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Evaluating couch polyurethane foam for a potential passive sampler of semivolatile organic compounds

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Abstract

Background/Objective: Polyurethane foam (PUF), a proven sampling medium for measuring air concentrations of organic compounds, is widely used in upholstered home furniture. We evaluated the potential utility of couch PUF as a passive sampler and as a reservoir for non-flame retardant semivolatile organic compounds (SVOCs).

Methods: We collected PUF samples from 13 California home couches, measured concentrations (C_{PUF}) of 64 SVOCs at three different depths (i.e., top, top-middle, and middle from couch

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Declaration of interests
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval and consent to participate
All recruitment and data collection protocols were approved by the Institutional Review Board for the University of California Davis (UC Davis). Participants provided written informed consent before collection of any data.

surfaces facing outward), and examined concentration changes with depth. To calculate the PUF-air partition coefficient ($K_{PUF\text{-}air} = C_{PUF}/C_{air} = C_{PUF} \times K_{dust\text{-}air}/C_{dust}$), we used the calculated dust-air partition coefficient ($K_{dust\text{-}air}$) with the octanol-air partition coefficient (K_{oa}) and dust concentrations (C_{dust}) simultaneously collected and measured. We used $K_{PUF\text{-}air}$ to compute fugacity capacity of PUF and chemical mass distribution among various indoor compartments and PUF.

Results: Among 29 detected compounds, 11 compounds were detected in more than 50% of the samples at all depths. Among the 11 compounds, concentrations of phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate decreased with depth. Among the studied SVOCs, more than 20% of the total mass was distributed to couch PUF for phenol and compounds in skin-applied products (i.e., 2-benzylideoneoctanal, galaxolide, and homosalate).

Conclusions: Our results showed that couch PUF can absorb many SVOCs and may be an important reservoir for some SVOCs. However, it may not be an effective passive sampling medium for those that have relatively high K_{oa} values. Direct dermal contact with couch seats may be an important exposure route for non-users of skin-applied compounds.

Graphical Abstract

Graphical Abstract
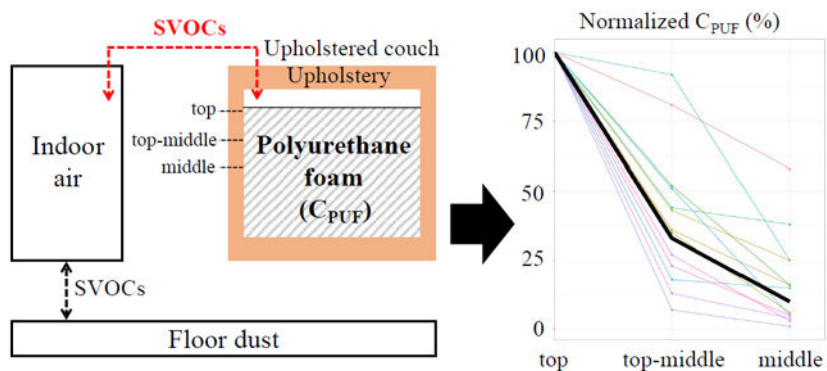
Keywords

couch; partition coefficient; polyurethane foam; sampling medium; semivolatile organic compound

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1. Introduction

Reliable sampling methods for assessing exposure to semivolatile organic compounds (SVOCs) in indoor environments are being demanded in environmental health studies (Dodson et al., 2019). Exposure to SVOCs released indoors is of concern because their levels are typically several orders of magnitude higher indoors than outdoors (Bennett et al., 2002; Shin et al., 2012) and SVOCs have been shown to be toxic or to be associated with adverse health effects such as neurotoxicity (Kamel and Hoppin, 2004; Colt et al., 2005; Colt, 2006; Colt et al., 2009; Munoz-Quezada et al., 2013; Viel et al., 2015), carcinogenicity (Knafla et al., 2006; Shi et al., 2018), and endocrine disrupting potential (Jacobson and

Jacobson, 1996; Howdeshell, 2002; Iwasaki et al., 2002; Birnbaum and Staskal, 2004; Sharpe, 2005). Polyurethane foam (PUF), a class of lightweight porous materials (Gama et al., 2018), has been widely used as a sampling medium for SVOCs in both active (Gouin et al., 2005; Moeckel et al., 2009; Hayward et al., 2010; Newton et al., 2016) and passive (Shoeib and Harner, 2002a; Jaward et al., 2004; Strandberg et al., 2018) air sampling systems due to its high sorption capacity for organic compounds (Zhao et al., 2004; Tromp et al., 2019). PUF-based active or passive samplers are used to measure concentrations of SVOCs in indoor air (Shoeib and Harner, 2002b; Newton et al., 2016). However, deploying purposeful active or passive samplers for SVOCs can add significant cost to studies in terms of extra trips to homes and may also create burdens for residents living in the homes. In addition, passive air samplers utilizing PUF material need to be deployed for a long period of time for SVOCs because of slow equilibrium of SVOCs in PUF disk (Shoeib and Harner, 2002a).

PUF is also widely used in pillows, beds, and chair and sofa cushions in office and home environments. There is evidence that PUF in the home furniture can absorb other SVOCs commonly detected indoors. For example, PUF in an infant crib mattress is not known as a source of plasticizers, but among the ten used infant crib mattresses that were in contact with mattress covers with detectable plasticizers, at least one plasticizer was detected in nine mattress PUF samples (Boor et al., 2015). This shows that mattress PUF can absorb plasticizers from its cover. Moreover, because PUF in upholstered home furniture is treated with flame retardants (FRs) to lower its flammability (Blum and Ames, 1977; Hale et al., 2002; Alaee et al., 2003; Stapleton et al., 2009; Hammel et al., 2017), PUF in the home furniture is an emission source of FRs in indoor environments (Keimowitz et al., 2016; Stubbings et al., 2018) and PUF-based couch cushions are one of the flame-retarded home furniture items with frequent skin contact. Thus, if couch PUF can be sampled, it may provide a simple way to assess chemical exposure with minimal cost (i.e., no extra trips to deploy and collect samplers).

