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Letter

Young Children's Exposure to Chemicals of Concern in Their Sleeping Environment: An In-Home Study

Sara Vaezafshar, Sylvia Wolk, Victoria H. Arrandale, Roxana Sühring, Erica Phipps, Liisa M. Jantunen, and Miriam L. Diamond*

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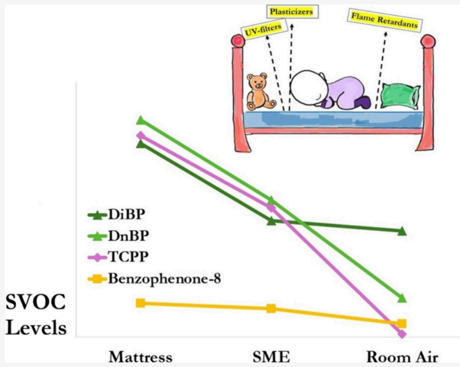
ABSTRACT: Sleeping microenvironments (SMEs) can expose young children to chemicals of concern. Using passive samplers, we measured the concentrations of ortho-phthalates (PAEs), organophosphate esters (OPEs), and UV-filters (benzophenones, salicylates, and phenolic benzotriazoles) in the bedroom air, SME, and released from mattresses in 25 bedrooms of children aged 6 months to 4 years in Toronto and Ottawa, Canada. We detected 28, 31, and 30 compounds in bedroom air, SME air, and mattresses, respectively. SME exceeded bedroom air concentrations, indicating elevated exposure while sleeping and sources from SME contents, with two exceptions. Higher concentrations of two PAEs and five OPEs (including isomers) in mattress versus SME samplers indicated that mattresses were a source. Bedding items were likely sources of tris(2-butoxyethyl) phosphate (TBOEP) where SME concentrations were significantly higher than those in mattress samplers. Older mattresses had higher concentrations of di-2-ethylhexyl phthalate (DEHP) and benzyl butyl phthalate (BzBP). These results indicate children's exposure to a range of chemicals of concern while sleeping, at higher concentrations than in their bedrooms. Practical steps to reduce exposure include limiting items in SMEs such as toys and frequently washing bedding. Also, these results should prompt stricter regulations and greater producer responsibility regarding harmful chemicals used in mattresses and SME articles.



KEYWORDS: *Children's mattresses, sleeping microenvironment, children's bedroom air, ortho-phthalates, organophosphate esters, UV-filters, early life exposure, chemical regulation of children's products*

INTRODUCTION

Children experience elevated exposure to environmental contaminants, including semivolatile organic compounds (SVOCs), compared to adults due to, for example, their 10-times higher inhalation rate, 3-times larger skin surface area per body weight compared to adults, and their unique behaviors such as frequent hand-to-mouth contact and mouthing objects.[1−7] Concerns have arisen due to children's higher exposures, particularly regarding ortho-phthalate esters (referred to as "phthalates" or PAEs) and certain organophosphate esters (OPEs) due to associations with higher incidences of childhood asthma, negative effects on cognitive function, and other outcomes associated with endocrine disruption.[8−12]

Infants and young children, from newborns to 4 years old, sleep up to 18 h per day, much of which is in a sleeping microenvironment (SME).[3,13−15] SMEs are defined as the conditions within a person's sleeping area, encompassing bedding items, mattress, the air in an individual's breathing zone, and their buoyant thermal plume.[16,17] SMEs can expose individuals to a wide range of chemicals, including SVOCs, volatile organic compounds (VOCs), and biological contaminants like fungi, mites, and bacteria.[17−22]

Mattresses, the largest physical item in SMEs, can be a source of plasticizers, flame retardants, and stain repellents.[18,23−27] Children's mattresses typically consist of a thick foam layer and an easy-to-clean outer cover. Adult mattresses, which may be used by children over 2 years of age, also contain foam with a textile cover. In Canada, flammability regulations for adult mattresses specify a cigarette (smolder) test whereas infants' cribs, cradles, and bedding items must pass a standardized small open flame test with a flame spread time of no more than seven seconds.[28,29] Additive flame retardants are not necessary to pass either the smolder or the small open flame test which can be met using a low flammability barrier.[30−33] However, the foam of children and adult mattresses may contain 1−30% brominated and/or OPE flame retardants.[34−37] PAEs, such as di(2-ethylhexyl) phthalate (DEHP), which can constitute up to 40% by weight of a

