

Per- and polyfluoroalkyl substances (PFAS) and total fluorine in fire station dust

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Supplemental Information (SI)

Fire Station Characteristics

All the stations employed career firefighters rather than volunteers. At the stations, the firefighters perform activities such as training, writing reports, preparing and eating meals, resting, and sleeping. Station floors were generally reported to be cleaned on a daily or weekly basis (with some compliance issues) by either sweeping, mopping, vacuuming, or hosing. Eight stations conduct training with foam in-house once or twice per year but use a reportedly fluorosurfactant-free foam. The remaining stations either conduct annual training off-site with foam or not at all; one even uses dish detergent instead of foam for training. Most of the station buildings were built between the 1930s and 1980s, except one in 2014 and one in the 1880s.

Methods for Fluorine Analysis

The fire station dust samples were analyzed for total fluorine by the Nuclear Science Laboratory at the University of Notre Dame (Notre Dame, IN). For each dust sample, we manually removed the dust from its cellulose extraction thimble and sieved the dust through a 2 mm stainless steel mesh. The dust samples were then placed into small, thin walled (0.05 mm) polyethylene bags from Uline (Pleasant Prairie, WI). Prior to dust sample collection, a media blank for the cellulose extraction thimble was determined by the laboratory to not contain fluorine.

For fluorine measurements with particle-induced gamma ray emission (PIGE) as described previously,¹ the sample bags were mounted on stainless steel target ladder frames. Each sample was irradiated by a beam of 4.0 MeV protons with an intensity between 40–60 nA for about 180 seconds. The intensity of the beam was monitored with periodic faraday cup measurements. For each measurement, the intensity was also normalized based on the 770 keV background-

corrected peak from the ^{40}Ar reaction in air. To measure the gamma rays produced, a 20% High-Purity Germanium detector (Mirion Technologies; Meriden, CT) was placed at about 70 degrees to the incident beam and less than 3 cm from the target. Two gamma rays at 110 keV and 197 keV for the ^{19}F reaction were background-corrected and integrated to determine fluorine content in the samples.

The method detection limit (MDL) was obtained from the linear fit to a series of five inorganic standards between 0 and 2000 $\mu\text{g/g}$ of sodium fluoride mixed with cellulose nitrate (Sigma Aldrich; St. Louis, MO) which was previously determined to be fluorine free. The slope and intercept of these external fluoride standards were used to convert the detected gamma rays associated with ^{19}F into a concentration, and the MDL was determined to be approximately 25 $\mu\text{g/g}$ in the dust samples.

The six field blanks had an average fluorine concentration of 6.8 $\mu\text{g/g}$, which was lower than any uncorrected levels of fluorine in the dust samples (minimum 13 $\mu\text{g/g}$). All fluorine data were blank-corrected using this average value prior to data analyses.

For quality assurance and quality control (QA/QC), 16 percent of the dust samples were routinely split into two and analyzed as replicates for fluorine measurements. These replicate samples had relative percent differences up to 11% due to natural dust heterogeneity and limits on instrument precision. In addition, 10% of all samples were measured twice with the instrument, which yielded 6% relative percent differences due to limits on instrument precision.

Methods for PFAS Analysis

Of the fire station dust samples collected in living rooms, locker areas, and bays ($n=39$), Vista Analytical Laboratory (El Dorado Hills, CA) analyzed the first 22 samples in February 2019 and we analyzed the remaining 17 dust samples as well as the six wipes of turnout gear at the Environmental Contaminants Laboratory of the Harvard Paulson School (Cambridge, MA) in May 2019. The sample selection methods are described more in the main manuscript.

