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

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are a class of over 4,700 fluorinated compounds used in industry and consumer products. Studies have highlighted use of aqueous film-forming foams (AFFFs) as an exposure source for firefighters, but little is known about PFAS occurrence inside fire stations, where firefighters spend most of their shifts. In this study, we aimed to characterize PFAS concentrations and sources inside fire stations. We measured 24 PFAS (using LC-MS/MS) and total fluorine (using particle-induced gamma ray emission) in dust from multiple rooms of 15 Massachusetts stations, many of which (60%) no longer use PFAS-containing AFFF at all and the rest of which only use it very rarely. Compared to station living rooms, turnout gear locker rooms had higher dust levels of total fluorine ($p < 0.0001$) and three PFAS: perfluorohexanoate (PFHxA), perfluoroheptanoate (PFHpA), and perfluorodecanoate (PFDoDA) ($p < 0.05$). These PFAS were also found on six wipes of station turnout gear. By contrast, the dominant PFAS in living rooms was N-ethyl perfluorooctane sulfonamidoacetic acid (N-MeFOSAA), a precursor to perfluorooctane sulfonate (PFOS) that still persists despite phase-outs almost two decades ago. The Σ_{24} PFAS accounted for less than 2% of fluorine in dust ($n=39$), suggesting the potential presence of unknown PFAS. Turnout gear may be an important PFAS source in stations due to intentional additives and/or contamination from firefighting activities.

INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are a class of over 4,700 anthropogenic highly fluorinated aliphatic chemicals used by diverse industries and in consumer products.^{1,2}

They are detectable in the serum of over 98% of Americans,³ and the perfluoroalkyl acids (PFAAs) do not appreciably degrade in the environment.^{4,5} Exposures to PFAS have been

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Conflict of Interest

The authors declare no conflict of interest.

linked to adverse health impacts such as thyroid dysfunction, immune system suppression, impaired fetal development, high cholesterol, obesity, and diabetes.^{1,6–9} High levels of PFAS exposure in occupational or contaminated communities have also been associated with kidney and testicular cancer.^{10–12}

Because of their stain- and water-repellant properties, PFAS are commonly found in non-stick cookware, disposable food packaging, carpet, upholstery, and outdoor clothing.^{13–16} PFAS are also used as thermally stable surfactants in aqueous film-forming foams (AFFFs) to extinguish hydrocarbon fuel-based fires during firefighting and training activities.¹⁷ In fact, AFFF use at airports and military bases is a major source of environmental water and soil contamination that exposes the general population.^{18,19}

Firefighters are an understudied, potentially vulnerable occupational population exposed to PFAS. Research has shown that firefighters may experience higher risks of kidney and testicular cancer compared to general or occupational populations, although the influence of PFAS exposures is uncertain.^{20–22} Several biomonitoring studies have found elevated PFAA exposures for firefighters compared to the general population.^{23–27} For example, perfluorodecanoate (PFDA) concentrations in serum from California firefighters were three times higher than the U.S. general population.²⁸ Responders to the 2001 collapse of the World Trade Center had two-fold higher levels of perfluorohexane sulfonate (PFHxS) and perfluorooctanoate (PFOA) in serum than the general population, with sources including contaminated smoke and firefighting foams.²⁹ One recent study found that dust collected in living areas of fire stations had a fifteen times higher median level of perfluorooctane sulfonate (PFOS) and three times higher median level of PFHxS than homes, with the sources thought to include their use as components in AFFF and stain-resistant carpet burned during fires.³⁰ However, most previous studies have focused on firefighter exposures to PFAS while out fighting fires or training, not potential sources of PFAS contamination specific to the fire station itself.

Many firefighters spend the majority of their working time at the fire station.³¹ Sources of indoor exposure could include PFAS-containing products stored at the station (e.g. AFFF, turnout gear, and consumer products) and residual PFAS contamination brought back to the station after firefighting activities (e.g. from smoke, gear, and AFFF use). For example, firefighting turnout gear must be heat stable and water resistant,³² a property often imparted to clothing and other products using PFAS.⁵ A recent study detected PFAS in several thermal liners, moisture barriers, and/or outer shells of used or unused turnout gear samples.³³ Prior studies have also found that many other types of volatile and semi-volatile chemicals can contaminate turnout gear during firefighting and then later off-gas or lead to cross contamination.^{34–36} This may extend to contamination of turnout gear by PFAS that firefighters encounter when exposed to smoke from fires involving consumer products or when using AFFF at a fire. Since turnout gear is stored inside fire stations, PFAS on or in the gear may be carried back to the stations and thus extend firefighter exposures to PFAS beyond their time while out fighting fires.

