

The Air That We Breathe: Neutral and Volatile PFAS in Indoor Air

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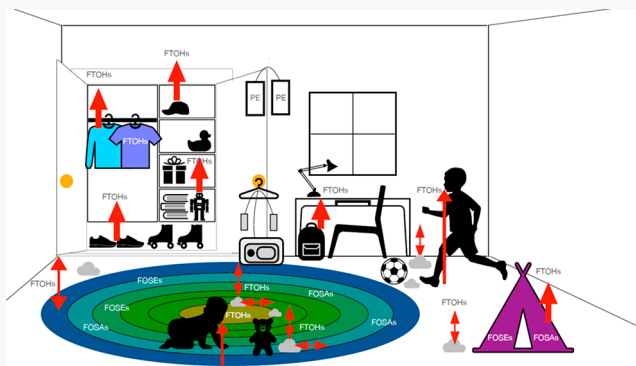


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Supporting Information

ABSTRACT: Sources of exposure to per- and polyfluorinated alkyl substances (PFAS) include food, water, and, given that humans spend typically 90% of their time indoors, air and dust. Quantifying PFAS that are prevalent indoors, such as neutral, volatile PFAS, and estimating their exposure risk to humans are thus important. To accurately measure these compounds indoors, polyethylene (PE) sheets were employed and validated as passive detection tools and analyzed by gas chromatography–mass spectrometry. Air concentrations were compared to dust and carpet concentrations reported elsewhere. Partitioning between PE sheets of different thicknesses suggested that interactions of the PEs with the compounds are occurring by absorption. Volatile PFAS, specifically fluorotelomer alcohols (FTOHs), were ubiquitous in indoor environments. For example, in carpeted Californian kindergarten classrooms, 6:2 FTOH dominated with concentrations ranging from 9 to 600 ng m⁻³, followed by 8:2 FTOH. Concentrations of volatile PFAS from air, carpet, and dust were closely related to each other, indicating that carpets and dust are major sources of FTOHs in air. Nonetheless, air posed the largest exposure risk of FTOHs and biotransformed perfluorinated alkyl acids (PFAA) in young children. This research highlights inhalation of indoor air as an important exposure pathway and the need for further reduction of precursors to PFAA.



INTRODUCTION

Human exposure to fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamides (FOSAs), perfluorooctane sulfonamidoethanols (FOSEs), and other precursors to perfluorooalkyl acids (PFAA) comes primarily from consumer and industrial products readily available in people's homes.^{1–4} FTOHs were the dominant polyfluorinated compounds in indoor air,⁵ where ~60% of detected per- and polyfluorinated alkyl substances (PFAS) were associated with the particle phase.² Because most people spend more than 90% of their time indoors,² indoor air and dust are important uptake pathways for human PFAS exposure⁶ in addition to the widely recognized exposure sources of diet and water.^{7–9} Indeed, correlations between increased indoor air exposure to precursors and increased PFAS serum concentrations have been reported.^{10,11}

The use of passive sampling, which can measure the concentration of freely dissolved or gas-phase trace organic contaminants, has been widely accepted as an effective detection tool.^{12,13} Single-phase polymers, such as polyethylene (PE) sheets, have been able to detect a wide range of nonpolar and moderately polar contaminants in the gas phase or dissolved in water.^{14,15} In addition, PE sheets are inexpensive, are easy to handle, and can be easily transported and deployed.¹³ Recently, neutral PFAS were successfully

measured in outdoor air and water using PE sheets.¹⁶ However, the partitioning of neutral PFAS into or onto the PE sheets indoors is not yet fully understood.

To further assess the role that indoor environments play as an exposure source of airborne PFAS in the gas phase and dust, the main objectives of this research were (i) to derive indoor PE–air partitioning coefficients (K_{PE-air}), (ii) to compare the volatile PFAS composition in different indoor environments using PE sheets as passive samplers, (iii) to evaluate the air–dust partitioning of PFAS in carpeted kindergarten classrooms, and (iv) to estimate daily intake (EDI) in 2–6-year-old children.

MATERIALS AND METHODS

Sampling of neutral PFAS was performed in carpeted kindergarten classrooms, residences, an outdoor gear and apparel store in northern California, university offices,

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classrooms, laboratories, and a carpet store in southern Rhode Island between 2018 and 2020. A total of 90 PE sheets were deployed in the indoor locations, in addition to eight radiello samplers (Sigma-Aldrich) with precleaned XAD-4 as the sorbent used for active sampling.

