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Perfluorinated Compounds in House Dust from Ohio and North Carolina, USA

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The perfluoroalkyl acids (PFAAs), including perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS), have come under increasing scrutiny due to their persistence, global distribution, and toxicity. Given that their human exposure routes remain poorly characterized, the potential role of house dust needs to be more completely evaluated. In this study, new methods for the analysis of 10 PFAAs and three fluorinated telomer alcohols (FTOHs) were developed for dust samples collected from homes ($n = 102$) and day care centers ($n = 10$) in Ohio and North Carolina in 2000–2001. FTOHs were measured by GC/MS and PFAAs were analyzed by LC-MS/MS. PFOS and PFOA were the most prominent compounds detected, occurring in over 95% of the samples at median concentrations of 201 and 142 ng/g of dust, respectively. Maximal concentrations of PFOS were 12 100 ng/g (95th percentile, 2240 ng/g), PFOA 1960 ng/g (95th percentile, 1200 ng/g), and perfluorohexanesulfonate (PFHS) 35 700 ng/g (95th percentile, 2300 ng/g). The 8:2 FTOH, which is volatile and can degrade to PFOA, had a maximum concentration of 1660 ng/g dust (95th percentile, 669 ng/g). These results indicate that perfluorinated compounds are present in house dust at levels that may represent an important pathway for human exposure.

Introduction

Perfluorinated compounds (PFCs) have been used in a growing variety of consumer and industrial products over the past 50 years (*1*). These compounds have many properties that make them useful in a range of applications, including resistance to degradation, thermal stability, and various surfactant properties. As a result of their widespread use and extraordinary persistence, these chemicals have been found to be globally distributed in a range of environmental and biological media (*2–5*). Unfortunately, comparatively little is known about the toxicity of most of these chemicals, with much of the published literature only focusing on PFOS, PFOA, and a small number of structurally related PFCs (*6–8*). The distribution and toxicity of many of the labile precursors and longer chain length PFCs (> C8) largely remain unknown.

The PFCs have been used in a large number of products that are found primarily in residential and indoor environments. One of the principal uses has been for stain-resistant coatings used in the carpet, upholstery, and textile industries (*1*). Other applications, such as in paper coatings, cosmetics, and insecticides are also primarily present in indoor residential environments. Given the stability of some of the PFCs, one might hypothesize that as these various consumer-use products wear and degrade, the applied PFC materials may slough off and accumulate in the dust and debris that remain. Moreover, one might further hypothesize that the residual volatile PFCs that are found in some products (*9*)] could volatilize and then become adsorbed onto dust within the home. Previous studies have reported that house dust is a repository for pesticides and polycyclic aromatic hydrocarbons (PAHs), representing a significant potential source of exposure to these compounds in the residential environment (*10–12*). Evidence is now mounting that this may also be the case for the PFCs. A recent study from Japan reported mean concentrations of PFOS and PFOA at 200 ng/g and 380 ng/g, respectively, in 16 house dust samples (*13*). Another recent study of house dust from Ottawa Canada ($n = 67$) measured mean concentrations of PFOS, PFOA, and perfluorohexane sulfonate (PFHS) of 444, 106, and 392 ng/g, respectively (*14*). An additional Canadian study involving volatile PFCs in house dust found mean concentrations of *n*-methylperfluorooctane sulfonamidoethanol (MeFOSE) and *n*-ethylperfluorooctane sulfonamidoethanol (EtFOSE) of 412 and 2200 ng/g, respectively (*15*). On a ng/g basis, the PFOS in dust concentrations are comparable to the mean levels that have been reported in the livers of top fish, bird, and mammal predators which are known to bioaccumulate PFOS (*2, 16, 17*). PFOA and the other PFCs are seldom reported in any matrix at levels that are comparable to what has been reported for house dust. As such, house dust may be one of the most significant potential pathways for human exposure to the PFCs.

