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Letter

Translating Silicone Wristbands to a Personal Exposure Estimate for Dermal Absorption: A Case Study with PCBs

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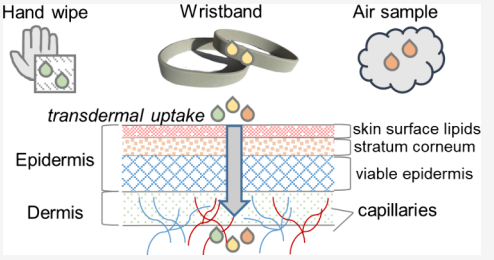
ABSTRACT: Dermal absorption is an exposure pathway frequently underestimated due to challenges in quantifying how exogenous chemicals pass through skin. Silicone wristbands have been used as effective personal passive sampling tools for semivolatile organic compounds (SVOCs) and serve as integrated measures of inhalation and dermal exposures. However, to-date, translating SVOC concentrations in wristbands to estimated doses has been a gap for their application in exposure assessment. Here, we utilized wristbands as proxy measures for skin-surface lipids to calculate estimated daily intakes (EDIs) from the dermal absorption pathway. EDIs were compared to dermal uptake estimates previously calculated from indoor air measurements and hand wipes, which were collected from residents of both polychlorinated biphenyl (PCB)-contaminated and noncontaminated buildings. Wristband-based estimates showed strong agreement with those from air and hand wipes ($r_s > 0.8$, $p < 0.0001$), particularly for lower chlorinated PCBs, with dermal uptake accounting for a third of exposed residents' PCB exposure ($\sum_7$PCB = 29.2 ng/kg bodyweight/day). All three dermal estimates were significantly associated with serum measurements among the lower chlorinated congeners. Our results demonstrate wristbands' potential for quantifying personal external exposure, building on their utility for ranking internal dose. This study further presents a method for using wristbands to calculate dermal absorption for personalized exposure assessment.



KEYWORDS: *polychlorinated biphenyl, indoor environment, transdermal uptake, passive sampling, estimated daily intake, hand wipes, indoor air*

INTRODUCTION

Humans encounter an extensive array of natural and synthetic chemicals daily. Common exposure routes include dietary and nondietary ingestion, inhalation, and dermal absorption. Of these, dermal absorption has been historically neglected and underestimated due to challenges in assessing how exogenous chemicals penetrate the skin.[1,2] Additionally, fractional absorption (i.e., an assumption that a somewhat arbitrary fraction of chemical concentration will penetrate the skin) has been commonly and erroneously used for estimating dermal uptake.[3] To address transdermal permeation of semivolatile organic compounds (SVOCs), Weschler and Nazaroff presented two methods for calculating dermal uptake: air-to-skin transport and hand wipes as measures of skin-surface lipid concentrations.[1] These methods have been used to estimate dermal exposure for several SVOC classes including pesticides, polycyclic aromatic hydrocarbons, phthalates, and polychlorinated biphenyls (PCBs).[1,2,4−6] They are described as frameworks for determining the dermal contribution to exposure within an order of magnitude; this is due to an explicit understanding that these dermal estimates rely on the accuracy of thermodynamic parameters.

Silicone wristbands have been used as passive sampling tools for evaluating personal exposure to industrial and consumer product chemicals primarily consisting of SVOCs.[7−9] Wristbands have been shown to effectively reflect internal dose as measured via biospecimens (i.e., urine and serum).[10−17] Previous work has also shown that wristbands are integrated measures of inhalation and dermal absorption, suggesting that wristbands could provide critical information on an oft-neglected exposure pathway.[18] However, it has been an ongoing challenge in exposure assessment to link a wristband concentration to a specific estimated dose. Recent work proposed using wristbands as generalized measures of skin adsorption across the entire skin surface; however, this dose estimate assumed that the measurement in the wristband would be the same as on the skin surface and did not consider uptake through the skin or the effects of skin washing on transdermal absorption.[19] O'Connell et al. conducted chamber studies with silicone wristbands to provide a generalized method for calculating air equivalency based on a wristband measurement;[20] however, chamber studies including only the silicone material are limited to gas-phase chemicals and do not

