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Article

Cloth–Air Partitioning of Neutral Per- and Polyfluoroalkyl Substances (PFAS) in North Carolina Homes during the Indoor PFAS Assessment (IPA) Campaign

Clara M. A. Eichler, Naomi Y. Chang, Elaine A. Cohen Hubal, Daniel E. Amparo, Jiaqi Zhou, Jason D. Surratt, Glenn C. Morrison, and Barbara J. Turpin*

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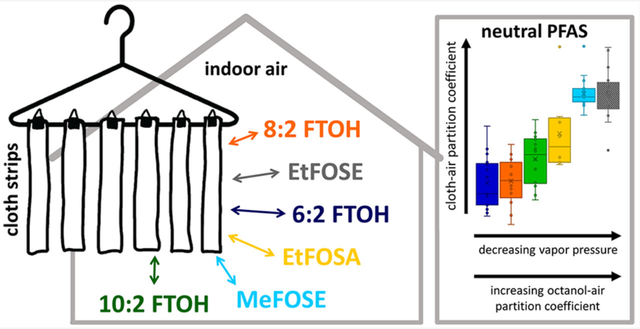

ABSTRACT: Partitioning of per- and polyfluoroalkyl substances (PFAS) to indoor materials, including clothing, may prolong the residence time of PFAS indoors and contribute to exposure. During the Indoor PFAS Assessment (IPA) Campaign, we measured concentrations of nine neutral PFAS in air and cotton cloth in 11 homes in North Carolina, for up to 9 months. Fluorotelomer alcohols (i.e., 6:2 FTOH, 8:2 FTOH, and 10:2 FTOH) are the dominant target species in indoor air, with concentrations ranging from 1.8 to 49 ng m^{-3}, 1.2 to 53 ng m^{-3}, and 0.21 to 5.7 ng m^{-3}, respectively. In cloth, perfluorooctane sulfonamidoethanols (i.e., MeFOSE and EtFOSE) accumulated most significantly over time, reaching concentrations of up to 0.26 ng cm^{-2} and 0.24 ng cm^{-2}, respectively. From paired measurements of neutral PFAS in air and suspended cloth, we derived cloth–air partition coefficients (K_{ca}) for 6:2, 8:2, and 10:2 FTOH; ethylperfluorooctane sulfonamide (EtFOSA); MeFOSE; and EtFOSE. Mean log(K_{ca}) values range from 4.7 to 6.6 and are positively correlated with the octanol–air partition coefficient. We investigated the effect of the cloth storage method on PFAS accumulation and the influence of home characteristics on air concentrations. Temperature had the overall greatest effect. This study provides valuable insights into PFAS distribution, fate, and exposure indoors.

KEYWORDS: *Indoor air quality, indoor exposure, clothing, sinks, residential, cotton, sorption, textile*

INTRODUCTION

The indoor environment can be an important contributor to population exposures to per- and polyfluoroalkyl substances (PFAS).[1,2] Exposures may occur via inhalation of airborne particles and gases; dermal exposure by direct contact with PFAS-containing materials or from the air; ingestion of food, water, and dust; and mouthing of materials.[3] PFAS are manufactured chemicals, many of which are persistent in the environment and bioaccumulative.[4−6] Currently, it is estimated that thousands of different PFAS exist, either for use in industry and product manufacture or as precursors or breakdown products.[1,6−9] PFAS are commonly used in consumer products and articles as water- and stain-resistant or nonstick coatings and as surfactants.[1,10−12] Examples of products treated with PFAS include furnishings, carpets, functional apparel, outdoor gear, cookware, food-contact materials, paints, and cleaning products. Exposure to several PFAS has been linked to a number of adverse health outcomes in humans and wildlife;[1,13] however, the toxic potential of the majority of PFAS has not been studied to date.[14] While diet and contaminated drinking water are often considered main pathways of human exposure to PFAS,[1] the colocation of people and PFAS-containing products and materials in homes, where most people spend the majority of their time,[15] makes the indoor environment a likely place for substantial inhalation and perhaps dermal PFAS exposure.

Existing studies measuring PFAS in residential indoor air (i.e., studies from Asia, Europe, and Canada and one study from the United States) have concluded that neutral PFAS dominate indoor PFAS concentrations; specifically, the fluorotelomer alcohols (FTOHs) 6:2 FTOH and 8:2 FTOH are present in indoor air with concentrations in the range of 1–100 ng m^{-3} in residential settings.[16−25] Other neutral PFAS, such as fluoroalkyl sulfonamides (FASAs), fluoroalkyl sulfonamidoethanols (FASEs), and fluorotelomer acrylates (FTACs), have also been measured in residential indoor air.[16−28] Biotransformation of neutral PFAS can result in

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intermediates and products that accumulate in the human body, as has been shown, for example, for 6:2 FTOH.[29,30] Previous studies[31,32] found positive associations between FTOH concentrations in indoor air and concentrations of perfluoroalkyl acids such as perfluorooctanoic acid (PFOA) and perfluorononanoic acid (PFNA) in human serum. PFOA and PFNA have been linked to a range of adverse health effects.[33] However, our knowledge of the concentrations, composition, occurrence, and fate of neutral PFAS in residential indoor air is still very limited, especially in the United States (U.S.).

Clothing could be a mediator of exposure to PFAS, as has been shown for a variety of semivolatile organic compounds (SVOCs; e.g., phthalates,[34,35] nicotine,[36] and other compounds[35,37,38]). People are in continuous and intimate contact with different types of clothing materials. Clean clothing can act as a barrier to exposures by serving as a sink for chemicals present in the indoor air, as has been shown for phthalates and nicotine, for example.[36,39] However, over time, these clothing items can accumulate SVOCs from the indoor air, and clothing can then become a secondary source with the potential to prolong exposure to the accumulated compounds, contributing to dermal exposure and to exposure via other pathways (e.g., ingestion via mouthing of fabrics and inhalation).[36,39,40] In addition, PFAS have been found in clothing that has been treated to repel water or stains.[41−47] These clothing items are likely additional sources of PFAS in indoor environments.[40,47,48]

Thus, it is likely that clothing plays an important role in human PFAS exposure; however, the partitioning of PFAS between air and initially PFAS-free, untreated clothing has not been studied to date. Cloth−air partition coefficients (K_{ca}) are key to advancing our understanding of the impact of clothing on exposure.[49,50] K_{ca} values have been derived or directly measured for phthalates, flame retardants, methamphetamine, and volatile organic compounds (VOCs) for partitioning to cotton, polyester, rayon, linen, and some blends, either in chamber experiments or by deploying clothing in realistic indoor settings.[34,35,38,51−54] The benefit of measuring K_{ca} in realistic environments is that the influence of changing environmental conditions and competition with other indoor species is reflected in the resulting partition coefficients, thus making them more directly applicable in exposure assessments. However, no K_{ca} values have been measured for PFAS to date.

