

UC Davis

UC Davis Previously Published Works

Title

Temporal variability of indoor dust concentrations of semivolatile organic compounds

Permalink

<https://escholarship.org/uc/item/8w56p00j>

Journal

Indoor Air, 31(3)

ISSN

0905-6947

Authors

Kim, Kyunghoon
Shin, Hyeong-Moo
Wong, Luann
[et al.](#)

Publication Date

2021-05-01

DOI

10.1111/ina.12759

Peer reviewed



Published in final edited form as:

Indoor Air. 2021 May ; 31(3): 693–701. doi:10.1111/ina.12759.

Temporal variability of indoor dust concentrations of semivolatile organic compounds

Kyunghoon Kim^a, Hyeong-Moo Shin^{a,*}, Luann Wong^b, Thomas M. Young^b, Deborah H. Bennett^c

^aDepartment of Earth and Environmental Sciences, University of Texas, Arlington, TX, USA

^bDepartment of Civil and Environmental Engineering, University of California, Davis, CA, USA

^cDepartment of Public Health Sciences, University of California, Davis, CA, USA

Abstract

The determinants of the temporal variability of indoor dust concentrations of semivolatile organic compounds (SVOCs) remains mostly unexplored. We examined temporal variability of dust concentrations and factors affecting dust concentrations for a wide range of SVOCs. We collected dust samples three times from 29 California homes during a period of 22 months and quantified concentrations of 47 SVOCs in 87 dust samples. We computed intraclass correlation coefficients (ICCs) using three samples collected within the same house. We calculated correlation coefficients (r) between two seasons with similar climate (spring and fall) and between two seasons with opposite climate (summer and winter). Among 26 compounds that were detected in more than 50% of the samples at all three visits, 20 compounds had ICCs above 0.50 and 6 compounds had ICCs below 0.50. For 19 out of 26 compounds, correlation coefficients between spring and fall ($r = 0.48–0.98$) were higher than those between summer and winter ($r = 0.09–0.92$), implying seasonal effects on dust concentrations. Our study showed that within-home temporal variability of dust concentrations was small ($ICC > 0.50$) for most SVOCs, but dust concentrations may vary over time for some SVOCs with seasonal variations in source rates, such as product use.

Keywords

concentration; dust; ICC; source rate; SVOC; temporal variability

1. Introduction

Exposure to semivolatile organic compounds (SVOCs) in indoor environments and its potential impact on human health have been receiving increased public attention, because people in developed countries spend over 80% of their time indoors [1] and SVOC levels are several orders of magnitude higher indoors than outdoors [2–4]. SVOCs are introduced into indoor residential settings in the form of consumer products, building materials, furnishings,

*Corresponding author: Hyeong-Moo Shin, Ph.D., University of Texas, Arlington, 500 Yates Street, Box 19049, Arlington, TX, 76019, hyeongmoo.shin@uta.edu, Voice: 949-648-1614, Fax: 817-272-2628.

Conflict of interest

The authors declare that they have no competing interest.

pesticides, and combustion by-products [5, 6]. When indoor SVOCs are released from their original sources, they are redistributed over time among the gas phase, airborne particles, settled dust and other indoor surfaces [6, 7]. Consequently, residents can be exposed to indoor SVOCs via inhalation, dermal uptake, and dust ingestion [8]. Of interest, for young children who crawl and play on the floor and have frequent hand-to-mouth activity, dust ingestion has been determined to be a major non-dietary exposure route for several classes of SVOCs including phthalates [9, 10], polybrominated diphenyl ethers (PBDEs) [10, 11], organophosphate flame retardants (OPFRs) [12], per- and polyfluoroalkyl substances (PFAS) [9, 13, 14], polycyclic aromatic hydrocarbons (PAHs) [15], and polychlorinated biphenyls (PCBs) [16].

The measured indoor dust concentrations have been found to have positive correlations with concentrations in biological samples (e.g., serum, urine) for PBDEs [17, 18], OPFRs [19–21], PCBs [22], bisphenol A [23], and pyrethroid pesticides [24]. Moreover, epidemiologic studies using indoor dust concentrations as surrogates for human exposure found positive associations between dust concentrations of tributyl phosphate and incidence of asthma and allergies [25] as well as di(2-ethylhexyl) phthalate (DEHP) and incidence of allergies [26]. It was also found that increasing levels of biocides (i.e., organochlorines, residential insecticides) in dust were known to be associated with increasing risk of non-Hodgkin lymphoma [27–29]. This suggests that for young children who are exposed to SVOCs primarily via dust ingestion, indoor dust concentrations collected from a single visit can be used as surrogates for exposure to some SVOCs, particularly if diet and personal care product use are not significant exposure pathways.

SVOCs are fairly persistent indoors [5, 30] and therefore the temporal variability of SVOC dust concentrations is typically smaller than that of concentrations in indoor air [31]. In addition, compared to volatile organic compounds that are typically released into air from dynamic indoor sources, many SVOCs are slowly released from building materials, furnishings and other indoor solid items and have a strong tendency to partition to settled dust. Thus, SVOC dust levels are relatively stable over time. However, several studies observed different degrees of temporal variability of SVOC dust concentrations across chemical classes or within the same chemical class [18, 31–33]. Episodic occupant activities (e.g., cooking, vacuuming, ventilation) and environmental factors (e.g., temperature, relative humidity, airborne particulate matter concentration) are also known to affect SVOC air and/or dust concentrations during a short period of time [34, 35]. Seasonal factors (e.g., different product use pattern, ventilation frequency) may also play a role in temporal variability in measured dust concentrations [36]. Thus, further studies are needed to extensively examine temporal variability of dust concentrations for a wide range of SVOCs detected in household dust and other determinants (e.g., chemical properties, seasonal difference in use patterns or emission sources) of the temporal variability of dust concentrations.

The objective of this study was to investigate temporal variability of dust concentrations for a wide range of SVOCs. Specifically, we quantified 47 compounds in dust samples repeatedly collected from 29 homes during a period of 22 months. The measured SVOCs represent a broad range of compounds across chemical properties and product use

categories, and thus are ideal for evaluating the sources of any differences in temporal concentration patterns. We also examined whether sampling season affected measured dust concentrations using samples collected at two seasons with similar climate (spring and fall) and two seasons with opposite climate (summer and winter).

