested through oral intake of contaminated particles adhered to food and objects. We compared the estimated daily intake of target compounds through these two exposure pathways. The results and the equations and parameters used in the estimation are listed in Tables C10 and C11. Median values of inhalation rates, dust ingestion levels, and body weights were used for 3-year-old children and adults, based on the EPA exposure factor handbook (US EPA, 2011). We assumed the average time spent at home to be 64% (Harrad et al., 2006). For all target SVOC compounds, the intake through ingestion of settled dust was significantly higher than via inhalation of suspended particles, especially for children. However, the association between target compounds in dust and asthma severity in children was found only for HVAC filter dust, but not settled dust. The results suggest that the impact of an SVOC compound on a particular health endpoint is related not only to the amount of intake but also to the route of exposure. As air travels through the respiratory tract via inhalation, contaminated particles may deposit in the respiratory tract and release (i.e., desorb) a fraction of SVOCs which then interact with the tissue. The subsequent effect depends on the type of the compound, the size of particles, and the region of the respiratory tract (Liu et al., 2017). Oie et al. (1997) found that inhalation exposure to particles containing DEHP was important due to its low clearance and extensive penetration into the pulmonary region. They reported that high local concentrations of DEHP can be created in the airways at the particle deposition site and the primary hydrolysis product of DEHP increased the risk of inducing inflammation in the airways, which is a characteristic of asthma. Therefore, to evaluate the health effects in respiratory tract such as asthma, it may be more direct, representative, and meaningful to use airborne particles than settled floor dust.

HVAC filter dust may become a useful sampling medium to measure SVOC concentrations in airborne particles. HVAC filters can be installed with minimal effort for a particular period of time and capture particles from large volumes of air, resulting in the collection of spatially and temporally integrated concentrations. The large volume of air that passes through the filters simply increases the sensitivity of sampling and analysis. This approach overcomes the limitation of traditional air sampling methods that only provides a "snapshot" of compound concentrations, which can vary significantly with time and within a building. Although filters may affect SVOC distribution in gas-, dust-, and filter-phases in a short term, because of sorption of gaseous species on the HVAC filter or filter cake or desorption of SVOCs from particles collected on the filter or the filter itself, it is expected that any impact will be minimal after the sorption equilibrium is established. When combined with HVAC system characterization such as system run-times, volumetric flow rates, and mass of dust collected, HVAC filter dust sampling provides a novel filter forensics methodology to detect and quantify particulate SVOC concentrations in indoor air. In contrast, settled floor dust can be used to assess SVOC exposures via inadvertent ingestion of dust, especially for young children who spend considerable time on the floor and frequently put their hands and other objects in their mouths. However, our findings show that when using house settled dust, it is important to note where the samples were collected from, because the type and level of SVOCs in floor dust may be directly related to local materials used in the house and could be different at other indoor locations. Additionally, since the age of settled dust is usually unknown, for SVOCs with high K_{oa} values, the mass fraction in settled dust may not have sufficient time to equilibrate with the gas phase concentration and thus may be underrepresented in settled dust. Seasonal variations were observed for both HVAC filter dust and settled dust, resulting from the combined effect of several factors discussed previously. The concentrations of some phthalates and organophosphates in settled dust in winter were generally higher than in summer, but the seasonal pattern was not well-defined for HVAC filter dust. More integrated and consecutive studies involving seasonal variations of SVOCs in HVAC filter dust and settled dust should be conducted in the future.

Limitations

There are several limitations of this study that should be considered. First, this study had a cross-sectional design and environmental measurements were conducted only twice. Seasonal and environmental factors may affect the composition of SVOCs in dust. Second, the sample size was relatively small, which prevented the use of certain statistical tests such as multivariate regression analysis. It is possible that, because of the sample size, some observed statistically significant relations were obtained by chance. Third, the study only included low-income housing with children; other homes may differ in regard to the quantity and quality of furniture, building materials, consumer products, and toys. However, the results of the associations between the type of floor materials and levels of phthalates and organophosphates are likely applicable to other situations. Fourth, environmental measurements were made only in participants' homes, while other indoor environments, such as daycare centers or schools, where a child spends extended periods of time may also contribute to their exposure meaningfully. Fifth, settled dust was only collected from the children's room in each house, which may be less relevant if the child spends the majority of time in the main living area of the house. Lastly, the presence of other SVOC compounds in dust may affect our results, and the limits of detection may not have been low enough to detect certain trace-level compounds. Therefore, more groups of SVOCs should be included and more sensitive detection approaches should be used in future studies.