In this study, new methods for the measurement of perfluoroalkyl acids (PFAAs, including both carboxylates and sulfonates), and fluorotelomer alcohols (FTOHs) were applied to dust samples collected from U.S. homes and daycare centers in Ohio and North Carolina. The results and implications for human exposure are evaluated and discussed below.

Materials and Methods

Chemicals and Standards. 2-(perfluorohexyl)ethanol (6:2 FTOH), 2-(perfluorooctyl)ethanol (8:2 FTOH), 3-(perfluorooctyl)propanol (FTOH internal standard, I.S.), 2-(perfluorodecyl)ethanol (10–2 FTOH), perfluoroundecanoic acid (PFUNA), and perfluorododecanoic acid (PFDoA) were purchased from Oakwood Products, Inc. (West Columbia, SC). Perfluorohexanoic acid (PFHxA) was obtained from Fluka Chemical Corp.(Milwaukee, WI). Perfluoroheptanoic (PFHpA), perfluorooctanoic acids (PFOA), perfluorononanoic acid (PFNA), and perfluorodecanoic acid (PFDA) were purchased from Aldrich Chemical Corp. Inc., (Milwaukee, WI). Perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHS), and perfluorooctane sulfonate (PFOS) were generously provided by 3M Company, (Saint Paul, MN). Other internal standards were purchased from Perkin-Elmer (Wellesley, MA) (1,2-$^{13}C_2$ perfluorooctanoic acid) or graciously provided by the Centers for Disease Control and Prevention (Dr. Antonia Calafat) ($^{18}O_2$-perfluorooctane sulfonate). Table 1 contains a listing of all target compounds, their acronyms, and chemical structures. All standards were greater than 90% purity. Hexane, diethyl ether, methanol, and acetonitrile were all pesticide grade purchased from Fisher Scientific (Hampton, NH).

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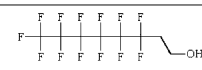
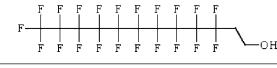
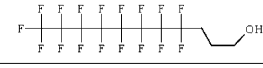
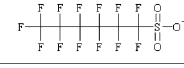
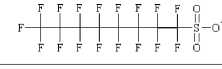
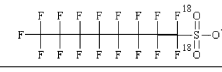
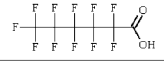
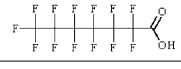
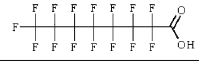
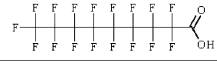
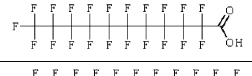
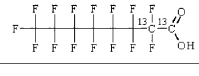
TABLE 1. Target Compounds, Acronyms, Structures, And Ions and Transitions Monitored for Analysis

Analyte	Acronym	Structure	MW	Ions (*m/z*)
2-(perfluorohexyl) ethanol	6:2 FTOH		364	363,344,314
2-(perfluorooctyl) ethanol	8:2 FTOH		464	463,444,405
2-(perfluorodecyl) ethanol	10:2 FTOH		564	563,544,505
3-(perfluorodecyl) propanol	Telomer I.S.		478	477,441,395
perfluorobutane sulfonate	PFBS		299	299 → 80
perfluorohexane sulfonate	PFHS		399	399 → 80
perfluorooctane sulfonate	PFOS		499	499 → 80
$^{18}O_2$-perfluoro - octane sulfonate	$^{18}O_2$-PFOS		503	503 → 84
perfluorohexanoic acid	PFHxA		314	313 → 269
perfluoroheptanoic acid	PFHpA		364	363 → 319
perfluorooctanoic acid	PFOA		414	413 → 369
perfluorononanoic acid	PFNA		464	463 → 419
perfluorodecanoic acid	PFDA		514	513 → 469
perfluoroundecanoic acid	PFUA		564	563 → 519
perfluorododecanoic acid	PFDoA		614	613 → 569
1,2 -$^{13}C_2$ perfluoro - octanoic acid	$^{13}C_2$-PFOA		416	415 → 370