2. Methods

2.1. Dust sample collection

We recruited households in Northern California as part of an effort to examine the overall decrease of flame retardant (FR) concentrations in indoor dust after replacing old couches (assumed to be the primary sources of flame retardants in participating homes). Under the assumption that dust concentrations of non-FR SVOCs were not affected by the replacement of old couches, we examined the temporal variability of non-FR SVOCs. Households were selected as part of one of two studies; one study where participants bought their couch by themselves and the other where the study team bought a couch. In total, 34 household replaced their couches, and of these, 29 completed three visits where a dust sample was collected over 22 months, from July 2015 to May 2018, for a total of 87 dust samples. The second visit was approximately 15.3 months apart from the first visit ($\sigma = \pm 2.6$ months), allowing us to examine variability of dust concentrations during a long period of time. The third visit was approximately 6.3 months apart from the second visit ($\sigma = \pm 0.8$ months), allowing us to examine variability of dust concentrations during a short period of time. 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.

For each home visit, samples were collected from the main living room under the assumption that the main living room is where dust concentrations are of interest for residential exposure assessment. One standardized protocol is to vacuum all surfaces except for under furniture and between cushions [37, 38], but we modified the protocol to also exclude vacuuming upholstered furniture. Dust was collected with a Eureka Mighty-Mite vacuum cleaner equipped with the standard crevice tool attachment (Model 3670), modified to capture dust in a 19×90 mm cellulose extraction thimble (Whatman Inc) [37, 38]. The thimbles containing the dust samples were wrapped in pre-cleaned aluminum foil, placed in 50 ml polypropylene vials, shipped in a cooler to the central repository, and stored at $-20\text{ }^{\circ}\text{C}$ at UC Davis until analysis. The dust was shaken from each thimble into a 100-mesh stainless steel sieve and then sieved to obtain the fraction of dust smaller than $150\text{ }\mu\text{m}$.

2.2. Target compounds

We used a target compound list developed through a project looking at widely-detected compounds in household dust [39, 40], including new compounds, originally detected through suspect screening or nontarget identification with high resolution mass spectrometry, for which we subsequently obtained standards. Using this expanded target compound list, we analyzed all compounds that were previously detected in dust via gas chromatography (GC). To find the most common and primary use category of studied compounds, we relied on use categorization in the U.S. Environmental Protection Agency

(EPA)'s Consumer Product Chemical Profiles database [41] and the U.S. National Library of Medicine's Household Product Database. Details of use categorization are available elsewhere [39]. The selected compounds include ultraviolet [UV] filters, fragrances, and other ingredients of personal care products (PCPs); insecticide ingredients; and a variety of other compounds widely detected in homes (phenols, phthalates, other plasticizers, PAHs, and skin oils, those lipids found in the skin such as squalene). We excluded 11 compounds used as flame retardants from this analysis because replacing couches would affect dust concentrations of flame retardants. The selected compounds represent a range of chemical properties, ranging from relatively volatile compounds (e.g., dimethyl phthalate [DMP]) to those with a high tendency to partition to dust (e.g., DEHP). Chemical properties including the octanol-air partition coefficient ($\log K_{oa}$) and use categories of our study compounds are listed in Table S1 of Supporting Information.

2.3. Sample analysis

Details of the analytical methods and settings are found elsewhere [39, 40]. Briefly, we quantified concentrations of 47 compounds using GC quadrupole time-of-flight (Q/TOF) mass spectrometry (MS) at UC Davis. Analysis was carried out on an Agilent 7890B gas chromatograph with a HP-5MS (30 m \times 0.25 mm, 0.25 μ m) column coupled to an Agilent Q/TOF 7200B instrument running in electron ionization (EI) mode. A 78 min run time with a linear temperature gradient from 35 to 325 $^{\circ}$ C was chosen to separate all selected compounds and all major peaks in the analysis of a dust extract. Absolute recovery and precision for each analyte are available in Table S1.

2.4. Statistical analysis

We performed all statistical analyses using STATA/IC 15.1 (StataCorp LLC, College Station, TX, USA) and R version 3.6.1. For all analyzed compounds, we provided summary statistics of dust concentrations for each of the three visits. For all other statistical analyses that require sufficient detection of the samples, we included compounds detected in more than 50% of the samples at all three visits. For concentrations below the limit of detection (LOD), we assigned a value of the LOD divided by the square root of 2 [42]. We compared the distributions of dust concentrations for all three visits and performed a paired t-test of log-transformed concentrations to compare means between two visits (i.e., 1st visit versus 2nd visit, 2nd visit versus 3rd visit, 1st visit versus 3rd visit) and adjusted p-values for multiple comparison of means with a false discovery rate method [43]. Sources of indoor SVOCs may vary with several household characteristics. Thus, we examined if there were significant differences in SVOC dust concentrations between groups of two household characteristics: household income (<\$100K, \$100K-\$150K, >\$150K) and presence of children (yes, no). We performed the Wilcoxon-Mann-Whitney test for a binary variable and the Kruskal-Wallis test for the other categorical variable.

To test for within-home temporal variability of SVOC dust concentrations during the three visits (~22 months), we computed the intraclass correlation coefficient (ICC) of individual compounds, a ratio of between-home variance to total variance (within-home variance + between-home variance). ICC ranges from 0 (no reproducibility of within-home measurements) to 1 (perfect reproducibility of within-home measurements) [44, 45]. We

used natural log (ln)-transformed concentrations to account for skewed distributions of dust concentrations and then calculated ICCs and 95% confidence intervals (CIs) using a mixed-effects model with sampling date for fixed effects [46]. We examined the relationship between ICCs and log K_{oa} within the same use category or chemical class. Because K_{oa} is a strong predictor of SVOC partitioning between indoor air and dust [7], it may affect within-home temporal variability of SVOC dust concentrations. For our study compounds, log K_{oa} values ranged from 5.68 for DMP to 11.74 for di-n-octyl phthalate (DnOP) and octocrylene.