The subsets of dust and wipe samples were analyzed for 24 per- and polyfluoroalkyl substances (PFAS), including perfluoroalkyl acids (PFAAs) and PFAA precursors listed in the main manuscript. Average sample MDLs are presented in Table S1 for both laboratories. The second phase of fire station dust samples and the six gear wipes were additionally analyzed by the Harvard Paulson School for both branched and linear isomers of PFHxS and PFOS, as well as three additional compounds whose measured concentrations in the dust samples were all less than 0.0286, 5.56, and 0.948 ng/g respectively: dodecafluoro-3H-4,8-dioxanonanoate (ADONA), perfluoro-1-hexane sulfonamide (FHxSA), and perfluoro-1-butane sulfonamide (FBSA). In the main data analyses of the manuscript, the isomers were integrated for total concentrations.

Methods for the Environmental Contaminants Laboratory

Dust and wipe samples were extracted following previously published methods with minor modifications.^{2,3} Briefly, dust samples were weighed out (~100 mg) into 15mL polypropylene tubes and for wipes, the entire wipe sample was extracted in a 15mL polypropylene tube. Samples were spiked with internal standards and/or native standards for sample recovery spikes. PFAS standards (27 native compounds and 19 isotopically labeled compounds) were purchased

from Wellington Laboratories (Guelph, Canada). Once spiked, samples were vortexed and left overnight to allow standard to equilibrate into sample matrix. For extraction, methanol was added to the sample (5 mL for dust and 10 mL for wipes), vortexed, shaken, sonicated (30 mins) and centrifuged (10 mins). Methanol was decanted into a second labeled tube and the extraction procedure repeated a second time. Methanol extracts were blown to dryness under nitrogen gas and then reconstituted in 0.75 mL methanol. The extract was mixed with 25 mg of Envi-carb and centrifuged for further clean-up. The supernatant (0.5 mL methanol) was mixed with 0.5 mL deionized water prior to analysis.

Samples were analyzed with an Agilent (Santa Clara, CA) 6460 triple quadrupole liquid chromatograph-tandem mass spectrometer (LC-MS/MS), equipped with an Agilent (Santa Clara, CA) 1290 Infinity Flexible Cube as described previously⁴ with minor modifications. LC-MS/MS blanks and calibration standards were prepared with 50:50 methanol:water and internal standard concentrations matching the samples. The 12-point calibration curve ranged from 1 to 20,000 ng/L and continuing calibration verification samples were included throughout the sample run. Branched PFOS and PFHxS isomers were quantified separately from linear PFOS and PFHxS isomers, and the sum of branched and linear is reported in the manuscript. The mobile gradient method was modified to encompass additional compounds with a 17.5-minute runtime. The column temperature was 50°C. The triple quadrupole tandem mass spectrometer was operated in dynamic multiple reaction monitoring (dMRM) mode using negative electrospray ionization (ESI) and a source temperature of 300°C.

QA/QC for the Environmental Contaminants Laboratory

For QA/QC, the laboratory analyzed 16 methanol instrument blanks, three method blanks, three method spikes, two dust sample spikes, two dust field blanks, and one wipe field blank. The instrument blanks were all less than the MDL for all PFAS analytes except for some that were less than the method quantification limit (MQL) of 0.0807 ng/g for PFTeDA (MDL: 0.0242 ng/g) and less than the MQL of 0.1696 ng/g for 6:2 FtS (MDL: 0.0509 ng/g). The method blanks, which were blank tubes that went through the extraction procedure, were also all less than the MDL, or MQL in the case of one blank for PFTeDA and one blank for PFOA (MQL: 0.7589 ng/g; MDL: 0.2277 ng/g). The method spikes consisted of three tubes spiked with 100, 1,000, and 10,000 ng/L of native PFAS that went through the extraction procedure. The recoveries for method spikes ranged from 96.6% to 166% for the 24 PFAS and from 96.6% to 142% for the ten PFAS detected in over 50% of our dust samples. The sample spikes consisted of two dust samples spiked with 1,000 and 10,000 ng/L of PFAS. For PFAS detected in over 50% of samples, the recoveries ranged from 48.3% to 130% for one sample spike and from 109% to 134% for the other sample spike. The variability in sample spikes likely results from the natural heterogeneity in the spiked and non-spiked dust. The dust field blanks had values all less than the MDL, or MQL in the case of PFOA and 8:2 FtS (MQL: 0.1261 ng/g; MDL: 0.0378 ng/g). The wipe field blank had values between 0 and 286 ng for the PFAS analytes, which were subtracted from the sample wipe levels. The highest detected level in the wipe blank (286 ng) was for 6:2 FtS, which for comparison was measured at up to 5220 ng (before blank correction) in the sample wipes.