CONCLUSIONS

This was the first study to measure concentrations of phthalates and organophosphates in HVAC filter dust collected from residential homes and to investigate their associations with settled dust, seasonal variation, building characteristics, and childhood asthma. We found that BBzP, DEHP, DnOP, TCPP, TPhP, and TDCPP were the most frequently detected compounds in both

HVAC filter dust and settled dust. The median level of TCPP in settled dust was 3- to 180-times higher than levels reported in other studies of residential homes, which was likely due to the use of TCPP in roof and wall insulation installed in low-cost housing units. No correlation between HVAC filter dust and settled dust was observed for the target compounds other than TCPP. Significantly higher concentrations in HVAC filter dust than in settled dust were observed for most of the frequently detected compounds in the summer and winter, except for several phthalates in winter, possibly due to the direct transfer of phthalates from floor surfaces to dust and the use of space heaters in winter. For settled dust, the concentrations of target compounds in winter were generally higher than in summer. In contrast, the seasonal trend was inconsistent for HVAC filter dust, with higher concentrations in summer than in winter for DEHP and DnOP, while the opposite trend was seen for TCPP. This pattern may be attributable to the fact that emission of SVOCs with low vapor pressures are more sensitive to temperature changes as compared to those with high volatilities. Settled floor dust samples from homes with vinyl flooring contained significantly higher levels of BBzP and DEHP as compared to homes with other types of floor material. The concentration of DEHP and TDCPP in settled dust also significantly correlated with the presence of carpet in homes. Cleaning activities to remove dust from furniture or hard floors may actually cause contaminated dust to resuspend into the air, thereby increasing the levels of some compounds in HVAC filter dust, while frequent vacuuming of carpets may help to decrease the concentration of certain compounds in settled dust. Additionally, the levels of certain SVOCs in dust correlated with the size and the age of a given house. A statistically significant association was found between DEHP concentration in HVAC filter dust in the summer and the severity of asthma in children with asthma diesease. Since this DEHP concentration also correlated with concentrations of indoor bacteria and fungi, DEHP in airborne particles may not be the only factor that exacerbates asthma symptoms in children. Overall, the results suggest that HVAC filter dust is a useful sampling medium to monitor indoor SVOC concentrations. When using settled dust, in addition to consideration of seasonal influences, it is important to know the sampling location and type of surface because the type and level of SVOCs may be related to local materials used in the house.

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SUPPLEMENTARY MATERIAL

Chemical analysis

Phthalates and organophosphates were analyzed using an GC-FID system equipped with a DB-5ht column (30m, 0.25mm, 0.1μm). The GC-FID system was operated using a 4:1 split injection, with an injection temperature of 275 °C. The GC temperature program was 120 °C, hold 2 min, ramp 12°C/min for 15 min, hold 3 min, then ramp 20 °C/min for 2 min, hold 2 min. The detector was set at 320 °C. Helium was used as a carrier gas with a flow rate of 1.2 ml/min. The instrumental and method detection limits are given in Table C3.

Table C3. Instrumental detection limits (IDLs) and method detection limits (MDLs) of target compounds.

Abbreviation	Name	IDLs (ng)	MDLs (µg/g)
Phthalates			
DMP	Dimethyl phthalate	1.43	0.86
DEP	Diethyl phthalate	1.35	0.81
DBP	Di-n-butyl phthalate	1.28	0.77
BBzP	Benzyl butyl phthalate	1.50	0.90
DEHP	Bis (2-Ethylhexyl) phthalate	2.97	1.78
DnOP	Di-n-octyl phthalate	1.17	0.70
Organophosphates			
TnBP	Tri-n-butyl phosphate	2.57	1.54
TCPP	Tris (1-Chloro-2-propyl) phosphate	1.98	1.19
TBEP	Tri (2-Butoxyethanol) phosphate	13.2	7.92
TPhP	Triphenyl phosphate	2.66	1.59
TDCPP	Tris (1,3-dichloroisopropyl) phosphate	7.06	4.23

Table C4. Detection rates and the concentrations of phthalates and orgaophosphates in HVAC filter and settled dust (µg/g).

