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Per- and Polyfluoroalkyl Substances in Dust Collected from Residential Homes and Fire Stations in North America

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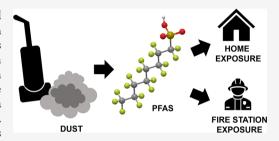
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ABSTRACT: Over the past few years, human exposure to per- and polyfluoroalkyl substances (PFAS) has garnered increased attention. Research has focused on PFAS exposure via drinking water and diet, and fewer studies have focused on exposure in the indoor environment. To support more research on the latter exposure pathway, we conducted a study to evaluate PFAS in indoor dust. Dust samples from 184 homes in North Carolina and 49 fire stations across the United States and Canada were collected and analyzed for a suite of PFAS using liquid and gas chromatography—mass spectrometry. Fluorotelomer alcohols (FTOHs) and di-polyfluoroalkyl phosphoric acid esters (diPAPs) were the most prevalent PFAS in both fire station and house dust



samples, with medians of approximately 100 ng/g dust or greater. Notably, perfluorooctanesulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexane sulfonate, perfluoronanoic acid, and 6:2 diPAP were significantly higher in dust from fire stations than from homes, and 8:2 FTOH was significantly higher in homes than in fire stations. Additionally, when comparing our results to earlier published values, we see that perfluoroalkyl acid levels in residential dust appear to decrease over time, particularly for PFOA and PFOS. These results highlight a need to better understand what factors contribute to PFAS levels in dust and to understand how much dust contributes to overall human PFAS exposure.

■ INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) have garnered attention as "forever chemicals" that are widely detected in humans and the environment. PFAS are used for their waterand stain-repellant properties and can be found in many kinds of consumer products. However, concerns over the persistence, bioaccumulation, and toxicity of some PFAS have motivated additional research to better understand human PFAS exposure.

PFAS are a large group of chemicals that include approximately 5000 different compounds¹ with several different subclasses such as perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonic acids (PFSAs), fluorotelomer alcohols (FTOHs), and polyfluoroalkyl phosphoric acid esters (PAPs).^{2,3} The most persistent PFAS compounds are the perfluoroalkyl acids (PFAAs), which include the PFCAs and PFSAs. Some PFAAs, particularly perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), are known to have long half-lives in humans and have been associated with a myriad of human health effects.⁴ Other PFAS such as the FTOHs and PAPs are less persistent because of their physicochemical properties; however, these compounds can degrade into the more persistent PFAAs and have been referred to as PFAA precursors.²

PFAS are amphiphilic compounds with both hydrophobic and lipophobic properties; thus, they are frequently used as

water and oil repellents. Various consumer products common to most homes contain PFAS⁵ and can be sources of PFAS to the indoor environment and dust. For example, PFAS can be found in nonstick cookware and food packaging, 6-8 in personal care products such as cosmetics and sunscreens, 9 and in impregnation sprays and agents used for textile protection, such as stain-repellant carpets and upholstery. 5,10

Data for PFAS concentrations in the indoor environment are important for better understanding the main exposure pathways for PFAS in humans, particularly as indoor sources are less well characterized than dietary sources. While a tolerable weekly intake for four PFAS in food has been established by the European Food Safety Authority, 11 house dust ingestion and indoor air may be another major pathway of PFAS intake for some people. 12 People spend a considerable amount of time indoors, leading to exposure to chemicals from indoor air and dust derived from building materials and consumer products in the home. For example, exposure to dust has been shown to be an important exposure pathway for

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chemicals such as flame retardants. ^{13–15} The United States Environmental Protection Agency estimates that adults ingest approximately 30 mg of indoor dust per day and children ingest approximately 60–100 mg. ¹⁶

Previous studies have measured PFAS in dust from homes, daycares, and business areas across several countries. ^{17–25} However, there are limited current data on PFAS in dust from homes in the United States, and no one to date has measured dust PFAS concentrations in fire stations as an occupational exposure pathway for firefighters.

