



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 (TCIPP), triphenyl phosphate (TPHP), and tris (1,3-dichloroisopropyl) phosphate (TDCIPP). The median level of TCIPP in settled dust was 3- to 180-times higher than levels reported in other studies of residential homes. 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 TDCIPP 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.

1. 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, 2014a). The global production rate of phthalates has increased to 6 million tons/year within a decade; currently, phthalates account for > 70% of the worldwide plasticizer market (Erickson, 2015; Rudel and Perovich, 2009).

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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 (Cooper et al., 2016; Gallen et al., 2014; Saito et al., 2007; Stapleton et al., 2009, 2011, 2012). Restrictions on the use of polybrominated diphenyl ether (PBDE) flame retardants have led to increased use of organophosphates as a replacement (Araki et al., 2014; Bergman et al., 2012; Castorina et al., 2017; Stapleton et al., 2011; van der Veen and de Boer, 2012). 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 and Hubal, 2009; Xu and Little, 2006) and are now ubiquitous indoor pollutants (Bergh et al., 2011a, 2011b; 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 (Rudel et al., 2010; Staaf and Östman, 2005).

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 linked to profound and irreversible changes in the development of the reproductive tract, especially in males (Høyer et al., 2018; Kay et al., 2013, 2014; Latini et al., 2006; Martino-Andrade and Chahoud, 2010; Matsumoto et al., 2008; Zarean et al., 2016). Two widely used organophosphates, tri(2-chloroethyl) phosphate (TCEP) and tri(1,3-dichloroisopropyl) phosphate (TDCIPP), 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 (Dishaw et al., 2011, 2014; Ferguson et al., 2016; Hoffman et al., 2017; Jeddi et al., 2015; Meeker et al., 2013; Meeker and Stapleton, 2010; Miodovnik et al., 2014; Oliveri et al., 2015; Patisaul et al., 2013; Preston et al., 2017; Schang et al., 2018; van der Veen and de Boer, 2012; Zhao et al., 2017). In contrast to these health effects, 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 (Ait Bamai et al., 2014b, 2016; Bornehag et al., 2004; Hsu et al., 2012; Jaakkola and Knight, 2008; Kolarik et al., 2008; Larsson et al., 2010). 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 (Bekö et al., 2015; Callesen et al., 2014a, 2014b).

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 (MW) SVOCs, such as phthalates with MW no less than DEHP, PBDEs with MW greater than BDE-99, and polycyclic aromatic hydrocarbons with MW > 252, the particle-phase fractions are > 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 (Jaakkola and Knight,

2008; Oie et al., 1997). 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. Lastly, dermal uptake of SVOCs from deposited particles on skin as well as gas absorption can also contribute to indoor SVOC exposures. Ingestion of house dust, inhalation of airborne particles, and dermal absorption have been identified as the major exposure pathways for phthalates and organophosphates (Abou-Elwafa Abdallah et al., 2016; Bekö et al., 2013; Frederiksen et al., 2018; Little et al., 2012; Mäkinen, 2009; Marklund et al., 2005; Mitro et al., 2016; Schreder et al., 2016; Xu et al., 2010; Xu and Hubal, 2009).

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; Liroy 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., 2012, 2013, 2016; Bergh et al., 2011a, 2011b; Blanchard et al., 2014a, 2014b; 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., 2008, 2009; 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., 2011a, 2011b; 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}) (Mandin et al., 2016; Wang et al., 2014; Zhang et al., 2014), 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. > 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 (Maestre et al., 2018a; Noris et al., 2009, 2011; Stanley et al., 2008) and heavy metals (Noris et al., 2009) but has been applied only in a limited number of studies on phthalates and organophosphates (He et al., 2016; Xu et al., 2014). 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 severity in children.

2. Materials and methods

2.1. 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. (2018a, 2018b) and Givhechi et al. (2018).

2.2. 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).

2.3. 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).