Winter	**HVAC filter dust**							**Settled dust**						
	N[a]	>MDL[b] (%)	Min	Median	Max	Mean	SD[c]	N[a]	>MDL[b] (%)	Min	Median	Max	Mean	SD[c]
Phthalates														
DMP	39	28	<MDL	<MDL	19.0	2.10	3.93	38	47	<MDL	<MDL	111	17.8	27.8
DEP	39	67	<MDL	2.27	163	7.60	25.9	38	5	<MDL	<MDL	6.64	0.61	1.02
DnBP	39	49	<MDL	<MDL	6200	162	979	38	45	<MDL	<MDL	823	154	242
BBzP	39	82	<MDL	26.2	2440	138	402	38	74	<MDL	46.1	2380	184	474
DEHP	39	100	15.8	113	3980	330	724	38	100	45.3	242	2120	454	460
DnOP	39	46	<MDL	<MDL	358	17.2	60.8	38	74	<MDL	27.1	355	44.5	64.0
∑ Phthalates	39		35.6	154	6370	657	1320	38		63.4	468	5420	856	969
Organophosphates														
TnBP	39	0	<MDL	<MDL	<MDL	-	-	38	8	<MDL	<MDL	18.3	1.47	2.97
TCPP	39	85	<MDL	77.9	4080	313	779	38	87	<MDL	16.8	418	32.3	67.2
TBEP	39	5	<MDL	<MDL	124	7.68	19.3	38	5.3	<MDL	<MDL	121	9.41	23.3
TPhP	39	41	<MDL	<MDL	1930	96.5	350	38	26	<MDL	<MDL	1190	33.4	191
TDCPP	39	10	<MDL	<MDL	47.7	3.41	7.21	38	26	<MDL	<MDL	122	12.9	26.9
∑ Organophosphates	39		8.24	103	5190	421	1090	38		8.24	36.1	1220	89.6	199

Summer	**HVAC filter dust**							**Settled dust**						
	N[a]	>MDL[b] (%)	Min	Median	Max	Mean	SD[c]	N[a]	>MDL[b] (%)	Min	Median	Max	Mean	SD[c]
Phthalates														
DMP	52	2	<MDL	<MDL	4.85	0.51	0.61	54	15	<MDL	<MDL	6.66	0.78	1.08
DEP	52	4	<MDL	<MDL	2.98	0.49	0.42	54	7	<MDL	<MDL	6.93	0.73	1.21
DnBP	52	52	<MDL	28.1	3370	754	1070	54	20	<MDL	<MDL	950	87.2	214
BBzP	52	81	<MDL	21.1	311	37.8	51.8	54	85	<MDL	12.9	2060	88.7	298
DEHP	52	96	<MDL	189	1100	267	217	54	100	12.8	110	643	142	128
DnOP	52	69	<MDL	7.66	22.0	7.58	6.10	54	74	<MDL	7.34	59.3	10.7	12.8
∑ Phthalates	52		2.91	370	1328	424	251	54		26.47	189	2840	330	426
Organophosphates														
TnBP	52	2	<MDL	<MDL	4.01	0.83	0.44	54	4	<MDL	<MDL	15.7	1.07	2.02
TCPP	52	96	<MDL	19.2	108	28.4	24.3	54	70	<MDL	2.16	23.54	4.18	5.01
TBEP	52	4	<MDL	<MDL	47.5	4.82	5.98	54	1.9	<MDL	<MDL	5.87	4.00	0.26
TPhP	52	44	<MDL	<MDL	42.7	8.85	12.7	54	43	<MDL	<MDL	82.5	5.08	13.2
TDCPP	52	12	<MDL	<MDL	20.6	2.88	2.96	54	44	<MDL	<MDL	22.4	6.01	5.94
∑ Organophosphates	52		8.24	41.3	115	45.7	26.0	54		8.24	14.6	101	20.3	17.4

[a] Number of homes sampled.

[b] The method detection limits (MDLs) for dust samples were reported in the Supplementary Material. Detection rates are reported as % above MDL.

[c] Standard deviation.

Table C5. Spearman’s rank correlation coefficients between concentrations of different compounds in HVAC filter dust. Values marked in bold are statistically significant with *p*-values < 0.05.