Two types of precleaned PE passive samplers differentiated by thickness (25 and 50 μm) were deployed for 14 days (validation study), 21 days (kinetic study), and 28 days (measurements). Active sampling was performed on days 1, 7, and 14 where the radiello samplers were attached to a QuickTake 30 SKC pump at a constant flow of 5 L min^{-1} for 240 min. All samples were kept in a freezer at $-20\text{ }^{\circ}\text{C}$ until extraction (for details, see the [Supporting Information](#)).

Instrumental Analysis. Samples were analyzed for nine neutral PFAS on an Agilent 7890B gas chromatograph coupled to an Agilent 5977A mass selective detector (MSD) device operating in positive chemical ionization mode using selected ion monitoring (for details, see the [Supporting Information](#)).

Interpretation of Data. The partitioning constants of neutral PFAS between PE and air ($K_{\text{PE-air}}$) were derived in the validation study as

$$K_{\text{PE-air}} = \frac{C_{\text{PE}}}{C_{\text{air}}} \quad (1)$$

where C_{PE} is the concentration in PE sheets (nanograms per gram of PE) and C_{air} is the gas-phase concentration (nanograms per cubic meter).

Active sampling was used in the $K_{\text{PE-air}}$ validation study only. For all other campaigns, C_{air} was calculated using [eq 1](#). Partitioning within the PE sheets was derived as the ratio of the 25 μm passive sampler (C_{25}) to the 50 μm passive sampler (C_{50}) amounts at equilibrium (for details, see the [Supporting Information](#)).

Daily Intake. The total estimated daily intake (EDI) of neutral PFAS via air and dust was calculated from PFAS concentrations measured here and dust concentrations reported elsewhere¹⁷ based on established methods^{18,19} (for details, see the [Supporting Information](#)).

QA/QC. Field blanks, matrix spikes, matrix blanks, and field duplicate samples were included with each sample batch. Matrix spikes were prepared by spiking 50 μL of an 80 $\text{pg}/\mu\text{L}$ native standard solution and 50 μL of an 80 $\text{pg}/\mu\text{L}$ mass-labeled standard solution into a clean (unused and never removed from the laboratory) sampler. Method detection limits (MDLs) were calculated as the blank average plus 3 times the standard deviation; however, when a compound was not detected in the blanks, instrumental limits of detection (ILODs) were used. Only values above limits of quantitation (LOQ) were reported (for details, see [Table S2](#)). Recoveries of the matrix spikes ranged from 81% (± 35) to 111% (± 19) for all compounds (for details, see [Table S2](#)).

RESULTS AND DISCUSSION

PE–Air Partitioning Constants. The results from the kinetic study showed that 6:2 FTOH and 8:2 FTOH reached equilibrium after 14 days (see [Figure S1](#)). Log $K_{\text{PE-air}}$ values were approximately 4–5 for the FTOHs and ~ 5 for 8:2 FTAc and increased with molecular weight ([Table 1](#)). Although 10:2 FTAc, FOSAs, and FOSEs were detected by PE sheets, none were detected by active sampling; calculating their equilibrium partitioning constant was not possible (see [Table S6](#)). There were only minor differences between the 25 and 50 μm $K_{\text{PE-air}}$

Table 1. Indoor Log $K_{\text{PE-air}}$ Values from the Validation Study for 25 and 50 μm PE Sheets

compound	molecular weight (g mol^{-1})	mean log $K_{\text{PE-air}}$ 25 (this study)	mean log $K_{\text{PE-air}}$ 50 (this study)
6:2 FTOH	364.1	4.4 ± 0.1	4.3 ± 0.0
8:2 FTOH	464.1	4.3 ± 0.1	4.5 ± 0.0
10:2 FTOH	564.1	5.0	5.0 ± 0.0
8:2 FTAc	518.1	4.9 ± 0.4	5.0 ± 0.2
10:2 FTAc	618.1	5.2^a	5.3^a
MeFOSE	527.2	5.1^a	5.2^a
EtFOSE	513.1	ND ^b	ND ^b
MeFOSE	571.2	5.2^a	5.3^a
EtFOSE	557.2	5.2^a	5.2^a

^a $K_{\text{PE-air}}$ values from this study were estimated on the basis of a correlation between those measured here and those reported by Dixon-Anderson and Lohmann.¹⁶ Estimated $K_{\text{PE-air}} 50 = 0.44K_{\text{PE-air,measured}}$ (Dixon-Anderson and Lohmann, 2018) + 1.30 ($\text{RSQ}_{\text{PE50}} = 0.67$). ^bNot detected.

results, indicating good reproducibility of PE sheets as passive samplers.