The partitioning relationship between PUF and air can be used to compute the chemical distribution between air and PUF for unmeasured SVOCs with known chemical properties. To date, SVOC partitioning between PUF and air was determined in chamber studies under the controlled laboratory conditions or outdoor field studies using purposefully-designed PUF samplers (Kamprad and Goss, 2007; Bidleman et al., 2016; Parnis et al., 2016; Abdollahi et al., 2017; Francisco et al., 2017; Saini et al., 2019; Tromp et al., 2019). Chamber studies can control environmental conditions (e.g., temperature, humidity) and various indoor human activities that could disturb chemical equilibrium between PUF and air, such as ventilation, cooking, walking, sitting, and cleaning. However, because chamber studies typically use bare PUF disks, the PUF-air partition coefficient ($K_{PUF-air}$) from chamber studies may not represent common conditions of home furniture PUF such as upholstery. Morever, because $K_{PUF-air}$ values vary with environmental conditions (Zhao et al., 2004; Francisco et al., 2017), those derived from outdoor field studies may not represent relatively invariant indoor temperature and humidity. Thus, $K_{PUF-air}$ with direct concentration measurements in upholstered home furniture PUF may improve our understanding of home furniture PUF's actual holding capacity for indoor SVOCs.

The objective of this study was to assess the potential utility of couch PUF as a passive sampling medium and as a reservoir for indoor non-FR SVOCs. Specifically, we collected couch PUF samples at three different depths inside couch cushions, measured SVOC concentrations (C_{PUF}), and examined concentration changes with depth. Because we previously measured SVOC concentrations in dust (C_{dust}) collected in the same home when PUF was collected (Kim et al., 2020) and the dust-air partition coefficient ($K_{dust-air} = C_{dust}/C_{air}$, where C_{air} is the air concentration) is a function of the octanol-air partition coefficient (K_{oa}), we calculated $K_{PUF-air}$ ($= C_{PUF}/C_{air} = C_{PUF} \times K_{dust-air}/C_{dust}$) using direct measurements of C_{PUF} and C_{dust}. Then, we explored predictive relationships of $K_{PUF-air}$ with K_{oa} or vapor pressure (VP). Lastly, we used $K_{PUF-air}$ to compute fugacity capacity of PUF and chemical mass distribution among air, carpet, vinyl flooring, walls, and couch PUF.

2. Methods

2.1. Participant recruitment and couch PUF sample collection

As part of an effort to examine the overall decrease of FR concentrations in household dust after replacing old couches (assumed to be the primary sources of FRs in participating homes) with new ones, we recruited 14 homes in San Jose, California in July 2016. On the day of couch replacement, the study team removed samples from 11 of the old couches by cutting the foam with pre-cleaned scissors and removing the foam segment with pre-cleaned forceps. Samples were wrapped in pre-cleaned foil and placed in a zip-top bag (polyethylene sample bag). The samples were placed in a cooler and then stored in a −20 °C freezer. The new furniture items were delivered after the foam samples were removed. The study team collected samples from the top to the bottom of the whole cushion of the old couch and labeled top and bottom on it. In the laboratory, we further cut it to obtain individual samples at each depth (i.e., top, top-middle, and middle from couch cushion surfaces facing outward). Because two different couches were removed in two homes, we collected samples from both, bringing the total number of sample sets (top, top-middle, and middle) to 13.

All recruitment and data collection protocols were approved by the Institutional Review Board for the University of California at Davis (UC Davis). Participants provided informed consent before collection of any data.

2.2 Target compounds

We analyzed both FRs and non-FRs in our samples. However, in this current study, we focused on SVOCs that are assumed not to originate from couch PUF but rather be absorbed by the PUF and included the results of FRs in the Supporting Information. We used a compound list developed through a previous project searching for widely-detected compounds in household dust (Moschet et al., 2018; Shin et al., 2020) and then analyzed for the compounds detected in our previous studies as described below. The selected compounds included ultraviolet (UV) filters, fragrance ingredients, and other ingredients of personal care products (PCPs); insecticide ingredients; and a variety of other compounds widely detected in homes (phenols, phthalates, other plasticizers, polycyclic aromatic hydrocarbons (PAHs), and squalene found in the skin). The selected compounds represent a range of chemical properties, ranging from relatively volatile compounds (e.g., phenol) to those with

a high affinity to dust (e.g., di(2-ethylhexyl) phthalate [DEHP]). Chemical properties and use categories of the studied compounds were listed in Table S1.

2.3. Sample analysis

At UC Davis, we quantified concentrations of 64 compounds in three sections (top, top-middle, and middle) of each couch PUF sample. The PUF samples were cut using a pre-cleaned cutter and subsections (approximately 100 mg) were sonication-extracted using hexane:acetone (3:1 v:v) followed by acetone (100%). The supernatant was collected and combined into an evaporation tube and evaporated to 1 mL under nitrogen using a Turbovap (Biotage). The extract was then filtered through a polytetrafluoroethylene filter (0.2 μm). Half of the 1 mL extract was spiked with internal standard dibromooctafluorobisphenol (DBOFB) and 1 μL was injected for analysis. An Agilent 7890B gas chromatograph using a HP-5MS (30 m × 0.25 m, 0.25 μm) column coupled to an Agilent 7200B gas chromatography quadrupole time-of-flight mass spectrometry (GC-QTOF-MS) was used to acquire samples in electron ionization (EI) mode. Samples were acquired using a 78-minute method with a linear temperature gradient from 35 °C to 325 °C. A 13-point calibration curve was used to quantify target analytes using the Agilent MassHunter Quantitative Analysis (version B.09).