Methods for Vista Analytical Laboratory

First, the Nuclear Science Laboratory prepared the samples by weighing the dust samples (~ 250 mg) into 15mL polypropylene tubes with 10 mL of methanol. The sample tubes were sonicated for eight hours and then filtered through a methanol-washed qualitative filter to remove particulates (Whatman). The eluents were then shipped to Vista Analytical Laboratory with parafilm and on ice for PFAS analysis.

At Vista, the dust sample extractions were spiked with 19 isotopically labeled compounds. For sample recovery spikes, the 19 isotopes and 24 native standards were purchased from Wellington Laboratories. The extracts were cleaned using Strata X 33u Polymeric Reversed Phase SPE at a rate of 10ml/min and eluted using methanol. The eluted extracts were concentrated using a steady stream of nitrogen before analysis. The dust extractions were analyzed for 24 PFAS by following Method 537.1 by the U.S. Environmental Protection Agency (EPA)⁵ with additional isotopes and with compliance to the Department of Defense (DoD) Quality Systems Manual (QSM) Table B-15.⁶ Samples were analyzed with a triple quadrupole LC-MS/MS using ESI and a source temperature of 150°C. LC-MS/MS blanks and the calibration curve were prepared with 50:50 methanol:water and internal standard concentrations matching the samples. The 5-point calibration curve ranged from 250-500,000 ng/L and calibration quality controls were included throughout the sample run.

QA/QC for Vista Analytical Laboratory

For QA/QC, the laboratory analyzed one methanol instrument blank, four method blanks, four method spikes, spikes in all dust samples, and two dust field blanks. The instrument blank and method blanks were less than the MDL for all PFAS analytes. The recoveries for the method

spikes ranged from 77.3% to 112% for the 24 PFAS analytes and from 30.1% to 98.5% for the 19 isotopes. The 22 dust samples had recoveries ranging from 26.0% to 128% for all 19 isotopes, or 65.7% to 128% for the ten isotopes for PFAS detected in over 50% of our dust samples. The field blanks had values all less than the MDL except for one blank with PFOA measured at 4.24 ng/g. PFOA concentrations in all dust samples were blank-corrected by subtracting the average of this 4.24 ng/g value and the three other field blanks <MDL (substituted with zero; including the field blanks analyzed at Harvard).

Summary Statistics of Fluorine and PFAS Concentrations

Geometric means and geometric standard deviations for concentrations of total fluorine and PFAS by room type are shown in Table S2.

Branched Isomers of PFOS and PFHxS in Dust

Results

Among two living rooms and six locker areas of the second phase of dust samples that had detectable levels of PFOS or PFHxS, the median fraction of branched PFOS was 30.7% (range: 26.3–48.0%) and the median fraction of branched PFHxS was 40.4% (range: 17.3–79.4%). The median concentrations of branched isomers were 6.52 ng/g (range: 1.48–20.8 ng/g) for PFOS and 1.07 ng/g (range: 0.496–2.28 ng/g) for PFHxS. PFOA, N-EtFOSAA, and PFDS were also present as branched isomers in the dust samples but could only be reported as integrated with the linear concentrations.

In the six gear wipes, branched isomers made up a median 55.6% of total combined PFOS (range: 0–88.7%) and a median 46.4% of total PFHxS (range: 19.8–65.2%). Branched isomers of N-EtFOSAA and PFDS were also present in the wipes but could not be reported separately.