Because we collected dust samples with different time intervals (i.e., ~15 months between the 1st and 2nd visits and ~6 months between the 2nd and 3rd visits), we also examined the effect of different sample collection time intervals on the magnitude of temporal variability of dust concentrations by computing Pearson correlation coefficients (r) of ln-transformed concentrations between two visits. To examine whether sampling season affected dust concentrations because of seasonal variations in ventilation or product use (e.g., UV filters, pesticides), we computed Pearson correlation coefficients of ln-transformed concentrations using samples collected (1) between spring (March to May) and fall (September to November) for similar climate ($n = 13$ homes) and (2) between summer (June to August) and winter (December to February) for opposite climate ($n = 16$ homes). To examine the differences of correlation coefficients (1) between two different time intervals (1st and 2nd visits versus 2nd and 3rd visits) and (2) between two different sets of seasons (spring and fall versus summer and winter), we used the 'cocor' function in R which compares two correlations based on either independent or dependent groups [47].

3. Results

3.1. Measured dust concentrations

Among 47 quantified compounds, 26 compounds were detected in more than 50% of the samples at all three visits and 15 compounds were detected in more than 90% of the samples at all three visits (Table S2). The highest median from all three visits was observed in acetyl tributyl citrate [ATBC] (7.9×10^6 ng/g), followed by dioctyl terephthalate [DOTP] (9.3×10^4 ng/g), DEHP (7.6×10^4 ng/g), squalene (4.9×10^4 ng/g), and cholesta-3,5-diene (1.8×10^4 ng/g). We observed a statistically significant difference of mean concentrations between visits for 6 compounds (p-value < 0.05, see Figure 1). For 13 SVOCs, dust concentrations were associated with household income or presence of children (Table S3). Homes with the lowest household income had the highest dust concentration for di-isobutyl phthalate (DiBP), DOTP, phenanthrene, pyrene, 2-benzylideneoctanal, galaxolide, tonalide and lilial, but had the lowest dust concentration for octocrylene (p-value < 0.05). Homes with children had higher dust concentrations than those without children for ATBC, DOTP, 2-benzylideneoctanal, galaxolide, tonalide, lilial, and triethyl citrate, but had lower dust concentrations for di-n-butyl phthalate (DBP), phenanthrene and pyrene (p-value < 0.05).

Among the 47 quantified compounds, 21 compounds were detected in less than 50% of the samples at least one visit (Table S2). Most of them were insecticide ingredients, PAHs, or phenols (i.e., 7 out of 9 insecticide ingredients, 8 out of 10 PAHs, and all 5 phenols) with detection frequencies ranging between 1% and 57% among a total of 87 dust samples. The detection frequency of bis(2-ethylhexyl) adipate [DEHA] increased during three visits (38%,

55%, 59%, respectively) and the 75th percentile concentrations were relatively similar during three visits (10.6, 9.3, 9.1 $\mu\text{g/g}$ of dust, respectively). P-cresol, phenol, and benzo(b)fluoranthene were detected in more than 50% of the samples in the first visit but were less detected in later visits (7% and 14% for p-cresol, 3% and 0% for phenol, 48% and 24% for benzo(b)fluoranthene, respectively).

3.2. Within-home variability during three visits

Among the 26 compounds, 5 compounds had ICCs above 0.75, 15 compounds had ICCs between 0.50 and 0.75, and 6 compounds had ICCs below 0.50 (Figure 2). Permethrin (used as an insecticide ingredient) had the highest ICC ($= 0.90$; 95% CI: 0.83, 0.95) and benzophenone (used as a UV filter) had the lowest ICC ($= 0.30$; 95% CI: 0.08, 0.54). Within the same use category or chemical class with only 2 or 3 compounds, ICCs were higher with higher values of $\log K_{\text{oa}}$ for non-phthalate plasticizers, insecticides and UV filters, whereas ICCs were lower with higher values of $\log K_{\text{oa}}$ for PAHs and skin oils. For phthalates, fragrance ingredients, and cosmetic ingredients, ICCs did not increase or decrease with increasing values of $\log K_{\text{oa}}$ within the same use category or chemical class.

3.3. Variability across four seasons

When examining the effect of sampling seasons (collected during similar climate or opposite climate) on the magnitude of temporal variability of dust concentrations, the correlation coefficients between spring and fall ($r = 0.48\text{--}0.98$) were higher than those between summer and winter ($r = 0.09\text{--}0.92$) for 19 compounds (Figure 3). The differences of correlation coefficients between spring-fall and summer-winter were statistically significant ($p\text{-value} < 0.05$) for DMP, DOTP, octocrylene, phenanthrene, and squalene. Among the 19 compounds, DMP, which is commonly used in insect repellents [48], showed the largest difference between the two coefficients (0.09 versus 0.79) and squalene showed the second largest difference between the coefficients (0.31 versus 0.90). All insecticide ingredients, UV filters, PAHs, and skin oils had consistently lower correlation coefficients between summer and winter, compared to those between spring and fall. When comparing the distributions across four seasons, two compounds (i.e., ATBC, squalene) had variations in dust concentrations between seasons ($p\text{-value} < 0.05$, Figure S1). Mean ATBC concentrations were lower during summer than other seasons, and mean squalene concentrations were the highest during winter and the lowest during spring.

3.4. Correlation between visits

Pearson correlation coefficients (r) of ln-transformed dust concentrations between two visits for the 26 compounds were positive: $r = 0.26\text{--}0.90$ between the 1st and 2nd visits and $r = 0.34\text{--}0.95$ between the 2nd and 3rd visits (Figure S2). Except for benzophenone and DnOP, the correlation coefficients between the first two visits with a long time interval ($\mu \pm \sigma = 15.3 \pm 2.6$ months) was statistically significant ($p\text{-value} < 0.05$) for the other 24 compounds. Similarly, except for benzophenone, the correlation coefficients between the 2nd and 3rd visits with a short time interval ($\mu \pm \sigma = 6.3 \pm 0.8$ months) were statistically significant for the other 25 compounds. When the sample collection time interval between visits increased, the correlation coefficients tended to decrease for 19 out of 26 compounds. However, the

differences of correlation coefficients between 1st-2nd visits and 2nd-3rd visits were not statistically significant (p -value > 0.05) for all 26 compounds.