A method blank was prepared by conducting an extraction with no foam in identical containers to those used during foam extractions, with the extract subsequently processed in the same manner as for foam samples. For any target chemicals that also appeared in the method blank, the LOD was adjusted so that it is at least 3 times the response in blank. To determine recoveries during the extractions, a surrogate solution containing labeled pesticides, a labeled phthalate, and selected labeled polybrominated diphenyl ether (PBDE)'s was added to samples prior to extraction. A few of the labeled pesticides had somewhat lower recoveries, ranging from 40% to 150%. Recovery of the labeled phthalate ranged from 60% to 121%. For the labeled PBDE's, there was an overloading of a co-eluting compound that prevented calculation of recoveries for those samples.

2.4. Statistical analysis

We performed all statistical analyses using R version 3.6.1. For all analyzed compounds, we provided summary statistics of measured concentrations in PUF samples for each depth (Table S2). For concentrations below the limit of detection (LOD), we assigned a value of the LOD divided by the square root of 2 (Hornung, 1990; Antweiler, 2015). For concentrations between the LOD and the limit of quantification (LOQ), we assigned a value of the LOQ divided by 2.

For compounds detected in more than 50% of the PUF samples at all three depths, we used natural logarithm (ln)-transformed concentrations in all statistical analyses because distributions of concentrations were right-skewed for these compounds. We normalized the top-middle and middle concentrations to the top concentrations and then examined concentration changes with depth. We also performed a paired t-test to compare mean PUF concentrations between two depths (i.e., top versus top-middle, top-middle versus middle, top versus middle). For compounds detected in more than 50% of the top PUF samples and

dust samples collected in the same home (Kim et al., 2020), we computed the Pearson correlation coefficients (r) between PUF (C_{PUF}) and dust concentrations (C_{dust}) and also computed the ratio of C_{PUF} to C_{dust}. For two couches that were removed in two homes, we compared distributions of the measured PUF concentrations (ng/g of PUF) at each depth (i.e., top, top-middle, middle) to examine the effect of upholstery materials (e.g., leather, microfiber, velvet) on PUF concentrations. Couch age may affect PUF concentrations, but age information was not available for those couches in the two homes.

2.5. Calculation of PUF-air partition coefficients ($K_{PUF\text{-}air}$)

To calculate $K_{PUF\text{-}air}$, we used the compounds detected in more than 50% of both the top PUF samples and dust samples. Because $K_{PUF\text{-}air}$ ($= C_{PUF}/C_{air} = C_{PUF} \times K_{dust\text{-}air}/C_{dust}$) is a function of $K_{dust\text{-}air}$ (m^3 of air/mg of dust), we calculated $K_{dust\text{-}air}$ using the following relationship (Weschler and Nazaroff, 2010; Little et al., 2012).

$$K_{dust-air} = \frac{C_{dust}}{C_{air}} = f_{om\_dust} \times K_{oa} \;/\; \rho_{dust}$$

where f_{om_dust} is the fraction of organic matter associated with settled dust (unitless), K_{oa} is the octanol-air partition coefficient (unitless), and ρ_{dust} is the density of settled dust (mg/m^3). We used 0.2 and 2×10^9 mg/m^3 for f_{om_dust} and ρ_{dust}, respectively, reported elsewhere (Weschler and Nazaroff, 2010; Little et al., 2012) and chemical-specific K_{oa} values available in the U.S. Environmental Protection Agency (EPA)'s CompTox Chemistry Dashboard (Williams et al., 2017). The unit of PUF concentration was converted from ng/g of PUF to ng/m^3 of PUF using the bulk density of PUF (2.2×10^3 g/m^3) (Zhao et al., 2004) before calculating $K_{PUF\text{-}air}$ (m^3 of air/m^3 of PUF). In addition, we regressed our calculated $K_{PUF\text{-}air}$ on K_{oa} or VP (in Pa) to formulate predictive equations for $K_{PUF\text{-}air}$.

Predictive equations of $K_{PUF\text{-}air}$ with K_{oa} or VP were developed in two chamber studies (Zhao et al., 2004; Francisco et al., 2017) and another indoor field study with purposefully-designed PUF samplers (Shoeib and Harner, 2002b). The $K_{PUF\text{-}air}$ values in those three studies are considered to be "true" partition coefficients, because they were calculated after "effective" (or 95% of) equilibrium was established between concentrations in air and PUF under the tight control on sampling conditions. On the other hand, those values calculated in the current study are considered to be "apparent" partition coefficients, because we could not confirm whether our system was at equilibrium at the time of sampling. Although the $K_{PUF\text{-}air}$ values in the current study may not be directly comparable to those in those three studies, we compared them each other to observe potential differences in PUF's holding capacity between upholstered couch PUF and bare PUF.

2.6. Calculation of holding capacity for SVOCs

The fugacity capacity is defined as the holding capacity of a material for a chemical substance based on the properties of both the material and the chemical (Bennett and Furtaw, 2004). Because little is known for holding capacity of couch PUF for SVOCs, we used the following relationship to calculate the fugacity capacity of couch PUF.

$$Z_{PUF} = \frac{K_{PUF-air}}{R \times T}$$

where Z_{PUF} is the fugacity capacity of couch PUF (mol/m^3·Pa), R is the ideal gas constant (8.314 Pa·m^3/mol·K), and T is the ambient temperature (298 K). Then, we calculated the amount of SVOCs sorbed in couch PUF by multiplying the fugacity capacity and the volume (m^3) of couch PUF. To compare chemical mass in couch PUF with that in other indoor compartments (i.e., air, carpet, vinyl flooring, and walls), we also multiplied the fugacity capacity and the representative volume of each compartment. Equations and parameters used in calculating the fugacity capacity and the amount of SVOCs for other indoor compartments are available in Tables S3, S4, and S5.