Studies have shown that firefighters have higher exposure to PFAS compared to the general population. 26-31 This is especially true for firefighters exposed to aqueous film-forming foams (AFFF), a type of firefighting foam that contains PFAS.³² AFFF is used to quickly extinguish fires and contains varying types of PFAS depending on the formulation. AFFF is especially used to contain petroleum-fuel-based fires at airports and military sites (i.e., class B fires); unfortunately, AFFF use has led to widespread PFAS contamination near these sites and at firefighter training areas. 33,34 In one study, firefighters who trained with firefighting foams had increased serum perfluorohexane sulfonate (PFHxS) and perfluorononanoic acid (PFNA) following training in comparison to baseline serum concentrations taken 2 weeks prior.²⁷ Exposure to PFAS through dust and smoke inhalation during a fire event is also possible. For example, first responders to the World Trade Center collapse on September 11, 2011 were found to have plasma levels of PFOA and PFHxS that were twofold higher than the general population, and especially high levels were found in first responders exposed to the greatest amounts of dust and smoke.²⁸ Similar results were seen in the plasma of California firefighters, with PFOA and PFNA being twofold higher than in the general United States population.² Additionally, the protective turnout gear used by firefighters may have PFAS coatings which could contribute to exposure.³⁵

PFAS sources unique to firefighters (e.g., AFFF and turnout gear) have raised questions about the extent of PFAS exposure in the fire stations where firefighters spend a substantial amount of their time while on duty. Therefore, the goal of this present study was to provide more information on the levels of PFAS in indoor dust for both the general public and for firefighters. To support this goal, we collected dust samples from fire stations (occupational exposure) and residential homes (general population exposure) and analyzed them for a suite of 17 PFAS (Table S1). Fire stations were hypothesized to have higher PFAS dust levels because of the likelihood of more frequent contact with potential PFAS sources (e.g., AFFF and turnout gear). A secondary goal of this study was to assess potential differences in PFAS levels between fire stations and residential settings and to determine if PFAS dust concentrations varied based on location, size, age of the buildings, or carpeting.

■ MATERIALS AND METHODS

Dust Collection and Survey Data Collection. Residential House Dust. Dust samples from 184 residential homes in North Carolina, United States, were collected between 2014 and 2016 as a part of the Toddlers' Exposure to Semivolatile organic contaminants in Indoor Environments (TESIE) study. The TESIE study population has been further described by previous studies. Briefly, home visits were conducted with families enrolled in the TESIE study to collect dust samples

and data about the home environment. Study protocols were approved by the Duke Institutional Review Board.

Dust extracts from homes were processed as previously described in Hammel et al. $(2019)^{37}$ and Phillips et al. $(2018)^{.38}$ Briefly, the entire exposed floor area of the main living room was vacuumed with a Eureka Mighty Mite vacuum fitted with a cellulose thimble within the hose attachment, ¹³ and each thimble was wrapped in aluminum foil, placed in sealed plastic bags, and stored at -20 °C after collection until analysis.

Participants were asked to complete questionnaires during the home visits as also previously described in Hammel et al. (2019)³⁷ and Phillips et al. (2018).³⁸ The age of the home and the square footage of the home and main living area were collected as a part of the questionnaire and verified with tax records where possible.

Fire Station Dust. Dust samples were collected from 49 fire stations in the United States in 2015³⁹ and in Canada in early 2018.⁴⁰ Fire stations were selected for participation in the study through a collaboration with the International Association of Fire Fighters (IAFF). Stations were selected if they met certain criteria: location in urban or suburban areas, active engagement in urban or suburban fires, a fire station as the residence for on-duty firefighters who return to the station after deployment to a fire for the duration of their shift, and fire stations and bays contained to one building.

The fire station dust samples have previously been analyzed for flame retardants. 39,40 Additional methodology information for the U.S. fire stations has been previously reported in Shen et al. (2018)³⁹ and for the Canadian fire stations in Gill et al. (2020).⁴⁰ In brief, participating stations were sent packets of information describing the study's purpose and instructions for collecting samples in only the living quarter areas (including kitchen and eating areas, sleeping areas, and television viewing areas). Fire stations in the U.S. were asked to use their own vacuum cleaners with new vacuum bags and to collect dust over the course of 1 month. Canadian fire stations were provided with new vacuum cleaners (Dirt Devil, Featherlite Bagged Upright) and a set of new vacuum cleaner bags. Stations were instructed to use the vacuum cleaner only in the living quarters for the purposes of the study and to refrain from mopping, sweeping, or other cleaning techniques in the living quarters during the month of the study. Vacuum cleaner bags from fire stations were sealed in polyurethane bags and stored at 4 °C until analysis. Surveys were collected from fire stations to obtain information on building location, year of construction, square footage, and the percentage of the living quarters containing carpeting.

Dust Sample Preparation, Extraction, and Analysis. Full method information and HPLC-tandem mass spectrometry and gas chromatography-mass spectrometry (GC-MS) conditions are included in the Supporting Information and Table S2.