4. Discussion

In this study, we examined temporal variability of dust concentrations for a wide range of SVOCs over a 22-month period and investigated the effect of sampling season, log K_{oa} , and sample collection time intervals on the magnitude of temporal variability of dust concentrations. Among the 26 compounds detected in more than 50% of the samples at all three visits, 20 compounds showed moderate ($0.50 < ICCs < 0.75$) to high ($ICCs > 0.75$) reproducibility of within-home measurements in dust concentrations, while the remaining 6 compounds showed relatively low reproducibility ($ICCs < 0.5$). Overall, dust concentrations of insecticide ingredients, PAHs, and fragrance ingredients were more stable over time, with those of skin oils (or constituents of skin surface lipids) being the least stable. For those compounds with high ICCs, dust concentrations collected from a single visit may serve as surrogates for exposure for approximately 2 years, particularly for infants and toddlers. We observed higher correlation coefficients of dust concentrations between spring and fall than those between summer and winter for 19 compounds, indicating seasonal effects on dust concentrations. When the sample collection time interval between visits increased, the correlation coefficients tended to decrease for 19 out of 26 compounds, but results were not statistically significant for any compounds.

Our study showed that dust concentrations may vary over time for some SVOCs with seasonal variations in source rates. Among the 6 compounds that had relatively low reproducibility of within-home measurements in dust concentrations ($ICCs < 0.5$), two compounds are associated with PCP use including benzophenone (UV filter) and isopropyl myristate (cosmetic ingredient) with a third related to skin sloughing, squalene (found in skin surface lipids). Among them, we observed that benzophenone had the lowest concentration during winter and squalene had the highest concentration during winter (see Figure S1). The high concentration of squalene in dust might be due to reduced levels of skin moisture during winter, which would result in more skin flaking [49]. Squalene had the second biggest difference in its correlation coefficients (0.90 between spring and fall versus 0.31 between summer and winter) among the 26 compounds. Compared to the correlation coefficients between spring and fall, those between summer and winter tended to be smaller for all UV filters, skin oils, and PAHs in our samples. The reason for the difference between seasons for PAHs is less clear, as there are multiple sources, including cooking, smoking, use of candles, infiltration of outdoor air, and residential heating. Potential reasons for the differences include seasonal differences in ventilation rates or increased PAH emissions from residential heating such as wood burning during winter [50]. Our results further support that seasonal variations in source rates, either through increased product use (e.g., UV filters), increased skin flaking (e.g., squalene), or increased transport from outdoors through ventilation or residential heating (e.g., PAHs), may have increased temporal variability of these compound classes in dust concentrations.

In our study, we observed relatively low reproducibility for two phthalates (DnOP, DMP) and one non-phthalate plasticizer (ATBC), although DnOP and DMP did not have seasonal

variations (see Figure S1 for the distributions across four seasons). For example, although we did not observe differences in means across four seasons for DMP, we observed a very low correlation coefficient between summer and winter ($r = 0.09$). Sensitivity to temperature changes is known to be another factor leading to temporal variations in the household dust concentrations of compounds with small K_{oa} [7, 32, 51] because K_{oa} is inversely proportional to temperature [52] and compounds with small K_{oa} have relatively high affinity for air rather than dust. DMP and DEP have the smallest value and the second smallest value of $\log K_{oa}$ (= 5.68 and 6.75, respectively) among phthalates and non-phthalate plasticizers. Thus, dust concentrations of these two compounds may vary over time, in part, due to intermittent human activities such as ventilation, combined with a greater fraction in air, resulting in poor reproducibility in dust concentrations. In addition, after equilibrium among indoor compartments (e.g., dust, gas phase, airborne particles) is broken due to vacuuming or ventilation, SVOCs with relatively high $\log K_{oa}$ values (DnOP = 11.7 and ATBC = 10.3, respectively) do not reach equilibrium within a short period of time [5]. Thus, intermittent human activities before sample collection may have caused increased temporal variability for DnOP and ATBC.

Comparing temporal variability of dust concentrations for a wide range of SVOCs allowed us to examine the degree of variability by the chemical property relevant to dust concentrations. K_{oa} is a strong predictor of SVOC partitioning between the gas phase and settled dust in indoor environments [8]. Although our comparison by chemical properties within the use category is limited due to the small number of compounds with >50% detection frequency, ranging from 2 to 7, we observed that $\log K_{oa}$ may play a role on the magnitude of temporal variability of dust concentrations for such SVOCs. However, we also observed that K_{oa} is not a governing factor determining the magnitude of temporal variability for other chemical classes or use categories. It is likely that dust concentrations of such SVOCs may vary over time due to other factors including intermittent human activities and seasonal differences in use patterns of consumer products. We found increasing reproducibility with increasing values of $\log K_{oa}$ among two non-phthalate plasticizers, two insecticides and three UV filters. Based on the U.S. EPA's CompTox Chemicals Dashboard [53], the compounds in these three use categories tend to have a primary use, compared to other chemical classes or use categories that did not show particular trends and tend to have multiple uses (e.g., phthalates). In other words, if emission sources or product use patterns were similar or relatively simple, reproducibility of dust concentrations is increasing with increasing values of $\log K_{oa}$. On the other hand, for compounds with multiple uses or emission sources, we did not observe the effect of $\log K_{oa}$ on the reproducibility of dust concentrations.

We observed that dust concentrations of some SVOCs were associated with household characteristics (Table S3). For example, lower income in our population, which were correlated with apartments or townhouses, were associated with higher PAH concentrations in dust. Potential reasons for higher PAH concentrations in homes with low income include close proximity to heavy traffic or industrial sources, or tobacco smoke [54]. Higher concentrations of three fragrance ingredients, which are added to personal care and cleaning products, were observed in households with child(ren). Perfumes are used more frequently by the U.S. younger adults with children than elders (aged 60+) [55]. It is also likely that

households with child(ren) are associated with increased use of cleaning products and personal care products.