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plastic, can be added to the cover of children's mattresses to increase flexibility.[38] Other SVOCs such as UV-filters (i.e., benzophenones, salicylates and phenolic benzotriazoles or benzotriazoles) are used in dyes, synthetic resins, and fibers to protect textiles from UV degradation.[39−42] UV-328 was listed as a Persistent Organic Pollutant in May 2023 by the Stockholm Convention for elimination (with specific exemptions).[43] SVOC additives are not typically chemically bonded to the host polymer leading to their release into the surrounding air, followed by partitioning to dust, children's skin, clothing, bedding items, and toys, and providing opportunities for exposure.[8,44−49]

Health concerns have prompted Canada, the US and the European Union to limit their use and set maximum concentrations of certain PAEs and OPEs (see Supporting Information SI1).[50−57] Previous research on SVOCs in 16 new Canadian children's mattresses found some exceedances of those regulations.[27] Testing of those mattresses also showed higher concentrations of mattress-derived SVOCs in the SME compared with room air, as found by others.[17,58]

Here we assessed in-home levels of SVOCs in SMEs and bedrooms of 25 children aged 6 months to 4 years in participating households, focusing on PAEs, OPEs, and UV-filters including benzophenones, salicylates, and benzotriazoles. We hypothesized that mattresses are significant sources of SVOC emissions in children's SMEs, leading to early life exposures from the SME.

■ METHODS AND MATERIALS

In 2022−2023, we recruited 21 parent participants with 25 children participants aged 6 months to 4 years (4 parents had >1 child), in the Greater Toronto Area and Ottawa, Canada, from mid to high socio-economic status (SES) households. Sampling kits were prepared and delivered in-person or by mail as home access was restricted due to the SARS-CoV-2 pandemic. The kits contained the samplers, necessary deployment materials, access to an instructional video with a written guide, and a questionnaire (see SI2). Ethical approval (protocol #41217) was obtained from the Research Ethics Board of University of Toronto.

Sampling Approach. We used polydimethylsiloxane (PDMS), also known as silicone rubber, passive samplers (Specialty Silicone Products, Inc., Ballston Spa, NY) with density of 1.1−1.2 g/cm^3 and thickness of 0.1 cm. PDMS samplers were 9 × 5.5 cm^2 sheets with a total surface area of 102 cm^2. Samplers were precleaned, stored in 250 mL precleaned glass jars, then deployed for a 7-day period.[59,60]

Participants were instructed to deploy three PDMS samplers in the child's bedroom: 1) Room Air sampler was hung on a metal stand to monitor the ambient air. 2) SME sampler was placed on the mattress under the bedsheet nearby where the child slept, exposed to the mattress and SME air. 3) Mattress sampler was placed on the mattress nearby where the child slept, under the bedsheet with the top surface covered with aluminum foil to isolate uptake from the mattress (Figure S1). Participants returned these three "field" samplers in their original jars along with sampling start and retrieval dates and the travel blank that remained in its jar. One laboratory blank per participant (n = 25) consisted of a precleaned PDMS sampler not exposed to the environment. See SI2.1 for detailed deployment procedures. A written questionnaire was provided to participants to collect detailed information (see SI2.2).

Analytical Methods. We analyzed 51 SVOCs: PAEs (n = 8), OPEs (n = 29 including isomers), benzophenones (n = 3), salicylates (n = 5), and benzotriazoles (n = 6) (Table S1). All deployed field samplers (n = 75) and laboratory (n = 25) and field blanks (n = 21) were transferred to 40 mL amber vials and spiked with 20 μL of 7 labeled surrogate compounds to monitor recoveries (Table S2). Samplers were extracted for 30 min in acetonitrile using a wrist-action shaker (Burrell Scientific, Model 75) followed by soaking in acetonitrile overnight, and concentrating to 0.5 mL.[61] An internal standard (mirex) was added to the final volume of each extract for time reference and volume adjustment. Analysis was performed by gas chromatography with a mass spectrometer (Agilent GC 7890-MS 5977) in electron ionization mode (see SI3−5, Table S3).