Dust Samples. House dust samples came from household vacuum cleaner bags that were collected in 2000–2001 during the U.S. Environmental Protection Agency's Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) study (*18*). The samples are from 102 homes and 10 daycare centers in North Carolina and Ohio (NC homes $n = 49$, daycare centers $n = 5$; OH homes $n = 53$, daycare centers $n = 5$). While CTEPP was designed to be a population-based study, the vacuum cleaner bags were only collected if they were available at each site. The results from this portion of the study are therefore not generalizable and do not necessarily represent conditions in either state. Samples were irradiated to eliminate microbiological activity and then sieved to remove materials greater than 150 μm in diameter. Material passing the sieve was stored in amber I-CHEM (Rochester, NY) glass containers and stored at room temperature prior to analysis. To our knowledge, nothing has been published concerning the storage stability of the target compounds in dust or the potential impact of irradiation.

Fluorotelomer Alcohols. FTOHs were extracted by placing 100 mg of dust in a 10 mL polypropylene centrifuge tube and adding 5.0 mL of hexane containing 20 ng of 3-(perfluorooctyl)propanol (I.S.) The tubes were then placed in an ultrasonic bath for 30 min, and then centrifuged in a Thermo IEC Centra CL2 centrifuge (Needham Heights, MA) for 10 min. The supernatant was then removed and loaded on to a Supelco Supelclean LC-Silica 3 mL solid phase extraction (SPE) tube (Bellefonte, PA) which had been previously conditioned with 3.0 mL of hexane. Vacuum was applied to allow the solvent to drip at an approximate rate of 1 drip/second throughout SPE cleanup. After loading, 6.0 mL of 10% diethyl ether in hexane was added to wash the SPE tube. The solutions that passed from conditioning, loading, and washing were discarded to waste. Next, 12.0 mL of 10% diethyl ether in hexane was added and eluted into a clean 15 mL glass centrifuge tube. This eluent was concentrated to approximately 1 mL under a N_2 gas stream using an Organomation Associates Inc., N-EVAP 112 evaporation system (Berlin, MA) equipped with an OA-SYS heated water bath set at 37 °C and then placed in a standard glass autosampler vial for GC/MS analysis.

Samples were analyzed using an Agilent 6890N gas chromatograph (GC) coupled with a 5973N mass spectrometer (MS) and a 7683 injector (Palo Alto, CA) using a Restek (Bellefonte, PA) RTX200 column (30 m × 0.25 mm i.d. , 0.1

TABLE 2. Recovery of Compounds Spiked into Dust and Precision of Replicate Analysis of Unknowns

	high spike[a]		low spike[a]		replicate analysis of unknowns[b]
	% recovery	CV	% recovery	CV	CV
6:2 FTOH	77.3	0.03	77.7	0.08	0.05
8:2 FTOH	85.2	0.03	82.1	0.06	0.16
10:2 FTOH	87.6	0.05	85.6	0.03	0.10
PFDoA	104	0.11	107	0.07	0.12
PFUA	102	0.09	107	0.08	0.12
PFDA	101	0.09	102	0.10	0.10
PFNA	39.5	0.10	47.9	0.04	0.08
PFOA	79.0	0.14	82.5	0.07	0.25
PFHpA	102	0.07	99.7	0.03	0.17
PFHxA	98.3	0.09	92.4	0.05	0.13
PFOS	104	0.07	103	0.09	0.06
PFHS	44.2	0.08	51.1	0.07	0.12
PFBS	95.1	0.05	98.9	0.05	0.31

[a] For fluoro-telomer alcohols, the high spike was 82.5 ng, and the low spike was 8.25 ng (n = 5 at each spike level); for perfluorinated acids and sulfonates, the high spike was 100 ng and the low spike was 10 ng (n = 9 at each spike level). [b] Based on replicate analysis of 10% of all unknown dust samples, nonzero samples only.