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consider processes such as particle deposition, direct contact, and skin excretion, which have been shown to likely impact accumulation into wristbands.[21,22] Similar evaluations have been done with silicone materials to determine partitioning between silicone and air[23−25] as well as uptake experiments conducted among individuals wearing wristbands for 5 days.[26,27]

Here, we hypothesized that the silicone wristband could serve as a proxy measure for deposition on and absorption through the skin. We calculated individualized dermal-based estimated daily intakes (EDIs) for PCBs from wristbands worn by residents of both PCB-contaminated and reference apartments. We compared these wristband-based dermal uptake estimates to our previous exposure pathway assessment using air samples and hand wipes as well as compared these three methods of estimating dermal uptake to paired serum measurements. Our work here proposes a method to provide a critical and quantitative link between wristband measurements and estimated dose for applications in personalized exposure assessment.

MATERIALS AND METHODS

Study Population. Participants were recruited from a Danish housing estate, Brøndby Strand Parkerne. Five of 12 high-rise apartment buildings were built with PCB-containing sealants which led to widespread indoor contamination. The exposed group included residents of these contaminated apartments, and the reference group resided in other apartments in the same estate. Concentration levels of ∼14 PCBs from active air samples, house dust, and surface wipes collected from each home,[28] and hand wipes, silicone wristbands, serum samples, and questionnaires collected from participants as well as sampling methods have been described in detail previously.[29,30] The most relevant parameters are summarized in the SI. Further, wristbands have been analyzed previously in both kinetic uptake and field experiments, providing equilibrium concentrations from individual wristbands.[30]

Calculation of Transdermal Uptake from Wristbands. These calculations were adapted from Weschler and Nazaroff (2012, 2014) and are based on transdermal permeation of SVOCs as measured on hands.[1,2] However, instead of a hand wipe, a wristband was used to estimate skin-surface lipid concentrations. These estimations rely on calculating transdermal flux, J, for transport from the skin-surface lipids through the stratum corneum and viable epidermis to enter the capillaries in the papillary dermis. Thus, the method estimating transport from skin-surface lipid to capillaries are identical for wristbands and hand wipes.

Transdermal flux is equivalent to the PCB concentration in the skin-surface lipids (C_l) multiplied by the permeability coefficient from the skin-surface lipid layer to the capillaries (k_{p_l}) at steady state.

$$J = C_l \times k_{p\_l} \tag{1}$$

Here, we estimated the PCB concentration in skin-surface lipids (C_l) from the equilibrium concentration in the wristband (C_s) and the partitioning coefficient between skin-surface lipids and silicone (K_{ls}).

$$C_l = C_s \times K_{ls} \tag{2}$$

The calculation of C_s [ng/g silicone] is based on Frederiksen et al. (described there as C_∞) and is estimated from the measured wristband concentration (C_{wb}), the congener-specific rate constant (k) [day^{-1}], and time (t) the wristband is worn (7 days).[30]

$$C_s\ [ng/g\ silicone] = \frac{C_{wb}}{1 - e^{-kt}} \tag{3}$$

For k, we used congener-specific estimates from the kinetic study in Frederiksen et al.[30] For higher chlorinated PCBs not estimated there, we used the model from O'Connell et al. to estimate k based on QSAR-based boiling points.[20] PCB boiling point data from the EPA CompTox[31] served as inputs.[20] C_s was then converted to PCB mass per wristband volume based on wristband density (1.08 ng/cm^3).[30]

Next, we estimated K_{ls} from the lipid-air (K_{la}) and silicone−air (K_{sa}) partitioning coefficient.

$$K_{ls} = \frac{K_{la}}{K_{sa}} \tag{4}$$

However, partitioning at the skin surface will occur at 32 °C (skin-surface temperature) while the wristband reflects the ambient environment (e.g., room temperature). Therefore, we retain K_{la} at the skin-surface temperature and K_{sa} at ambient temperature, resulting in a partitioning coefficient reflecting this difference, K'_{ls}.