2.4. 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 purchased from local stores and installed at each house for 1 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. Details on characterization of the HVAC systems were reported in Givhechi et al. (2018). 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 1. 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. A piece of unused 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 unused filter piece. Prior to analysis, the HVAC filter and settled dust samples were fortified with known amounts of surrogate standards (i.e., dibenzyl

Table 1
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
TCIPP	Tris (1-chloro-2-propyl) phosphate	13674-84-5
TBOEP	Tri (2-butoxyethanol) phosphate	78-51-3
TPHP	Triphenyl phosphate	115-86-6
TDCIPP	Tris (1,3-dichloroisopropyl) phosphate	13674-87-8

phthalate, diphenyl isophthalate, and diphenyl phthalate) to explore extraction efficiency. The samples were then ultrasonically extracted for 30 min with hexane, which was repeated three times per sample. Extracts were concentrated using a rotary evaporator (IKA RV-10) and a high purity N₂ 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.

2.5. 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%. Two types of dust recovery methods from HVAC filters, cutting pieces of the filter and vacuuming the surface of the filter, were evaluated. The average difference of measured SVOC concentrations in dust between the two methods was 17%. 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 < 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 S1 (Table and Figure numbers preceded by an “S” are in the Supplementary Material). 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 min, rinsed with methanol three times, and then baked in an oven prior to use.

2.6. 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 (TBOEP) 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. Results and discussion

3.1. 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 2, in which the data for winter and summer seasons were combined (the data for each season are reported in Table S2 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 (Fig. S1). 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 S3 and S4), 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 concentrations ranged from 26.4 to 5420 µg/g, with a median value of 288 µg/g. 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 whose metabolites were 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, 2014a, 2014b). 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, Fig. 1a 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., 2011a, 2011b; Blanchard et al., 2014a,

Table 2
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
TCIPP	91%	< MDL	33.3	4090	150	529	77%	< MDL	5.22	418	15.8	45.5
TBOEP	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
TDCIPP	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.

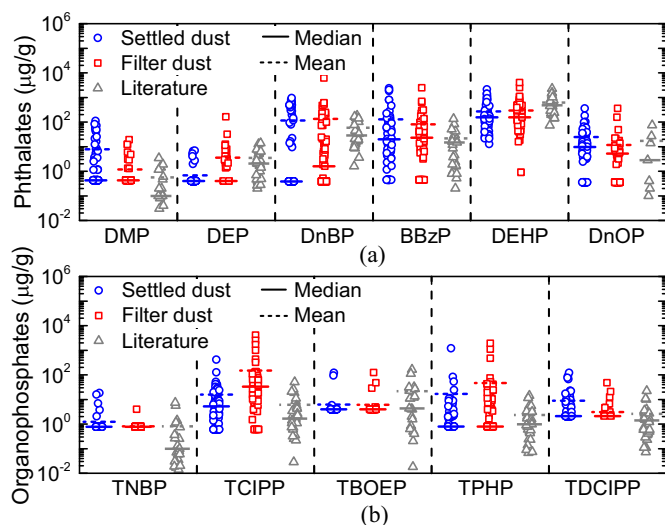


Fig. 1. Comparison of measured concentrations of (a) phthalates and (b) organophosphates in dust with median values reported in literature for residential homes (over 4000 homes in 42 studies).

2014b; 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 U.S. homes at other locations in the United States (Dodson et al., 2015; Guo and Kannan, 2011; Rudel et al., 2003; Shin et al., 2014), 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).

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. In HVAC filter dust, tris (1-chloro-2-propyl) phosphate (TCIPP) 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 TDCIPP

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 TDCIPP and TNBP. TCIPP is majorly applied in rigid polyurethane foams (PUFs) for building insulation (BAUA, 2008). 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). TDCIPP and TCIPP 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 TDCIPP, TPHP, and TCIPP were the most frequently detected biomarkers in humans (Butt et al., 2014; Cooper et al., 2011; Dodson et al., 2014; Jayatilaka et al., 2017; Meeker et al., 2013; Ospina et al., 2018; Stapleton et al., 2009). Fig. 1b 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., 2012, 2013, 2016; Araki et al., 2014; Bergh et al., 2011a, 2011b; Brommer and Harrad, 2015; Castorina et al., 2017; Cequier et al., 2014; Dodson and Perovich, 2012; Fan et al., 2014; He et al., 2016; Hoffmann 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., 2009, 2014; 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 TCIPP. The median level of TCIPP 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., 2011a, 2011b; 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 (Cequier et al., 2014; Dodson and Perovich, 2012; Schreder and La Guardia, 2014; Stapleton et al., 2009, 2014; Zhou et al., 2017), but was relatively lower than levels reported in two studies in Japan (Araki et al., 2014; Kanazawa et al., 2010). The high level of TCIPP 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 TCIPP is used in rigid PUFs, mainly for construction applications (BAUA, 2008).