Quality Assurance and Quality Control. Quality assurance and quality control (QA/QC) were addressed by analyzing laboratory processing blanks alongside the samples and by analyzing the house dust standard reference material (SRM 2585, National Institute of Standards and Technology (NIST), Gaithersburg, MD). Laboratory blanks (n = 5-6) and SRM 2585 (n = 5) were analyzed in each batch of dust samples, and five field blanks (n = 5) were processed along with the fire station dust samples. PFAS levels in samples were blank-corrected using the average laboratory processing blank.

Table 1. Detection Frequency, MDL, and Median and Maximum Concentrations for PFAS Analytes in Dust Samples a, b, c, d, e

	on p-value*	$^*p < 0.0001$				p = 0.1411	p < 0.0001						$^*p = 0.0075$	$p^*p = 0.0035$			$^*p < 0.0001$	*p < 0.0001
residential homes (2014–2016, $n = 184$)	maximum concentration (ng/g dust)	34,360	0889	7890	370	248,920	44,220	N/A	546	135	1380	713	2350	208	4130	320	694	2810
	median concentration (ng/g dust)	113	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>695</td><td>1435</td><td>N/A</td><td><mdl< td=""><td><mdl< td=""><td>8.5</td><td>8.9</td><td>7.9</td><td>3.3</td><td>6.2</td><td><mdl< td=""><td>2.0</td><td>4.4</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>695</td><td>1435</td><td>N/A</td><td><mdl< td=""><td><mdl< td=""><td>8.5</td><td>8.9</td><td>7.9</td><td>3.3</td><td>6.2</td><td><mdl< td=""><td>2.0</td><td>4.4</td></mdl<></td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>695</td><td>1435</td><td>N/A</td><td><mdl< td=""><td><mdl< td=""><td>8.5</td><td>8.9</td><td>7.9</td><td>3.3</td><td>6.2</td><td><mdl< td=""><td>2.0</td><td>4.4</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	695	1435	N/A	<mdl< td=""><td><mdl< td=""><td>8.5</td><td>8.9</td><td>7.9</td><td>3.3</td><td>6.2</td><td><mdl< td=""><td>2.0</td><td>4.4</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>8.5</td><td>8.9</td><td>7.9</td><td>3.3</td><td>6.2</td><td><mdl< td=""><td>2.0</td><td>4.4</td></mdl<></td></mdl<>	8.5	8.9	7.9	3.3	6.2	<mdl< td=""><td>2.0</td><td>4.4</td></mdl<>	2.0	4.4
	MDL (ng/g dust)	0.48	10.63	13.17	15.45	6.44	15.80	N/A	1.72	0.14	0.42	0.51	0.26	0.15	90.0	22.28	0.25	0.20
	detection frequency (%)	100	34	19	15	26	99.5	N/A	9.2	10	76	76	100	99.5	41	1.1	57	84
fire stations (2015 and 2018, $n = 49$)	maximum concentration (ng/g dust)	6270	2250	1110	17,540	86,060	65,170	183,730	213	1410	1150	382	791	203	137	2650	8280	74.370
	median concentration (ng/g dust)	287	99.3	30.2	76.6	756	216	84.9	4.6	<mdl< td=""><td><mdl< td=""><td><mdl< td=""><td>17.6</td><td>7.2</td><td>2.5</td><td><mdl< td=""><td>8.9</td><td>64.5</td></mdl<></td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td><mdl< td=""><td>17.6</td><td>7.2</td><td>2.5</td><td><mdl< td=""><td>8.9</td><td>64.5</td></mdl<></td></mdl<></td></mdl<>	<mdl< td=""><td>17.6</td><td>7.2</td><td>2.5</td><td><mdl< td=""><td>8.9</td><td>64.5</td></mdl<></td></mdl<>	17.6	7.2	2.5	<mdl< td=""><td>8.9</td><td>64.5</td></mdl<>	8.9	64.5
	MDL (ng/g dust)	2.54	9.63	2.00	5.00	48.90	7.23	5.00	1.03	1.20	10.30	6.77	1.60	0.47	1.49	8.56	0.97	1.44
	detection frequency (%)	100	94	88	65	96	92	82	84	41	41	37	82	96	63	12	98	96
	punoduoo	6:2 diPAP	8:2 diPAP	MeFOSE	EtFOSE	6:2 FTOH	8:2 FTOH	10:2 FTOH	PFBA	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFBS	PFHxS	PFOS
	class	diPAP		FOSE		FTOH			PFCA							PFSA		
		PFAA	precursors						legacy PFAAs									

^aBolded compounds were significantly different between fire stations and homes. b* denotes statistically significant differences between fire stations and homes by the two-tailed nonparametric Mann-Whitney test; no p-values are reported for compounds detected in less than 50% of samples. $^{c}N/A$: not applicable as it was not measured. d <MDL: below the method detection limit. c Values less than the MDL were imputed with dust mass-specific MDLs.