$$K'_{ls} = \frac{K_{la,32°C}}{K_{sa,21°C}} \tag{5}$$

Therefore, we use K'_{ls} to estimate C_l in a variation of eq 2, given that the concentrations in both the skin-surface lipids and silicone will reflect this temperature difference.

$$C_l = C_s \times K'_{ls} \tag{6}$$

As in Weschler and Nazaroff, stratum corneum lipids are assumed to partition to water in a similar manner as skin-surface lipids.[3] As such, the partitioning coefficient between skin-surface lipids and water (K_{lw}) is approximated from a modeled linear relationship with log K_{ow}, described in Bunge and Cleek (1995) as cited in Weschler and Nazaroff and in Anissimov et al. 2013.[1,32] Log K_{ow} and Henry's law constant (H) was calculated using SPARC at 32 °C.[33] R is the gas constant (0.0821 atm/M/K).

$$\log(K_{lw}) = 0.74 \log(K_{ow}) \tag{7}$$

Using partitioning to water as the common denominator, K_{la} (skin lipids-air) was calculated from K_{lw} and K_{aw} (air−water).

$$K_{la} = \frac{K_{lw}}{K_{aw}} \tag{8}$$

When combined with the ideal gas law, where $K_{aw} = (HRT)^{-1}$, K_{la} can be estimated as

$$\log K_{la} = 0.74 \log K_{ow} + \log H + \log RT \tag{9}$$

Values for K_{sa} were similar when calculated from the kinetic uptake study in a contaminated apartment[30] and in chamber studies.[23] We utilized the K_{sa} from Frederiksen et al. for the lower chlorinated PCBs[27] then Tromp's model for the remaining congeners.[20] Log K_{oa} values for the Tromp model were sourced from experimentally derived values from Harner and Bidleman.[34] Relevant physical-chemical properties are presented in Table S1. Thereby C_l can be estimated from eq 6. Calculations of k_{p_l} follow the equations and assumptions