Our measured dust concentrations reflected regulations of some phthalates in consumer products. For example, when comparing pooled SVOC concentrations in the U.S. dust collected between 2000 and 2015 by Mitro et al. [9] and our dust concentrations collected in California between 2015 and 2018, we observed lower dust concentrations in our study for most phthalates (Table S4). Our lower concentrations may reflect the changes of phthalate use in consumer products due to the Consumer Product Safety Improvement Act that was enacted in the U.S. in 2008 to prohibit the use of several phthalates including benzyl butyl phthalate, DBP, and DEHP in children's items [56]. Moreover, as DiBP is used as a substitute for DBP [57], we observed higher DiBP dust concentrations than Mitro et al. We also observed increased detection frequencies of DEHA over three visits from 38% to 59% in our study, potentially due to increased use of consumer products that replace DEHP with DEHA.

5. Conclusions

We observed relatively low temporal variability of dust concentrations for most SVOCs with higher temporal variability for some specific SVOCs, implying that dust concentrations may vary over time for SVOCs with seasonal variations in product use, or for those with significant outdoor sources that change by ventilation rates. For compounds for which household dust concentrations are used as surrogates for human exposure in indoor environments, understanding temporal variability of dust concentrations is an important step for developing optimal sampling strategies. For example, for compounds with low temporal variability in dust concentrations over time, dust concentrations collected from a single visit may represent average exposure. In addition, for compounds that are slowly released from building materials, furnishings or other indoor solid items where dust or the indoor environment are generally important in exposure (e.g., PBDEs, OPFRs, PFAS, and PCBs), one measure of dust levels may be sufficient. On the other hand, for compounds with high temporal variability in dust concentrations, future studies may need to collect multiple dust samples to account for temporal variability to minimize bias in estimating average exposure from dust concentrations. Further studies are needed to confirm our findings by comparing exposures reconstructed from dust concentrations and those inferred from biomarkers.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

Research reported in this publication was supported by the U.S. Environmental Protection Agency (EPA-G2013-STAR-K1) and the UC Davis Superfund Research Center, National Institutes of Health, NIEHS award (P42-ES004699).

References

1. Klepeis NE, et al., The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. *Journal of Exposure Analysis and Environmental Epidemiology*, 2001. 11(3): p. 231–252. [PubMed: 11477521]
2. Bennett DH, et al., Intake fraction for multimedia pollutants: A tool for life cycle analysis and comparative risk assessment. *Risk Analysis*, 2002. 22(5): p. 905–918. [PubMed: 12442988]
3. Nazaroff WW, Inhalation intake fraction of pollutants from episodic indoor emissions. *Building and Environment*, 2008. 43(3): p. 269–277.
4. Shin HM, McKone TE, and Bennett DH, Intake Fraction for the Indoor Environment: A Tool for Prioritizing Indoor Chemical Sources. *Environmental Science & Technology*, 2012. 46(18): p. 10063–10072. [PubMed: 22920860]
5. Weschler CJ and Nazaroff WW, Semivolatile organic compounds in indoor environments. *Atmospheric Environment*, 2008. 42(40): p. 9018–9040.
6. Bennett DH and Furtaw EJ, Fugacity-based indoor residential pesticide fate model. *Environmental Science & Technology*, 2004. 38(7): p. 2142–2152. [PubMed: 15112818]
7. Weschler CJ and Nazaroff WW, SVOC partitioning between the gas phase and settled dust indoors. *Atmospheric Environment*, 2010. 44(30): p. 3609–3620.
8. Little JC, et al., Rapid Methods to Estimate Potential Exposure to Semivolatile Organic Compounds in the Indoor Environment. *Environmental Science & Technology*, 2012. 46(20): p. 11171–11178. [PubMed: 22856628]
9. Mitro SD, et al., Consumer Product Chemicals in Indoor Dust: A Quantitative Meta-Analysis of U.S. Studies (vol 50, pg 10661, 2016). *Environmental Science & Technology*, 2016. 50(24): p. 13611–13611. [PubMed: 27993061]
10. Pelletier M, et al., Indoor residential exposure to semivolatile organic compounds in France. *Environment International*, 2017. 109: p. 81–88. [PubMed: 28950160]
11. Wilford BH, et al., Polybrominated diphenyl ethers in indoor dust in Ottawa, Canada: Implications for sources and exposure. *Environmental Science & Technology*, 2005. 39(18): p. 7027–7035. [PubMed: 16201626]
12. Zhou L, Hiltcher M, and Puttmann W, Occurrence and human exposure assessment of organophosphate flame retardants in indoor dust from various microenvironments of the Rhine/Main region, Germany. *Indoor Air*, 2017. 27(6): p. 1113–1127. [PubMed: 28556503]
13. Lorber M and Egeghy PP, Simple Intake and Pharmacokinetic Modeling to Characterize Exposure of Americans to Perfluorooctanoic Acid, PFOA. *Environmental Science & Technology*, 2011. 45(19): p. 8006–8014. [PubMed: 21517063]
14. Egeghy PP and Lorber M, An assessment of the exposure of Americans to perfluorooctane sulfonate: A comparison of estimated intake with values inferred from NHANES data. *Journal of Exposure Science and Environmental Epidemiology*, 2011. 21(2): p. 150–168. [PubMed: 20145679]
15. Gevao B, et al., Polycyclic aromatic hydrocarbons in indoor air and dust in Kuwait: Implications for sources and nondietary human exposure. *Archives of Environmental Contamination and Toxicology*, 2007. 53(4): p. 503–512. [PubMed: 17571201]
16. Li L, Arnot JA, and Wania F, Revisiting the Contributions of Far- and Near-Field Routes to Aggregate Human Exposure to Polychlorinated Biphenyls (PCBs). *Environmental Science & Technology*, 2018. 52(12): p. 6974–6984. [PubMed: 29771504]
17. Johnson PI, et al., Associations between brominated flame retardants in house dust and hormone levels in men. *Science of the Total Environment*, 2013. 445: p. 177–184.
18. Bennett DH, et al., Polybrominated diphenyl ether (PBDE) concentrations and resulting exposure in homes in California: relationships among passive air, surface wipe and dust concentrations, and temporal variability. *Indoor Air*, 2015. 25(2): p. 220–229. [PubMed: 24832910]
19. Dodson RE, et al., Urinary Biomonitoring of Phosphate Flame Retardants: Levels in California Adults and Recommendations for Future Studies. *Environmental Science & Technology*, 2014. 48(23): p. 13625–13633. [PubMed: 25388620]

