Semi-volatile Organic Compounds in Indoor Settled and HVAC filter dust: Association with Seasons, Childhood Asthma and Building characteristics

Chenyang Bi\textsuperscript{1}, Juan Maestre\textsuperscript{1}, Kerry Kinney\textsuperscript{1}, Jeffrey Siegel\textsuperscript{2}, Sharon Horner\textsuperscript{1} and Ying Xu\textsuperscript{1,*}

\textsuperscript{1}The University of Texas at Austin, Austin, U.S.A.
\textsuperscript{2}University of Toronto, Toronto, Canada.

\textsuperscript{*}Corresponding email: xuying@mail.utexas.edu

SUMMARY
In previous studies, settled dust samples were frequently used to quantify levels of semi-volatile organic compounds (SVOCs) for the estimation of exposure. However, the age of the settled dust is usually unknown. Instead, the heating, ventilation and air conditioning (HVAC) filters dust samples may potentially be used as an alternative approach to evaluate the levels of SVOCs in the indoor environments. A field study that enrolled 54 households, including families with children diagnosed with asthma, was conducted to measure six phthalates, five phosphorus flame retardants in HVAC filter dust and settled dust in both summer and winter. The results showed that SVOC concentration in filter dust has less season variance than those in settled dust. SVOC levels in settled dust were found to be related with the types of flooring material. In addition, no association was found between children’s asthma and SVOC concentration.

KEYWORDS
Semi-volatile organic compounds, Dust, Seasonal variation, Flooring, Asthma.

1 INTRODUCTION
Phthalates and organophosphates are wildly used as additives in building materials and consumer products such as vinyl flooring, carpeting, wall covering, electronics, and furniture, as plasticizer and flame retardants, respectively. Exposure to those semi-volatile organic compounds (SVOCs) are found to be related with the asthma and allergic symptoms in children. Oie et al. (1997) first reported that bis (2-Ethylhexyl) phthalate (DEHP) concentration in house dust might be associated with asthma in children. As reviewed by Bornehag and Nanberg (2010), a variety of studies have reported that phthalate exposure is an important risk factor on the incidence of children’s asthma or allergic symptoms (Bornehag et al., 2004). Compared with phthalates, organophosphates attracted less attention by researchers for the relationship with asthma. Araki et al. (2014) showed that organophosphates concentration in dust are related with asthma and allergic disorders while another study did not find any relationship between them (Canbaz et al., 2015). However, none of these studies was conducted in the United States. Therefore, it is important to study the phthalate and organophosphate exposure in the indoor environments and investigate their association with children’s asthma in the United States.

In those previous studies mentioned above and other field studies (Rudel et al., 2003; Zhang et al., 2013), settled dust samples were frequently used to quantify levels of those SVOCs for the
estimation of exposure. However, the age of the settled dust is usually unknown, and contaminants associated with larger-size particles may be overemphasized in settled dust samples. The heating, ventilation and air conditioning (HVAC) filters installed in homes serve as long-term samplers of particle-bound SVOCs in homes. Therefore, HVAC filter dust samples may potentially be used as an alternative approach to evaluate the levels of SVOCs in the indoor environments.

Thus, the goal of this research was to assess the variation of SVOC levels in settled and filter dust in different season and determine the accuracy to use dust as a metric to estimate indoor exposure to SVOCs. The specific objectives were to (1) compare SVOC concentrations between settled and filter dust among 54 homes in Texas; (2) Analyse seasonal variance of SVOC levels in dust; (3) investigate the association between SVOC levels in dust and building characteristics, and (4) study the association between childhood asthma and SVOC concentrations in dust.

2 MATERIALS/METHODS
Selection of Homes. We recruited a total of 40 homes of children with asthma and 20 homes of children without asthma in this study. Among those 60 homes, six of them were not sampled. These homes are located in rural central Texas and are associated with two school districts.

Building Investigations. Building investigations were performed during June 2014 to February 2015. 54 homes participated in this study were sampled in summer from June to September. Then a second sampling to 39 of the 54 homes was conducted in winter from December to February. A survey was conducted during our first visit of each home. The survey covered a fairly extensive list of 119 questions about environmental triggers associated with asthma exacerbation and building characteristics, including questions related with smoking in the home, pets, flooring types and cleaning practices. We also measured the air exchange rate and monitored the operation of air conditioning system (e.g., real-time temperature and humidity) for each home site.

SVOCs in Dust. Sampling of settled dust was performed in the child’s room on the floor. The dust, approximate 0.1g for each home, 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. Those cellulose thimble filters were stored in cooler and transported to the lab for chemical analysis.