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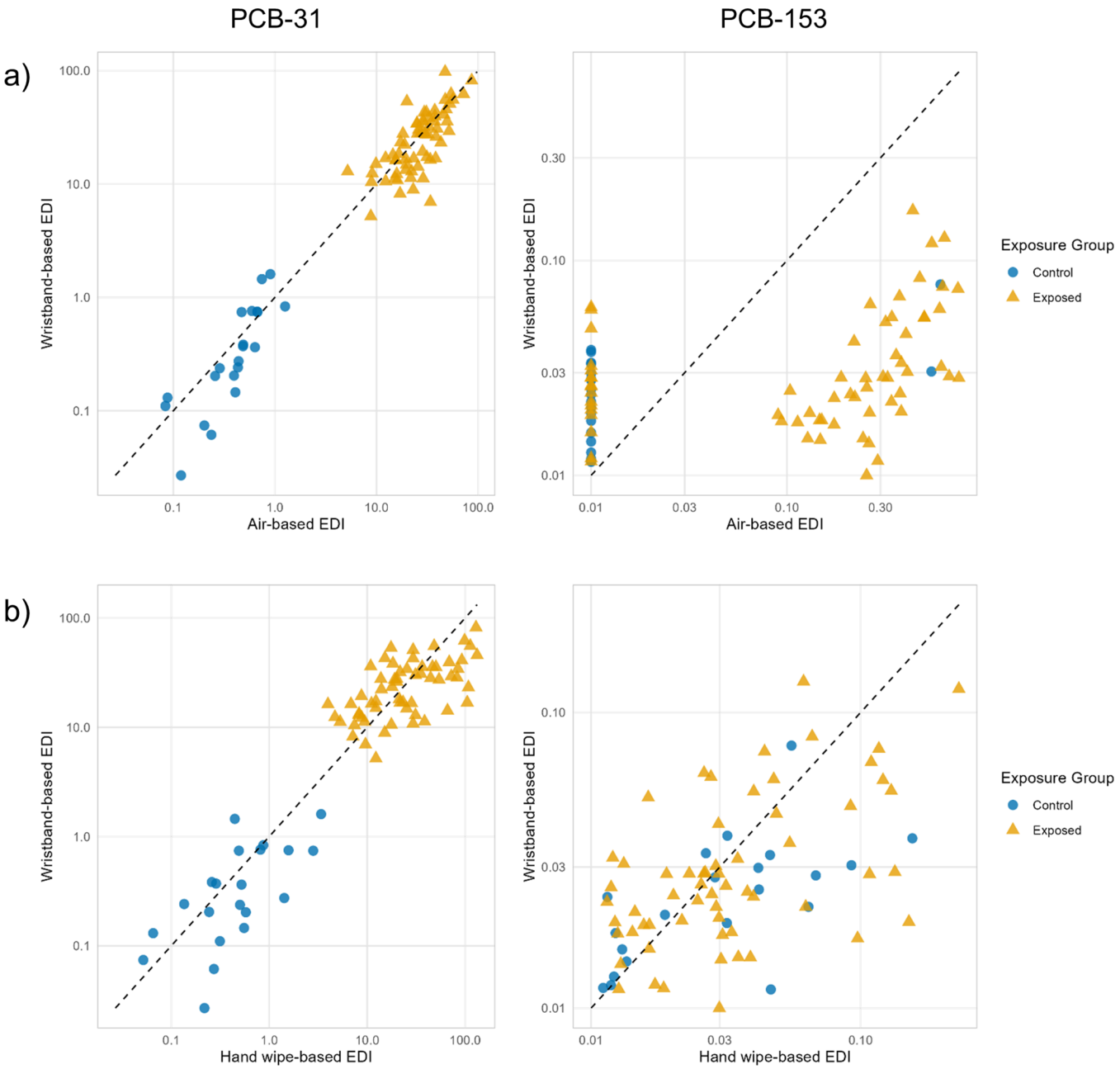

Figure 1. Comparison of individualized dermal EDIs (ng/kg body weight/day) for PCB-31 and −153 comparing wristband-based estimates to (a) air-based estimates and (b) hand wipe-based estimates (n = 85). The control group is plotted as blue circles exposed group is plotted as golden triangles. A 1:1 line is plotted for comparisons between methods.

made by Weschler and Nazaroff (Section S2).[1,5] Together, these can be used to calculate J [ng/m^2/h] (eq 1).

We applied J to the total body surface area, estimated by age and sex of each participant from the US EPA Exposure Factors Handbook (range = 1.69−2.15 m^2).[35] Estimated daily intakes (EDIs) via dermal uptake were calculated assuming a 24 h/day exposure duration and self-reported weight. Missing body-weight data (n = 1) was imputed to 70 kg.[36]

$$EDI_{dermal} = \frac{J \times total\ body\ surface\ area \times exposure\ duration}{body\ weight} \quad (10)$$

Comparison of Dermal Uptake Estimates and to Serum Measurements. Transdermal uptake was calculated from both air samples and hand wipes in a similar manner to above and have been described previously.[5] Here, we included the 14 congeners which overlap across samples for comparisons: PCB-8, −18, −28, −31, −44, −52, −66, −99, −101, −105, −118, −138, −153, and −180. Comparisons between individualized dermal estimates for air and wristbands were conducted using Spearman correlations (r_s) due to their non-normal distributions. Linear regression models were used for additional comparisons between the matrices. All variables were $\log_{10}$-normalized to approach normality with nondetects replaced with LOQ/2. Congeners were only assessed with ≥ 50% detection frequencies. Skin-surface lipid PCB concentrations from hand wipes was compared to those from wristbands to assess the slope and intercept. Comparison of

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the three dermal uptake estimates to serum were also conducted with linear regressions that were adjusted for exposure category (exposed or reference). Fold-change in serum per 10-fold increase in dermal estimate were calculated per matrix and congener. Stratified analyses by exposure category are also shown in SI.