20. Fromme H, et al., Organophosphate flame retardants and plasticizers in the air and dust in German daycare centers and human biomonitoring in visiting children (LUPE 3). *Environment International*, 2014. 71: p. 158–163. [PubMed: 25033099]
21. Meeker JD, et al., Urinary Metabolites of Organophosphate Flame Retardants: Temporal Variability and Correlations with House Dust Concentrations. *Environmental Health Perspectives*, 2013. 121(5): p. 580–585. [PubMed: 23461877]
22. Rudel RA, Seryak LM, and Brody JG, PCB-containing wood floor finish is a likely source of elevated PCBs in residents' blood, household air and dust: a case study of exposure. *Environmental Health*, 2008. 7.
23. Liu M, et al., The occurrence of bisphenol plasticizers in paired dust and urine samples and its association with oxidative stress. *Chemosphere*, 2019. 216: p. 472–478. [PubMed: 30388685]
24. Trunnelle KJ, et al., Urinary pyrethroid and chlorpyrifos metabolite concentrations in Northern California families and their relationship to indoor residential insecticide levels, part of the Study of Use of Products and Exposure Related Behavior (SUPERB). *Environ Sci Technol*, 2014. 48(3): p. 1931–9. [PubMed: 24422434]
25. Araki A, et al., Phosphorus flame retardants in indoor dust and their relation to asthma and allergies of. *Indoor Air*, 2014. 24(1): p. 3–15. [PubMed: 23724807]
26. Bamai YA, et al., Exposure to phthalates in house dust and associated allergies in children aged 6–12 years. *Environment International*, 2016. 96: p. 16–23. [PubMed: 27588698]
27. Colt JS, Residential Insecticide Use and Risk of Non-Hodgkin's Lymphoma. *Cancer Epidemiology Biomarkers & Prevention*, 2006. 15(2): p. 251–257.
28. Colt JS, et al., Organochlorine exposure, immune gene variation, and risk of non-Hodgkin lymphoma. *Blood*, 2009. 113(9): p. 1899–905. [PubMed: 19066394]
29. Colt JS, et al., Organochlorines in carpet dust and non-Hodgkin lymphoma. *Epidemiology*, 2005. 16(4): p. 516–25. [PubMed: 15951670]
30. Shin HM, et al., Indoor Residence Times of Semivolatile Organic Compounds: Model Estimation and Field Evaluation. *Environmental Science & Technology*, 2013. 47(2): p. 859–867. [PubMed: 23244175]
31. Egeghy PP, et al., Determinants of temporal variability in NHEXAS-Maryland environmental concentrations, exposures, and biomarkers. *Journal of Exposure Analysis and Environmental Epidemiology*, 2005. 15(5): p. 388–397. [PubMed: 15602583]
32. Cao ZG, et al., Differences in the seasonal variation of brominated and phosphorus flame retardants in office dust. *Environment International*, 2014. 65: p. 100–106. [PubMed: 24480750]
33. Whitehead TP, et al., Carpet-dust chemicals as measures of exposure: Implications of variability. *Emerging Themes in Epidemiology*, 2012. 9.
34. Kristensen K, et al., Sources and dynamics of semivolatile organic compounds in a single-family residence in northern California. *Indoor Air*, 2019. 29(4): p. 645–655. [PubMed: 31004533]
35. Wei W, Ramalho O, and Mandin C, A long-term dynamic model for predicting the concentration of semivolatile organic compounds in indoor environments: Application to phthalates. *Building and Environment*, 2019. 148: p. 11–19.
36. Cao ZG, et al., Spatial, seasonal and particle size dependent variations of PAH contamination in indoor dust and the corresponding human health risk. *Science of the Total Environment*, 2019. 653: p. 423–430.
37. Allen JG, et al., Critical factors in assessing exposure to PBDEs via house dust. *Environment International*, 2008. 34(8): p. 1085–1091. [PubMed: 18456330]
38. Rudel RA, et al., Phthalates, alkylphenols, pesticides, polybrominated diphenyl ethers, and other endocrine-disrupting compounds in indoor air and dust. *Environmental Science & Technology*, 2003. 37(20): p. 4543–4553. [PubMed: 14594359]
39. Shin HM, et al., Measured concentrations of consumer product chemicals in California house dust: Implications for sources, exposure, and toxicity potential. *Indoor Air*, 2019.
40. Moschet C, et al., Household Dust as a Repository of Chemical Accumulation: New Insights from a Comprehensive High-Resolution Mass Spectrometric Study. *Environmental Science & Technology*, 2018. 52(5): p. 2878–2887. [PubMed: 29437387]