Quality Assurance/Quality Control. Prior to extractions of field samplers, native and isotopically labeled compound recoveries were assessed by spiking known concentrations on four replicates of precleaned PDMS samplers (Table S4). Recoveries of isotopically labeled compounds in all field blanks and samples were 77−115% which was deemed acceptable (Table S5).[62] Data were recovery and blank corrected according to details provided in SI6.[63] The limit of detection (LOD) was calculated as the average of target SVOC concentrations in 21 field blanks plus 3 standard deviations (SI7, Table S6). Values < LOD were substituted with 1/2 LOD for calculating descriptive statistics of analytes with detection frequencies (DFs) > 50%.

Data Analysis. Nonparametric statistical tests were conducted since raw and transformed data were not normally distributed (chi-square test). Descriptive summary statistics were calculated and visualized using Microsoft Excel 365 (Version 2307) and RStudio (Version 2022.02.0). The similarity in SVOC patterns in Room Air and SME samples was determined using the cosine similarity method for analytes with DFs > 50%.[64] Mann−Whitney U-tests were used to compare concentration differences between the Room Air and SME samplers as well as between the SME and Mattress samplers. Spearman's rank tests evaluated correlations between qualitative data from questionnaires (which were assigned numerical values corresponding to categorical responses) and SME concentrations (SI8, Table S7). Concentrations were compared according to analyte mass per sampler surface area, specifically 102 cm^2 for SME and Room air samplers (both sides and edges of the sampler), and 52.4 cm^2 for Mattress samplers (bottom and edges).

■ RESULTS AND DISCUSSION

SVOCs Detected in Samplers. Room Air samplers detected 28 out of 51 SVOCs with DFs ranging from 4% for two UV-filters to 96% for benzophenone (Table S8). The highest median concentrations measured within each SVOC class, with DFs > 50%, were for diethyl phthalate (DEP), tris(chloropropyl)phosphate, sum of 3 isomers (∑TCPP), benzophenone (BP), and methyl salicylate (M-SAL). Benzotriazoles had DFs < 50%. For SME samplers, we measured 31 SVOCs with DFs of 4% for UV-327 to 88% for DEP, tris(2-butoxyethyl)phosphate (TBOEP), and BP. Here, the highest median concentrations per class were for DEP, TBOEP, BP and phenyl salicylate (P-SAL). Finally, for Mattress samplers, 30 SVOCs were detected with DFs ranging from 8% for UV-327 and UV-350 to 88% for TBOEP. The highest median

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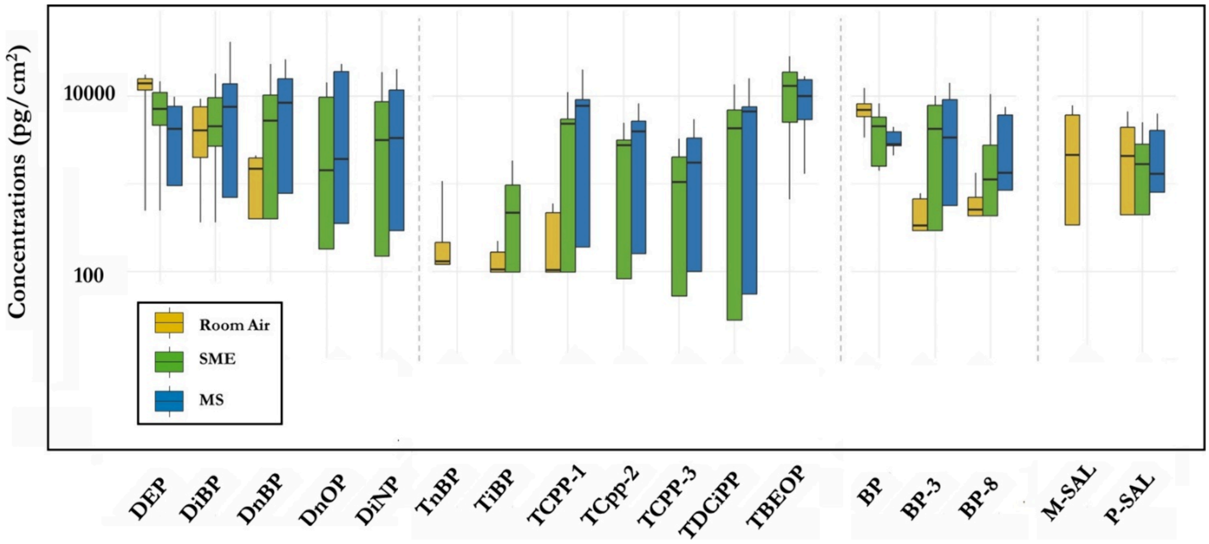