Mean log $K_{\text{PE-air}}$ values from the FTOHs of this study were approximately 3 log units lower than those reported for outdoors (see [Table S3](#)),¹⁶ where break-through and environmental factors could have affected the partitioning of the compounds. Missing $K_{\text{PE-air}}$ values were derived on the basis of a correlation between previously reported¹⁶ and currently measured PE–air partitioning constants. Further studies are needed to corroborate the partitioning coefficients of the FOSAs and FOSEs.

PE–Air Partitioning Ratios. The partitioning ratios of the weight-normalized neutral PFAS between 25 and 50 μm thick PE sheets were ~ 1 (see [Figure S2](#)), implying absorption as the mechanism of partitioning. The greater mass of the 50 μm PE sheets for the same size resulted in easier detection and is thus preferable for future studies.

Neutral PFAS Indoor Air Concentrations. Indoor air concentrations were derived from the PEs for neutral PFAS in (1) California kindergarten classrooms ([Table S4](#)), (2) offices, classrooms, and laboratories at a university and a nearby carpet store in southern Rhode Island ([Table S5](#)), and (3) a storage room at an outdoor clothing store in California ([Table S6](#)) (see [Figure 1](#)). Neutral PFAS were present at all locations, dominated by FTOHs, which is consistent with previous results.^{5,20,21} PFAS profiles and concentrations varied between locations, though, likely driven by the different PFAS-containing products present. These results indicated that PE sheets can be used to determine differences in PFAS profiles and concentrations in various indoor air settings.

In the California kindergarten classrooms, 6:2 FTOH dominated with concentrations ranging from 10 to 600 ng m^{-3} (accounting for 29–96% of the sum of nine PFAS), followed by 8:2 FTOH (2–160 ng m^{-3} , 3–54% of total PFAS) ([Figure 1](#) and [Table S2](#)). In three kindergarten classrooms (KG2, KG3, and KG5), concentrations of 8:2 FTOH exceeded those of 6:2 FTOH. In all kindergarten classrooms, EtFOSE was present at low concentrations while MeFOSE was below method detection limits (MDLs) ([Figure 1](#) and [Table S2](#)). EtFOSE, 8:2 FTAc, and 10:2 FTAc were not detected ([Table S3](#)).

When detected, 6:2 FTOH (with a detection frequency of 83% and ranging from <MDL to 1900 ng m^{-3}) and 8:2 FTOH (17%, <MDL–270 ng m^{-3}) also dominated total PFAS in the