3. Results

3.1. Measured PUF concentrations

Among 64 studied compounds, 29 compounds were detected in at least one of the 39 PUF samples (Table 1 and Table S2). Among the 29 detected compounds, 15, 13, and 11 compounds were detected in more than 50% of the samples in the top, top-middle, and middle sections, respectively, and 11 compounds were detected in more than 50% of the samples at all three depths. For 26 out of the 29 detected compounds, detection frequency (%) was the highest at the top section. The highest geometric mean (GM) concentration at the top section was observed in acetyl tributyl citrate (7.9×10^4 ng/g of PUF), followed by phenol (3.3×10^4 ng/g of PUF), homosalate (1.2×10^4 ng/g of PUF), 2-benzylideneoctanal (9.9×10^3 ng/g of PUF), and galaxolide (7.7×10^3 ng/g of PUF).

Among 64 studied compounds, 35 compounds were not detected in any samples and 18 compounds were detected in less than 50% of the samples at least from one depth (Table S2). Most of the non-detected compounds were insecticide ingredients (i.e., 16 out of 20), phenols (i.e., 13 out of 18), and PAHs (i.e., 5 out of 10). Among those detected in less than 50% of the dust samples in the same home (Kim et al., 2020), all were not detected or detected in less than 50% of the PUF samples as well, except for phenol and 4-chloro-3-methylphenol. Among those detected in more than 50% of the dust samples, 7 compounds (i.e., benzyl butyl phthalate, di-n-octyl phthalate, acetyl tributyl citrate, bifenthrin, permethrin, octocrylene, squalene) were not detected or detected in less than 50% of the PUF samples.

3.2. Changes in PUF concentrations inside couch cushions

Among the 11 compounds detected in more than 50% of the samples at all three depths, the GM PUF concentrations of 6 compounds (i.e., diethyl phthalate (DEP), phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide and homosalate) decreased from top to middle, whereas those of ortho-cresol + meta-cresol [cresol (o, m)] increased from top to middle (Figure 1). The GM concentrations of the other 4 compounds did not appear to decrease or increase with depth. Similarly, the mean concentrations of 5 compounds (i.e., phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate) were different between two

depths (p-value < 0.05) and decreased from top to middle (Figure S1). Cresol (o, m) had lower mean PUF concentrations in the top section than in the middle section, and its difference between two depths was borderline significant (p-value = 0.06).

For PUF concentrations measured in two different couches in the same home, differences of the measured PUF concentrations were within one order of magnitude between two couches in the top section for most compounds (Figure S2). Compared to the top section, the differences of the measured PUF concentrations between two couches were higher in the top-middle and middle sections for most compounds. In one of the two homes (Home B in Figure S2), concentrations were higher in a velvet-upholstered couch than a microfiber-upholstered couch for most compounds.

3.3.   Association between PUF and dust concentrations

Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, 4 compounds (i.e., di-isobutyl phthalate [DiBP], dimethyl phthalate [DMP], galaxolide, homosalate) showed statistically significant positive correlations between concentrations in the two media (r = 0.66-0.91, p-value < 0.05, Table 1). Except for phenol and pyrene, correlation coefficients for the other compounds (i.e., DEHP, di-n-butyl phthalate [DBP], diethyl phthalate [DEP], acetyl tributyl phthalate, phenanthrene, 2-benzylideneoctanal, tonalide) were generally positive, but not statistically significant (r = 0.05-0.51, p-value > 0.05). For the same 13 compounds above, the ratios of C_{PUF} to C_{dust} varied by use category or chemical class. The ratios ranged from 0.004 to 0.30 for 5 phthalates and 1 other plasticizer and ranged from 3.4 to 5.7 for four skin applied compounds (3 fragrance ingredients and 1 UV filter). Phenol and 4-chloro-3-methylphenol had the highest ratio (= 36.1) and the second highest ratio (= 10.2), respectively, among all detected compounds.

3.4.   Calculated $K_{PUF\text{-}air}$

Among the 13 compounds detected in more than 50% of both the top PUF samples and dust samples, calculated log $K_{PUF\text{-}air}$ values varied from 2.46 (DMP) and 7.80 (homosalate) (Table S6). The calculated log $K_{PUF\text{-}air}$ values increased with increasing log K_{oa} values (log $K_{PUF\text{-}air}$ = 0.64 log K_{oa} + 0.01, r^2 = 0.62) (Figure 2). The slopes of the regression equations and r^2 values increased when regressing by individual use categories (e.g., r^2 = 0.92 for 6 phthalates/plasticizers and r^2 = 0.99 for 4 skin-applied compounds such as fragrance ingredients and UV filters) (Figure S3). Our regression slope for all 13 compounds was similar to those obtained in two previous studies: 0.64 for the current study, 0.64 for Shoeib and Harner (2002), and 0.66 for Francisco et al. (2017). Our calculated log $K_{PUF\text{-}air}$ values were on the same magnitude of those of other studies for 4 skin-applied compounds (3 fragrance ingredients and 1 UV filter) but were approximately 1 to 3 orders of magnitude smaller than those of the other studies for other compounds (Table S6). On the other hand, the calculated log $K_{PUF\text{-}air}$ values decreased with increasing log VP values (log $K_{PUF\text{-}air}$ = −0.60 log VP + 3.98, r^2 = 0.47) (Figure 2). Our calculated log $K_{PUF\text{-}air}$ values were approximately 1 to 3 orders of magnitude lower than those of Zhao et al. (2004), except for phenol, 2-benzylideneoctanal, galaxolide, and homosalate.