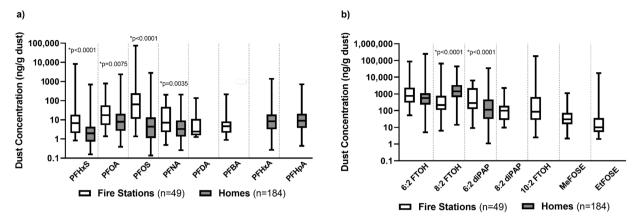


Figure 1. PFAS dust concentrations in fire stations and homes. Dust concentrations for (a) legacy PFAAs and (b) PFAA precursors in both fire stations and residential homes. Boxplots represent the minimum, 25th percentile, median, 75th percentile, and maximum concentrations for each analyte. Analytes are only represented as boxplots if they were detected in more than 50% of samples as listed in Table 1. Concentrations are reported on a log scale. Significance is determined by the Mann—Whitney test.

Method detection limits (MDLs) were calculated using 3 times the standard deviation of the average laboratory blank levels if the analyte was detected in the blanks or by using a value equal to 10 times the noise if absent in the lab blanks. MDLs were normalized to the average mass of dust extracted (0.1 g). If dust concentration values were below the MDL, the value was imputed with dust mass-specific MDL values. PFAS measurements for the extracted SRM and reference values for house dust SRM 2585^{17,41,42} are provided in Table S3.

Statistical Analyses. Data were analyzed and graphed using GraphPad Prism (version 8.4.0) and Microsoft Excel. Nonparametric statistical tests were used as the data were not normally distributed, as determined by the Shapiro–Wilk normality test. Statistical analyses were only performed on individual analytes that were detected in more than 50% of samples. The Mann–Whitney test (two-tailed) was used to determine statistical significance at $\alpha < 0.05$. Some survey questionnaires were incomplete or missing; thus, the sample size for some endpoints varied.

■ RESULTS AND DISCUSSION

PFAS Concentrations in Dust Samples. In this study, we report on PFAS concentrations measured in both residential dust samples and dust collected from living quarters of fire stations within the United States and Canada. PFAS were detected in all dust samples. Detection frequencies and MDLs for each PFAS are listed in Table 1, along with maximum and median concentrations. In general, the shorter-chain PFAAs (perfluorobutanoic acid (PFBA), perfluorobutane sulfonate (PFBS), and perfluoropentanoic acid (PFPA)) were less frequently detected than the longer-chain PFAAs and the precursor compounds. The FTOHs were the most abundant group of the PFAS quantified in both homes and fire stations. In contrast, the median concentrations for many of the PFAAs were below 20 ng/g dust.

There was considerable variation in the distribution of dust concentrations for individual PFAS analytes, as seen by the range of concentrations depicted in Figure 1. For example, the maximum concentrations for some PFAS (e.g., PFOS, FTOHs, and perfluorooctane sulfonamido ethanols (FOSEs)) were orders of magnitude higher than the median concentrations. Based on median dust concentrations, the most abundant PFAS was 8:2 FTOH in residential homes (1440 ng/g dust)

and 6:2 FTOH in fire stations (760 ng/g dust). Of the PFAAs, perfluoroheptanoic acid (PFHpA) was the most abundant in homes (a median concentration of 9.0 ng/g dust), while PFOS was the most abundant in fire stations (a median concentration of 65 ng/g dust). Median levels for dust PFAS reported here ranged from tens to hundreds of nanograms per gram (ng/g), which are generally lower than the levels of organophosphate ester flame retardants (OPEs)³⁸ and phthalates³⁷ we measured in these same house dust samples in previous studies.