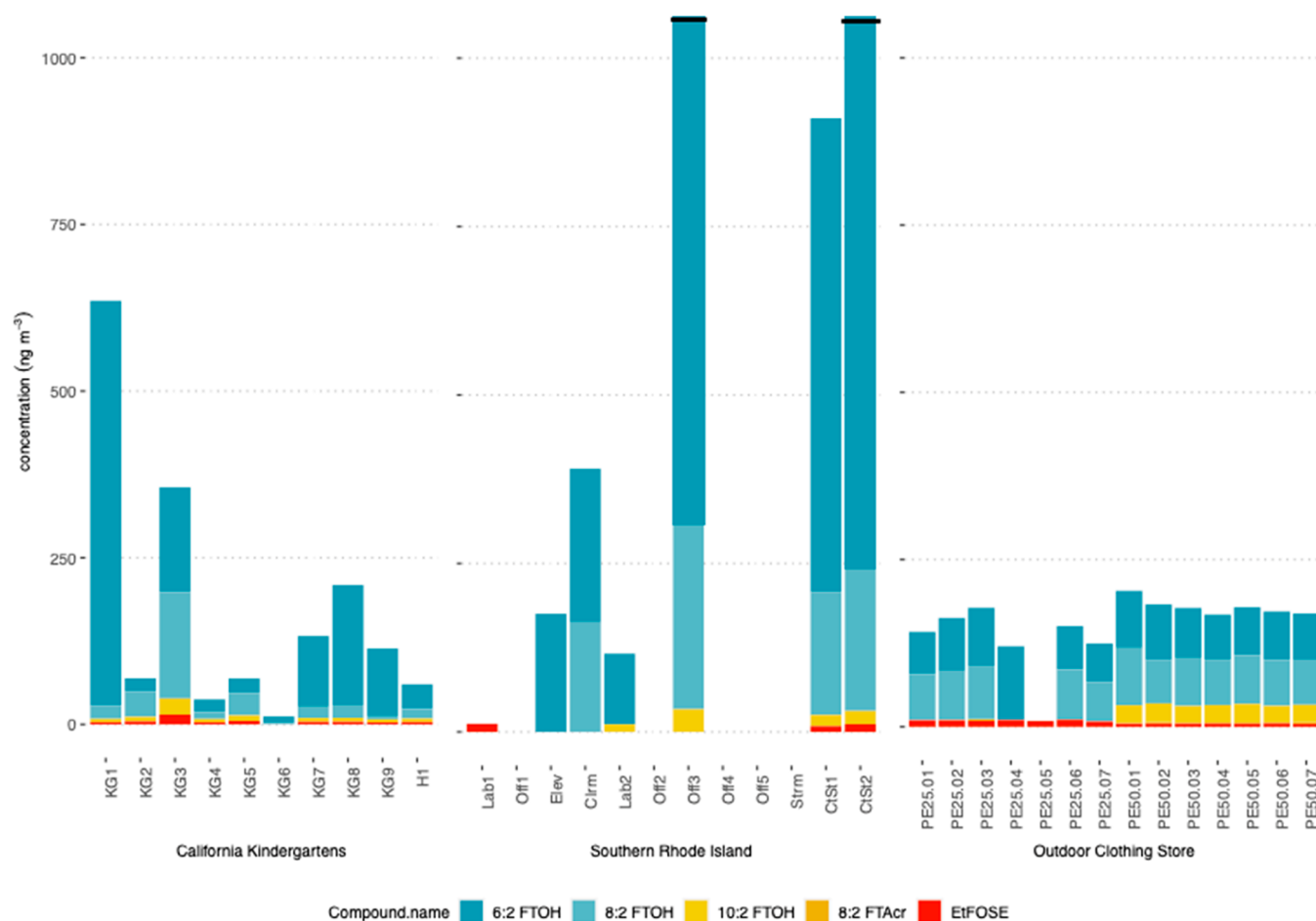


Figure 1. Indoor air concentrations measured in California kindergarten classrooms, an outdoor clothing store, university classrooms, offices and laboratories, and a carpet store in southern Rhode Island. Abbreviations: H, home; KG, kindergarten classrooms; Lab, laboratory; Off, office; Elev, elevator; Clrm, classroom; Ctst, carpet store; Strm, storage room. Numbers (i.e., KG7) are indicative of separate/individual samples. Off3 (2220) and CtSt2 (1040) have concentrations of $>1000 \text{ ng m}^{-3}$.

university rooms (Figure 1 and Table S5). FTOHs were detected only in carpeted rooms and in the analytical laboratory (Table S5). The detection of 10:2 FTOH was sporadic, with concentrations of $\leq 33 \text{ ng m}^{-3}$ (Figure 1 and Table S5). MeFOSE, EtFOSE, and MeFOSE were at or below MDL at all sites, while EtFOSE and FTAcr were rarely above MDLs (Table S5).

Volatile PFAS were present in all replicates from the outdoor clothing store (Figure 1). FTOHs were the most abundant and dominant group, consistent with previous studies of the composition of PFAS in various indoor environments.^{5,20,21} The most abundant compound was 8:2 FTOH, with an average concentration of $\sim 200 \text{ ng m}^{-3}$, followed by 6:2 FTOH and 10:2 FTOH with average concentrations of 70 and 30 ng m^{-3} , respectively (Figure 1). The dominance of 8:2 FTOH is concerning because this and other longer-chain PFAS have been phased out by PFAS producers in the United States, European Union, and Japan.²² These results show that these compounds are still being used for textiles and possibly point to the imports of textiles from other countries where PFAS are poorly regulated.²³

Fraser et al.¹¹ reported concentrations of FTOHs ranging from $<\text{MDL}$ to 11 ng m^{-3} (6:2 FTOH), $0.3\text{--}70 \text{ ng m}^{-3}$ (8:2 FTOH), and $0.14\text{--}12 \text{ ng m}^{-3}$ (10:2 FTOH) in multiple office spaces in Boston, MA, similar to results reported here and in

other studies.^{5,21} A study in Ottawa, ON, in 2005 reported concentrations of MeFOSE, EtFOSE, and EtFOSE in indoor air of ~ 7 , 2, and 0.1 ng m^{-3} , respectively,² and even lower in Vancouver, BC, in 2011, at 0.4, 0.06, 0.02 and 0.03 (MeFOSE) ng m^{-3} , respectively.²¹ In the study presented here, although present in many locations, FOSEs rarely exceeded concentrations of 0.001 ng m^{-3} . FOSEs were detected even fewer times. The difference in the concentrations of the FOSEs and FTOHs in different locations across North America could reflect geographic differences of indoor sources. Additionally, the difference between older and newer data could point to the phase out of PFOS-based chemicals, including FOSEs and FTOHs since 2002, whereas the use of replacement FTOHs in North America has increased since 2000.^{21,24,25}