Discussion

The median fractions 31% and 40% of branched isomers for PFOS and PFHxS in a subset of dust samples suggest that the presence of these chemicals likely originated from manufacture through electrochemical fluorination (ECF), as opposed to the relatively newer process of telomerization which only yields linear isomers. ECF was used historically by the major global manufacturer until the early 2000s, although some other manufacturers may still employ the process.^{7,8} Therefore, PFOS and PFHxS in the fire station dust samples may reflect legacy sources from the presence or use of older products; this also makes sense given that the levels of these two chemicals were relatively lower than found in past studies. PFOS and PFHxS were also present as branched isomers in the gear wipe samples at median fractions 56% and 46% respectively, although they were not the primary components. One possibility is that these two particular chemicals contaminated the gear from combustion of older consumer products during fires. The other PFAS present in the samples could not be analyzed separately for branched and linear isomers.

Table S1. Average sample method detection limits (MDLs) in ng/g for measurement of 24 PFAS in the fire station dust samples by the Harvard Environmental Contaminants Laboratory and Vista Analytical Laboratory.

Analyte	Average Harvard MDL	Average Vista MDL
PFBA	3.6302	1.243
PFPeA	1.7876	1.795
PFHxA	1.4143	1.803
PFHpA	0.5058	1.820
PFOA	0.2277	2.097
PFNA	0.1191	1.580
PFDA	0.1365	2.273
PFUnDA	0.0546	3.145
PFDoDA	0.0658	2.450
PFTTrDA	0.0285	1.083
PFTeDA	0.0242	1.758
PFBS	0.8478	3.224
PFPeS	0.4983	4.353
PFHxS	0.4108	2.753
PFHpS	0.3003	1.510
PFOS	0.2612	7.506
PFNS	0.2410	12.71
PFDS	0.1644	1.785
4:2 FtS	0.2891	4.387
6:2 FtS	0.0509	2.034
8:2 FtS	0.0378	2.532
N-MeFOSAA	0.1537	2.683
N-EtFOSAA	0.3229	2.851
FOSA	0.0595	2.016

Table S2. Geometric means (GM) and geometric standard deviations (GSD) for concentrations of total fluorine ($\mu\text{g/g}$; $n=88$) and PFAS^a (ng/g ; $n=39$) in dust samples from different rooms in 15 fire stations in Massachusetts.