μm df trifluoropropylmethyl polysiloxane). Samples (2 μL) were injected in pulsed splitless mode at 225 °C and separated chromatographically with a program starting at 60 °C, 3 °C/min. to 125 °C, 50 °C/min. to 250 °C. The MS was operated in electron impact (EI) mode using selected ion monitoring (SIM). SIM regions monitored at least three high mass fragments unique to the analyte of interest (Table 1). The chromatograms were manually integrated for each FTOH and the I.S (Figure S1, Supporting Information).

Six point standard curves for quantitation of the FTOHs were prepared by spiking a blank dust (~100 mg) with between 3.33 –167 ng of the 6:2, 8:2, and 10:2 FTOHs. The spiked dust standards were then subjected to the same extraction, SPE cleanup, and concentration procedures used for the unknowns. The blank dust was chosen from among all of dust samples available after preliminary measurements indicated the analytes of interest were below the limit of detection in this sample. Standard curves (all with $r^2 > 0.99$) were created for each FTOH by plotting the analyte/I.S. ratio versus the mass of FTOH spiked into the dust. Check standards, spiked at 8.25, 16.7, 33.3, 83.3 ng/ 100 mg, were analyzed with each subsequent batch of 10 unknowns to ensure that the original standard curves maintained validity throughout the analysis. Moreover, at least 10% of the unknowns were randomly selected for duplicate analysis and the coefficient of variation (CV) was determined for all nonzero responses (6:2 FTOH, n = 4; 8:2 FTOH, n = 8; 10:2 FTOH, n = 5, Table 2).

Perfluorinated Alkyl Acids. The PFAAs were extracted by placing 100 mg of dust into a 15 mL polypropylene centrifuge tube and adding 5.0 mL of acteonitrile containing 50 ng of the internal standards ($^{13}C_2$−PFOA and $^{18}O_2$−PFOS). After vortexing for 30 s, the samples were shaken on a horizontal shaker for 10 min and then placed in an ultrasonic bath for 30 min. The tubes were then centrifuged and an aliquot of the supernatant was combined 50:50 (vol/vol) with 2 mM ammonium-acetate in a clean autosampler vial for liquid chromatography-mass spectrometry (LC-MS/MS) analysis.

LC-MS/MS Analysis for PFAAs. Samples were analyzed using an Agilent 1100 liquid chromatograph equipped with an Applied Biosystems API 3000 triple quadrupole mass spectrometer operated in negative ESI mode. Analytes were separated chromatograhically over an 8 min run using Phenomenex C18(2) Luna 50 × 4.6 mm column (Torrence, CA) with an isocratic mobile phase consisting of 50:50 2 mM ammonium acetate:acetonitrile flowing at 200 μL/min. The mass spectrometer was operated in the MS/MS mode using negative-ion TurboIonSpray ionization under the following conditions: curtain gas (N_2) nine arbitrary units (au), nebulizer gas (N_2) 8 au, turbo dryer gas (zero air) 8 L/m at 350 °C, and ion spray voltage, 1500 V. Ionization and collision cell parameters were optimized for each individual analyte. The mass transitions monitored for each analyte are listed in the Table 1.

Because all dust samples were found to contain measurable amounts of the PFAAs, standard curves were constructed using solvent standards prepared to give calibration points at the 0, 1, 10, 25, 50, 100, 250, and 500 ng/g level for all unknowns. The analyte/IS area count ratio was plotted against the mass of analyte spiked into the solution as discussed above.

Unknown samples (10%) were randomly selected for duplicate analysis and the CV was determined using all nonzero responses. Method accuracy was assessed by measuring solvent spikes that were prepared at three different levels (5, 50, and 500 ng) over the course of the analysis.