3.2. 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 TCIPP ($r_s = 0.41$, $p = 0.0001$) was observed, suggesting their common sources inside the house. Currently, the vast majority of TCIPP is used in rigid PUFs as construction materials and a small fraction is used in flexible PUFs for furniture and upholstery (BAUA, 2008; WHO, 2004). Therefore, it is possible that a large amount of TCIPP 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 TCIPP 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. TCIPP 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., 2011a, 2011b; 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 Fig. 2, 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 $368 \mu\text{g/g}$, respectively, which were higher than those in settled dust (0.39, 112, and $195 \mu\text{g/g}$, respectively). Similarly, for organophosphates, the concentrations of TCIPP, TPHP, and total organophosphates in HVAC filter dust were significantly higher than in settled dust in both summer and winter (Fig. 2). In summer, the median levels of TCIPP and total organophosphates in HVAC filter dust were 18.7 and $41.3 \mu\text{g/g}$, respectively; while in settled dust, TCIPP and total organophosphate concentrations were 2.22 and $14.7 \mu\text{g/g}$, respectively. In winter, the median levels of TCIPP and total organophosphates in HVAC filter dust were 78.6 and $104 \mu\text{g/g}$, respectively; while in settled dust were 16.2 and $34.3 \mu\text{g/g}$, respectively.

One possible reason for the higher SVOC concentrations 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 Givchchi et al. (2018). The mean value of particle volume median diameters is $32.3 \mu\text{m}$ ($\text{SD} = 4.78 \mu\text{m}$). 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 particle diameters of settled dust were reported in other indoor studies, with a range of $86\text{--}685 \mu\text{m}$ (Al-omran and Harrad, 2016; Cao et al., 2014a; Gustafsson et al., 2018; Lewis et al., 1999; Morawska and Salthammer, 2003). Previous studies (Weschler and Nazaroff, 2008, 2010) have demonstrated that the equilibration of an SVOC in dust is largely controlled by