New HVAC filters, which have been tested for backgrounds in target SVOCs, were installed for each house one month upon sampling. Those filters were collected during sampling after operating in the homes for one month, stored in 4°C room and taken for lab analysis. Two pieces of filters, with an area of 10 × 10 cm each, were then cut with metal scissors and weighted before extraction. The filter pieces were then extracted for chemical analysis. After extraction, the filter pieces were dried in the oven for 24 hours and weighted again. The mass of dust on the filter pieces was determined by calculating the difference of weight before and after extraction.

Sample Extraction and Chemical Analysis. The settled dust and filter dust samples were ultrasonically extracted with hexane, concentrated, and then analyzed for dimethyl phthalate (DMP), Diethyl phthalate (DEP), di-n-octyl phthalate (DnOP), Benzy1 butyl phthalate (BBzP), DEHP, Tributyl phosphate (TNBP), tris(1-Chloro-2-propyl) phosphate (TCPP), tri(2-
Butoxyethanol) phosphate (TBEP), Triphenyl phosphate (TPHP) and Tris(1,3- dichloroisopropyl)phosphate (TDCPP) using a GC-FID system (Agilent 7890A) equipped with a DB-5ht column (30 m, 0.25 mm, 0.1 μm). The detailed analysis method was described in Bi et al. (2015).

**Statistical Methods.** Concentrations below the limit of detection were assigned half of the detection limit. We performed analyses of potential associations between concentrations of SVOCs in dust and building characteristics using nonparametric tests (Mann-Whitney U-test). Wilcoxon signed-rank test was used to compare the SVOC levels between winter/summer and filter dust/settled dust.

### 3 RESULTS AND DISCUSSIONS

**Occurrence and Concentration Ranges.** The concentrations of phthalates and organophosphates were summarized in Table 1, with the abundant rank order as follows: DEHP>BBzP>TCPP>TPHP>DnOP>TDCPP>TBEP>TNBP~DMP~DEP. In this study, the most frequently detected compounds are BBzP, DEHP and TCPP, which are detected more than 70% in both filter dust and settled dust. DEHP was detected at the highest concentration of 294±502 and 271±347 μg/g in filter and settled dust, respectively. Figure 1 showed the measured SVOC concentration in filter and settled dust. The results indicate that no significant differences were observed for phthalates between filter and settled dust while TPHP and TCPP have higher concentration in filter dust than those in settled dust. TNBP and TBEP were not included for comparison and further analysis because TNBP was detected only once in filter dust and TBEP was detected in less than five samples.

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Winter</th>
<th></th>
<th>Summer</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Filter dust</td>
<td></td>
<td>Settled dust</td>
<td></td>
</tr>
<tr>
<td></td>
<td>NHS</td>
<td>%&gt;DL</td>
<td>min</td>
<td>median</td>
</tr>
<tr>
<td>DMP</td>
<td>39</td>
<td>28.2</td>
<td>0.43</td>
<td>0.43</td>
</tr>
<tr>
<td>DEP</td>
<td>39</td>
<td>66.7</td>
<td>0.40</td>
<td>2.27</td>
</tr>
<tr>
<td>BBzP</td>
<td>39</td>
<td>82.1</td>
<td>0.45</td>
<td>26.3</td>
</tr>
<tr>
<td>DEHP</td>
<td>39</td>
<td>100</td>
<td>15.8</td>
<td>112</td>
</tr>
<tr>
<td>DnOP</td>
<td>39</td>
<td>46.2</td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td>TNBP</td>
<td>39</td>
<td>0</td>
<td>N.A.</td>
<td>N.A.</td>
</tr>
<tr>
<td>TCPP</td>
<td>39</td>
<td>84.6</td>
<td>0.60</td>
<td>77.9</td>
</tr>
<tr>
<td>TBEP</td>
<td>39</td>
<td>5.13</td>
<td>3.96</td>
<td>3.96</td>
</tr>
<tr>
<td>TPHP</td>
<td>39</td>
<td>41.0</td>
<td>0.80</td>
<td>0.80</td>
</tr>
<tr>
<td>TDCPP</td>
<td>39</td>
<td>10.3</td>
<td>2.12</td>
<td>2.12</td>
</tr>
</tbody>
</table>

**Table 1. Summary statistics for household dust samples (μg/g)**

- NHS is the number of home sampled.
- DL is the method detection limit.
The results were further compared with the median values of SVOC concentration in residential house dust reported in the literature (Araki et al., 2014; Bergh et al., 2011; Cequier et al., 2014; Kolarik et al., 2008; Rudel et al., 2003), as shown in Figure 1. For phthalates and organophosphates, the results in current study agreed well with the medians reported in the literature except that TDCPP concentration in the current study is lower than the levels reported in the literature. Because the sample size in the current study is relatively small and the detection frequency of organophosphates was significantly less than those of phthalates, it is possible that for some of organophosphates have concentrations different from median values reported in the literature.