Recovery. The recovery of the PFCs was determined by spiking blank dust (or low level dust samples for the acids and sulfonates) with a low (8.25 ng) and high (82.5 ng) level of FTOHs or low (10 ng) and high (100 ng) level of PFAAs (Table 2). Extraction and cleanup was conducted as described above. Recovery was calculated by comparing the analyte/IS ratio determined above with the analyte/IS ratio of a blank dust elution that was spiked with the standards after extraction and cleanup steps were completed.

Statistical Analysis. Mean, median, and range were determined using MicroSoft Excel. To allow inclusion of all samples in the analyses, concentrations below the (limit of quantitation) LOQ were assigned a value of LOQ/$\sqrt{2}$. All other statistical analyses were performed using SAS/STAT software (SAS Institute, Cary, NC) with the level of significance set at 0.05.

Results and Discussion

Method Performance and Quality Control. The limits of detection (LODs) for the FTOHs were designated as peaks with signal-to-noise ratios of 5:1, thereby leading to LODs of 10, 14.7, and 15.9 pg on column for the 6:2, 8:2, and 10:2 FTOHs, respectively. The corresponding LOQs were set as the lowest calibration standard above the LOD, giving LOQs of 29.1, 28.5, and 30.8 ng of FTOH per g house dust for the 6:2, 8:2, and 10:2 FTOHs, respectively (Table 3). Average accuracy for FTOHs check standards at four spike levels between 8.33 and 83.3 ng was 99.2 ± 7.6% (n = 11). Duplicate analysis of 12 unknown dusts gave CVs ranging from 5.0 to 15.5% (Table 2). Supporting Information Figure S1 is a representative chromatogram of FTOH analysis.

For the PFAAs, peaks with a signal-to-noise ratio of 5:1 were deemed to be at the LOD, resulting in values of approximately 0.5 pg on column for all the perfluorinated sulfonates and carboxylic acids. The LOQ was set as the lowest point on the standard curve above the LOD, leading to values of ~10 ng/g for each compound (Table 3).

The average accuracy of all perfluorinated carboxylic acids and sulfonates was assessed by measurement of matrix-free solvent spikes. Average accuracy for three spike levels over 3 orders of magnitude (5.0–500 ng) was 100 ± 8.9% for the acids and 103 ± 8.0% for the sulfonates (n = 54). Over 10% of the samples were run in duplicate giving coefficients of variation (CV) ranging from 6.2% (PFOS) to 30.9% (PFBS), with the average CV for duplicate analysis for perfluorinated

TABLE 3. Summary of Perfluorinated Compounds in Dust Samples (ng/g)

analyte	LOQ	mean[a]	med.[a]	95th percentile	max.	% above LOQ
6:2 FTOH	29.1	74.9	23.5	285	804	43.7
8:2 FTOH	28.5	167	32.9	669	1660	53.6
10:2 FTOH	30.8	95.8	30.6	333	883	50.9
PFHxA	10.0	117	54.2	412	1250	92.9
PFHpA	12.5	109	50.2	389	1150	74.1
PFOA	10.2	296	142	1200	1960	96.4
PFNA	11.3	22.1	7.99	57.0	263	42.9
PFDA	9.40	15.5	6.65	47.4	267	30.4
PFUA	10.7	30.4	7.57	101	588	36.6
PFDoA	11.0	18.0	7.78	31.0	520	18.7
PFOS	8.93	761	201	2240	12 100	94.6
PFHS	12.9	874	45.5	2300	35 700	77.7
PFBS	12.5	41.7	9.11	150	1150	33.0
ΣPFCs		2624	917	8210	52 900	

[a] Calculation based on values below the LOQ being assigned the value of LOQ/$\sqrt{2}$.

acids and sulfonates at 14.6% (Table 2). Supporting Information Figure S2 is a representative chromatogram of PFAA analysis.

In general, the recoveries for samples spiked with FTOHs and PFAAs were 100 ± 20% (Table 2). Some recoveries were lower (e.g., 39.5% for PFNA and 44.2% for PFHS) but the precision for all of these determinations was uniformly high, with the highest CV for replicate analyses being 14% for PFOA.