Figure 1. Concentrations of SVOCs with DF > 50% in Room Air (yellow), Sleeping Microenvironment (SME; green) and Mattress (MS; blue) (pg/cm^2). The horizontal lines in the boxplots indicate median (50th percentile) concentrations. The boxes indicate the 25th and 75th percentiles, and the whiskers indicate the 5th and 95th percentiles. Some whiskers are not shown where the 5th and 25th percentiles were the same due to low sample sizes. Box and whiskers are not shown for those SVOCs with DF < 50% for the specific sampler type.

concentrations for each class were for di-*n*-butyl phthalate (DnBP), ∑TCPP, benzophenone-3 (BP-3) and P-SAL.

Although concentrations of UV-328 were low in all three samplers, the DFs were the highest compared to other benzotriazoles. We also measured some SVOCs of concern, with low DFs: DEHP, triphenyl phosphate (TPhP), UV-234, and UV-350. Other OPEs were also detected with low DFs in Mattress, SME and Room Air samplers, notably tris(2-chloroethyl) phosphate (TCEP) and tris(1,3-dichloro-2-propyl)phosphate (TDCiPP).

Air Concentrations. To put the bedroom air concentrations into context, we compared concentrations measured here with those of studies conducted using passive sampling methods within the past 6 years. Air concentrations were calculated from the Room Air samplers using the generic passive sampling rate for PDMS of 1.5 ± 1.1 $m^3.day^{-1}.dm^{-2}$ (Table S9).[60]

We observed comparable median concentrations of DEP but lower concentrations of diisobutyl phthalate (DiBP), DnBP, and ∑TCPP in children's bedrooms compared to adult bedrooms in a study using the same methods, with typically lower DFs which could be an artifact of the 7- (this study) vs 60-day deployment period.[60] All PAEs were lower in children's bedrooms compared to those in the most-used rooms in low-income social housing in Toronto. We note that the air concentrations of an OPE and six PAEs were over 10-fold higher in social housing than in higher income housing.[59]

The maximum air concentrations of BP were ∼40 times lower than those reported by Dodson et al.,[65] who measured indoor air concentrations using active and passive sampling methods. However, BP air concentrations were within an order of magnitude of maximum levels measured in indoor air of low-income housing collected before and after renovations using active air sampling.[66] Additionally, we observed lower levels of BP-3 compared with literature reports.[65−67] We did not find studies reporting the air concentrations of other UV-filters.

Finally, we compared the SVOCs measured in children's bedroom air samplers with those from the experimental testing of new mattresses in a university office.[27] We measured higher concentrations in children's bedroom air of DiBP (∼3 times), BP (∼10 times), and P-SAL (∼3 times) compared to the office air which contained one mattress at-a-time for testing.

Exposure and Sources in Room Air and SME. We compared the potential exposure from SME versus Room Air by comparing the concentrations of 9 SVOCs detected in both sample types with DFs > 50% (Figure 1, Table S8). We were aware of the potential bias in this comparison since SME samplers could have a lower uptake rate versus Room Air samplers due to less air flow, which could be counterbalanced by greater uptake of the SME sampler placed directly on the mattress.[68]

SVOCs were higher in SME samplers indicating greater exposure from the SME than from room air, except for DEP and BP. Differences were significant for DEP ($p < 0.01$), DnBP ($p < 0.05$), TCPP-1 ($p < 0.01$), BP ($p < 0.01$), and BP-3 ($p < 0.01$, Mann−Whitney U test, Table S10). The significantly higher concentrations of DEP and BP in room air suggested sources other than SME items (see below). However, higher concentrations of DnBP, TCPP-1, and BP-3 in SME samplers suggested sources within the SMEs including mattresses.[27]

Cosine similarity values indicating the similar patterns of 9 SVOCs with DFs > 50% ranged from 15 participants with moderate, to 10 participants with high similarity (Table S11). The similar patterns between Room Air and SME are consistent with SVOCs release over time from primary source(s), followed by partitioning to other indoor media including surfaces, according to their physicochemical properties.[44−46,49] As such, the SVOCs in a room become well-mixed (equifugacity), making identification of the primary source(s) challenging without product testing.