41. Goldsmith MR, et al., Development of a consumer product ingredient database for chemical exposure screening and prioritization. *Food and Chemical Toxicology*, 2014. 65: p. 269–279. [PubMed: 24374094]
42. Hornung RWe.a., Estimation of average concentrations in the presence of nondetectable values. *Applied Occupational and Environmental Hygiene*, 1990. 5(1): p. 46–51.
43. Benjamini Y, et al., Controlling the false discovery rate in behavior genetics research. *Behav Brain Res*, 2001. 125(1–2): p. 279–84. [PubMed: 11682119]
44. Adibi JJ, et al., Characterization of phthalate exposure among pregnant women assessed by repeat air and urine samples. *Environmental Health Perspectives*, 2008. 116(4): p. 467–473. [PubMed: 18414628]
45. Rosner B, ed. *Fundamentals of Biostatistics*, 5th Ed. 2000, Duxbury, Pacific Grove, CA.
46. Shrout PE and Fleiss JL, Intraclass correlations: uses in assessing rater reliability. *Psychol Bull*, 1979. 86(2): p. 420–8. [PubMed: 18839484]
47. Diedenhofen B and Musch J, cocor: a comprehensive solution for the statistical comparison of correlations. *PLoS One*, 2015. 10(3): p. e0121945. [PubMed: 25835001]
48. Metcalf RL, in *Insect Control*. 2014.
49. Engebretsen KA, et al., Changes in filaggrin degradation products and corneocyte surface texture by season. *British Journal of Dermatology*, 2018. 178(5): p. 1143–1150.
50. Finardi S, et al., Seasonal variation of PAHs concentration and source attribution through diagnostic ratios analysis. *Urban Climate*, 2017. 22: p. 19–34.
51. Bi CY, et al., Phthalates and organophosphates in settled dust and HVAC filter dust of US low-income homes: Association with season, building characteristics, and childhood asthma. *Environment International*, 2018. 121: p. 916–930. [PubMed: 30347374]
52. Shoeib M and Harner T, Using measured octanol-air partition coefficients to explain environmental partitioning of organochlorine pesticides. *Environmental Toxicology and Chemistry*, 2002. 21(5): p. 984–990. [PubMed: 12013145]
53. Phillips KA, et al., High-throughput screening of chemicals as functional substitutes using structure-based classification models. *Green Chemistry*, 2017. 19(4): p. 1063–1074. [PubMed: 30505234]
54. Chuang JC, et al., Polycyclic aromatic hydrocarbon exposures of children in low-income families. *Journal of Exposure Analysis and Environmental Epidemiology*, 1999. 9(2): p. 85–98. [PubMed: 10321348]
55. Statista, Frequency of use of fragrances (perfume, eau de toilette) among consumers in the United States as of May 2017, by age group. 2017.
56. Zota AR, Calafat AM, and Woodruff TJ, Temporal Trends in Phthalate Exposures: Findings from the National Health and Nutrition Examination Survey, 2001–2010. *Environmental Health Perspectives*, 2014. 122(3): p. 235–241. [PubMed: 24425099]
57. Wittassek M, et al., Internal phthalate exposure over the last two decades - A retrospective human biomonitoring study. *International Journal of Hygiene and Environmental Health*, 2007. 210(3–4): p. 319–333. [PubMed: 17400024]

Practical Implications:

For studies where household dust concentrations are used as surrogates for human exposure in indoor environments, understanding temporal variability of dust concentrations is a key for developing optimal sampling strategies. We found that temporal variability of dust concentrations was relatively low for most SVOCs. Thus, dust concentrations collected from a single visit may represent average exposure for those with low temporal variability. We expect that our findings may support future work that includes dust collection.

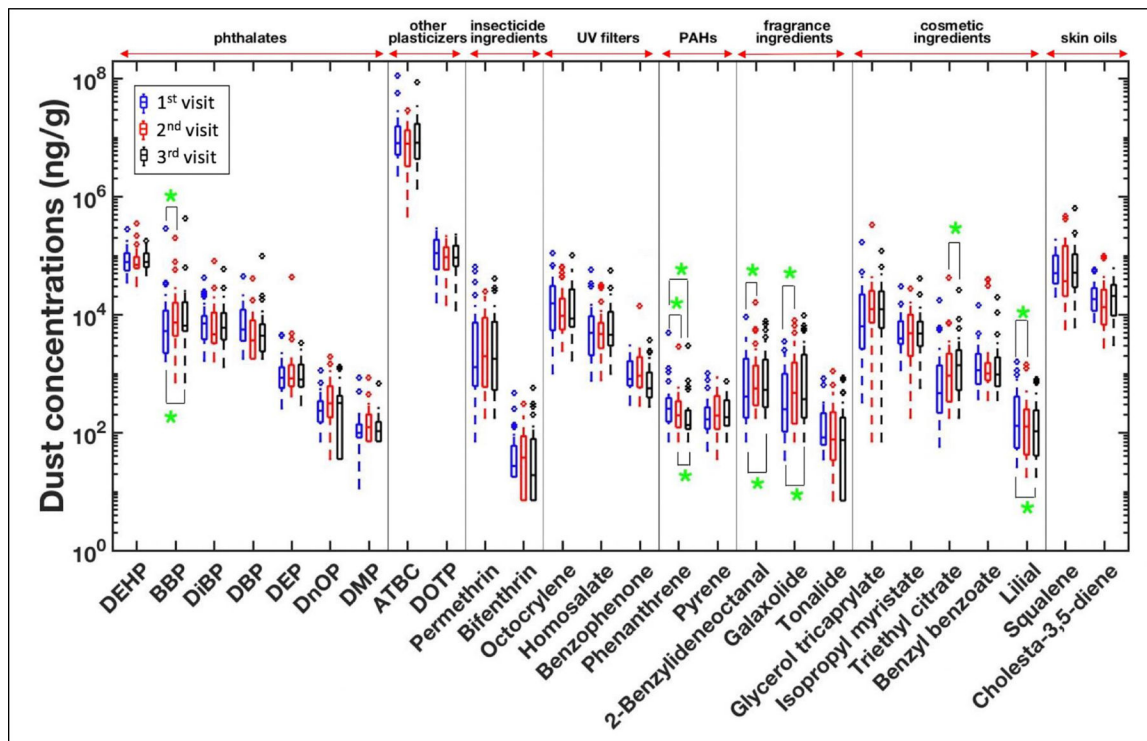


Figure 1.

Distributions of dust concentrations (ng/g of dust) for 26 organic compounds detected in more than 50% of the samples at all three visits. P-values < 0.05 are marked (*).