3.5. Holding capacity of indoor compartments for SVOCs

Among the 13 compounds for which $K_{PUF\text{-}air}$ values were calculated in this study, the fugacity capacity of couch PUF (mol/m^3·Pa) ranged from 1.2×10^{-1} (DMP) to 2.6×10^4 mol/m^3·Pa (Homosalate) (Table S7). The fugacity capacity of couch PUF was relatively high for 4 skin-applied compounds (3 fragrance ingredients and 1 UV filter), compared to other chemical classes or use categories. The fugacity capacity of couch PUF was 3 to 8 orders of magnitude higher than that of air for all compounds and was up to 3 orders of magnitude lower than that of carpet and vinyl flooring for most compounds, except for galaxolide and homosalate. However, when comparing chemical mass distribution among five indoor compartments (i.e., couch PUF, air, carpet, walls, vinyl flooring), more than 20% of the total mass was distributed to couch PUF for phenol, 2-benzylideoneoctanal, galaxolide, and homosalate while less than 10% of the total mass was distributed to couch PUF for the other compounds (Figure 3).

4. Discussion

To examine the potential utility of couch PUF as a passive sampling medium and as a reservoir for indoor non-FR SVOCs commonly detected in the home, we measured concentrations of 64 non-FR SVOCs in couch PUF samples at varying depths and examined concentration changes with depth. Among the studied non-FR compounds, 29 compounds were detected in at least one of the 39 samples, and 11 compounds were detected in more than 50% of the samples at all three depths. In addition, detection frequency (%) was the highest at the top section for 26 out of the 29 detected compounds and the geometric mean PUF concentrations decreased from top to middle for 6 compounds, supporting that couch PUF can absorb many SVOCs from air. We observed that our calculated $K_{PUF\text{-}air}$ increased with K_{oa} and decreased with VP, which is consistent with previous studies, and that K_{oa} (r^2 = 0.62) was a stronger predictor of $K_{PUF\text{-}air}$ than VP (r^2 = 0.47). For compounds whose our calculated $K_{PUF\text{-}air}$ are comparable to those in chamber studies, we observed that more than 20% of the total mass can be distributed to couch PUF, indicating that couch PUF may be an important reservoir for these compounds.

We evaluated the potential utility of couch PUF as a passive sampling medium for indoor SVOCs by comparing our $K_{PUF\text{-}air}$ values calculated with direct concentration measurements in couch PUF with those measured or calculated in two chamber studies under controlled laboratory conditions (Zhao et al., 2004; Francisco et al., 2017) or in an indoor field study with purposefully-designed PUF air samplers (Shoeib and Harner, 2002b). We observed that for most compounds, our calculated $K_{PUF\text{-}air}$ values were approximately 1 to 3 orders of magnitude smaller than those estimated using predictive relationships developed in the other three studies. One potential reason for these discrepancies is that equilibrium might not be reached between couch PUF and air in our system. The increasing or decreasing concentration profiles in the cushion (Figure 1) are a sign that for many of the compounds, equilibrium was not reached between PUF and air or inside the cushion. Interestingly, for four compounds (3 fragrance ingredients and 1 UV filter) that people "wear" or apply on the skin, our calculated $K_{PUF\text{-}air}$ values were on the same order of magnitude of those of the other three studies (Figure 2). Skin-applied compounds can be

transferred by both dermal contact and diffusive mass-transfer from air, whereas phthalates and PAHs that people mostly do not wear or apply on the skin are transferred to couch PUF mostly by diffusive mass-transfer from air. Thus, it is likely that these skin-applied compounds had higher mass-transfer rates to couch PUF than phthalates or PAHs with similar K_{oa} values. Other possible reasons include the effect of upholstery and other indoor sinks or removals. For example, mass-transfer of SVOCs from indoor air to couch PUF may be limited by couch upholstery because it acts as resistance to mass-transfer between air and PUF (Klopffer and Flaconneche, 2001). In addition, compared to chamber studies where PUF competes for SVOCs primarily with air, couch PUF in our study competes with other indoor sinks (e.g., airborne particles, dust, surface film, and other indoor surfaces) (Weschler and Nazaroff, 2008, 2010; Little et al., 2012; Weschler and Nazaroff, 2017) and removal processes (e.g., ventilation, vacuuming and surface cleaning) (Shin et al., 2012; Shin et al., 2013). Lastly, as people continuously sit on couches, air pockets inside couch PUF are likely to be pushed out, resulting in lower concentrations of gas-phase chemicals in couch PUF than in the PUF of chamber studies. Thus, physically altered PUF in our study may partly contribute to the discrepancies in $K_{PUF\text{-}air}$ values between our study and previous chamber studies.

Among the 13 non-FR compounds detected in more than 50% of both the top PUF samples and dust samples, we observed positive correlation coefficients between the two media for DMP, DiBP, galaxolide, and homosalate (p-value < 0.05). DMP, with the lowest log K_{oa} value (= 5.68) among the five phthalates (i.e., DEHP, DiBP, di-n-butyl phthalate, DEP, DMP), had the highest correlation coefficient ($r = 0.78$). Its low K_{oa} value may accelerate mass-transfer between air and dust and between air and PUF (Schripp et al., 2010; Dodson et al., 2015). Because two fragrance ingredients (galaxolide and homosalate) are likely to be transferred (1) to couch PUF via direct skin contact with couch surfaces and (2) to carpet dust via skin flaking (Weschler et al., 2011), it is likely that we observed positive correlation coefficients of these two compounds. For four skin applied compounds (3 fragrance ingredients and 1 UV filter), we observed relatively high ratios of C_{PUF} to C_{dust} from 3.4 to 5.7, implying high transfer rates of these compounds from skin to couch PUF. On the other hand, the other 9 compounds did not show statistically significant correlation coefficients, potentially due to high within-home variability of dust concentrations of these compounds or a small sample size. For example, the correlation coefficients were relatively small for phenanthrene and pyrene ($r = 0.05$, -0.10, respectively), presumably because of the seasonal variations in source rates of indoor PAHs including increased infiltration of outdoor air during summer and increased residential heating such as wood burning during winter.