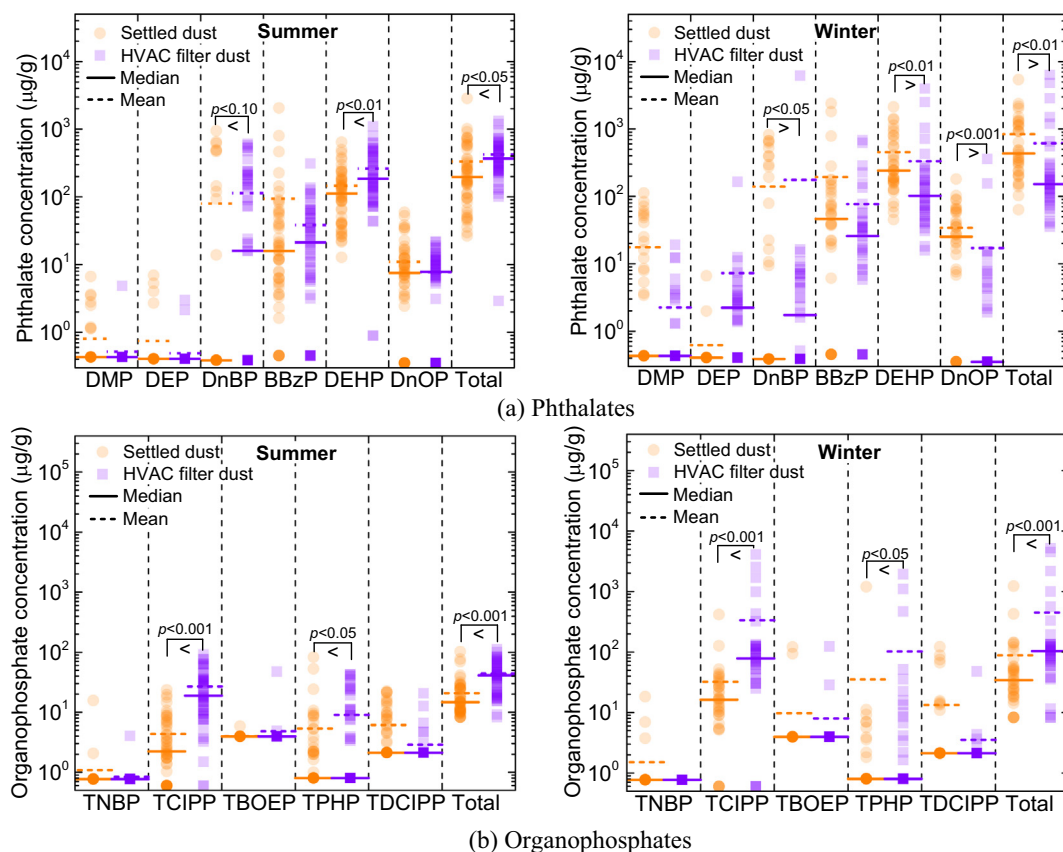


Fig. 2. Comparison of SVOC concentrations between HVAC filter dust and settled dust in summer ($N = 51$) and winter ($N = 36$).

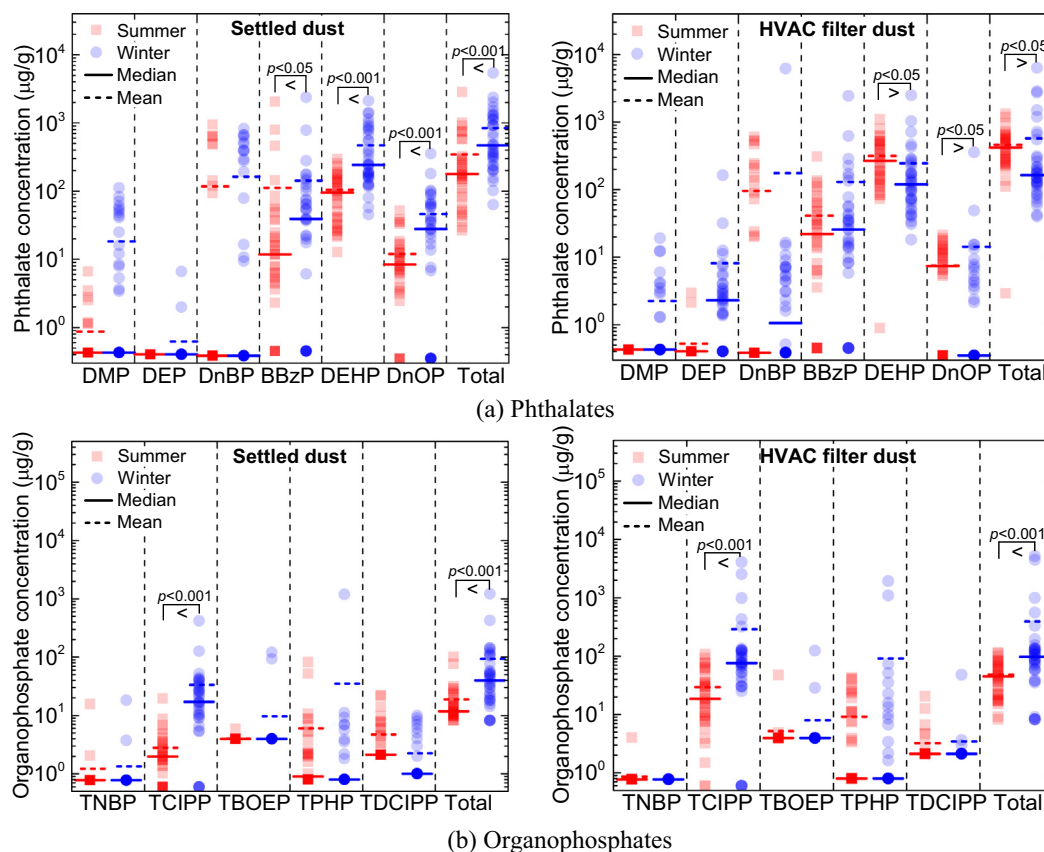


Fig. 3. Seasonal concentration variations of (a) phthalates and (b) organophosphates in HVAC filter dust and settled dust ($N = 36$).