Unknown Sample Concentrations. The average dust sample from this study had more than seven different target analytes at levels that were higher than their respective LOQs, with 96.4% of the samples having quantifiable levels of PFOA, 94.6% with PFOS, and 92.9% with PFHxA (Table 3). Median compound concentrations (calculated using LOQ/$\sqrt{2}$ for samples below the LOQ) ran from a low of 6.65 ng/g for PFDA to a high of 201 ng/g for PFOS. PFOA, with median concentrations of 142 ng/g, and PFHxA at 54.2 ng/g, were the second and third most prominent compounds, respectively, in this assessment. The median of the sum of all individual concentrations was 917 ng/g (Table 3), indicating that typical samples contained nearly 1 μg of PFC material per gram of dust.

Maximum concentrations were approximately 1–2 orders of magnitude higher than the medians with the 8:2 FTOH at 1660 ng/g, PFOA at 1960 ng/g, PFOS at 12 100 ng/g, and PFHS at 35 700 ng/g. Supporting Information Figure S3 contains box and whisker plots summarizing all of the data generated in this study. The FTOHs ranged from a low for the 6:2 FTOH of 29.1 ng/g to a high of 1660 ng/g for the 8:2 FTOH. The carboxylic acids ranged from PFHxA 10.0 ng/g to PFOA at 1960 ng/g. The sulfonates ranged from 8.93 ng/g for PFOS to 35 700 ng/g for PFHS.

A Wilcoxon's sum rank test indicated no difference in individual target compound concentrations between all samples collected in North Carolina and Ohio ($p > 0.262$). In general, the median levels of the PFCs were present in the same order of magnitude as the organophosphate and pyrethroid pesticides and the PAHs that were measured as part of the wider CTEPP study (*18*).

Spearman rank correlations indicate that, in general, almost all of the compounds had significant correlations with each other except PFBS (Table S1). The FTOHs were highly correlated with each other ($r > 0.87$, $p < 0.0001$) suggesting a common source for these compounds. Among the carboxylic acids, those with molecular weights at and above C9 tended to be highly correlated; those at and below C8 appeared to form another group, possibly suggesting two different sources of these materials. It is also interesting to note that PFOS had a strong correlation with PFOA, PFHpA, and PFHxA. ($r > 0.82$, $p < 0.0001$). Supporting Information Figure S4 is a regression of the log normalized PFOS and PFOA concentrations, illustrating the strength of the relationship between these compounds ($r^2 > 0.73$).

Few studies have documented the presence of perfluorinated compounds in the indoor environment. But given the wide range of consumer and residential products that contain or have been treated with PFCs, it is reasonable to hypothesize that as these products age and degrade, their dust and debris will accumulate indoors. Perhaps the most obvious potential source would be antistain agents used on carpets and upholstery (*1, 19*). As these materials are worn from the surface by routine use and abrasion, or as they volatilize directly from product surfaces, they may accumulate in any dust that is not removed by normal cleaning. Previous studies have shown house dust to be a sink for many contaminants including metals, pesticides, and PAHs, which in turn represent sources of potential exposure indoors (*10, 11, 20*). The current study and a handful of previous studies now indicate that house dust may also be a reservoir of PFC material in homes which may be available for human exposures.

In what appears to be the first published report on PFCs in house dust, Moriwaki et al. reported median concentrations of PFOS at 24.5 ng/g and PFOA at165 ng/g in 16 samples collected in Japan (*13*). The present study's median PFOS level (201 ng/g) was approximately 8 times higher, whereas PFOA levels were roughly comparable (148 ng/g). Moriwaki et al., also found a high correlation between PFOS and PFOA ($r^2 = 0.99$), but this relationship was strongly influenced by a single point representing a dust with PFOS and PFOA at 2500 and 3700 ng/g, respectively. After removing this point from their analysis the r^2 for the regression drops to 0.35. The current study, however, confirms this relationship (Supporting Information Figure S4) with a much greater sample size. It is worth noting that PFOA was always at a higher concentration than PFOS in all of the Japanese dust samples. In the current U.S. study, 78% of the samples had PFOS higher than PFOA (mean ratio PFOS/PFOA = 2.6), suggesting some distinct difference in sources between the U.S. and Japan.