To address these knowledge gaps, we conducted a field campaign with the first concurrent residential measurements of neutral PFAS in air and untreated cloth. Below, we describe the variations in concentrations and compositions of nine neutral PFAS in air and cloth sampled over up to 9 months in 11 U.S. (North Carolina) homes. We quantify K_{ca} for six neutral PFAS based on paired measurements in indoor air and cloth. This analysis was designed to further inform PFAS exposure models that can provide population-based exposure predictions and contribute to more comprehensive policies for PFAS exposure mitigation.

■ MATERIALS AND METHODS

The Indoor PFAS Assessment (IPA) Campaign. The overall goal of the IPA Campaign was to improve our understanding of the mechanisms that govern PFAS behavior, fate, and transport indoors. Thus, we designed a study that included a comprehensive suite of measurements across multiple indoor environmental compartments and seasons. A list of all samples collected during the IPA Campaign is provided in the Supporting Information (SI), Table S1. To the best of our knowledge, this campaign is the most comprehensive residential indoor PFAS measurement campaign to date, in terms of the number of indoor compartments sampled concurrently and over an extended period of time. Herein, we focus on nine neutral PFAS measured in total-air and gas-phase samples, as well as in cloth and clothing samples. These nine neutral target species have been found in indoor air in previous studies.[18,20,21,24,26] Subsequent publications will address other aspects of the IPA Campaign.

Eleven nonsmoking, detached, single-family homes in the Chapel Hill/Durham area of North Carolina were sampled between July 4, 2021 and May 20, 2022 (UNC Chapel Hill IRB 20−2771), with staggered start dates. Initially, a convenience sample of 11 homes was recruited; one home (Home 82) left the field campaign after one month, so that 10 homes completed the study. Each home was visited multiple times over a 6−9-month period, as illustrated in Figures S1 and S2. Before sampling, informed consent was given and a home survey was administered to collect information about building characteristics and typical occupant activities (Figure S3). Temperature and relative humidity (RH) were measured continuously throughout each home's 6−9-month study participation. At the end of each home's participation, a Home Exit Survey (Figure S4) was conducted to address any changes that may have occurred in the home during the study.

Active 6-day sampling periods took place three times in each home: at the beginning of each home's study participation (t = 0), after 3 months, and after 6 months (Figure S1). During active sampling, CO_2 was measured continuously, and participants completed daily activity checklists (Figure S5), detailing their cooking, cleaning, laundering, and heating/cooling/ventilation behaviors as well as occupancy. At the end of each 6 day sampling period, an activity survey (Figure S6) was conducted to capture additional information about activities and behaviors potentially related to PFAS emissions, concentrations, or removal during sampling.

Key characteristics of the 11 study homes (taken from surveys and measured environmental conditions) are provided in Table S2. Home sizes were approximately 70−240 m^2 (740−2620 ft^2). All homes had central heating and air conditioning (HAC) systems, 1−4 occupants, and 0−3 pets. Estimated air change rates (ACH) for each home averaged 0.2−0.6 h^{-1}, with variations (within home) of 0.02 h^{-1} (10%) to 0.3 h^{-1} (66%). Window opening behavior (Table S3) was quite variable, with most window opening (for more than 15 min) occurring in the fall and winter months. Several occupants did not open their windows at all during the sampling periods, and one reported opening windows on 14 occasions in late summer during the first 6-day sampling period. Carpet or rugs were found in all homes, and hardwood flooring, including bamboo, was the most common flooring type (Table S2). Additional survey data will be reported in subsequent papers.

Chemicals. Names, CAS RNs, and vendors of the nine neutral PFAS analytes and six mass-labeled neutral PFAS standards are listed in Table S4. Table S5 summarizes relevant physicochemical properties. We included estimates of vapor pressures and octanol−air partition coefficients derived with the OPEn structure−activity/property Relationship App (OPERA), as implemented in the U.S. Environmental Protection Agency's CompTox Chemicals Dashboard, because of its excellent performance in cross-model evaluation.[55] HPLC-grade methanol (99.9% purity) and GC-grade hexane (99.9% purity) were purchased from Fisher Scientific (Waltham, MA).

Air Sampling. PFAS in indoor air were sampled 1 m above floor level in the main living area of each home on three occasions by pulling air (0.3 m^3 h^{-1}) for ~72 h (21.2 m^3 average sample volume) through a polyurethane foam (PUF)-XAD2-PUF sandwich cartridge (ORBO 1500 Precleaned Small PUF/Amberlite XAD-2/PUF Cartridge, Catalog No. 21233-U, Supelco, Bellefonte, PA). There were two exceptions: Home 82 was sampled only on one occasion, and Home 01 was sampled on two occasions. In some cases, a quartz-fiber filter (QFF, 32 mm diameter, Catalog No. 21038, Supelco, Bellefonte, PA; prebaked at 550 °C for 12 h) was attached upstream of the PUF-XAD2-PUF cartridge to remove particles, so that the PUF-XAD2-PUF cartridge collected only the gas phase. Further details regarding air sampling and sample handling and storage can be found in section S1. In total, 21 total-air samples (PUF-XAD2-PUF alone) and 25 gas-phase samples (prefiltered) as well as field blanks, replicates, and breakthrough samples were collected (Table S6).