Our study showed that PUF in upholstered home couches can absorb SVOCs because a wide range of SVOCs which are not known to be added to couch PUF were detected in our samples, including phthalates, other plasticizers, insecticide ingredients, PAHs, fragrance ingredients, UV filters, and skin oils (Table 1). We observed the highest detection frequency in the top section for most compounds, supporting that home couches absorb SVOCs from air. Among the 11 compounds detected in more than 50% of the samples at all depths, concentrations of phenanthrene, 2-benzylideneoctanal, galaxolide, tonalide, and homosalate decreased with depth (Figure 1 and S1), showing potential inward diffusion of these compounds from air to couch PUF. We also observed that phenol and three skin-applied

compounds were distributed to couch PUF with relatively high percentage (21.2 – 60.5%) of the total mass among five indoor compartments (Figure 3). These compounds were measured in relatively high concentrations in couch PUF (Table 1) and our predicted $K_{PUF\text{-}air}$ values of these compounds were similar to those in other chamber studies. Thus, couch PUF may act as an important holding reservoir for these compounds. Moreover, we found that SVOC concentrations in PUF were higher in a velvet-upholstered couch than a microfiber-upholstered couch for most compounds (Figure S2). The reason for this difference is less clear but may include the differences in age of couches or materials of upholstery (Boor et al., 2015; Keimowitz et al., 2016). The age of couches in the home was not available for this study. We also observed that seven compounds, detected in more than 50% of the dust samples collected in the same home, were not detected or detected in less than 50% of the PUF samples. Because these seven compounds have relatively high K_{oa} values (e.g., log K_{oa} > 9.5), it is likely that compounds with high K_{oa} values may not be absorbed by the PUF well enough to be detected in the PUF.

Our study also showed that PUF in home couches may release some phenolic compounds including phenol, 4-chloro-3-methylphenol, and cresol (o, m). For example, among the same 11 compounds above, phenol had the highest geometric mean PUF concentration (32,832 ng/g of foam) and a relatively low geometric mean dust concentration (910 ng/g of dust) (Table 1), resulting in the highest ratio of C_{PUF} to C_{dust} (= 36.1). The geometric mean PUF concentration of 4-chloro-3-methylphenol (7,186 ng/g of foam) was the fifth highest, whereas it was detected in none of our dust samples. Furthermore, we observed a decreasing trend in PUF concentrations of cresol (o, m) from the middle section to the top section (p-value = 0.06, Figure S1). Based on the U.S. EPA's CompTox Chemistry Dashboard (Williams et al. 2017), phenol has a product or use category of 'furniture and furnishings', and 4-chloro-3-methylphenol has a chemical functional use of 'flame retardants'. Those couches with relatively high phenol concentrations tended to decrease from middle to top sections. These showed potential outward diffusion of these phenolic compounds from couch PUF to indoor air, suggesting that cresol (o, m) may have been present in the foam as manufactured.

We noted two major strengths of this study. Because upholstery may act as resistance of mass-transfer between air and couch PUF, direct concentration measurements in upholstered couch PUF allowed us to observe differences in PUF's holding capacity between upholstered couch PUF and a bare PUF disk. In addition, because a wide range of SVOCs are detected in indoor environmental media (e.g., airborne particles, dust, indoor surface), our novel sampling of upholstered couch PUF with different depths and subsequent chemical analysis helped understand its role on the absorption and release of SVOCs. However, some limitations should be noted. First, we did not differentiate the effect of different upholstery materials (e.g., leather, fabric) on couch PUF's holding capacity due to the limited sample size. Second, our predictive equations for $K_{PUF\text{-}air}$ versus K_{oa} may not be applicable to all SVOCs, because when regressing by individual use categories or chemical classes, the slope of $K_{PUF\text{-}air}$ against K_{oa} was approximately 30% steeper for skin-applied compounds than plasticizers. It is likely that these skin-applied compounds were primarily transferred to couch PUF via direct skin contact or skin flaking rather than molecular diffusion or deposition from air to couch PUF. Third, because only 4 out of the 11

compounds were correlated between the top PUF concentrations and dust concentrations, our calculated $K_{PUF\text{-}air}$ using C_{PUF} and C_{dust} may not be valid for those that were not correlated each other. Fourth, because of a small sample size and low detections in the current study, our predictive equations should be interpreted with caution. Fifth, we also analyzed 16 FRs in our samples. However, the results are difficult to interpret, and thus are presented and discussed in the Supporting Information. We were not always able to ascertain whether the couch PUF was manufactured with each particular FR or if the FR was initially in the fabric. For example, we observed four FRs that had generally low concentrations in all three sections and very high concentrations in the top section (Figure S4). In addition, we could not decisively determine if the couches may have absorbed these compounds or if they may have these compounds in upholstery fabric. For example, they were not frequently detected in fabric (i.e., less than 15%), limiting interpretation and confirmation of our results. Lastly, we did not measure the density of the couch PUF samples in this study, although different densities of PUF are known to affect its chemical uptake (Chaemfa et al., 2008; Chaemfa et al., 2009).