	DnBP	BBzP	DEHP	DnOP	TCPP	TPhP	TDCPP
DnBP							
BBzP	-0.103						
DEHP	**-0.364**	**0.267**					
DnOP	0.145	0.100	0.000				
TCPP	**-0.260**	0.073	-0.023	**-0.243**			
TPHP	**-0.236**	0.042	**0.226**	0.151	-0.107		
TDCPP	-0.105	0.035	-0.007	0.077	0.003	**0.214**	

Table C6. Spearman’s rank correlation coefficients between concentrations of different compounds in settled dust. Values marked in bold are statistically significant with *p*-values < 0.05.

	DnBP	BBzP	DEHP	DnOP	TCPP	TPhP	TDCPP
DnBP							
BBzP	0.092						
DEHP	0.162	**0.388**					
DnOP	**0.263**	-0.102	0.100				
TCPP	0.145	**0.299**	**0.509**	0.045			
TPHP	-0.062	0.066	-0.052	-0.025	-0.041	-0.014	
TDCPP	-0.125	0.173	-0.010	-0.237	-0.014	0.170	

Table C7. Spearman’s rank correlation coefficients between SVOC concentrations in HVAC filter dust and in settled dust. Values marked in bold are statistically significant with *p*-values < 0.05.

	DnBP	BBzP	DEHP	DnOP	∑ Phthalates	TCPP	TPhP	TDCPP	∑ Organophosphates
Correlation coefficient	0.051	0.096	-0.166	-0.069	-0.126	**0.405**	0.151	0.001	0.258
p-value	-0.219	0.379	0.125	0.522	0.244	**0.0001**	0.163	0.990	0.016

Table C8. Spearman's correlation coefficients between the age and area of house and the concentrations of phthalates and organophosphates in dust. Values marked in bold are statistically significant with *p*-values < 0.05.

	Age of house (years)		Area of house (ft^2)	
	HVAC filter dust	Settled dust	HVAC filter dust	Settled dust
Winter				
Phthalates				
DnBP	-0.023	0.149	-0.067	-0.232
BBzP	-0.087	0.215	-0.206	**-0.351**
DEHP	0.116	0.352	0.058	-0.224
DnOP	0.006	0.258	-0.028	0.034
Organophosphates				
TCPP	-0.253	-0.078	-0.016	**-0.359**
TPhP	**0.353**	-0.225	-0.114	-0.147
TDCPP	-0.058	-0.177	0.146	0.101
Summer				
Phthalates				
DnBP	0.195	0.209	**-0.497**	**-0.404**
BBzP	0.097	-0.236	-0.102	-0.121
DEHP	0.257	**-0.344**	**-0.441**	0.161
DnOP	0.218	0.001	-0.071	-0.074
Organophosphates				
TCPP	**-0.399**	**-0.547**	0.041	0.113
TPhP	0.064	0.082	**-0.471**	-0.034
TDCPP	-0.106	-0.005	-0.247	-0.003

Table C9. Relationships between housing characteristics and concentrations of phthalates and organophosphates in dust. Mean levels and standard deviations (std) of target compounds in dust (µg/g) for each binary category are presented and significant variables are bolded and indicated with asterisks (* $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$). The *p*-values were calculated using the Mann–Whitney *U*-test.

Building factors	N	DnBP		BBzP		DEHP		DnOP		Phthalates		TCPP		TPhP		TDCPP		Organophosphates	
		mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Settled dust																			
Stuffed toy																			
Yes	47	136	258	129	402	256	267	16.2	23.8	545	610	18.2	60.8	4.50	12.4	10.5	21.4	40.4	65.4
No	43	96.6	200	127	380	294	431	34.7	61.0	564	906	13.0	21.6	31.0	182	7.13	14.8	59.0	184
Pet																			
Yes	60	127	236	96.9	324	308	406	26.3	54.5	568	816	13.1	20.1	3.77	11.0	8.88	19.7	34.4	35.9
No	30	98.1	226	192	496	206	202	22.5	22.2	525	650	20.9	75.5	43.9	218	8.98	16.4	79.3	229
Vacuum on carpets																			
<once a week	9	75.9	217	340	689	216	142	16.6	20.0	653	904	9.62	8.42	3.51	3.93	**22.3*** **	**25.8**	40.1	32.3
>=once a week	42	115	238	127	383	304	392	25.5	61.7	580	887	12.9	21.5	4.20	12.9	**8.99**	**20.9**	35.8	39.4
Clean hard floors																			
<once a week	14	50.8	132	20.5	26.0	163	102	19.4	27.2	256	178	7.55	8.35	**1.25**	**1.64**	11.4	19.7	25.2	25.7
>=once a week	76	129	244	148	421	294	379	26.1	49.0	609	814	17.2	50.1	**20.1***	**137**	8.44	18.4	53.8	147
Clean furniture dust																			
Seldom or sometimes	37	75.5	193	**22.6**	**32.6**	251	286	28.9	60.2	386	427	9.64	12.0	35.2	196	10.9	22.2	63.8	198
Always or often	47	127	235	**218*** **	**524**	317	411	20.9	33.4	695	954	21.8	62.7	4.92	13.9	6.46	13.1	40.7	65.2
Change HVAC filters																			
<=once a month	39	99.0	193	49.1	81.6	264	314	21.0	27.9	442	431	20.7	66.6	33.0	191	6.34	12.2	70.7	202
>once a month	51	131	259	189	506	281	383	28.1	56.4	639	934	11.9	19.9	4.99	13.6	10.8	22.2	33.0	33.9