![Figure 1. SVOC concentration in filter and settled dust and comparisons to published data](image)

**Seasonal Variation.** The average indoor temperature of the sampled homes in winter and summer are 14.6±0.30 and 28.1±1.94 °C, respectively. This result suggest that the temperature in the sampled homes have distinct differences between winter and summer.

Figure 2 showed the concentrations of phthalates detected in settled dust and filter dust in summer and winter. In general, the abundant rank order of phthalate dust levels is as followed: settled dust (winter)>filter dust (summer)>filter dust (winter)>settled dust (summer). The results showed that phthalates in settled dust had a significantly higher concentration in winter than those in summer [142±408 vs 111±365 µg/g (n=36), p=0.010 for BBzP; 469±475 vs 104±73.6 µg/g (n=36), p<0.001 for DEHP; 46.2±66.3 vs 12.0±12.3 µg/g (n=36), p<0.001 for DnOP, Wilcoxon signed-rank test]. In contrast, in filter dust, DEHP and DnOP had slightly higher concentration in summer than those in winter (316±235 vs 245±437 µg/g (n=36), p=0.012 for DEHP; 7.52±6.54 vs 14.3±59.6 µg/g (n=36), p=0.039 for DnOP, Wilcoxon signed-rank test). No statistical difference was observed for BBzP concentrations in filter dust between the two seasons.
Figure 2. Seasonal variation of phthalate levels in dust

Less seasonal variation of organophosphates was observed as compared with seasonal variation of phthalates. In settled dust, TCPP was the only compound which showed a statistical difference between different seasons. The results showed that TCPP had a higher concentration in winter than those in summer [33.6±69.8 vs 2.80±3.44 µg/g (n=36), p<0.001 for TCPP; Wilcox signed-rank test]. In filter dust, no statistical differences were observed for organophosphate concentrations between winter and summer.

**Type of Flooring Material.** The results for summer samples showed that only BBzP in settled dust had a significantly higher concentration in homes with PVC flooring than those with non-PVC flooring [183±469 (n=21) vs 27.8±49.2 µg/g (n=33), p=0.019 for BBzP; Manning-Whitney U test]. Furthermore, because for those homes with carpet, we did not investigate whether there was vinyl flooring underneath their carpet, it is necessary to exclude those homes with carpet in the statistical test. Therefore, the same statistical test was performed again after excluding the homes with carpet. The results showed that both BBzP and DEHP in settled dust had a significantly higher concentration in homes with PVC flooring than those with non-PVC flooring [183±469 (n=21) vs 20.5±36.0 µg/g (n=17), p=0.009 for BBzP; 155±134 (n=21) vs 74.8±60.0 µg/g (n=17), p=0.007 for DEHP; Manning-Whitney U test]. In winter, BBzP and DEHP in settled dust also had significantly higher levels with the presence of PVC [371±789 (n=16) vs 37.5±60.1 µg/g (n=13), p=0.026 for BBzP; 681±545 (n=16) vs 335±366 µg/g (n=13), p=0.009 for DEHP; Manning-Whitney U test; homes with carpet were excluded]. In contrast, no correlation was found between the concentration of DEHP and BBzP in filter dust and the presence of PVC flooring in any of those statistical tests.

The results suggest that the PVC flooring materials can enhance the levels of phthalates in settled dust, which collected on the floor of the children’s room. Because PVC flooring materials usually contain phthalates up to 30% by mass, it is possible that the mass transfer of those phthalates between settled dust and flooring can be enhanced thus resulting in a higher equilibrium concentration.

**Asthma.** No significant difference of phthalates or phosphorus flame retardants was observed between the asthma group and control group. Although previous studies have reported that DEHP and TNBP may be associated with the development of asthma (Bornehag and Nanberg 2010; Bornehag et al. 2004; Araki et al. 2014). However, in this study, we cannot find any
relationship between phthalates/organophosphate and asthma. It is possible that a relatively low economic income may create other factors which can cause asthma thus weakening the relationship between target SVOCs and asthma.

4 CONCLUSIONS

The results suggested that phthalate levels in filter dust have less seasonal variance and they are less likely to be influenced by PVC flooring materials compared with those in settled dust. Therefore, filter dust might be an alternative and better metric for estimation of phthalate exposure. In addition, no statistical differences were found between children’s asthma target compounds levels in this study.

5 ACKNOWLEDGEMENT

The funding was provided by the U.S. Department of Housing and Urban Development and National Science Foundation.

6 REFERENCES