Cloth Sampling. Two types of passive cotton cloth samplers (samplers A and B) were deployed during the first sampling visit (t = 0) at each home, allowed to accumulate PFAS, and retrieved at several time points across each home's 6−9-month study participation. A precleaned, 100% cotton cloth (TS Designs, Burlington, NC; areal density 0.016 g cm^{-2}, thickness 0.058 cm, bulk density 0.27 g cm^{-3}) was used for this purpose. Details regarding the cloth's preparation, deployment, sampling, and storage are listed in section S2. Briefly, sampler A consists of six clean cotton cloth strips (~28 cm × 3.8 cm) hanging freely from a stainless-steel clothes hanger below an aluminum roof to minimize the deposition of large particles via gravitational settling (Figure S7A). At each home, two hangers (12 cloth strips total) were placed in the closet of the master bedroom. During deployment of the hangers, two cloth strips per home were brought to each home and immediately retrieved as "t = 0" samples. Then, two (duplicate) cloth strip samples were retrieved after 24 h (t = 1), 6 days (t = 2), 1 month (t = 3), 3 months (t = 4), 6 months (t = 5), and at the end of each home's participation in the field campaign (t = 6). A total of 143 cloth strip samples (incl. 71 duplicates) and 27 cloth strip field blanks were collected and subsequently analyzed.

Sampler B consists of 14 clean cotton pieces (~28 cm × 15 cm) that were folded once and distributed between stacked clothing items, most commonly in a dresser drawer in the master bedroom (Figure S7B). During deployment, two additional cloth pieces were brought to each home and immediately retrieved as "t = 0" samples without placement between clothing. Then, two cloth pieces were retrieved after 24 h (t = 1), 6 days (t = 2), 1 month (t = 3), 3 months (t = 4), 6 months (t = 5), and at the end of each home's participation in the field campaign (t = 6). Additional clean cotton pieces were brought to the homes on one to three occasions per home to serve as field blanks. In total, 146 cloth piece samples and 13 field blanks were collected; 91 cloth piece samples and 13 field blanks were analyzed.

All study participants also donated one piece of 100% cotton clothing for which the label did not indicate that it was treated with PFAS and that had been laundered and stored in their home for at least three months without being worn. In total, 11 pieces of household clothing items were collected (see section S2 for details).

Indoor and Outdoor Environmental Conditions. Carbon dioxide (CO_2; 6-day sampling periods only; Extech SD800, Extech Instruments, Nashua, NH), temperature, and RH (entire campaign; 5 min intervals; HOBO UX100−011A, Onset, Bourne, MA) were measured in the main living area of each home. ACH estimates were derived from the CO_2 data using the approach described by Bekö et al.[56] Outdoor temperature and RH for July 2021 to May 2022 were retrieved from the weather station at Raleigh-Durham International Airport (KRDU) using the University of Utah's MesoWest Web site (mesowest.utah.edu).

Sample Processing and Analysis. Extraction and analysis of PUF-XAD2-PUF cartridges, cloth strips, sections of the cloth pieces, and sections of clothing items as well as blanks, duplicate samples, PUF-XAD2-PUF breakthrough samples, and positive storage controls of cloth samples are described in detail in section S3. Briefly, all samples and blanks were spiked with 20 μL of a mass-labeled recovery standard mixture (5 ng μL^{-1}) and extracted twice by sonication (20 min) by using a 3:1 (v/v) hexane/methanol solvent mixture. A 50 mL polypropylene (PP) syringe (BD, Franklin Lakes, NJ) was used to squeeze remaining solvent from each substrate (i.e., cloth, clothing, PUF-XAD2-PUF) before extract cleanup with ENVI-Carb (Supelclean ENVI-Carb SPE Bulk Packing, Supelco, Bellefonte, PA). The extracts of each sample were combined and immediately concentrated under a gentle stream of high-purity nitrogen. PUF-XAD2-PUF extracts were reduced to ~1000 μL, and a 200 μL aliquot was used for analysis. Cloth and clothing extracts were concentrated to ~300 μL and analyzed. Details on quality control measures, including extraction efficiencies, breakthrough, detection limits, and precision, are reported in section S4 and Tables S7−S13. Method detection limits (MDLs, Table S9) range from 0.03 ng m^{-3} to 0.2 ng m^{-3} for PUF-XAD2-PUF air samples and 0.001 ng cm^{-2} to 0.1 ng cm^{-2} for cloth and clothing samples. MDL is given by the larger of the instrument detection limit (IDL) and the field-blank-based detection limit (FDL). The IDL is the mean concentration plus three times the standard deviation of repeat injections of the lowest calibration standard used (0.001 ng μL^{-1}). The FDL is the mean plus three standard deviations of the field blank values for that type of sample, if available.

We analyzed the samples for nine neutral PFAS analytes and six neutral mass-labeled PFAS standards in selected-ion monitoring (SIM) mode using an Agilent 8890 gas chromatograph (GC) with an Agilent DB-WAX column (30 m length, 0.25 mm ID, 0.25 μm film thickness) and an Agilent 5977B mass-spectrometry detector (MSD) in electron ionization (EI) mode (Table S14). Samples were injected using pulsed splitless injection (injection volume 2 μL). A subset (18% and 15% of air and cloth sample extracts, respectively) was analyzed in replicate. Each analyte and mass-labeled standard was quantified using authentic standards and a seven-point calibration curve ranging from 0.001 ng μL^{-1} to 1.0 ng μL^{-1}. Peak integration was performed using Agilent Enhanced ChemStation (Version F.01.03.2357) software. Reported concentrations were corrected for recoveries of the mass-labeled standards (section S4); they were not corrected for field blanks or breakthrough.

Calculation of Accumulation Rates and Partition Coefficients. Accumulation rates R_i (ng h^{-1} m^{-2}) of each

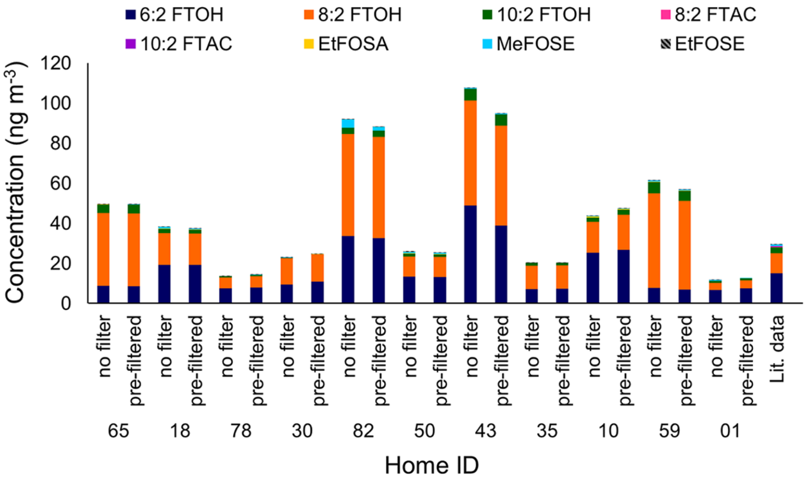

Figure 1. Concentration profiles of neutral PFAS measured in air samples in 11 homes during the first sampling visit. The label "no filter" refers to total-air samples, and "pre-filtered" refers to gas-phase samples. The bar showing the results for "Lit. data" is based on multiple studies.[16−18,20−25] MeFOSA was not detected in any samples and, therefore, not shown. Results are shown in the order in which samples were collected at the homes.