RESULTS AND DISCUSSION

In total, 85 wristbands were collected from 64 participants living in 52 contaminated apartments and 21 living in 18 reference apartments. For each wristband, we calculated a personalized exposure estimate for dermal uptake based on previously conducted kinetic experiments and modeled data from chamber studies. The lower chlorinated congeners dominated among exposed participants, with PCB-31 having the highest median dermal exposure estimate (median = 25.75 ng/kg bodyweight/day; Figure 1, Table S2). For the sum of seven indicator PCBs ($\sum_7$PCB; PCB-28, −52, −101, −118, −138-, 153, −180), which is a commonly used sum in European exposure assessment, the wristband-based median estimate of dermal absorption was 29.2 ng/kg bw/day (Table S2). This closely reflected the magnitude of estimates calculated from both air and hand wipes (median $\sum_7$PCB of 29.3 and 36.0 ng/kg bw/day, respectively). Notably, these summed estimates for this group are dominated by the lower chlorinated PCBs (PCB-28, −52) with the higher chlorinated congeners being similar across all participants.

Overall, our results demonstrated strong agreement between the dermal uptake estimates calculated from wristbands compared to air (i.e., air-to-skin transport) and hand wipes. When comparing individual dermal EDIs calculated from air and wristbands across all participants, there was strong agreement for lower chlorinated congeners up to PCB-101 (r_s = 0.81−0.87, p < 0.0001; Figures 1, S1), with a spread around the 1:1 line which demonstrated likeness between the two estimates and a slight bias of higher values among air-based estimates. PCB-105 to −180 had more limited detections in air, particularly among the reference population, but were still positively and significantly associated for the exposed population among congeners with >50% (PCB-105, −118, −153; r_s = 0.45−0.63, p < 0.0001; Figure 1). However, exposure to higher chlorinated PCBs like PCB-153 is generally attributed to diet, which would not be captured by the matrices presented here.[5] This is clear from Figure 1 when comparing PCB-31 and −153 from the wristband EDI to air and hand wipe EDIs. The plots for the lower chlorinated PCB clearly differentiate the two exposure groups while the higher chlorinated PCB shows a mixing (Figure S1, S2). In addition, the higher chlorinated PCBs are more likely to be particle-bound at room temperature; while the active air samples captured both gas and particle phases, these higher chlorinated congeners were less likely to be in the air altogether. A recent study indicated that for chemicals with logK_{oa} > 9, a cutoff which falls within the tetra-CBs, mass accumulation of SVOCs in wristbands is not explained solely by gas−solid partitioning and likely includes other processes such as particle deposition and direct contact.[22] Thus, wristband concentrations of higher chlorinated PCBs here could be due in part to contact with enriched surfaces.

A similar relationship was observed between the dermal estimates of wristbands and hand wipes (Figure 1, S2). Comparison of skin-surface lipid concentrations calculated from hand wipes to those from wristbands showed a strong linear relationship, with the linear regression between the two $\log_{10}$-transformed matrices having a slope of ∼1 (range = 0.96−1.36) for nearly all lower-chlorinated PCBs (Table S3). Excluding PCB-66 which had a significant y-intercept, wristband-based skin lipid estimates had a 9.1- to 14.1-fold change per 10-fold increase in hand wipe estimates (Table S3). This suggests that the skin-surface lipid estimates tend to fall consistently within the same order of magnitude (∼10-fold change in one per 10-fold increase in the other), thereby yielding similar dermal dose estimates despite using different partitioning parameters. Given that both air-to-skin transport and hand wipes have been described as validated dermal measures for several classes of SVOCs, this suggests that a wristband as a proxy measure for the skin-surface lipids can also produce a representative dermal exposure estimate.