To assess whether mattresses were likely sources of SVOCs to the SME and by extension, to room air, we compared SVOC masses in Mattress and SME samplers with DFs > 50% (Figure 1, Table S8). We assumed the same uptake rate for both samplers which is reasonable since both had restricted air flow. Thus, this is a comparison of fugacities in these two samplers which, as an equilibrium criterion, indicates the direction of

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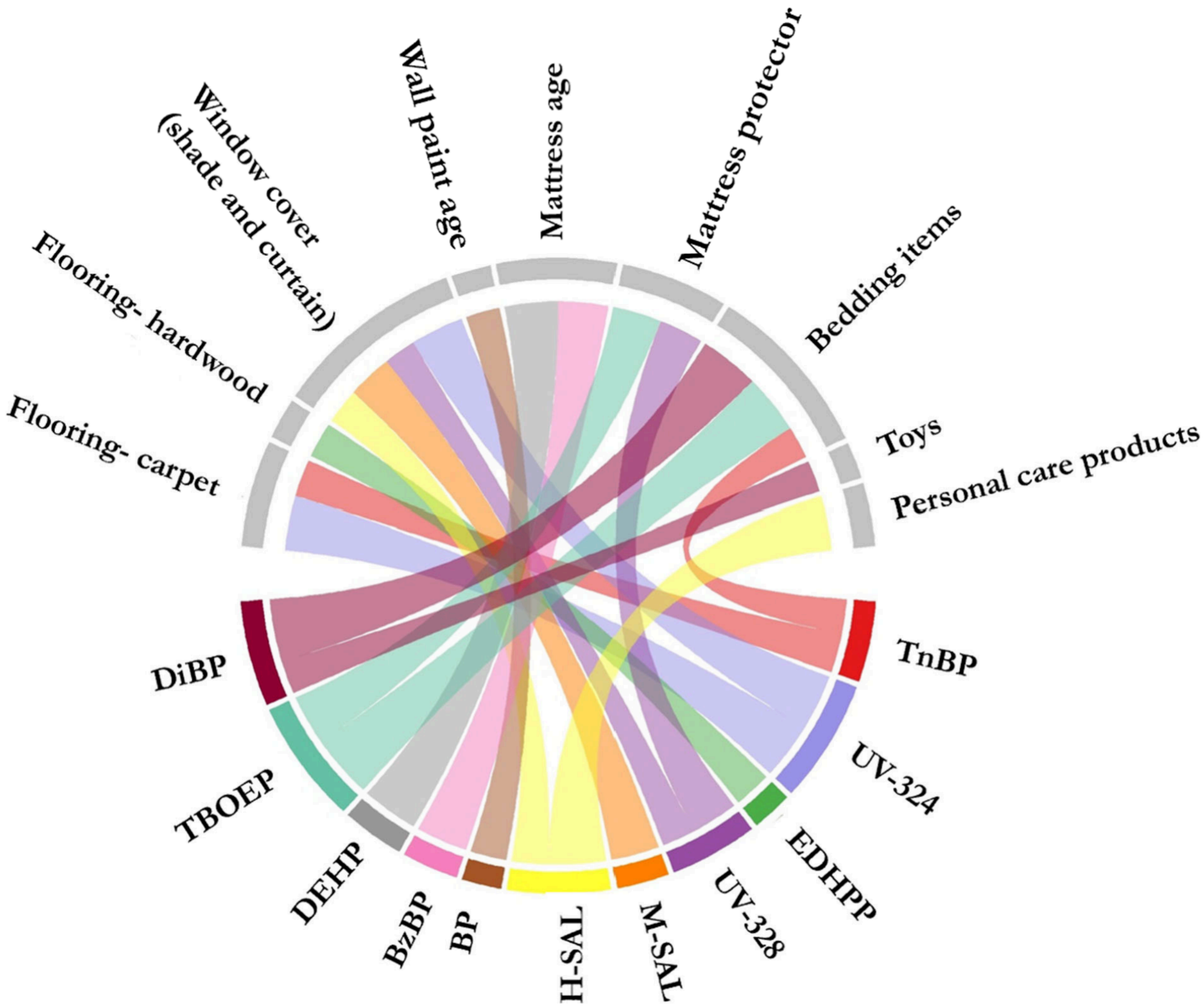