neutral PFAS i in the suspended cloth strips were calculated for each home and each sampling time step as

$$R_{i,t} = \frac{c_{i,t} - c_{i,t-1}}{d_t - d_{t-1}} \tag{1}$$

where $c_{i,t}$ and $c_{i,t-1}$ (ng cm^{-2}) are the area-based concentrations of species i in the cloth at sampling times t and $t - 1$, respectively, and $d_t - d_{t-1}$ (h) indicates the number of hours in between the two sampling times. Positive rates show an accumulation in the cloth, while negative rates indicate a loss.

The cloth−air partition coefficient, K_{ca}, is defined as the concentration of a chemical in the cloth divided by the gas-phase concentration of the chemical at equilibrium. For cloth, the partition coefficient can be normalized by area, mass, or volume of the cloth material.[38] The mass-normalized partition coefficient, $K_{ca,mass}$ (m^3 g^{-1}), is used here as the basis to derive K_{ca} (unitless) for each PFAS i as follows:[38]

$$K_{ca,mass,i} = \frac{c_{i,t}/\rho_{area}}{c_{g,t}} \tag{2}$$

where $c_{i,t}$ (ng cm^{-2}) is the area-based concentration of species i in the cloth at time t, ρ_{area} is the areal density of the cotton cloth (g cm^{-2}), and $c_{g,t}$ is the gas-phase concentration (ng m^{-3}) at time t. If a species was detected but concentrations were below the MDL, the measured value, rather than zero or MDL/2, was used. If a species was not detected, then the concentration was set to zero. $K_{ca,i}$ is then calculated as

$$K_{ca,i} = K_{ca,mass,i} \cdot \rho_{bulk} \tag{3}$$

where ρ_{bulk} is the bulk density of the cotton cloth (g m^{-3}).

Across-Home and within-Home Variability. Across-home variability (AHV) and within-home variability (WHV) of measured PFAS concentrations in indoor air were calculated as pooled coefficients of variation (%) for either paired ($n = 2$) or $n > 2$ samples. Details can be found in the SI, section S5.

RESULTS AND DISCUSSION

Neutral PFAS in Indoor Air and Suspended Cloth. FTOHs are the dominant measured PFAS in all indoor air samples, accounting for 94% (on average) of the mass of the nine measured neutral PFAS in the total-air (PUF-XAD2-PUF, no filter) samples (Figures 1 and S8; Table S15); in contrast, FOSEs are the dominant species in the suspended cloth strips, accounting for 62% (on average) of the mass of measured neutral PFAS (Figures 2 and S9; Table S16).

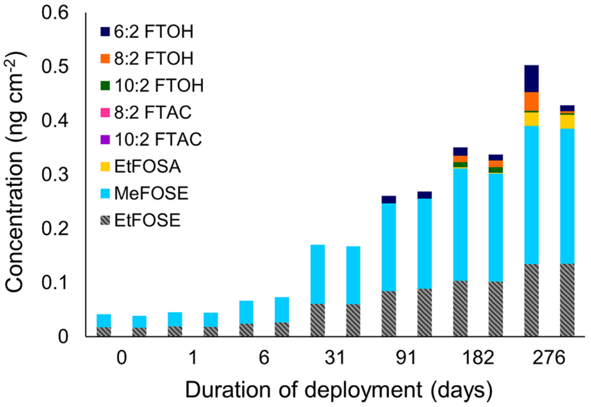

Figure 2. Concentration profiles of neutral PFAS measured in suspended duplicate cloth strips (sampler A), sampled from Home 18 over the duration of the field campaign. MeFOSA was not detected in any samples and is therefore not shown.

The FTOHs (6:2 FTOH, 8:2 FTOH, and 10:2 FTOH), MeFOSE, and EtFOSE were present above the MDL in 100% of the total-air samples (Table S15). Concentrations of 6:2 FTOH and 8:2 FTOH in total-air samples range from 1.2 to 53 ng m^{-3} (means: 13 ng m^{-3} and 15 ng m^{-3}, respectively), whereas concentrations of 10:2 FTOH are an order of magnitude lower (mean: 2.2 ng m^{-3}). Mean MeFOSE and EtFOSE concentrations in the total air are 0.60 ng m^{-3} and 0.19 ng m^{-3}, respectively. In contrast, 8:2 FTAC, 10:2 FTAC, and EtFOSA are above MDL in one-third or less of total-air samples, with mean concentrations in the range of 0.06 ng m^{-3} (10:2 FTAC) to 0.18 ng m^{-3} (8:2 FTAC). MeFOSA was not detected in any air sample. Overall, the concentrations are on the same order of magnitude and follow similar trends as reported in previous studies on PFAS in indoor air, with high detection frequencies of FTOHs and FOSEs, and 6:2 FTOH and 8:2 FTOH as the dominant neutral species.[16−18,20−25]

There is no statistically significant difference between total-air and gas-phase samples (Figures 1 and S8) for six of the eight detected PFAS, according to paired two-sided *t*-tests or Wilcoxon signed-rank tests, depending on the sample distribution (95% confidence intervals; section S6; Table S17). QFFs collect particles with >99.99% efficiency and are known to adsorb some organic gases.[57−60] Therefore, based on the difference between total-air and gas-phase concentrations, at a minimum, 88% of neutral PFAS found in total-air samples are associated with the gas phase. Statistically significant differences are observed only for MeFOSE and EtFOSE, which are less volatile than the other detected PFAS (Table S4), indicating some removal of FOSEs by the upstream QFF. On average, MeFOSE and EtFOSE account for 3.7% of the sum of measured neutral PFAS in total air ($\sum(\text{neutral PFAS})_{\text{air}}$) and 3.6% in the gas phase ($\sum(\text{neutral PFAS})_{\text{gas}}$). Removal of FOSEs by filters has also been reported in the literature, while FTOHs have not been found in the airborne particle phase indoors.[61]