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., 2014). The high-velocity airflow pulled by a fan in the HVAC system further enhances the mass transfer coefficient between gas and particle phases (Clack, 2017; EI-Amin, 2011). 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 (Fig. 2a). The median concentrations of DEHP, DnOP, and total phthalates in settled dust were 242, 25.3, and 434 $\mu\text{g/g}$, respectively, which were significantly higher than those in HVAC filter dust (101, 0.35, and 151 $\mu\text{g/g}$, respectively). Although no statistically significant difference was observed for BBzP, its median and mean levels in settled dust (46.1 and 184 $\mu\text{g/g}$, respectively) were

also higher than those in HVAC filter dust (26.2 and 138 $\mu\text{g/g}$, 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 $< 5\%$, 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, 2014b), while the K_{oa} value of DEHP, which is proportional to the dust/gas partition coefficient, decreased from $10^{11.5}$ to $10^{10.4}$ (Bi et al.,

2015; Gobble et al., 2014; Weschler and Nazaroff, 2010; Xiao and Wania, 2003). 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. 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., 2014b). However, only a few studies of seasonal effects have been performed (Allen et al., 2008; Batterman et al., 2009; Cao et al., 2014b, 2015; Harrad et al., 2008; Vorkamp et al., 2011; Yu et al., 2012), and most of these were based on limited sampling sizes. The average daily outdoor air temperature in central Texas changed from $14.6 \pm 0.3^\circ\text{C}$ in winter to $28.1 \pm 1.9^\circ\text{C}$ in summer, while the average daily indoor air temperature changed slightly from $21.9 \pm 1.9^\circ\text{C}$ in winter to $24.1 \pm 1.6^\circ\text{C}$ in summer. Fig. 3 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 $468 \mu\text{g/g}$, respectively, which were approximately three times higher than those in the summer (11.8, 95, 8.39, and $177 \mu\text{g/g}$, respectively). For organophosphates, higher median concentrations of TCIPP and total organophosphates were also observed in settled dust in winter (17.1 and $39.9 \mu\text{g/g}$, respectively) than in summer (1.97 and $11.7 \mu\text{g/g}$, respectively). The seasonal trend in the present work is similar to the few previous studies available (Cao et al., 2014b, 2015; Yu et al., 2012), 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 $420 \mu\text{g/g}$, respectively) than in winter (119, 0.35, $163 \mu\text{g/g}$, respectively), while TCIPP and total organophosphates showed higher median concentrations in winter (76.1 and $97.8 \mu\text{g/g}$, respectively) than in summer (18.5 and $44.7 \mu\text{g/g}$, 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 $\text{PM}_{2.5}$ in summer as compared to winter, while Mandin et al. (2016) observed opposite results.

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 $C_{\text{dust}} = C_g \times K_{\text{oa}} \times f_{\text{om}} / \rho_{\text{dust}}$ (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, 2014b), 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 and Hubal, 2009). However, ventilation inside residential buildings depends mainly on infiltration, weather conditions, and occupant behaviors (e.g., window opening) (Melymuk et al., 2016). 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 1 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 (Butte and Heinzow, 2002; Edwards et al., 1998). 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 TCIPP, 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.

3.4. 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 Fig. 4a, significantly higher median concentrations of BBzP and DEHP in settled dust were observed in homes with PVC flooring (BBzP: 39.8 and $13.6 \mu\text{g/g}$ in homes with and without PVC flooring,

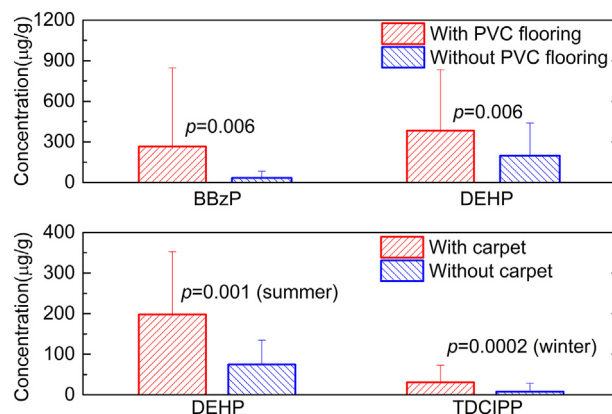


Fig. 4. Phthalate and organophosphate concentrations in settled dust in relation to the presence of PVC flooring (a) and carpet (b). The p -values were calculated using the Mann-Whitney U test and $p < 0.05$ indicates a statistically significant difference.