Comparing Dust PFAS Concentrations in Fire Stations and Homes. Overall, precursor PFAS were found at higher concentrations than legacy PFAAs in both fire stations and homes, and both locations were dominated by diPAPs and FTOHs. This suggests that PFAS exposure via dust in fire stations versus residential settings is not drastically different, although a few analytes show some differences. PFOS, PFOA, PFHxS, PFNA, and 6:2 diPAP were significantly higher in fire station dust than in residential dust. Median dust levels in fire stations were approximately 15 times higher for PFOS (*p < 0.0001), approximately 3 times higher for PFHxS (*p < 0.0001), and approximately 2.5 times higher for 6:2 diPAP (*p < 0.0001) compared to median dust levels in homes. Only 8:2 FTOH was significantly higher in house dust than in fire station dust (approximately 7 times higher, *p < 0.0001).

PFAS profiles in house and fire station dust were both dominated by diPAPs and FTOHs, as shown in Figure S1. Dust samples from the fire stations and homes were both collected from the living areas, so it is unsurprising that the overall relative proportions of PFAS in dust are similar. This likely suggests that the sources of PFAS to the living quarter dust are from similar products or similar construction materials used in both homes and fire stations.

However, fire station dust PFAS levels did somewhat differ from home dust PFAS levels as fire stations had higher median concentrations of PFOS, PFOA, PFHxS, PFNA, and 6:2 diPAP. The higher levels of PFOS and PFHxS in fire stations may be linked to the use or storage of AFFF by firefighters. PFOS and PFHxS have been noted to be components of some AFFF formulations. ^{31–33,43} High PFOS and PFHxS serum levels in firefighters have been hypothesized by some to be due to use of these foams or exposure to burning stain-resistant carpeting. ^{31,32,44,45} Information on the types of AFFF used by

the various stations was not obtained as a part of this present study. There may also be differences between fire stations and homes because of how the living quarters are utilized in each environment. Fire stations likely have a greater flow-through of people than homes in addition to frequent turnovers as firefighters arrive and leave for shifts; the fire stations in our study had an average of seven firefighters per shift.

PFAS Concentrations in Fire Station Dust by Country. A notable result from this present study is that fire stations in the United States had significantly higher Σ PFAS dust concentrations than fire stations in Canada for the subset of 17 PFAS analytes we measured. As shown in Figure 2, U.S. fire

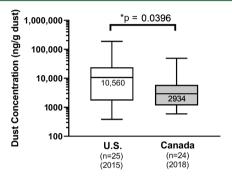


Figure 2. Dust ΣPFAS in fire stations by country. The ΣPFAS concentration in dust was significantly higher in U.S. fire stations than Canadian fire stations (Mann–Whitney test, two-tailed, *p = 0.0396). The ΣPFAS concentration represents the sum of all 17 PFAS analytes measured in a dust sample. Boxplots represent the minimum, 25th percentile, median, 75th percentile, and maximum concentrations. Median concentration values are listed inside the box. Concentrations are reported on a log scale.

stations (n = 25) had a median Σ PFAS concentration of 10,600 ng/g dust, which is approximately 3.5 times higher than the Canadian fire stations' median Σ PFAS concentration of 2900 ng/g dust (n = 24).

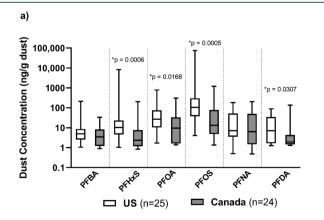
Different trends and regulations regarding building codes, construction materials, and fire station practices in the use of PFAS-containing products may result in different PFAS dust levels between the U.S. and Canada. However, it must be noted that the fire station samples in this present study were collected at different times (2015 for the U.S. stations and

2018 for the Canadian stations); thus, the Canadian fire station dust samples may have lower PFAS levels solely because they were collected more recently. We also cannot rule out the possibility that differences in the sample collection methodology may contribute to the observed differences between countries.

Analyses were also conducted on individual PFAS to determine which PFASs were driving the difference in ΣPFAS between U.S. and Canadian fire stations. Median concentrations of individual PFAS captured by our analytical methods were generally higher in U.S. fire station dust samples compared to those in Canadian fire station dust samples, as seen in Figure 3; a total of 12 of 13 analytes had higher median dust concentrations in the U.S. samples with only 8:2 diPAP being higher in Canadian fire stations. PFOS, PFOA, PFHxS, perfluorodecanoic acid (PFDA), 10:2 FTOH, and EtFOSE were significantly higher in U.S. fire station dust compared to those in Canadian fire station dust. The median PFOS dust concentration in the U.S. fire stations was 104 ng/g dust, which is 7 times greater than the Canadian concentration of 13 ng/g dust. Median and maximum concentrations by country are listed in Table S4.