Another published report on PFCs in dust ($n = 67$) from select homes in Ottawa, Canada reported median concentrations of PFOS at 37.8 ng/g and PFOA at 19.7 ng/g (*14*). The present study's median PFOS and PFOA levels were more than 5 times and 7 times higher respectively, whereas the overall ranges of concentrations were similar between the two studies. PFOS and PFOA were detected in 67% and 63% of the houses, respectively, roughly 30% less frequently than the current U.S. study. This observation is probably not related to the different method LOQs from each study, as the Canadian effort actually had lower LOQs for both compounds (PFOS = 4.5 ng/g, PFOA = 7.29 ng/g vs ~10 ng/g for both compounds for the current study). Together these observations clearly indicate that these compounds are more prominent in the U.S. dust from the current study both in terms of occurrence and concentration.

This Canadian study also found that the amount of carpeting in the house was positively correlated with PFOS, PFOA, and PFHS concentrations in the dust. Older homes, which had less carpeting, also had significantly lower levels of PFOS and PFOA. This lead the authors to speculate that carpets and the use of carpet surface treatment products, such as antistain agents, might be responsible for the elevated PFC concentrations they observed.

The current study is in general agreement with the preceding Japanese and Canadian studies in terms of the general ranges of PFOS and PFOA that can be found in house dust, but the current work adds more data on other PFCs

that have previously not been reported indoors. For example, to the best of our knowledge this is the first published data on FTOHs, which were quantifiable in roughly half of the samples in the same general range of concentration as the acids and sulfonates. Median levels were 23.5 ng/g for the 6:2 FTOH, 32.9 ng/g for the 8:2 FTOH, and 30.6 ng/g 10:2 FTOH. Maximum concentrations of 804 ng/g, 1660 ng/g, and 883 ng/g dust were determined for the 6:2 FTOH, 8:2 FTOH, and 10:2 FTOH, respectively (Table 3). The FTOHs are volatile (*21*) and can be oxidized in various biological systems to yield corresponding perfluorinated carboxylic acids, including PFOA (*22, 23*). Other volatile perfluorinated compounds, *n*-methylperfluorooctane sulfonamidoethanol (MeFOSE) and *n*-ethylperfluorooctane sulfonamidoethanol (EtFOSE), were shown to be in indoor air and dust samples from Ottawa, Canada homes with geometric mean concentrations of 113 and 138 ng/g for MeFOSE and EtFOSE, respectively (*15*). These compounds are known to be metabolized to PFOS in liver microsome systems (*24*). It is perhaps surprising to find that these volatile and reactive PFCs are associated with indoor dust, but other volatile materials including pesticides and PAHs are known to partition onto house dust (*10, 20*) making them potentially available for exposures indoors.

Potential for Human Exposure. The presence of PFCs in human blood has been well documented in many countries around the world, but the relative importance of the various potential routes of exposure remain almost completely unknown (*25, 26*). Leading hypotheses about potential exposure routes suggest that food and food packaging (*27–29*), water (*30*), house dust (*15*), and airborne sources (*31*) might all be significant exposure routes, but few data are available to allow adequate evaluation any of these hypotheses at this time.

The USEPA Exposure Factors Handbook suggests default ingestion rates of 50 and 100 mg of dust/soil for adults and children, respectively (*32*). Using these assumptions, if we use the median concentration of total PFC of 917 ng/g from the current study (Table 3), this leads to median daily intake 92 ng of total PFCs for children, and 46 ng for adults. This further breaks down to median intakes of 20.1 ng PFOS per day and 14.2 PFOA per day for children ingesting 100 mg of the median dust from this study.