5. Conclusions

Our study showed that couch PUF can absorb many SVOCs. However, because most SVOCs were less frequently detected in couch PUF and had low correlation coefficients between concentrations in dust and couch PUF, couch PUF may not be an effective passive sampling medium for SVOCs. We observed that phenolic compounds and skin-applied compounds were detected higher in couch PUF than in dust (i.e., a ratio of C_{PUF}/C_{dust} was larger than 3.0), implying that direct skin contact with couch surfaces or inhalation while sitting on couches might be an important exposure route of these compounds, particularly for infants and young children who did not apply the compound to their skin. We also found that more than 20% of the total mass was distributed to couch PUF for phenol and three skin-applied compounds, implying that couch PUF is an important holding reservoir for these compounds. As different upholstery materials may have different degrees of resistance of mass-transfer between air and PUF, additional studies are needed to examine the effect of various materials on couch PUF's holding capacity and to analyze for SVOCs on the surfaces of home furniture with frequent skin contact.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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- Measured concentrations of 64 SVOCs in couch PUF samples at varying depths.

- Detection frequency was the highest at the top section for most detected compounds.

- Observed difference and similarity in $K_{PUF-air}$ between upholstered PUF and bare PUF.

- Couch PUF may be an important reservoir for some compounds.

- Dermal contact with couch seats may be an important exposure route for skin-applied compounds.
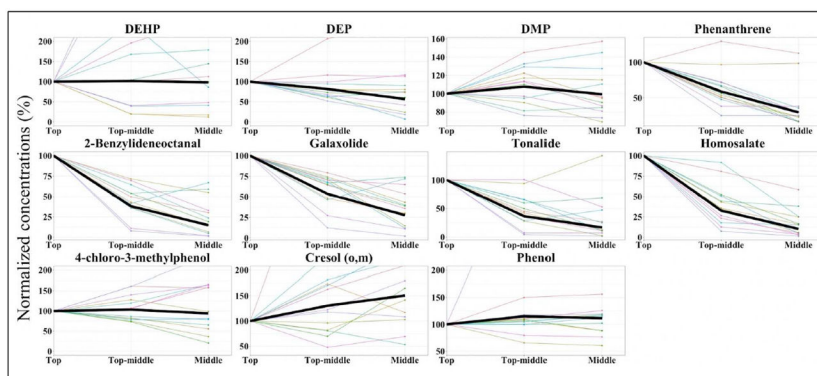
Figure 1.
Measured PUF concentrations at three depths of 13 couch cushions (i.e., top, top-middle, middle) after normalizing the top-middle and middle concentrations to the top concentrations. Phenol and 4-chloro-3-methylphenol had geometric mean (GM) concentrations higher than 6,000 ng/g of PUF in at least two of three depths, while others included in this figure had GM concentrations less than 6,000 ng/g of PUF in at least two of three depths. Black lines in bold represent normalized GM concentrations.
Abbreviation: di(2-ethylhexyl) phthalate (**DEHP**), diethyl phthalate (**DEP**), dimethyl phthalate (**DMP**), ortho-cresol + meta-cresol (**Cresol (o, m)**)
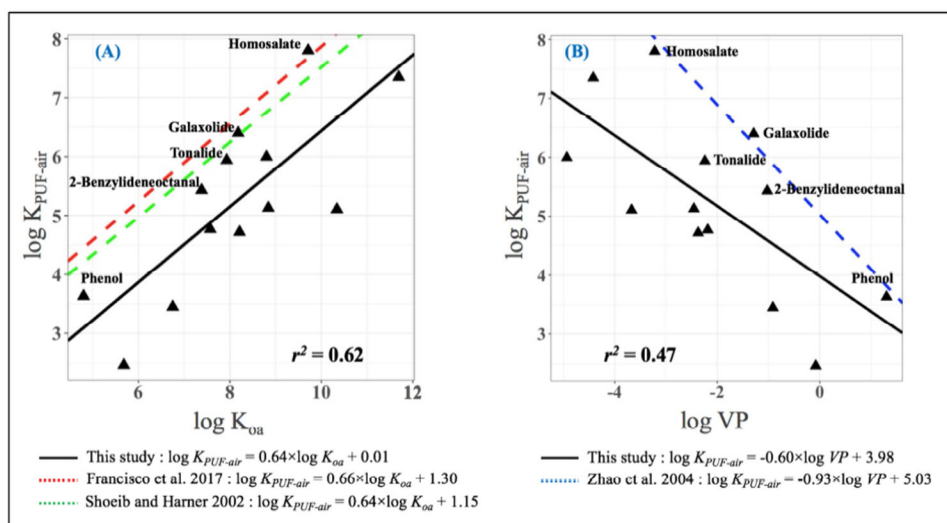
Figure 2.
Calculated median log $K_{PUF\text{-}air}$ versus log K_{oa} (A) or vapor pressure (B) for 13 compounds detected in more than 50% of both the top PUF samples and dust samples. Regression equations from other three studies are shown in dotted lines. Compound names shown in the plots are fragrance ingredients, UV filters or phenol. All $K_{PUF\text{-}air}$ values are expressed in the unit of m^3 of air/m^3 of PUF.
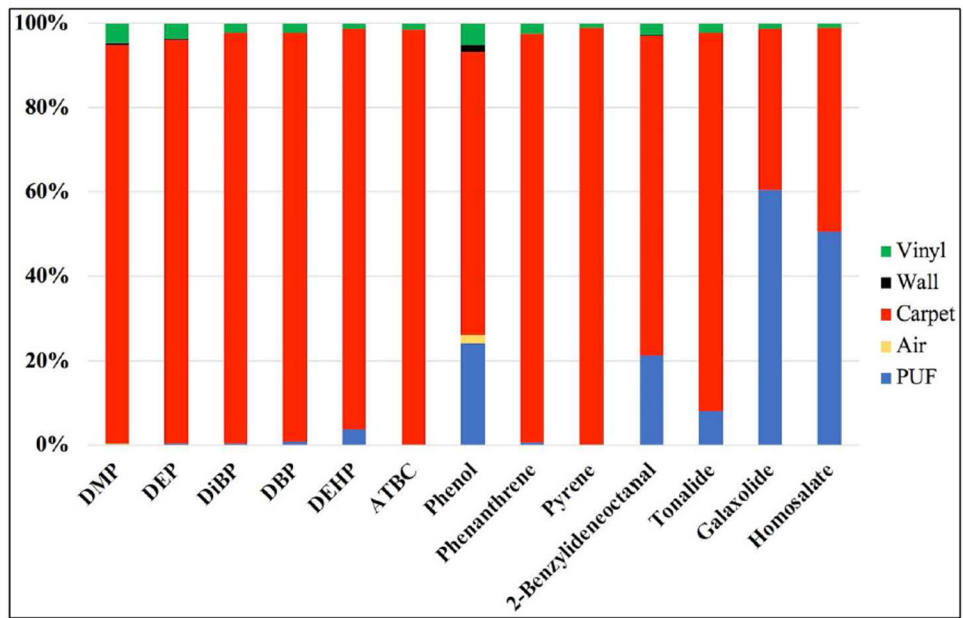
Figure 3.
Mass distribution (%) of 13 compounds among five indoor compartments.