Fire station dust samples were collected from several different states and provinces. The Σ PFAS dust concentrations by region (state and province) are shown in Figure S2 to illustrate regional variability. The Σ PFAS for homes represents the sum of 16 PFAS compounds (without 10:2 FTOH), while Σ PFAS for fire stations represents the sum of 17 PFAS compounds. The observed ranges of Σ PFAS dust concentrations displayed wide variability, even within a single region (e.g., within New Hampshire). No large differences in Σ PFAS dust levels were observed between regions in this limited assessment, although because of the low sample size per region for the fire station samples (n=4-9), statistical analysis by region was precluded.

PFAS Dust Concentrations and Survey Responses. Information on building characteristics was collected via surveys during the study. Table S5 lists survey data from fire stations, and Table S6 lists survey data from homes. The total number of samples was small, and there was wide variability in the recorded age of each building, so dust samples were grouped into two categories: construction pre- and post-1970. PFAS dust concentrations stratified by building age are shown



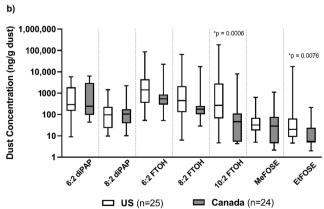


Figure 3. Dust PFAS concentrations by analyte between U.S. and Canadian fire stations. Results for (a) legacy PFAAs and (b) PFAA precursors. Significance is determined by the two-tailed Mann—Whitney test. Boxplots represent the minimum, 25th percentile, median, 75th percentile, and maximum concentrations. Concentrations are reported on a log scale.

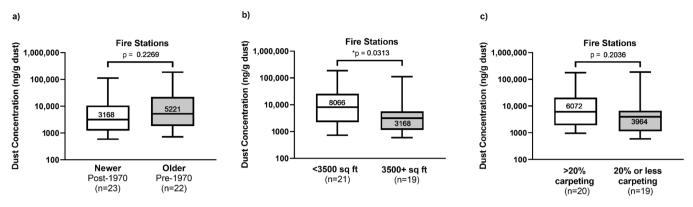


Figure 4. Σ PFAS dust concentrations and survey responses for fire stations. Differences in dust Σ PFAS concentrations in fire stations due to (a) year of construction, (b) square footage of fire station living quarters, and (c) percentage carpeting in the living quarters. Significant differences were seen in Σ PFAS dust concentrations based on square footage in fire stations (*p = 0.0313, two-tailed Mann–Whitney test). Sample sizes are variable because of missing survey data. Median concentrations are listed inside the box. Boxplots represent the minimum, 25th percentile, median, 75th percentile, and maximum concentrations. Concentrations are reported on a log scale. The Σ PFAS concentration represents the sum of all 17 PFAS analytes measured in an individual fire station dust sample.

in Figure 4a for fire stations and Figure S3a for U.S. homes. No significant differences in dust Σ PFAS were observed because of building age in this present study. However, it is important to note that we did not record whether any renovations might have occurred within the building.

The square footage of home and fire station living quarters was also recorded and analyzed. Because of the small sample size, the square footage was dichotomized near the median and categorized into either less or greater than 3500 square feet for fire stations and less or greater than 1720 square feet for homes. Within fire stations, $\Sigma PFAS$ dust concentrations were significantly higher in stations that had living quarters with a smaller square footage (*p=0.03, Mann–Whitney test) (Figure 4b). This difference appears to be driven by differences in several PFAS, including PFBA, PFHxS, PFOA, PFOS, PFDA, 6:2 FTOH, and 10:2 FTOH (Figure S4). For homes, the square footage was not significantly associated with $\Sigma PFAS$ dust levels (Figure S3b).

Analyses were also conducted considering the presence or absence of carpeting in the vacuumed room (generally the main living room). For fire stations, we recorded the percentage of carpeting, and for homes, we recorded the presence of carpeting or rugs in the main living area of the home (i.e., the area vacuumed). Within fire stations, the percentage of carpeting in the living quarters was not significantly associated with Σ PFAS dust levels (Figure 4c); a similar nonsignificant result for Σ PFAS was found for homes (Figure S3c). However, when further delving into these data by individual analytes, several of the FTOHs were found to be significantly higher in the dust samples from carpeted areas in both fire stations and homes (Figure S5). This suggests that carpeting may be a source of the FTOHs observed in these dust samples.