Building factors	N	**Filter dust**																	
		DnBP		BBzP		DEHP		DnOP		Phthalates		TCPP		TPhP		TDCPP		Organophosphates	
		mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Stuffed toy																			
Yes	49	196	885	108	364	276	565	9.14	22.8	596	1140	159	620	39.5	168	2.55	1.68	206	788
No	40	62.5	156	50.2	76.1	322	440	15.0	55.9	453	508	143	423	56.7	304	3.83	7.70	213	712
Pet																			
Yes	56	174	831	52.6	107	282	542	7.73	20.8	519	1030	88.8	255	17.4	63.4	3.59	6.66	115	309
No	33	72.1	146	132	429	321	459	18.7	61.7	552	666	260	815	97.7	380	2.34	0.71	369	1160
Vacuum on carpets																			
<once a week	8	47.8	82.7	27.4	38.9	178	175	3.11	3.48	258	192	39.2	24.3	5.88	11.9	2.12	0.00	55.1	31.0
>=once a week	43	74.9	151	51.4	71.9	216	172	6.14	8.48	352	254	71.2	149	7.43	12.7	3.36	6.97	86.7	148
Clean hard floors																			
<once a week	13	59.0	87.2	35.6	50.6	224	164	10.1	13.2	332	197	**24.3**	**23.4**	6.96	13.5	2.12	0.00	38.1	26.7
>=once a week	76	149	718	90.0	296	309	548	12.0	44.1	565	977	**174***	**580**	54.1	256	3.30	5.74	239	809
Clean furniture dust																			
Seldom or sometimes	37	243	1020	99.0	398	189	194	7.27	8.61	543	1090	**42.9**	**73.6**	7.54	15.0	**2.19**	**0.43**	**60.7**	**92.3**
Always or often	47	66.0	150	69.9	139	336	592	8.31	22.7	486	727	**178****	**631**	42.2	171	**3.95***	**7.24**	**230****	**801**
Change HVAC filters																			
<=once a month	37	210	1020	132	418	331	657	9.50	25.2	691	1300	225	722	50.3	193	2.84	3.07	285	910
>once a month	52	83.2	167	46.3	64.3	272	379	13.4	49.4	418	445	100	353	45.0	266	3.33	6.48	156	617

Table C10. Equations and parameters used to calculate exposure to phthalates and organophosphates through inhalation of suspended particles and ingestion of settled dust

Exposure pathway	Daily exposure dose (µg/kg-bw/day)	Parameters	Parameter values
Inhalation of particle-phase SVOCs	Exposure dose = $\frac{C_{FD}\times TSP\times InhR\times ED}{BW}$	C_{FD}= particle-phase concentrations of SVOCs measured in filter dust (µg/g)	Mean concentration of SVOCs in filter dust measured in this study
		TSP = total suspended particle concentrations (µg/m^3)	20 µg/m^3. [a]
		$InhR$ = inhalation rate (m^3/day)	Child: 10.3; adult: 16. [b]
		ED = exposure duration (-)	0.64. [c]
		BW= body weight (kg)	Child: 18.6; adult: 80. [b]
Ingestion of settled dust	Exposure dose = $\frac{C_{SD}\times IngR\times ED}{BW}$	C_{SD}= concentrations of SVOCs measured in settled dust (µg/g)	Mean concentration of SVOCs in settled dust measured in this study;
		$IngR$ = dust ingestion rate (mg/day)	Child: 60; adult: 30. [b]
		ED = exposure duration (-)	0.64. [c]
		BW= body weight (kg)	Child: 18.6; adult: 80. [b]