Neutral PFAS accumulated in the suspended cloth strips (sampler A) as they were exposed in homes during the campaign (Figures 2 and S9). The highest measured sum of neutral PFAS concentration ($\sum(\text{neutral PFAS})_{\text{susp}}$), 0.52 ng cm^{-2}, was measured in Home 50 after 6 months of deployment. The increase in PFAS concentrations on the cloth strips is mostly due to the accumulation of MeFOSE and EtFOSE. MeFOSE (76% > MDL) and EtFOSE (63% > MDL) dominate $\sum(\text{neutral PFAS})_{\text{susp}}$ concentrations in most homes, with $\sum(\text{neutral PFAS})_{\text{susp}}$ concentrations that increase from 0 to 0.13 ng cm^{-2} after 1 day of exposure to 0.16−0.47 ng cm^{-2} after (on average) 244 days of exposure. In contrast to the air samples, 6:2 FTOH concentrations (1% > MDL) in suspended cloth strips remained below the MDL in most homes even after several months of deployment. We found that 8:2 FTOH, 10:2 FTOH, and EtFOSA were detected above the MDLs of 20%, 34%, and 5%, respectively, in the cloth strips. FTACs and MeFOSA were not detected in any of the strips. The characteristic time for 6:2 FTOH indicates that it takes only a few hours to reach equilibrium between air and cloth (Table S18), but its high vapor pressure (Table S5) suggests that partitioning favors the gas phase.

While FOSEs dominate $\sum(\text{neutral PFAS})_{\text{susp}}$ concentrations on average, this is not true for Home 78. In this unusual case, 8:2 FTOH and 10:2 FTOH concentrations are on the same order of magnitude as MeFOSE and EtFOSE concentrations and the $\sum(\text{neutral PFAS})_{\text{susp}}$ peak after 3 months (0.25 ng cm^{-2} on average). The highest $\sum(\text{neutral PFAS})_{\text{air}}$ and $\sum(\text{neutral PFAS})_{\text{gas}}$ concentrations in Home 78 were also measured at the 3-month time point. MeFOSE and EtFOSE air concentrations are generally lower in Home 78 than in other homes. It was also observed that due to the small size of the closet in Home 78, strips frequently touched other items kept in the closet, some of which may have been FTOH sources.

Equilibration Time of Suspended Cloth Strips. PFAS accumulated in the suspended cloth strips (sampler A) over a period of months, accumulating quickly at first and more slowly at longer deployment times. Figure 3 shows the mean accumulation rates of six neutral PFAS over the 8 month (on average) study period. The average accumulation rate of the FTOHs becomes negative between 1 and 6 days, indicating that equilibrium was reached during that time period. Fluctuations in the accumulation rates afterward (e.g., 6:2 FTOH) indicate that FTOHs react within hours or days to changes in air concentrations and environmental conditions, consistent with the expectation that the characteristic times to equilibrium for 6:2 FTOH, 8:2 FTOH, and 10:2 FTOH are on the order of hours to days (Table S18), based on their estimated K_{oa} values (Table S5). In contrast, mean accumulation rates of the FOSEs approach zero but remain positive until between 6 and 8 months of accumulation. This indicates that the FOSEs in the cloth need, on average, at least 6 months to come to equilibrium with the indoor air, which is in agreement with their lower volatility and higher K_{oa}, resulting in characteristic times (τ) of up to 3 months and 6 months for MeFOSE and EtFOSE, respectively (Table S18).[62] To calculate τ, K_{ca} was approximated by K_{oa}, assuming that the cotton acts as a uniform organic phase.[62] It will be shown in the following section that this approximation is reasonable and that K_{ca} is very close to K_{oa} for the PFAS and cotton investigated in this study.

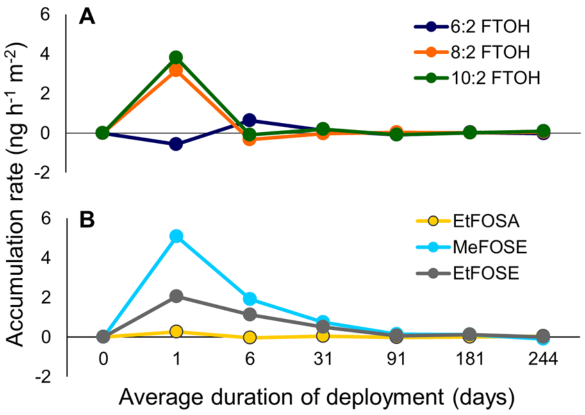

Figure 3. Mean accumulation rates of neutral PFAS in suspended cloth strips (sampler A) based on average concentrations in the cloth strips from the 11 IPA Campaign homes, for (A) 6:2 FTOH, 8:2 FTOH, and 10:2 FTOH and (B) EtFOSA, MeFOSE, and EtFOSE.

Cloth−Air Partition Coefficients. Cloth−air partition coefficients (as mean $\log(K_{\text{ca}})$) range from 4.7 (6:2 FTOH) to 6.7 (MeFOSE) and the partitioning of PFAS from air to cloth increases with increasing $\log(K_{\text{oa}})$ and decreasing vapor pressure (Figure 4; Tables 1 and S19). Concentrations of 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, EtFOSA, MeFOSE, and EtFOSE in paired suspended cloth strips (sampler A) and air samples, collected at t = 3 months and t = 6 months, were used to calculate K_{ca} (see eqs 2 and 3). For t = 3 months, total-air data were used in the calculation of $\log(K_{\text{ca}})$, while for t = 6 months, gas-phase data were used because of data availability. As described earlier, the difference between total-air and gas-phase concentrations is overall relatively small. Note that for 6:2 FTOH and EtFOSA, the majority of concentrations measured in the cloth and used for calculating $\log(K_{\text{ca}})$ are below the MDL.