respectively, $p = 0.006$; DEHP: 189 and 127 $\mu\text{g/g}$ 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 $\mu\text{g/g}$ in homes with and without PVC flooring, respectively, $p = 0.03$; DEHP: 553 and 187 $\mu\text{g/g}$ in homes with and without PVC flooring, respectively, $p = 0.009$) and in summer (BBzP: 18.9 and 5.2 $\mu\text{g/g}$ in homes with and without PVC flooring, respectively, $p = 0.009$; DEHP: 138 and 50.6 $\mu\text{g/g}$ in homes with and without PVC flooring, respectively, $p = 0.007$). However, no such relationship between concentration in HVAC filter dust and floor type was found for any target compounds. 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 di-isobutyl phthalate (DiBP) 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 (Ait Bamai et al., 2014a; Bi et al., 2015; Bornehag et al., 2005; Sukiene et al., 2017).

Fig. 4b shows that TDCIPP levels in settled dust were significantly associated with the presence of carpets (median values of 14 and 2.1 $\mu\text{g/g}$ 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 $\mu\text{g/g}$ 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 materials 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 (Causer et al., 2008; Yiin et al., 2007). 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, TCIPP, TPHP, and TDCIPP 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 (Jonas et al., 2015) reported that organophosphates detected in carpet were typically at low levels ($< 0.01\%$ 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 S6 and S7. The average ($\pm$ SD) floor area and house age were 160 ($\pm$ 55) m^2 and 18 ($\pm$ 13) years, respectively. Higher concentrations of DnBP, BBzP, DEHP, TCIPP, 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 TCIPP 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 TDCIPP 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 TCIPP, TDCIPP, 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.

3.5. 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 (Hsu et al., 2012; Kolarik et al., 2008). However, results were not always consistent. Two studies conducted in Denmark and Japan (Callesen et al., 2014a, 2014b; 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 (Bekö 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 Bekö et al.

(2015) and Deuschle 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 (Bekö et al., 2015; Bornehag and Nanberg, 2010; Franken et al., 2017; Kimber and Dearman, 2010; Larsen et al., 2002, 2003; Larsen and Nielsen, 2008; Oie et al., 1997). 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., 2011a, 2011b). 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 (Dannemiller et al., 2016; Mendell et al., 2011; Michel et al., 1996; Zambelli-Weiner et al., 2004).

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 (Liang et al., 2008; Nakamiya et al., 2005; Nalli et al., 2006a, 2006b; Pradeep et al., 2013; Webb et al., 2000). 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., 2018a, 2018b). 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., 2006a, 2006b), which has been associated with nasal symptoms (Wieslander et al., 1999) or asthma (Kanazawa et al., 2010; Norbäck et al., 2000) 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.

3.6. 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, because HVAC filter dust was dominated with particles suspended in indoor air, with a small portion of coarse particles re-suspended from indoor surfaces (Mukai et al., 2009). In contrast, those measured in settled dust, which consists of a mixture of large particles including deposited indoor aerosols, soil particles tracked inside by foot, and biologically derived material such as skin flakes, was assumed to be digested through oral intake of contaminated particles adhered to food and objects (Özkaynak et al., 2011). 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 S8 and 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, although it is limited to regions where centralized HVAC systems are installed in buildings. 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.

3.7. 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. For example, the average runtime for HVAC systems in winter was 10%, which is much shorter than in summer (31%). It may result in lower dust loading and shorter dust age on winter filters. However, the impact of dust loading and age on the concentrations of SVOCs in HVAC filter dust remains unclear. 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.

4. 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, TCIPP, TPHP, and TDCIPP were the most frequently detected compounds in both HVAC filter dust and settled dust. The median level of TCIPP in settled dust was 3- to 180-times higher than levels reported in other studies of residential homes. 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 TCIPP. This pattern may be attributable to the fact that emission of SVOCs with low vapor pressures is 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 TDCIPP 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 re-suspend 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 disease. 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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2018.09.013>.

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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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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 S1.

Table S1. Instrumental detection limits (IDLs) and method detection limits (MDLs) of target compounds.