Analyte	Geometric Mean (Geometric Standard Deviation)			
	All Samples	Living Room	Gear Locker Area	Apparatus Bay
<i>In $\mu\text{g/g}$:</i>	$n=88^b$	$n=15$	$n=15$	$n=15$
Total Fluorine	136 (2.75)	79.2 (3.16)	291 (1.42)	227 (2.91)
<i>In ng/g:</i>	$n=39^c$	$n=11$	$n=14$	$n=14$
Σ_{24} PFAS	131 (2.94)	204 (4.16)	175 (2.38)	69.3 (1.89)
Σ_6 Precursors ^d	59.5 (4.17)	98.7 (6.91)	83.0 (3.12)	28.7 (2.55)
Σ_{11} PFCAs ^e	31.7 (2.29)	30.8 (2.26)	49.4 (2.53)	20.9 (1.61)
Σ_7 PFSA ^f	21.2 (2.46)	28.9 (3.07)	25.8 (2.10)	13.7 (2.07)
6:2 FtS	11.9 (4.42)	11.6 (5.49)	18.9 (4.45)	7.72 (3.50)
N-EtFOSAA	13.3 (7.99)	41.4 (14.6)	13.5 (6.35)	5.38 (3.66)
8:2 FtS	7.55 (3.26)	6.31 (3.08)	12.8 (3.87)	5.12 (2.42)
PFOA	4.64 (3.47)	4.85 (4.19)	7.75 (3.20)	2.68 (2.69)
PFOS	7.40 (2.75)	8.99 (3.11)	10.8 (2.82)	4.35 (1.93)
PFNA	2.25 (2.81)	2.11 (2.49)	3.56 (3.43)	1.49 (2.09)
PFHxA	4.45 (3.61)	3.47 (3.86)	9.19 (3.30)	2.63 (2.80)
PFHpA	2.25 (2.56)	1.93 (2.49)	3.88 (2.84)	1.46 (1.77)
PFDA	2.32 (2.32)	1.80 (2.04)	4.18 (2.77)	1.57 (1.25)
PFUnDA	1.80 (2.29)	1.72 (1.83)	2.49 (2.47)	1.34 (2.32)
PFDoDA	1.69 (1.99)	1.62 (1.86)	2.43 (2.28)	1.22 (1.45)
PFTTrDA	.701 (2.39)	.680 (2.09)	.916 (2.80)	.550 (2.16)
PFTeDA	1.04 (1.88)	1.08 (1.89)	1.33 (1.97)	.796 (1.64)
PFBA	2.42 (2.81)	2.35 (2.81)	3.31 (3.43)	1.82 (2.14)
PFDS	1.24 (3.83)	2.52 (6.68)	1.32 (2.28)	.664 (2.67)
PFPeA	2.15 (2.27)	2.56 (2.48)	2.82 (2.55)	1.43 (1.49)
FOSA	.569 (3.49)	.814 (3.67)	.520 (3.08)	.470 (3.88)
N-MeFOSAA	.777 (3.19)	1.13 (2.85)	.786 (2.75)	.573 (3.89)
PFHxS	1.80 (1.95)	2.11 (1.86)	2.26 (2.18)	1.27 (1.54)
PFBS	1.71 (1.98)	1.77 (1.83)	2.21 (2.09)	1.28 (1.86)
PFPeS	1.28 (2.82)	1.68 (2.86)	1.14 (2.79)	1.17 (2.94)
PFHpS	.572 (2.19)	.734 (2.24)	.525 (2.16)	.513 (2.21)
4:2 FtS	1.06 (3.69)	1.34 (3.80)	.998 (3.67)	.928 (3.90)
PFNS	1.67 (7.06)	2.75 (6.56)	1.20 (7.61)	1.58 (7.38)

^a Sorted by detection frequency.

^b Samples analyzed for total fluorine included locker areas, bays, living rooms, kitchens, and sleeping quarters from all 15 stations, as well as gym samples from 13 stations.

^c All bay, locker, and living room dust samples that had sufficient mass of dust were further analyzed in the laboratory for PFAS.

^d Precursor compounds included FOSA, 4:2 FtS, 6:2 FtS, 8:2 FtS, N-MeFOSAA, and N-EtFOSAA.

^e PFCAs included PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTTrDA, and PFTeDA.

^f PFSA^s included PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, and PFDS.

Table S3. Summary of PFAS concentrations (ng/g) in dust from this fire station study compared to other U.S. studies.⁹⁻¹⁶