To the best of our knowledge, these are the first measurements of K_{ca} for PFAS. However, we can use related PFAS partition coefficients to place these results in context. Shoeib et al.[61] measured PUF−air and sorbent-impregnated PUF (SIP)−air partition coefficients for FTOHs and FOSEs. These partition coefficients follow the same trend as the K_{ca} values reported in this study—smaller partition coefficients for the FTOHs and higher, very similar partition coefficients for MeFOSE and EtFOSE.[61] In another study, partitioning of

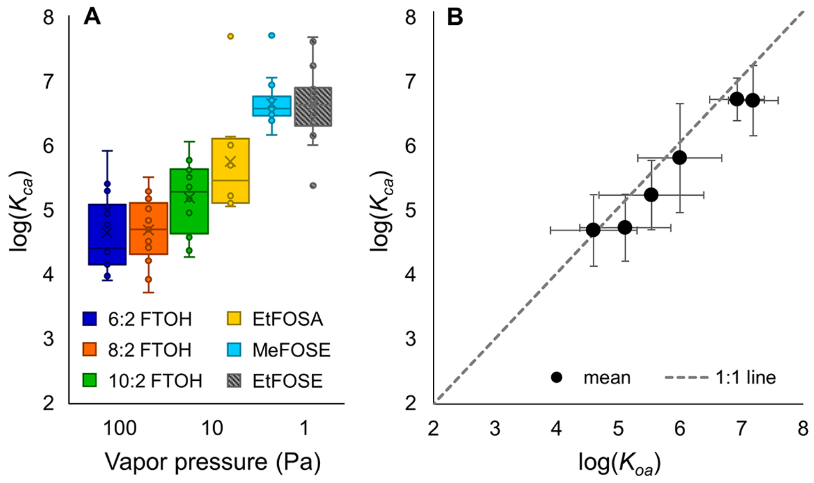

Figure 4. (A) Boxplot showing the log(K_{ca}) of 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, EtFOSA, MeFOSE, and EtFOSE with decreasing vapor pressure. The box indicates the 25th and 75th percentiles. The line inside the box is the median. The × represents the mean, and the whiskers indicate the minimum and maximum. Points outside the range of the whiskers are considered outliers. (B) Log(K_{ca}) based on 3-month and 6-month measurements over log(K_{oa}) derived from measured and predicted literature values (geometric mean and standard deviation, R^2 = 0.98).[28,63−69]

Table 1. Geometric Means and Standard Deviations of log(K_{ca}) for Neutral PFAS, Calculated from Measurements of PFAS in Indoor Air and Cloth

	PFAS					
log(K_{ca})	6:2 FTOH	8:2 FTOH	10:2 FTOH	EtFOSA	MeFOSE	EtFOSE
mean ± std. dev.	4.7 ± 0.6	4.7 ± 0.5	5.2 ± 0.5	5.8 ± 0.8	6.7 ± 0.3	6.6 ± 0.5
median	4.4	4.7	5.3	5.5	6.6	6.6

neutral PFAS to polyethylene (PE) sheets was studied, and the PE−air partition coefficients also followed the same trend.[16] These comparisons, in combination with the relatively small standard deviations, increase the confidence in the K_{ca} values presented here (Table 1).

Figure 4B shows a positive correlation (R^2 = 0.98) between log(K_{ca}) and mean log(K_{oa}) values from the literature (Figure 4B; Table S5).[28,63−69] This is not unreasonable, given that cotton fibers consist predominantly of organic material. Previous studies have shown positive correlations between log(K_{ca}) and log(K_{oa}) for polybrominated diphenyl ethers (PBDEs),[35,37] phthalates, some PCBs,[35,38] and homologous series of VOCs.[54] However, in these cases, log(K_{ca}) was either below the 1:1 line (for PBDEs, phthalates, and PCBs) or above the 1:1 line (for VOCs; Figure S10). Five of the six neutral PFAS log(K_{ca}) values increase with log(K_{oa}); the log(K_{ca}) values of MeFOSE and EtFOSE are nearly identical despite an evident, yet small, difference in log(K_{oa}). The two smallest FTOHs (6:2 FTOH and 8:2 FTOH) also have very similar log(K_{ca}) values; however, the log(K_{ca}) of 6:2 FTOH has larger uncertainties because a larger portion of the concentrations in cloth are below MDL.

Correlations of log(K_{ca}) with the vapor pressure (Figure S11) and temperature (Section S7; Figures S12 and S13) were also examined. For vapor pressure, one major issue is the lack of consistent data for the vapor pressure of neutral PFAS, as shown by the large error bars in Figure S11, especially for the FOSEs. Despite this, the general trend that can be observed is a decrease in log(K_{ca}) with increasing vapor pressure, leading to reduced partitioning of more volatile species to the cotton cloth. A similar inverse relationship between log(K_{ca}) and vapor pressure has also been observed for PDBEs and VOCs.[37,54] Further, we observed an inverse relationship between log(K_{ca}) for all neutral PFAS except EtFOSA and indoor temperature, indicating that these species favor the gas phase over the cloth at elevated temperatures (Section S7; Figures S12 and S13). Although the correlations are modest, they are significant (95% confidence interval) for 6:2 FTOH and MeFOSE, with R^2 values of 0.24 and 0.55 and p values of 0.0152 and 0.0002, respectively. A similar relationship was observed for phthalates by Eftekhari and Morrison.[53] An inverse relationship of log(K_{ca}) with air concentration can also be observed for all species. However, as shown for 6:2 FTOH and MeFOSE in Figure S14, this relationship is likely caused by the temperature-dependence of both log(K_{ca}) and the air concentration, with higher temperatures resulting in lower partition coefficients and higher air concentrations.

Log(K_{ca}) values have been reported for several groups of compounds, including SVOCs such as phthalates, PCBs, and methamphetamine,[34,35,38,51−53] and most recently for VOCs, including carbonyls, carboxylic acids, aromatics, and hydrocarbons.[54] For SVOCs, log(K_{ca}) values for cotton clothing are in the range of 5 to 7.5 and are located below the 1:1 line of log(K_{ca}) and log(K_{oa}); i.e., log(K_{ca}) is usually about 1 order of magnitude lower than log(K_{oa}) (Figure S10). VOCs, on the other hand, appear to have cotton log(K_{ca}) values that are roughly 1 order of magnitude higher than their respective log(K_{oa}) values.[54] The log(K_{ca}) data measured for neutral PFAS included in this study lay close to the 1:1 line between VOCs and SVOCs, with the less volatile species, MeFOSE and EtFOSE, more closely aligned with the SVOCs and the FTOHs following the VOC trends more closely. These findings reinforce the perception of neutral PFAS as a set of chemicals that exists at the intersection of VOCs and SVOCs;

the behavior of neutral PFAS does not fall neatly into either group. This work validates, in part, the use of K_{oa} as a predictor for K_{ca} for neutral PFAS and for use in describing the partitioning of neutral PFAS in air to cotton cloth.