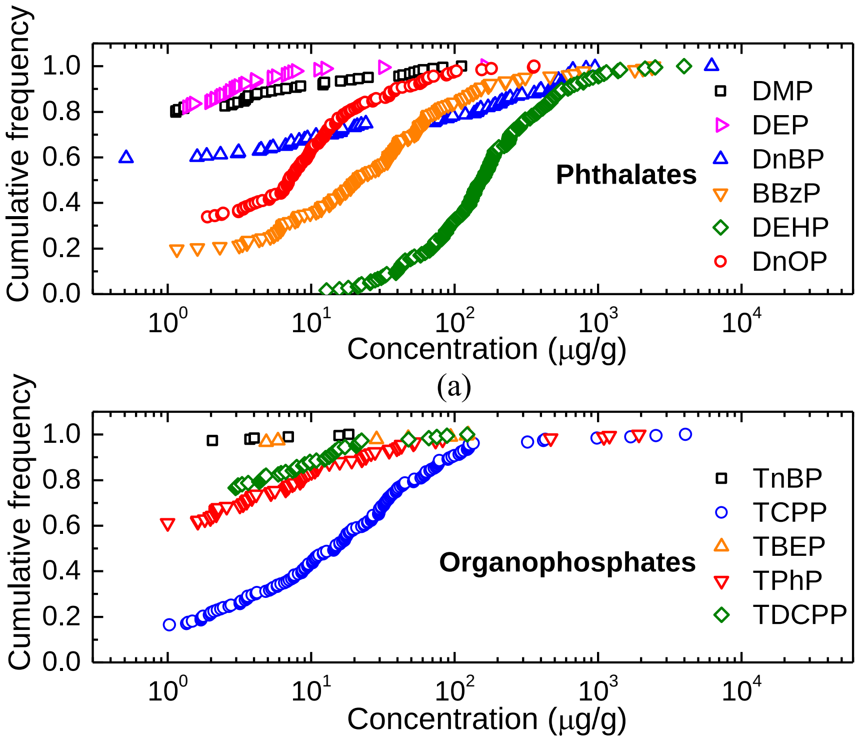
[a] Weschler et al. (2008)

[b] EPA exposure handbook. (2011)

[c] Harrad et al. (2006)

Table C11. Mean daily intake (ng/kg-bw/day) to target compounds through inhalation of suspended particles and ingestion of settled dust for 3-year-old children and adults. (Values in parenthesis represent standard deviation of the mean).

	DnBP	BBzP	DEHP	DnOP	TCPP	TPhP	TDCPP
3-year-old child							
Inhalation of particles	0.95	0.57	2.08	0.08	1.07	0.33	0.02
	(4.64)	(1.92)	(3.56)	(0.29)	(3.75)	(1.66)	(0.04)
Ingestion of dust	237	264	559	50.9	32.6	34.7	18.3
	(471)	(792)	(718)	(93.9)	(94.0)	(256)	(37.7)
Adult							
Inhalation of particles	0.341	0.207	0.753	0.03	0.385	0.119	0.008
	(1.67)	(0.693)	(1.28)	(0.103)	(1.35)	(0.599)	(0.013)
Ingestion of dust	27.6	30.7	65.0	5.92	3.79	4.04	2.13
	(54.8)	(92.1)	(83.4)	(10.9)	(10.9)	(29.8)	(4.38)
Adult, using the lowest ingestion rate (2.5 mg/day) found in Wilson et al. (2013)							
Inhalation of particles	0.341	0.207	0.753	0.03	0.385	0.119	0.008
	(1.67)	(0.693)	(1.28)	(0.103)	(1.35)	(0.599)	(0.013)
Ingestion of dust	2.30	2.57	5.42	0.49	0.32	0.34	0.18
	(4.57)	(7.68)	(6.96)	(0.91)	(0.91)	(2.49)	(0.37)



(a)

(b)

Figure C5. Cumulative frequency distribution of measured concentrations of (a) phthalates and (b) organophosphates in dust samples. Distributions are truncated at the method detection limit of target compounds.

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