Differences in building use and construction may be important factors for PFAS dust concentrations. For example, Zheng et al. (2020)²⁴ recently reported PFAS dust concentrations in U.S. childcare centers. Similar to this present study, Zheng et al. (2020)²⁴ found that FTOHs were the most abundant class, and PFAAs were the least abundant class measured in dust. However, median PFAS dust levels in the childcare centers appear to be lower than the levels reported in this present study and may reflect differences in building use and construction.

Time Trends in PFAS Dust Levels and Comparisons to Those of Previous Studies. Previous studies have measured PFAS in house dust from the United States ^{19,25,46,47} and across the world. ^{14,17–19,48–56} Studies reporting PFAS levels in U.S. house dust are listed in Table S7 and compared to the present study. As shown in Figure 5, the median

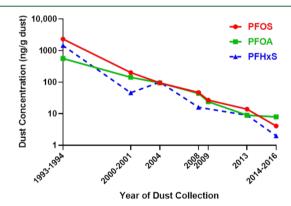


Figure 5. Decline in PFAS concentration in U.S. residential dust over time as reported in the studies cited in Table S7.

concentrations of PFOA, PFOS, and PFHxS in U.S. house dust samples appear to be decreasing over the past 20 years. In 2000, the 3M company began phasing out its production of PFOS, and in 2006, several other companies pledged to phase out the use of PFOS and PFOA in the U.S.. While PFOS production and PFOA production have declined over the past 2 decades in the U.S., other PFAS replaced them. We would expect to see increases in these newer replacement PFAS in dust as they are identified and as analytical and sampling methods improve over time. Data on PFAS precursors in U.S. house dust studies are limited and could not be examined thoroughly for trends over time.

Karásková et al. ¹⁹ measured house dust collected in 2013 and found a higher median concentration for PFOS in U.S. homes than in Canadian homes (14 vs 9 ng/g), similar to our results in U.S. and Canadian fire stations. Strynar and Lindstrom ⁴⁶ analyzed U.S. house dust from 2000 to 2001 and found higher dust concentrations for PFAAs but lower 6:2 FTOH and 8:2 FTOH when compared to our house dust samples collected in 2014–2016. We compared house dust

concentrations from the 102 U.S. homes in Strynar and Lindstrom⁴⁶ to the house dust samples in our present study in Figure S6 and found that PFHxA, PFHxS, PFHpA, PFOA, and PFOS were significantly lower in our study, suggesting that levels have significantly decreased over time. In contrast, 8:2 FTOH significantly increased and was detected more frequently in our more recent house dust samples, and 6:2 FTOH followed a similar trend. Knobeloch et al.⁴⁷ analyzed U.S. house dust collected in 2008 and measured lower median PFAA dust concentrations than in Strynar and Lindstrom⁴⁶ and higher PFAA concentrations than the present study. These results, and their differences over time (Figure 5), may reflect the phase-out of PFOS and PFOA from consumer products since 2000.

Winkens et al.¹⁷ measured PFAS in floor dust from children's bedrooms in Finland and found that FTOHs and diPAPs dominated the dust samples, similar to the results of this current study. However, U.S. residential dust concentrations were higher than Finnish samples for 6:2 diPAP (113 vs 54 ng/g) and for 8:2 FTOH (1440 vs 46 ng/g). FTOHs have been found at high levels in waterproofing agents, carpets, and textiles,⁵⁷ and this may explain why FTOHs are the most abundant PFAS class measured in these dust samples collected from living quarters.

Limitations. There are several limitations to this present study. House dust samples were only collected in North Carolina, and it is possible that there are regional differences in home construction or building practices that influence PFAS dust levels. Furthermore, we measured 17 PFAS compounds using our liquid chromatography and GC techniques in this study, but there are thousands of different PFAS. At the time when the house dust samples were analyzed, an analytical standard for 10:2 FTOH was not available in our laboratory, and as a consequence, 10:2 FTOH was quantified in the fire station dust but not in house dust. Additionally, we did not measure 6:2 fluorotelomer sulfonate (6:2 FTS), a PFAS that has been found in some AFFF formulations.⁵⁸ Another limitation is that PFBS had a relatively high detection limit in our study compared to the other analytes; this may explain why PFBS had a low detection frequency in our study despite PFBS being used to replace longer-chain PFAS.