Table 1.

Summary of measured concentrations for 29 SVOCs detected in PUF samples ($n = 39$) and dust samples ($n = 11$).

Use category or chemical class	Compound name	Couch PUF concentration (ng/g of PUF)		GM[a]			Dust concentration[b] (ng/g of dust)				r[c]	C_PUF/C_dust[d]
		LOD	LOQ	Top	Top-middle	Middle	LOD	DF (%)	GM (n=11)			
Phthalates	Benzyl butyl phthalate	500	1,000	-	-	-	1,000	100	7,715	-	-	
	Di(2-ethylhexyl) phthalate	1,000	1,000	1,447	1,469	1,425	10,000	91	66,778	0.10	0.02	
	Di-isobutyl phthalate [e]	250	1,000	1,710	594	301	1,000	100	8,883	0.71*	0.19	
	Di-n-butyl phthalate [e]	500	1,000	850	354	-	1,000	100	8,213	0.32	0.10	
	Di-n-octyl phthalate [e]	100	500	-	-	-	100	73	143	-	-	
	Diethyl phthalate	50	100	574	469	325	100	100	1,890	0.20	0.30	
	Dimethyl phthalate [e]	10	25	30	33	30	50	100	122	0.78*	0.25	
Other plasticizers	Acetyl tributyl citrate	10,000	25,000	79,029	-	-	10,000	100	2.1×10^7	0.20	0.004	
	Bis(2-ethylhexyl) adipate	1,000	5,000	-	-	-	10,000	55	13,088	-	-	
Insecticide ingredients	Bioallethrin	100	500	-	-	-	100	0	-	-	-	
	Esfenvalerate	100	250	-	-	-	500	0	-	-	-	
	Permethrin	250	250	-	-	-	100	91	1,031	-	-	
	Phenothrin	50	100	-	-	-	100	9	-	-	-	
Phenols	2-Nitrophenol	100	100	-	-	-	1,000	0	-	-	-	
	4-Chloro-3-methylphenol	100	250	7,186	7,431	6,752	1,000	0	-	-	-	
	Cresol (o, m)	100	100	168	219	253	500	0	-	-	-	
	para-Cresol	1,000	1,000	-	-	-	1,000	73	1,663	-	-	
	Phenol	250	250	32,832	37,774	36,642	1,000	91	910	-0.12	36.1	
Polycyclic aromatic hydrocarbons	Anthracene	10	50	-	-	-	100	0	-	-	-	
	Benzo(a)anthracene	10	25	-	-	-	100	36	-	-	-	
	Fluorene	10	50	-	-	-	100	9	-	-	-	
	Phenanthrene	10	50	226	133	66	100	100	268	0.05	0.84	
	Pyrene	5	25	123	-	-	50	100	168	-0.10	0.73	

Use category or chemical class	Compound name	Couch PUF concentration (ng/g of PUF)					Dust concentration [b] (ng/g of dust)			r [c]	C_{PUF}/C_{dust} [d]
		LOD	LOQ	GM [a]			LOD	DF (%)	GM (n = 11)		
				Top	Top-middle	Middle					
	2-Benzylideneoctanal	100	250	9,850	3,745	1,448	500	100	2,062	0.46	4.78
Fragrance ingredients	Galaxolide	5	25	7,712	4,130	2,124	50	100	1,363	0.66*	5.66
	Tonalide	10	50	1,092	399	183	100	91	324	0.51	3.37
Ultraviolet filters	Homosalate	25	50	12,041	3,936	1,236	250	100	2,416	0.91*	4.98
	Octocrylene	100	250	-	-	-	100	100	6,624	-	-
Skin oils	Squalene	1,000	1,000	-	-	-	1,000	100	71,984	-	-

[a] Geometric means (ng/g) at each depth (i.e., top, top-middle, middle) from 13 PUF samples.

[b] Data from Kim et al. (Kim et al., 2020).

[c] Pearson correlation coefficients (r) of ln-transformed concentrations between PUF (C_{PUF}) and dust (C_{dust}) for 13 compounds detected in more than 50% of both the top PUF samples and dust samples. P-values < 0.05 are marked with asterisk (*).

[d] Ratios of couch PUF concentrations (C_{PUF}) to dust concentrations (C_{dust}).

[e] For samples below the LOQ or between the LOD and the LOQ, we substituted them by dividing the LOQ by 2.

Abbreviation: limit of detection (**LOD**), limit of quantification (LOQ), detection frequency (**DF**), geometric mean (**GM**), ortho-Cresol + meta-Cresol (**Cresol (o, m)**))