Figure 2. Cord diagram showing significant Spearman correlations (ρ > 0.5) between measured SVOCs in SME samplers and factors including room characteristics, SME contents, and variables related to children. The width of each ribbon is proportional to the Spearman's (ρ) correlation coefficient.

chemical movement from high to low fugacity, i.e., from mattress to SME or vice versa. Median Mattress sampler concentrations tended to exceed SME samplers for all PAEs (except for DEP), ∑TCPP, TDCiPP, and benzophenone-8 (BP-8) with only ∑TCPP reaching statistical significance ($p < 0.05$, Table S12). This result is consistent with the use of ∑TCPP as a flame retardant added to mattress foam.[23,27] Conversely, TBOEP was significantly higher ($p < 0.05$) in SME than the Mattress samplers. Although TBOEP is used in mattress covers,[27] stronger emissions likely emanate from other sources in the SME such as bedding and other textiles since it is used as a defoamer in the textile industry.[69,70] TBOEP concentrations are consistent with previously reported elevated concentrations in dust sampled from children's SMEs, which were associated with an increased incidence of asthma.[12]

These findings, as well as those of Vaezafshar et al.,[27] support the conclusion that mattresses are a source of some SVOCs within the SME, notably PAEs (except DEP) and OPEs (except TBOEP), contributing to elevated exposures of children in the SME.

Factors Affecting SVOC Concentrations in SME. We investigated factors in addition to mattresses that could influence SVOC concentrations, as shown in Figure 2 that displays the magnitude of Spearman's rank correlation coefficients ρ>0.5 between SME sampler concentrations and participant responses to questionnaires. For example, positive associations were found between concentrations of UV-328 (ρ = 0.75) and TBOEP (ρ = 0.98) with mattress protectors.[69−71] The presence of bedding items, including pillows, sheets, blankets, and mattress protectors, were strongly positively correlated with DiBP (ρ = 0.95) and TBOEP (ρ = 0.95) which could be due to its use in mattress protectors used by 60% of participants. DiBP, other PAE and nonortho-phthalate plasticizers could be added to items containing polyvinyl chloride, such as pillow protectors.[72]

DEHP (ρ = 0.57) and BzBP (ρ = 0.62) were correlated with mattress age, with higher concentrations in older mattresses, which is consistent with decreased usage because of regulatory restrictions.[4,73−76] The natural aging of foam also can lead to higher emissions of additives.[77] Mattress foam is typically made of a flexible polyether-based urethane polymer which degrades over time due to its porous structure, allowing for oxidation which causes the depolymerization process of the polyol chain.[77−80] UV-filters are presumably added to slow this aging process. Benzophenones are also widely used as a UV-stabilizer and photoinitiator in building materials such as paints, coatings, and stains.[81,82] We found a significant, negative correlation between BP (ρ = 0.60) and the age of

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wall paints. Additionally, benzophenones are used in the textile industry (e.g., room draperies) to prevent discoloration caused by UV radiation.[83]

We found a significant positive correlation between the application of skincare products and H-SAL levels (ρ = 0.87, Figure 2) which is widely used in fragrances and skin and other personal care products.[84] Although DEP is commonly found in products such as perfumes, fragrances and cosmetics,[85] we did not find a significant correlation between concentrations and reported use of personal care products.

Among room-related factors, in addition to the negative correlation between BP and wall paint age, we found a positive correlation between UV-234 and carpeted flooring (ρ = 0.75), explained by its use as a UV-stabilizer in carpet backing.[41] We also identified correlations between textile-based window coverings (shades and curtains) and several UV-filters that are used as UV-stabilizers in synthetic fabrics.[71,86] Finally, we found a positive correlation between ethylhexyldiphenyl phosphate (EHDPP: ρ = 0.60) and hardwood flooring, consistent with its use as an adhesive additive.[87,88]

Implications. Our study confirmed that children are exposed to SVOCs while in their SME, where most of the SVOCs targeted in this study originated from the SME contents. The results, combined with the previous detection of certain SVOCs in children's new mattresses, underscore the role of children's mattresses as a significant source of SVOCs in the SME.[18,25,27] These results are particularly concerning given the substantial amount of time children spend sleeping each day, their tendency for higher exposures and increased susceptibility to harm due to their developmental stage.[3]