Effect of Cloth Storage Method. The method of storage influences the distribution and concentrations of PFAS in the cloth as well as the accumulation rate. However, regardless of whether cloth is stored in a drawer or on hangers in a closet, measurable accumulation of PFAS can occur in as little as 24 h. Neutral PFAS were detected with similar frequency in the folded cloth pieces (sampler B) compared to the suspended cloth strips (sampler A), and total measured concentrations were typically on the same order of magnitude, with concentrations in the suspended strips being slightly higher, on average (Table S20; Figure S15). However, the FTOH-to-FOSE ratios were typically higher in the folded cloth pieces than in the strips. The FTOH-to-FOSE ratio is particularly high for Homes 01, 35, and 78. This indicates the potential presence of FTOH sources in the drawer near the folded cloth pieces. PFAS concentrations in collocated, concurrently sampled folded cloth pieces were also more variable than the concentrations in duplicate suspended cloth strips. This can be seen by comparing the Home 18 time series for duplicate suspended cloth strips (Figure 2) with that for duplicate folded pieces, as well as for samples from Home 10 (Figure S15). The greater concentration variability for folded cloth may point to the potential presence of PFAS sources in the drawers as well as the potential influence of kinetic factors on transfer to the folded cloth. It may also reflect the reduced influence of airborne PFAS or the variable degree of disturbance as clothing is placed into or removed from the drawer.

Household Clothing Items. Neutral PFAS were found in all household-donated clothing items, showing directly that presumably untreated clothing can serve as a sink and potential mediator of exposure for PFAS (Figure S16, Tables S21 and S22, and section S8). Homes with a higher sum of neutral PFAS in the folded cloth ($\sum(\text{neutral PFAS})_{\text{folded}}$) typically also have a higher sum of PFAS in the household-donated clothing ($\sum(\text{neutral PFAS})_{\text{clothing}}$; e.g., Homes 01, 18, and 82 compared to Home 78). Interestingly, FTOHs were more frequently detected and at higher concentrations (mass-normalized) in the household-donated clothing items ($\sum(\text{FTOH})_{\text{clothing}}$) than in the 3- or 6-months folded cloth pieces. In many samples, $\sum(\text{FTOH})_{\text{clothing}}$ concentrations contribute more than 50% to $\sum(\text{neutral PFAS})_{\text{clothing}}$ concentrations. On average, $\sum(\text{neutral PFAS})_{\text{clothing}}$ is higher than the 6-month $\sum(\text{neutral PFAS})_{\text{folded}}$ concentrations (in ng g^{-1} of cloth), although they are on the same order of magnitude. $\sum(\text{neutral PFAS})_{\text{clothing}}$ range from 2.3 ng g^{-1} (Home 78) to 34 ng g^{-1} (Home 65). Concentrations of the three FTOHs, MeFOSE, and EtFOSE are greater than the MDL in 70% or more of the samples. EtFOSA is above MDL in about one-fifth of the samples (18% > MDL). The FTACs and MeFOSA were not detected. Overall, these findings suggest that the folded cloth pieces serve as a reasonable proxy for clothing stored in homes; however, they may have not been the most suitable fabric to assess the full range of neutral PFAS that accumulate in household clothing. More volatile species (i.e., 6:2 and 8:2 FTOH) might have been less likely to accumulate in the cloth used in the field campaign because the fabric was thinner with a smaller available surface area, and it was selected to be free of surface treatments that may enhance partitioning of volatiles to the cloth.

Factors Influencing PFAS Concentrations in Air. We expect PFAS concentrations across homes to vary because of differences in furnishings, building materials, home age (and other factors affecting mean home air change rates), and activities that occur very regularly (i.e., daily). We additionally expect within-home variability in PFAS concentrations as a result of changes in occupant activities related to sources (e.g., cooking), indoor conditions (e.g., temperature), and within-home variations in air change rates, such as those that result from window opening, indoor−outdoor temperature differences, or variations in wind speed. Thus, we calculated the within-home variability (WHV) and across-home variability (AHV) in PFAS air concentrations and investigated correlations between air concentrations and building characteristics, environmental factors, air change rates, and window opening behavior in order to better understand the drivers of PFAS concentrations in indoor air.

For all detected compounds except MeFOSE, the WHVs (74−334%) were greater than the respective AHVs (67−311%) for PFAS air concentrations, as shown in Table S23. For example, the WHV (150%) was nearly twice the AHV (84%) for 8:2 FTOH. This suggests a significant role for the influence of occupant activities or environmental conditions in the emission, partitioning, or dilution of these PFAS compounds. In contrast, MeFOSE was more variable across homes (138%) than within homes (37%). This suggests that MeFOSE is likely associated with a different, more permanent type of source, such as building materials or furnishings.

To investigate these relationships further, we present correlations between PFAS air concentrations and building characteristics, window opening behavior, occupancy, and environmental conditions in Tables S24 and S25. $\sum(\text{neutral PFAS})_{\text{air}}$ and $\sum(\text{neutral PFAS})_{\text{gas}}$ are moderately positively linearly correlated with average indoor temperature ($R^2 = 0.3$ and 0.4, respectively, Figure S17) and also with the outdoor temperature and inversely correlated (moderate) with indoor−outdoor temperature difference (Tables S24 and S25). The correlation of PFAS with indoor temperature may be due to a shift of PFAS partitioning toward the gas phase (Figures S12, S13, and S14) as well as reflect the increased emission (or partitioning) of neutral PFAS from indoor sources or reservoirs to the air at higher temperatures, as demonstrated by Liu et al.[42] for FTOHs in chamber experiments. Similar behavior has been shown in several studies for SVOCs[70,71] and VOCs.[72] Home age is inversely correlated (moderately) to the air concentration of MeFOSE. Together with the stronger AHV for MeFOSE, this finding suggests that newer home construction or newer furnishings are associated with sources of MeFOSE. In contrast, 8:2 FTAC in air is positively correlated with home age, associating 8:2 FTAC more strongly with older homes. The type of flooring is correlated (weak-to-moderate) with 6:2 FTOH. Specifically, houses with laminate in the main living area were more likely to have higher 6:2 FTOH air concentrations. Note that rugs were present in all homes except Home 18, which had a carpeted section. For this reason, rugs were excluded from the correlations. EtFOSA in air is moderately correlated with the air change rate, but we did not observe any correlation with the window opening behavior. Overall, these findings indicate that for most neutral PFAS, occupant activities and indoor conditions, especially changes in temperature (Figure S17), are more important factors influencing variations in total neutral PFAS concentrations in air than building materials and furnishings.