Regarding the relationships between the dermal estimates and internal dose for the lower chlorinated PCBs, the three matrices pointed to a 1.2- to 3.9-fold increase in serum per 10-fold increase in dermal estimate, reaching statistical significance for all except PCB-99 in wristbands (Figure S3). This relationship was even clearer when assessed only among the exposed, demonstrating that all three dermal estimates were significantly related to internal dose and verifying the contaminated indoor environment as a source of PCB exposure (Figure S3). The lack of relationships in the reference population points to the fact that those individuals were very limitedly exposed to PCBs from their indoor environment. For lower chlorinated PCBs, the closeness of three matrices' EDIs in magnitude and significant relationships to serum measurements indicate that all three measures can be used interchangeably to estimate transdermal absorption. However, this interchangeability could rely on the fact that this study population spent most of their time in their apartments and had lived in their homes for 18 years, on average.[29] As such, their exposure would distinctly reflect their homes, and their skin-surface lipids were likely in equilibrium with their home environment. This suggests, however, that if someone spent less time at home (or in any dominant microenvironment), indoor air samples and hand wipes could have less utility for predicting a person's individualized dose. Our study here provides an ideal comparison group, specifically among the exposed group, to test the wristband's utility for estimating dermal exposure. Since the wristband samples from all the microenvironments encountered during a sample period, our study indicates that we could calculate a cumulative dose of dermal absorption solely based on the wristband measure.

Our results should be considered within the context of some limitations. We assumed that the wristband surface reflects ambient room temperature, but this could change based on how tightly the wristbands fit. Proximity and constant contact with the skin could increase the wristband temperature, although the outer wristband surface will likely maintain a cooler temperature than the skin surface. Additionally, there is uncertainty about how clothing affects accumulation in wristbands. Our previous work in occupational settings did not show significant differences between wristbands that were covered by clothing or not;[37] however, clothing coverage could affect the temperature at which the wristband samples, thereby changing the dermal estimates. Further, PCBs have been widely studied as legacy contaminants, which provided a strong literature base for deriving experimental values for physical-chemical and partitioning characteristics. Other SVOCs may

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have more limited data available, and therefore calculations could rely more heavily on models to derive critical parameters, many of which are estimated within an order of magnitude. The framework used here was best fit for chemicals with molecular weight <400 g/mol (Section S2), which limits applications to some higher chlorinated PCBs as well as larger SVOCs.[1] Other skin permeability models may be required for these larger chemicals, since this parameter relies heavily on a chemical's molecular weight, lipophilicity, and hydrogen-bonding capacity.[38] As legacy contaminants, background environmental exposure to lower-chlorinated PCBs is very low and rather rare. This particular case study presents an exposure scenario with two distinct exposure categories: one with high exposure and the other with more typical background levels. While this limits generalizability to the general population's exposure to PCBs specifically, this provides a definitive examination of the role of the indoor environment in contributing to personal exposure and thus could provide a framework for evaluating other ubiquitous environmental contaminants. Lastly, we acknowledge uncertainty regarding the role of particle-bound PCBs. Previous studies have demonstrated that <4% of the lower chlorinated PCBs will be particle-bound up to PCB-101, indicating gas-phase dominance.[39,40] Here, any sorbed particles on wristbands were not physically removed prior to extraction, so all PCBs, both gas- and particle-phase, are included in wristband concentrations. While this likely reflects how chemicals come in contact with the skin, further research is necessary to examine how particles and particle-bound SVOCs partition through the skin.

Here, we presented a calculation of EDIs for dermal absorption from wristbands, which showed strong agreement with our previous estimates from paired air and hand wipes. This suggests the potential for using wristbands in a quantitative manner to estimate personal daily dose beyond their utility as ranking tools representing internal body burden. It further presents the wristbands as effective tools for measuring dermal absorption so that this pathway can be more strongly incorporated into exposure assessment, given its prominent contribution to SVOC exposure.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.5c00346.

Additional information about sample collection, permeability coefficient calculations, relevant physical-chemical properties, and tabulated results and plots comparing the matrices in regression analyses (PDF)

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Notes

The authors declare no competing financial interest.

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