To mitigate children's exposures to SVOCs in the SME, parents can adopt practices such as frequently washing bedding items and children's clothing which can act as barriers to SVOC exposure because of the high sorptive capacity of textiles, although as seen here, some textiles may also be a source of contamination.[47,89,90] We also recommend minimizing the number of items in the bed, as we found correlations between the presence of bedding items, toys, and mattress protectors, with concentrations of certain PAEs and OPEs. Additionally, mattress protectors, often used as waterproof barriers, may contain additional chemicals of concern, such as per- and polyfluoroalkyl substances (PFAS).[91]

Some of the SVOCs measured here are of concern because of their association with adverse health effects (e.g., DnBP, BzBP, DEHP, TCEP, TDCiPP), which has prompted regulatory efforts to reduce their use in new children's products. Our results reinforce the need for manufacturers and retailers of children's products to comply with regulatory restrictions, e.g., banned use of TCEP which was detected in 28 and 40% of Room Air and SME samplers, respectively. However, more effective restrictions are also needed such as updating Canadian phthalate regulations to restrict the use of DiBP, diisononyl phthalate (DiNP), and di-*n*-octyl phthalate (DnOP) in mattresses and not just toys and passing regulatory restrictions on TCPP and TDCiPP use in foam products which were proposed in 2016. Such regulations need timely implementation because of the long lag time between passage and evidence of decreasing product usage by families.

In closing, reducing children's exposure to harmful chemicals should include a focus on their sleeping environment given the number of hours spent there, achieved by the timely implementation of coherent and evidence-based regulations and vigilance on the part of manufacturers and retailers.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.5c00051.

Description of experimental design, participant questionnaire, list of analytes (with CAS numbers, molecular weight, log Koa and vapor pressure), list of surrogate and internal standards, details of instrumental parameters, list of quantifier and qualifier ions for GC-MS analysis, details of QA/QC measures taken, results (native and surrogate standard recoveries) from PDMS spike testing and field blanks and standards, method detection limits, conversion codes for categorical data collected in questionnaire, masses of analytes in PDMS samplers (descriptive statistics), ranges of estimated bedroom air concentrations obtained from bedroom passive sampler, results of statistical comparison between SME and room air sampler masses, results from cosine similarity test for analytes with DFs > 50% in SME and room air within the same home, results of statistical comparison between SME and mattress sampler, and references (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Miriam L. Diamond − *Department of Earth Sciences, University of Toronto, Toronto, Ontario M5S 3B1, Canada; School of Environment, University of Toronto, Toronto, Ontario M5S 3E8, Canada; Dalla Lana School of Public Health, University of Toronto, Toronto, Ontario M5T 3M7, Canada;* orcid.org/0000-0001-6296-6431; Email: miriam.diamond@utoronto.ca

Authors

Sara Vaezafshar − *Department of Earth Sciences, University of Toronto, Toronto, Ontario M5S 3B1, Canada;* orcid.org/0009-0004-9834-0704

Sylvia Wolk − *Department of Earth Sciences, University of Toronto, Toronto, Ontario M5S 3B1, Canada*

Victoria H. Arrandale − *Dalla Lana School of Public Health, University of Toronto, Toronto, Ontario M5T 3M7, Canada;* orcid.org/0000-0002-9587-4207

Roxana Sühring − *Department of Chemistry and Biology, Toronto Metropolitan University, Toronto, Ontario M5B 2K3, Canada*

Erica Phipps − *Canadian Partnership for Children's Health and Environment, c/o Canadian Environmental Law Association, Toronto, Ontario M5J 2H7, Canada*

Liisa M. Jantunen − *Air Quality Processes Research, Environment and Climate Change Canada, Egbert L0L 1N0, Canada; Department of Earth Sciences, University of Toronto, Toronto, Ontario M5S 3B1, Canada*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.5c00051

Author Contributions

Credit: **Sara Vaezafshar**: conceptualization, data curation, formal analysis, investigation, methodology, validation, visualization, writing − original drafts, review and editing. **Sylvia Wolk**: investigation. **Victoria Arrandale**: conceptualization, writing − review and editing. **Roxana Sühring**: conceptualization, writing − review and editing. **Erica Phipps**: conceptu-

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alization, writing − review and editing. **Liisa Jantunen**: validation, writing − review and editing. **Miriam L. Diamond**: conceptualization, funding acquisition, project administration, supervision, visualization, writing − review and editing.

Notes

The authors declare no competing financial interest.

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