Abbreviation	Name	IDLs (ng)	MDLs (µg/g)
<i>Phthalates</i>			
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
<i>Organophosphates</i>			
TNBP	Tri-n-butyl phosphate	2.57	1.54
TCIPP	Tris (1-Chloro-2-propyl) phosphate	1.98	1.19
TBOEP	Tri (2-Butoxyethanol) phosphate	13.2	7.92
TPHP	Triphenyl phosphate	2.66	1.59
TDCIPP	Tris (1,3-dichloroisopropyl) phosphate	7.06	4.23

Table S1. Detection rates and the concentrations of phthalates and organophosphates in HVAC filter and settled dust ($\mu\text{g/g}$).

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
<i>Phthalates</i>														
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
Σ Phthalates	39		35.6	154	6370	657	1320	38		63.4	468	5420	856	969
<i>Organophosphates</i>														
TNBP	39	0	<MDL	<MDL	<MDL	-	-	38	8	<MDL	<MDL	18.3	1.47	2.97
TCIPP	39	85	<MDL	77.9	4080	313	779	38	87	<MDL	16.8	418	32.3	67.2
TBOEP	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
TDCIPP	39	10	<MDL	<MDL	47.7	3.41	7.21	38	26	<MDL	<MDL	122	12.9	26.9
Σ 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
<i>Phthalates</i>														
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
Σ Phthalates	52		2.91	370	1328	424	251	54		26.47	189	2840	330	426
<i>Organophosphates</i>														
TNBP	52	2	<MDL	<MDL	4.01	0.83	0.44	54	4	<MDL	<MDL	15.7	1.07	2.02
TCIPP	52	96	<MDL	19.2	108	28.4	24.3	54	70	<MDL	2.16	23.54	4.18	5.01
TBOEP	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
TDCIPP	52	12	<MDL	<MDL	20.6	2.88	2.96	54	44	<MDL	<MDL	22.4	6.01	5.94
Σ 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 S3. 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	TCIPP	TPHP	TDCIPP
DnBP							
BBzP	-0.103						
DEHP	-0.364	0.267					
DnOP	0.145	0.100	0.000				
TCIPP	-0.260	0.073	-0.023	-0.243			
TPHP	-0.236	0.042	0.226	0.151	-0.107		
TDCIPP	-0.105	0.035	-0.007	0.077	0.003	0.214	

Table S4. 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	TCIPP	TPHP	TDCIPP
DnBP							
BBzP	0.092						
DEHP	0.162	0.388					
DnOP	0.263	-0.102	0.100				
TCIPP	0.145	0.299	0.509	0.045			
TPHP	-0.062	0.066	-0.052	-0.025	-0.041	-0.014	
TDCIPP	-0.125	0.173	-0.010	-0.237	-0.014	0.170	

Table S5. 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	Σ Phthalates	TCIPP	TPHP	TDCIPP	Σ 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 S6. 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				
<i>Phthalates</i>				
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
<i>Organophosphates</i>				
TCIPP	-0.253	-0.078	-0.016	-0.359
TPHP	0.353	-0.225	-0.114	-0.147
TDCIPP	-0.058	-0.177	0.146	0.101
Summer				
<i>Phthalates</i>				
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
<i>Organophosphates</i>				
TCIPP	-0.399	-0.547	0.041	0.113
TPHP	0.064	0.082	-0.471	-0.034
TDCIPP	-0.106	-0.005	-0.247	-0.003