Implications for Human Exposure. Concentrations of several neutral PFAS measured in indoor air in this study are in the range of typical SVOCs like di(2-ethylhexyl)adipate (DEHA), di(2-ethylhexyl) phthalate (DEHP), butylbenzyl phthalate (BBzP), and tris(chloropropyl)phosphate (TCPP).[62,73,74] Because of the ubiquitous presence and relatively high concentrations of neutral PFAS in indoor air, consideration of inhalation exposure as an important pathway of exposure of the general U.S. population is warranted, especially for FTOHs. Biotransformation of neutral PFAS compounds (e.g., FTOHs and FOSEs) to potentially more toxic, persistent, or bioaccumulative intermediates (e.g., 5:3 fluorotelomer carboxylic acid) or products (e.g., PFOA and PFOS) in the human body increases the toxicological relevance of the neutral PFAS inhalation pathway, especially in locations where drinking water sources are not highly contaminated.[1,20,29,30,32,75,76]

This study shows that neutral PFAS accumulate in initially PFAS-free clothing. Thus, clothing as a secondary PFAS source could potentially facilitate dermal PFAS exposures and prolong indoor inhalation exposures. Generally, dermal absorption of PFAS has been considered a minor route of exposure compared to other pathways, i.e., dietary intake and drinking water, because dermal permeability has been thought to be low for PFAS.[1,24,48,77,78] However, a recent study found that direct transdermal absorption of PFAS from air may be of greater relevance than inhalation exposure for multiple neutral PFAS.[79] Based on the estimates of the dermal uptake-to-inhalation (D/I) ratio reported by Kissel et al.[79] and the criteria established by Weschler and Nazaroff,[80] dermal absorption is not expected to exceed inhalation exposure but is expected to contribute non-negligibly to MeFOSE and EtFOSE uptake. Such dermal absorption from air can be mediated by PFAS sorbed to clothing. These findings emphasize the importance of including dermal PFAS exposure in exposure assessments. The cloth−air partition coefficients reported here and their relationship to physicochemical properties and temperature can be used in such exposure models to incorporate the influence of clothing.

Limitations. The IPA Campaign measured PFAS in multiple indoor compartments in 11 homes over 9 months. To the best of our knowledge, this is the first study to report concurrent measurements of neutral PFAS in air and cloth across multiple homes in the U.S. It is also the first study to report cloth−air partition coefficients for PFAS. However, the study has several limitations. The 11 single-family homes constituted a convenience sample, and the homes studied were located in a relatively small area in North Carolina. Additional measurements in more types and ages of homes and different climactic regions, with occupants spanning across a broader socioeconomic range, would provide a more complete understanding of the factors that influence residential PFAS exposures. Paired measurements of indoor air and cloth were not conducted in the same room, thus, introducing larger uncertainty in the cloth−air partition coefficients. We further assumed that the air concentrations of the target species are lower outdoors than indoors. We are confident this is true because we "cleaned" all cloth strips and pieces by hanging them outside for 3−4 months, and we found that concentrations of target species in the field blanks and $t = 0$ samples (essentially outdoor samples) were low compared to cloth exposed to indoor air, particularly for MeFOSE and EtFOSE. The literature also supports this assumption. For example, Shoeib et al.[81] reported 2 orders of magnitude lower concentrations of MeFOSE and EtFOSE in outdoor air compared to indoor air, and Ahrens et al.[82] reported FTOH and FOSE concentrations in the low pg m^{-3} range in the outdoor air near Toronto, Canada. Also, this study measured nine neutral PFAS. However, thousands of PFAS exist,[7] and the indoor concentrations of most are unknown. Although detection limits still pose challenges, total organic fluorine measurements[83] and analytical advancements such as real-time chemical ionization mass spectrometry (CIMS)[84,85] in combination with integrated measurements of ionic PFAS may prove useful in approaching mass closure. Overall, there is great potential for future research to close knowledge gaps regarding PFAS emissions, partitioning, and exposure indoors.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.3c04770.

> Overview of collected samples; sampling schedules; surveys and checklist; home characteristics; window opening; PFAS analytes and mass-labeled standards; additional sampling, sample processing, and sample analysis details; quality assurance and quality control measures and results; GC-MS parameters; across-home and within-home variabilities; concentration profiles of neutral PFAS in air and cloth; statistical analysis details and results; characteristic times and measured equilibration times; cloth−air partitioning coefficients; comparison and temperature-dependency of log(K_{ca}); details regarding household clothing items; temperature dependency of air concentrations; and Spearman rank correlation coefficients (PDF)

AUTHOR INFORMATION

Corresponding Author

Barbara J. Turpin − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States;* orcid.org/0000-0003-4513-4187; Email: bjturpin@email.unc.edu

Authors

Clara M. A. Eichler − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States;* orcid.org/0000-0002-1282-3608

Naomi Y. Chang − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States*

Elaine A. Cohen Hubal − *U.S. EPA, Center for Public Health and Environmental Assessment, Research Triangle Park, North Carolina 27711, United States;* orcid.org/0000-0002-9650-3483

Daniel E. Amparo − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States*

Jiaqi Zhou − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of*

Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States

Jason D. Surratt − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States; University of North Carolina at Chapel Hill, College of Arts and Sciences, Department of Chemistry, Chapel Hill, North Carolina 27599-3290, United States;* orcid.org/0000-0002-6833-1450

Glenn C. Morrison − *University of North Carolina at Chapel Hill, Gillings School of Global Public Health, Department of Environmental Sciences and Engineering, Chapel Hill, North Carolina 27599-7400, United States;* orcid.org/0000-0001-6876-7185

Complete contact information is available at:
https://pubs.acs.org/10.1021/acs.est.3c04770

Notes

The authors declare no competing financial interest.

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