A study by Shoeib et al. also suggests that ingestion of house dust could be an important source of human exposure, particularly for MeFOSE and Et FOSE (*15*) . Under their assumption that adults ingest approximately 100 mg of house dust per day, they estimated median exposures to perfluorinated alkyl sulfonamides (primarily MeFOSE and EtFOSE) via ingestion of house dust to be 20 ng/day for adults, and about 44 ng/day for children, who were assumed to ingest almost twice as much dust as adults. To our knowledge, the toxicological significance of this potential exposure is not well-known, and the extent to which these materials could be transformed to PFOS by human metabolism remains to be described.

To provide further context for the interpretation of this study's results, it is informative to compare these data with a recent dietary exposure study by Tittlemier et al. (*29*). As part of the Canadian Total Diet Study, they measured PFC levels in 54 solid food composites that were said to be representative of the typical Canadian (12 years and older) diet. Their analysis, coupled with typical food consumption estimates for adults, lead to a conservative estimate of 250 ng/day of total PFCs via food sources. This further breaks down to 110 ng/day of PFOS and 70 ng/day for PFOA, with the balance of the total being ascribed to other homologous carboxylic acids of various lengths. Tittlemier et al. then went on to compare this calculated dietary exposure with other previously published estimates. They found that after ingestion of food, the second most important potential source of exposure was calculated to be from solution-treated carpeting, which has a maximum reasonable aggregate adult exposure of 120 ng/day for total PFAAs (*19*). While water and airborne sources were judged to be negligible, Tittlemier et al. used the results from the study by Kubwabo et al. (*14*) to estimate total PFAA exposure via dust to be 28 ng/day. This is very close to the 46 ng/day of total PFAA estimated above for adults in the present study, which is approximately 18% of the total PFAA thought to be ingested by via food.

One caution with the interpretation of these results is that the dust was collected at a time when PFOS was being withdrawn from the U.S. market by its principle manufacturer. Moreover, since that time, additional agreements have made with industry to limit the use of materials that can degrade to PFOS and minimize the residual PFOA and other related materials in consumer use items (*33*). This suggests that the composition of dust-related sources is expected to change as nature of the PFCs in consumer-use products change. It is important to note that studies with pesticides have shown that materials that have been removed from the market may remain present in house dust for decades after their production is discontinued (*10*). Given the persistence of the PFCs, it is very likely that PFOS, PFOA, and similar compounds will remain available in house dust for potential exposures for some time to come.

In summary, this study confirms previous observations which indicate that PFCs are present in house dust and that this material will be available in residences for potential exposure. Compared to dust samples collected from other countries, samples from this study show distinctive patterns of compounds, higher concentrations, and a greater prevalence of some of the most common compounds that have been measured indoors. Moreover, with volatile and nonvolatile PFCs present in dust at median levels from 6 to 201 ng/g, it is possible that dust may represent an important exposure route that could contribute to personal exposure to these materials. Given the range of industrial and consumer products that have used PFCs in the past, and the changing composition of the materials found in these products, a great deal of further work will be required to characterize the true nature of these potential exposures.

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Supporting Information Available

Table S1. Correlation matrix between all perfluorinated compounds in dust samples. Figure S1. (A) GC/MS select ion monitoring (SIM) chromatogram showing analytes of interest (~167 pg on column injection), a 300–600 m/z scan for (B) 6:2 FTOH, (C) 8:2 FTOH, (D) 10:2 FTOH, and (E) FTOH IS. Figure S2. LC/MS/MS chromatogram of perfluorinated carboxylic and sulfonic acids. Figure S3. Distribution of PFCs in house-dust samples. Boxes show 25th, and 75th percentile and median. Whiskers show 5th and 95th percentile. Symbols indicate outliers. Figure S4. Log normal distribution plot of PFOS vs PFOA concentration (ng/g) in all house dust samples. This material is available free of charge via the Internet at http://pubs.acs.org.

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