Table S7. Relationships between housing characteristics and concentrations of phthalates and organophosphates in dust. Mean levels and standard deviations (std) of target compounds in dust ($\mu\text{g/g}$) 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		TCIPP		TPHP		TDCIPP		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		TCIPP		TPHP		TDCIPP		Organophosphates	
		mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
<i>Stuffed toy</i>																			
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
<i>Pet</i>																			
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
<i>Vacuum on carpets</i>																			
<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
<i>Clean hard floors</i>																			
<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
<i>Clean furniture dust</i>																			
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
<i>Change HVAC filters</i>																			
<=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 S8. 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 ($\mu\text{g/kg-bw/day}$)	Parameters	Parameter values
Inhalation of particle-phase SVOCs	$\text{Exposure dose} = \frac{C_{FD} \times TSP \times InhR \times ED}{BW}$	Particle-phase concentrations of SVOCs measured in filter dust ($\mu\text{g/g}$)	Mean concentration of SVOCs in filter dust measured in this study
		C_{FD} = particle-phase concentrations of SVOCs measured in filter dust ($\mu\text{g/g}$)	20 $\mu\text{g/m}^3$. ^a
		TSP = total suspended particle concentration ($\mu\text{g/m}^3$)	
		$InhR$ = inhalation rate (m^3/day)	Child: 10.3; adult: 16. ^b
		ED = exposure duration (-)	0.64. ^c
Ingestion of settled dust	$\text{Exposure dose} = \frac{C_{SD} \times IngR \times ED}{BW}$	BW = body weight (kg)	Child: 18.6; adult: 80. ^b
		C_{SD} = concentrations of SVOCs measured in settled dust ($\mu\text{g/g}$)	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 S9. 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	TCIPP	TPHP	TDCIPP
3-year-old child							
Inhalation of particles	0.95 (4.64)	0.57 (1.92)	2.08 (3.56)	0.08 (0.29)	1.07 (3.75)	0.33 (1.66)	0.02 (0.04)
Ingestion of dust	237 (471)	264 (792)	559 (718)	50.9 (93.9)	32.6 (94.0)	34.7 (256)	18.3 (37.7)
Adult							
Inhalation of particles	0.341 (1.67)	0.207 (0.693)	0.753 (1.28)	0.03 (0.103)	0.385 (1.35)	0.119 (0.599)	0.008 (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 (1.67)	0.207 (0.693)	0.753 (1.28)	0.03 (0.103)	0.385 (1.35)	0.119 (0.599)	0.008 (0.013)
Ingestion of dust	2.30 (4.57)	2.57 (7.68)	5.42 (6.96)	0.49 (0.91)	0.32 (0.91)	0.34 (2.49)	0.18 (0.37)

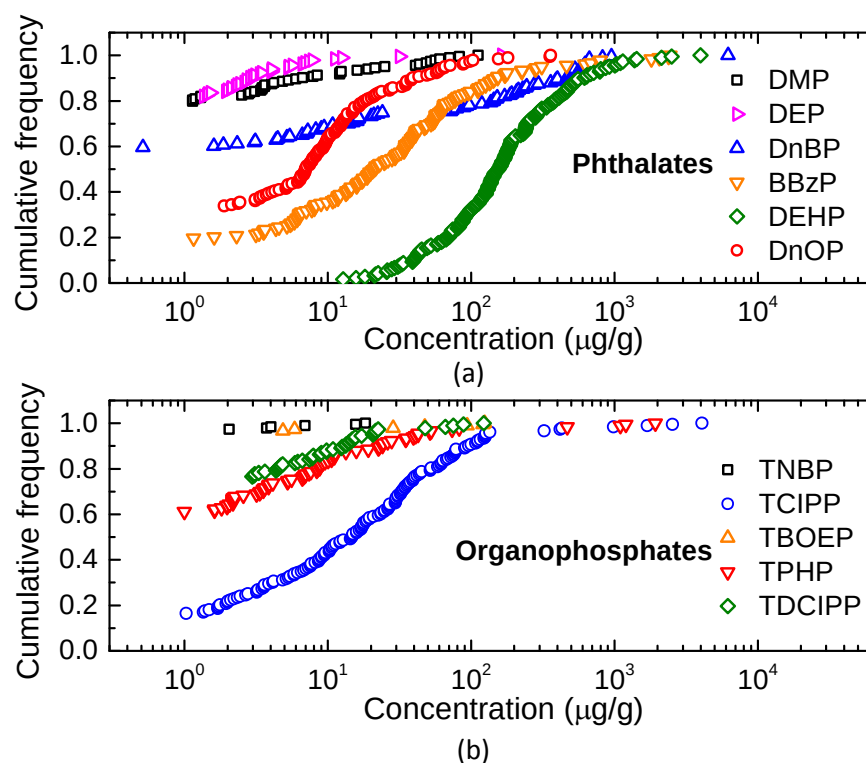


Figure S1. Cumulative frequency distribution of measured concentrations of (a) phthalates and (b) organophosphates in dust samples. Distributions are truncated at the method detection limit of target compounds.