	This study	This study	Fraser (2013)	Fraser (2013)	Goosey (2011)	Hall (2020)	Hall (2020)	Kárasková (2016)	Knobeloch (2012)	Scher (2019)	Strynar (2008)	Wu (2015)
Sample Size	11	11	31	31	10	49	184	20	39	19	112	42
Region	MA, US	MA, US	MA, US	MA, US	CO, US	US; Canada	NC, US	IN, US	WI, US	MN, US	NC/OH, US	CA, US
Environment	Fire station living rooms	Fire station locker areas	Offices	Homes	Homes	Fire station living areas	Homes	Homes	Homes	Homes (at-risk water)	Homes, some daycares	Homes of older adults
Year of Samples	2018	2018	2009	2009	2007-9	2015-18	2014-16	2013	2008	2010	2000-1	2008-9
Measure of Center	Median	Median	GM	GM	Median	Median	Median	Median	Median	Median	Median	Median
Center [Max]:												
6:2 FtS	9.85 [316]	20.1 [310]	--	--	--	--	--	--	--	--	--	--
N-EtFOSAA	87.5 [1800]	7.84 [299]	--	--	--	--	--	--	--	--	--	--
8:2 FtS	6.56 [66.1]	11.6 [131]	--	--	--	--	--	--	--	--	--	--
PFOA	5.77 [30.6]	9.69 [60.0]	32.0 [336]	23.7 [894]	240 [1800]	17.6 [791]	7.9 [2350]	9 [318]	44 [420]	51 [970]	142 [1960]	48.05 [728]
PFOS	7.57 [78.9]	7.97 [91.5]	14.6 [98.2]	26.9 [280]	310 [930]	64.5 [74370]	4.4 [2810]	14.1 [239]	47 [1100]	67 [2000]	201 [12100]	34.05 [1040]
PFNA	1.59 [15.1]	2.79 [29.8]	63.0 [639]	10.9 [1420]	--	7.2 [203]	3.3 [208]	3.9 [62.9]	12 [280]	26 [450]	7.99 [263]	11.85 [883]
PFHxA	1.82 [76.9]	16.9 [40.9]	10.8 [102]	8.65 [1380]	--	ND [1150]	8.5 [1380]	6.5 [190]	ND [180]	29 [240]	54.2 [1250]	--
PFHpA	1.53 [11]	4.88 [22.6]	27.6 [388]	12.0 [586]	--	ND [382]	8.9 [713]	3.6 [86.7]	17 [140]	23 [260]	50.2 [1150]	--
PFDA	1.87 [4.54]	3.24 [42.0]	46.5 [492]	ND [26.8]	--	2.5 [137]	6.2 [4130]	1.8 [64.0]	5.7 [60]	13 [370]	6.65 [267]	8.20 [355]
PFUnDA	2.11 [3.17]	2.10 [17.5]	19.0 [373]	ND [39.4]	--	--	--	1.2 [13.1]	3.1 [48]	7.2 [67]	7.57 [588]	--
PFDoDA	1.92 [2.83]	1.67 [19.7]	40.0 [481]	ND [13.3]	--	--	--	0.6 [9.0]	5 [41]	8.2 [190]	7.78 [520]	--
PFTrDA	.849 [2.68]	.688 [7.03]	21.6 [768]	ND [10.3]	--	--	--	ND [2.1]	1.2 [11]	--	--	--
PFTeDA	1.38 [1.77]	1.13 [7.83]	18.6 [367]	ND [11.2]	--	--	--	0.8 [3.0]	3.7 [24]	--	--	--
PFBA	2.57 [18.4]	3.50 [27.5]	ND [148]	13.9 [999]	--	4.6 [213]	ND [546]	--	3.5 [64]	24 [200]	--	--
PFDS	1.41 [185]	1.13 [12.5]	--	--	--	--	--	2.8 [9.8]	--	--	--	--
PFPeA	1.51 [14.2]	2.27 [16.6]	ND [27.5]	ND [249]	--	ND [1410]	ND [135]	--	--	6.2 [66]	--	--
FOSA	1.58 [2.27]	.865 [1.74]	--	--	69 [110]	--	--	--	--	--	--	--

N-MeFOSAA	1.80 [2.70]	1.24 [2.32]	--	--	--	--	--	--	--	--	--	--
PFHxS	2.17 [6.24]	1.79 [12.2]	ND [18.5]	ND [430]	240 [560]	6.8 [8280]	2.0 [694]	8.7 [84.4]	16 [1000]	18 [790]	45.5 [35700]	5.55 [1050]
PFBS	2.17 [3.25]	2.15 [7.48]	ND [12.0]	ND [4.98]	--	ND [2650]	ND [320]	0.9 [2.6]	1.8 [31]	ND [58]	9.11 [1150]	--
PFPeS	2.93 [4.38]	1.91 [3.76]	--	--	--	--	--	--	--	--	--	--
PFHpS	1.09 [1.52]	.884 [1.30]	--	--	--	--	--	ND [2.9]	9.9 [37]	--	--	--
4:2 FtS	2.95 [4.42]	2.64 [3.79]	--	--	--	--	--	--	--	--	--	--
PFNS	8.56 [12.8]	3.97 [11.0]	--	--	--	--	--	--	--	--	--	--

GM = geometric mean; ND = not detected.

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