Abbreviation: acetyl tributyl citrate (**ATBC**), benzyl butyl phthalate (**BBP**), diethyl phthalate (**DEP**), bis(2-ethylhexyl) phthalate (**DEHP**), di-isobutyl phthalate (**DiBP**), dimethyl phthalate (**DMP**), di-n-butyl phthalate (**DBP**), di-n-octyl phthalate (**DnOP**), dioctyl terephthalate (**DOTP**)

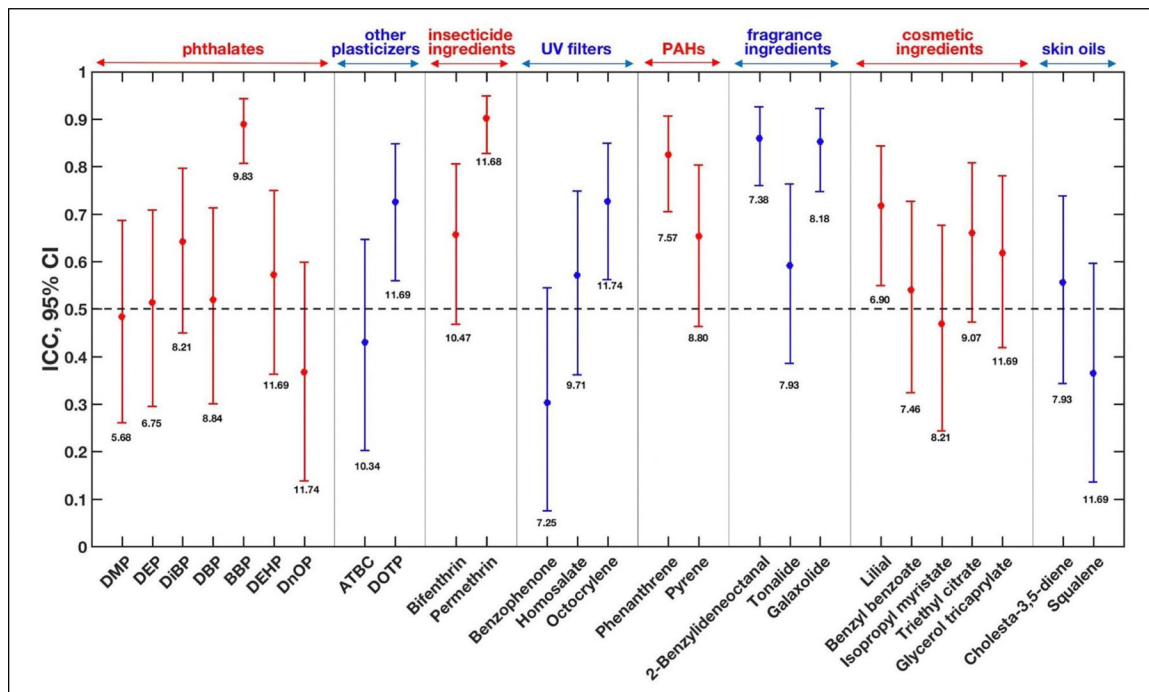


Figure 2. Intraclass correlation coefficients (ICCs) and 95% CIs of ln-transformed dust concentrations of 26 compounds across three sampling time points in the increasing order of the octanol-air partition coefficient (K_{oa}) within each class. The log K_{oa} value of each compound is placed below the lower bound of 95% CI.

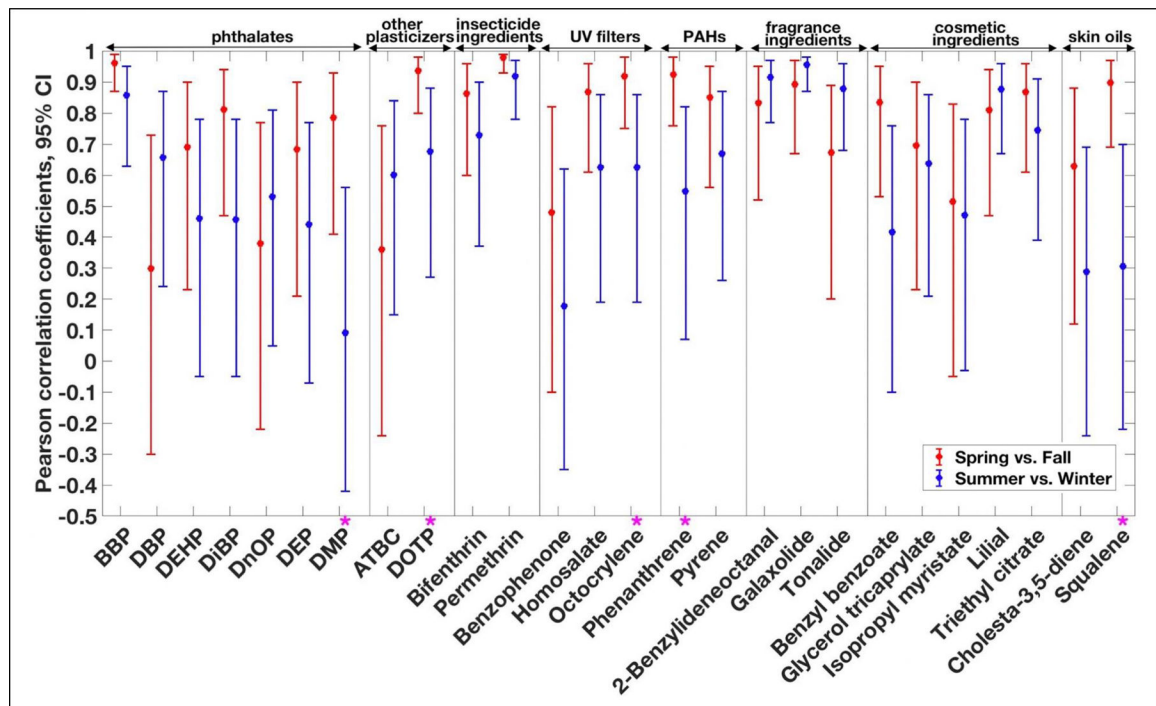


Figure 3.

Pearson correlation coefficients of ln-transformed dust concentrations and 95% CIs between spring and fall ($n = 13$ homes) and between summer and winter ($n = 16$ homes). Compounds whose coefficients between spring-fall and summer-winter were different (p -value < 0.05) are marked (*).