Air–Dust–Carpet Partitioning. Concentrations of neutral PFAS in the dust and carpet of the same kindergarten classrooms were measured by Wu et al.¹⁷ (see Table S7). Strong correlations ($\text{RSQ} > 0.7$; $P \leq 0.05$) were observed between different FTOHs in air and dust, and air and carpet [and dust and carpet from Wu et al.¹⁷ (see Table S8)], except for 6:2 FTOH in air and carpet. On the contrary, FOSEs were not strongly correlated in air, dust, or carpet.

The distributions of PFAS between indoor air and floor dust were reported to be controlled by partitioning between the gas phase and PFASs sorbed to the organic phases in the dust.²⁶

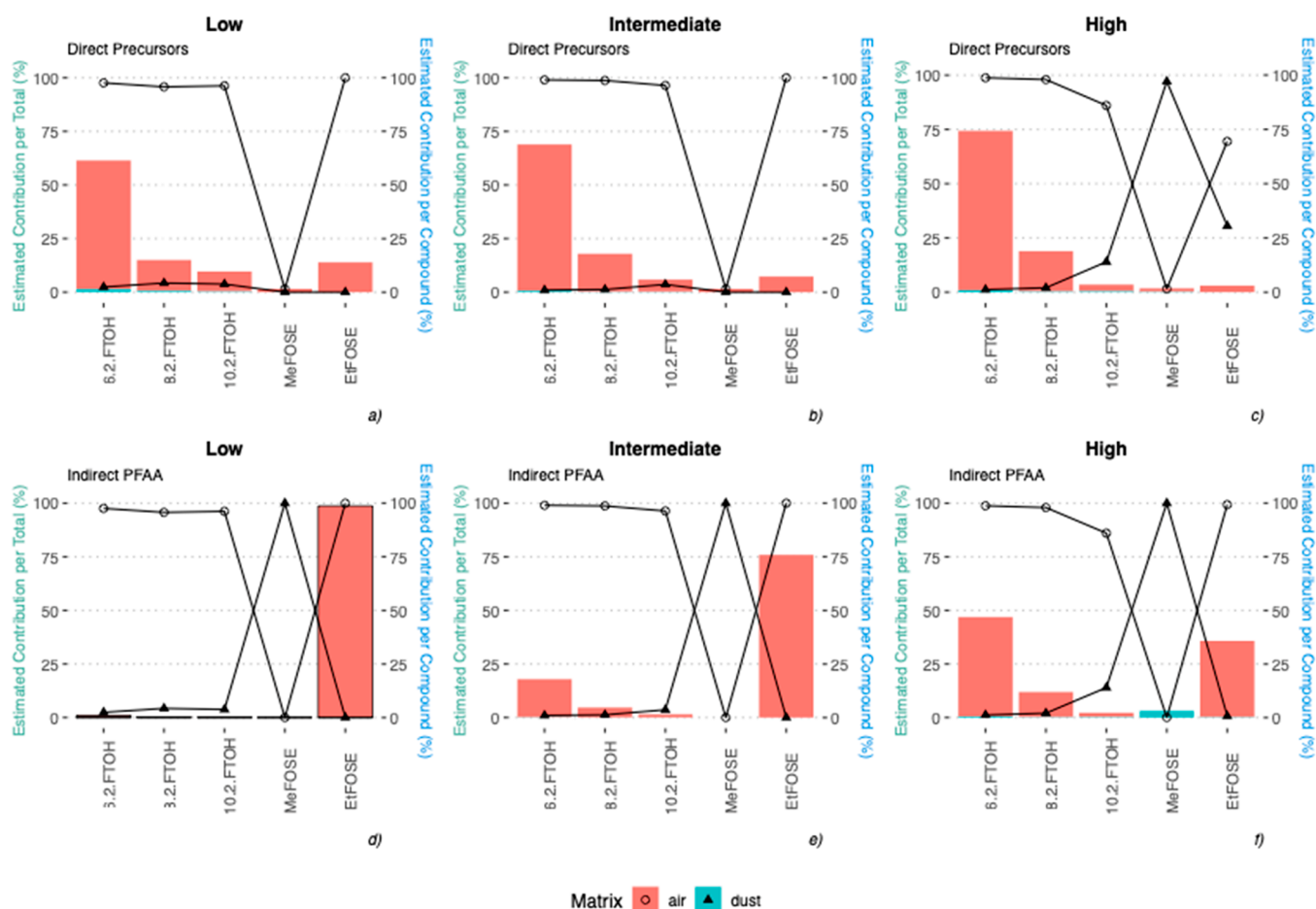


Figure 2. Percent of volatile and neutral PFAS (top panels) and indirect or biotransformed PFAA (bottom panels) intake via air inhalation (pink) and dust ingestion (blue) for 2–6-year-old children. Bars represent the relative contribution of individual precursors to total PFAS (left axes); bars are differentiated by color for both matrices. Lines represent the percent estimated contribution for each compound in air and dust (right axes). MeFOSE was detected at low concentrations in dust and <MDL in air.

Our results corroborated that neutral PFAS were present in air and partitioned to dust. Given that the origin of volatile PFAS in air in the (carpet-free) outdoor clothing storage room was likely to be textiles, it is possible that multiple products in the kindergarten classrooms were in fact the source of these compounds that also partitioned into carpet and dust. Previous work demonstrated that FTOHs, FOSAs, and FOSEs degrade in the atmosphere into more stable PFAA.^{27,28} Significant associations between precursors in air and PFCAs in dust have been reported (e.g., 6:2 FTOH and PFHxA).²⁹ Similarly, significant associations were observed between FOSAs and FOSEs in air and PFOS and PFDS in house dust.²⁸

In contrast to FTOHs, there were no significant correlations between the FOSAs and FOSEs in air, dust, and carpet from this study, suggesting that the sources of FOSAs and FOSEs were different and likely not linked to carpets or textiles. A previous study did not find significant correlations between the FOSEs in kindergarten classrooms either, but did find strong associations in offices,³⁰ implying that there were common sources of these sulfonamidoethanols in items associated with office spaces that perhaps were not usually found in kindergarten classrooms. Additionally, as previously mentioned, the production of FOSAs and FOSEs has been largely phased out of production since 2002,²⁴ and thus, their low concentration or absence is expected.³¹