Fire station dust was not retrieved using the same standardized techniques or equipment as the house dust. Fire station samples were collected by firefighters rather than trained researchers, so we cannot guarantee that collection methodologies were consistent. For example, Canadian and U.S. fire stations used different vacuuming equipment, and a few fire stations in British Columbia collected combined dust from the interiors of their fire engine trucks as well as their living quarters. In contrast, the house dust was collected in a standardized way by a small team of researchers. A metaanalysis by Mitro et al. (2016)⁵⁹ demonstrated the variability among indoor dust studies in the collection method and storage conditions. Mitro et al. (2016) noted that methodological consistency may be improved by using extraction thimbles in the crevice tool of vacuum cleaners instead of sampling from used bags.⁵⁹ Different brands of vacuum cleaners and bags could also contribute to dust data variability.60

In addition, house dust samples were collected at only a single point in time (i.e., during one home visit), while fire station dust was sampled and combined over the period of 1 month. This present study also only examined dust found in

the main living quarters of homes and fire stations, which may not represent PFAS exposure in other rooms of the buildings. While this present study explored some of the differences between PFAS dust concentrations in fire stations and homes, the relatively low number of fire stations somewhat limits generalizability. With these limitations, it is difficult to draw conclusive information regarding the factors influencing dust PFAS concentrations.

The lack of strong associations between the survey data and dust PFAS concentrations highlights a need for more research in understanding what drives PFAS dust concentrations. Survey data in this study were limited by participation, and questionnaires used at fire stations were originally designed to explore predictors of flame retardants, not necessarily PFAS. Our results suggest that fire stations in the United States have higher dust PFAS concentrations than Canadian fire stations. However, it is unclear if this difference could be driven by building materials or use of specific products used within those facilities (e.g., different types of AFFF) or by differences in how and when the dust was collected.

Future Implications. Moving forward, it will be important to consider the impact of precursors on PFAS exposure as these were found at the highest concentrations in our dust samples. Identifying the sources of FTOHs in the dust is an important issue. Previous studies have detected FTOHs in durable water-repellant clothing, 61 so it is possible that firefighting turnout gear or other textiles contribute FTOHs to indoor dust. Additionally, we detected FTOHs at significantly higher levels in samples with greater amounts of carpeting (Figure S5), suggesting that carpeting may be a source of FTOHs. FTOHs are used to treat paper and textiles and have been found at high levels in carpets, textiles, and waterproofing and cleaning agents.^{5,57} 6:2, 8:2, and 10:2 FTOHs have been found in impregnating agents and some AFFFs. 62 Notably, current commercial mixtures of AFFF contain 6:2 FTOH and 8:2 FTOH, while AFFF produced before 2010 did not contain them.⁶² FTOHs are also used in the synthesis of fluorotelomer-based products, and some estimate that residual FTOHs may remain in these products at about 2% by weight.⁶³ Fluorotelomer-based products (FTOHs and PAPs) are thought to be indirect sources of PFCAs in the environment through several means, namely, by product degradation, by PFCA impurities, or through fluorotelomer-based AFFF.⁶³ There is also evidence that atmospheric degradation of 8:2 FTOH and other FTOHs are a source for global PFCA pollution.^{64,65}

The dust concentrations reported in this present study show that FTOHs and diPAPs are abundant in residential dust and thus have the potential to lead to PFAA exposure. Additionally, the maximum values we report for the FTOHs, diPAPs, FOSEs, and PFOS were orders of magnitude higher than the median concentrations, indicating greater exposure potential for some residents and firefighters. Differentiating sources of PFAS in dust should also be explored in more depth in future studies. Comparing the relative amounts of different PFAS subclasses or branched versus linear isomers 66 could give better insight into which sources of PFAS are most contributive. More research is needed to understand the links between PFAA precursors in indoor environments and their contribution to PFAS in dust and how this relates to overall PFAS exposure in people.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c04869.

Additional methods; list of PFAS analytes measured in dust; MS conditions (m/z ions) for quantifying PFAS analytes in dust samples; average dust levels in house dust SRM 2585 and relative standard deviation; fire station dust concentrations stratified by country; characteristics of 49 fire stations included in this present study; characteristics of 184 residential homes included in this present study; summary of median PFAS dust concentrations in U.S. house dust reported in the published literature and the present study; PFAS profiles in dust from homes and fire stations; ΣPFAS concentrations in dust samples by region; $\Sigma PFAS$ dust concentration and survey responses for residential homes; square footage area of fire station living quarters and PFAS dust concentrations; carpeting and FTOH dust concentrations in fire stations and homes; and PFAS concentrations in house dust in 2000-2001 and 2014-2016 (PDF)

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Notes

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