Estimated Daily Intake of Volatile PFAS through Air and Dust. To assess the relevance of volatile and neutral PFAS in indoor air for 2–6-year-old children, the estimated daily intake (EDI) was calculated (Table S8) for three exposure estimates [low, medium, and high (see the Supporting Information)]. Biotransformation constants for each compound were used to calculate their contribution to Σ PFAA intake (Table S8).

The total EDI (Table S9) was 1.5 ng (kg of body weight)^{−1} day^{−1} for low exposure, 14 ng (kg of body weight)^{−1} day^{−1} for intermediate exposure, and 150 ng (kg of body weight)^{−1} day^{−1} for high exposure. Compounds that were regularly detected in both air and dust were 6:2 FTOH, 8:2 FTOH, and, to a lesser extent, 10:2 FTOH, while MeFOSE and EtFOSE appeared to have significantly larger contributions in dust than air (Figure 2 and Table S9). Volatile and neutral PFAS measured in air contributed 4.9–62% to Σ PFAA intake, while ionic PFAS measured in dust contributed 34–95% (Table S9). These results are similar to other studies that found precursors contributing 41–68% to Σ PFOS uptake via all investigated exposure pathways,³² and precursors responsible for 90% of the Σ PFOS intake in air (Figure 2).^{26,33} Our results imply that air inhalation was a major exposure pathway for FTOHs, while dust ingestion was dominant for FOSEs in children, similar to prior results.³⁰

Given the potential for precursors to be biotransformed into more stable PFAA, estimations of PFAA indirect exposure were also calculated as 1.2, 75, and 2800 ng (kg of body weight)⁻¹ day⁻¹ for the low-, intermediate-, and high-exposure scenarios, respectively (Table S9). The major contributors to indirect PFAA exposure were 6:2 FTOH and 8:2 FTOH in air and MeFOSE in dust (Figure 2). This study demonstrated that volatile neutral PFAS, such as FTOHs, are major contributors to exposure in air.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.1c00481>.

Additional details about the analysis, data interpretation, and EDI calculation (PDF)

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Notes

The authors declare no competing financial interest.

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