The prior firefighter biomonitoring studies have only analyzed 8–20 targeted PFAS, so these studies could underestimate total PFAS exposure. The production and use of different PFAS

in consumer products and AFFF have shifted over time.³⁷ Although two widespread legacy PFAS (PFOS and PFOA) were phased out of AFFFs in the early 2000's, stockpiles of older AFFF formulations continue to be used, and newer AFFF formulations frequently contain large quantities of unidentified replacement PFAS.^{38–40} New screening methods, such as particle-induced gamma ray emission (PIGE), provide an alternative to be able to assess total concentrations of elemental fluorine, which captures both known and unknown PFAS, and to estimate the fraction of unidentified PFAS in a sample. Several studies have used PIGE to non-destructively, rapidly measure total fluorine on solid-phase surfaces, including PFAS-containing firefighter gear, food packaging, paper, and textiles.^{33,41–43} To date, there are no published studies on measurements of total fluorine in indoor dust, which is a reservoir for chemicals, an important route of exposure, and an indicator of indoor environmental quality.^{44,45}

The main objective of this pilot study was to evaluate potential sources of firefighter exposures to PFAS inside fire stations. Specifically, we aimed to better understand: 1) the burden of 24 targeted PFAS and total fluorine in dust samples from 15 Massachusetts fire stations, many of which no longer (or rarely) use AFFF that contains PFAS, 2) factors contributing to differences in PFAS concentrations and profiles by room type and station policies, and 3) the relationship between profiles of PFAS in dust and in six wipes of turnout gear from stations.

METHODS

Dust and Wipe Sample Collection

Dust and wipe samples were collected at 15 career (non-volunteer) fire stations in Massachusetts, USA. We leveraged an existing partnership with the Boston Fire Department⁴⁶ to select the first eight stations, and seven others volunteered after a broad invitation for fire departments around Massachusetts. Our study included stations with varying building types, emergency call loads, and neighborhood types (Table 1).

We collected 89 dust samples from different room types at fire stations between June and December 2018. At each station, we sampled the following rooms: apparatus bays (where trucks are kept), turnout gear locker areas, kitchens, living rooms, sleeping quarters, and gyms. One station did not have a gym and another gym had insufficient dust mass for analyses. We collected each dust sample by vacuuming the floor for 10 minutes. To avoid contact between the dust and vacuum, the dust was collected in a cellulose extraction thimble (Whatman International; Maidstone, UK) secured in a crevice tool attachment on a vacuum cleaner (Dyson CY18; Chicago, IL) with a nitrile rubber o-ring around the thimble. At a given station, a separate clean crevice tool was used for each sample. Equipment was cleaned in between station sampling with isopropyl alcohol wipes and hot tap water and left to air dry in the laboratory. Thimbles were placed in polypropylene centrifuge tubes, sealed in polyethylene bags, and stored at -13°C . Our dust collection followed previously published protocols.^{47,48} After shipment to laboratories, samples were stored at room temperature. As field blanks, six unopened centrifuge tubes with thimbles were carried into the stations and treated in the same manner as samples. Characteristics of the buildings from walk-throughs as well as information on station policies and practices

(such as cleaning, gear washing, training, and AFFF use) from interviews with officers are provided in the supplemental information.

At each station, we collected a wipe sample of one set of turnout gear present in a truck in the apparatus bay following previously published methods.⁴⁹ While wearing nitrile gloves, we wiped a 36 in² area of one arm of a turnout gear jacket using a 3×3 in² sterile gauze pad absorbed with 3 mL isopropyl alcohol. Wipes were stored in polypropylene containers at -13°C. Field blanks were treated in the same manner as samples but not used to wipe any surface. For pilot testing of PFAS in wipes, a subset of six wipe samples were randomly selected equally across terciles of apparatus bay fluorine concentrations (along with one field blank). We only analyzed six wipes as a pilot test because at the time, there were no published studies on PFAS in wipes to our knowledge.

Fluorine Analysis

Dust samples were sieved with a 2-mm stainless steel mesh and placed in 0.05-mm thick polyethylene bags at the Nuclear Science Laboratory at the University of Notre Dame. The samples were analyzed for total fluorine in February 2019 by taking measurements with PIGE spectroscopy through the thin walls of the bags.^{42,43} The average method detection limit (MDL) was 25 µg/g. Total fluorine measurements capture aggregate contributions (without differentiation) from both inorganic fluoride and organofluorine compounds, including PFAS. Additional information on the methods and quality assurance and quality control (QA/QC) is provided in the supplemental information.

Targeted PFAS Analysis

We measured a suite of 24 PFAS using targeted liquid chromatograph-tandem mass spectrometer (LC-MS/MS) techniques. A subset of dust samples ($n=39$) was selected for PFAS analysis from the apparatus bay, gear locker area, and living room for each fire station when possible. The apparatus bays and gear locker areas were chosen because of regular contact with firefighting personal protective equipment (like turnout gear) and potential proximity to AFFF storage. Living rooms were chosen as a reference because these rooms should have lower PFAS contamination from firefighting products than locker or bay areas and lower contamination from food packaging and cookware than kitchens, and dust from sleeping quarters usually had insufficient mass for analysis. All but one station had bay and locker dust samples with sufficient mass, and all but four had living room samples with sufficient mass.

Vista Analytical Laboratory (El Dorado Hills, CA) analyzed the first 22 dust samples in February 2019, and the remaining 17 samples were analyzed in the Environmental Contaminants Laboratory of the Harvard John A. Paulson School of Engineering and Applied Sciences (Cambridge, MA) in May 2019. To ensure comparability in sample measurements within stations, the two laboratories analyzed dust samples from distinct fire stations, except one station had to be split. Stations in the first phase of analysis were randomly selected for an equal distribution across terciles of bay fluorine concentrations. Both laboratories followed strict QA/QC protocols and employed parallel analytical methods using a triple quadrupole LC-MS/MS with negative electrospray ionization (ESI) after

sample extraction with methanol, as described in the supplemental information. Each laboratory analyzed two field blanks. The six gear wipes were analyzed for targeted PFAS at Harvard University.

The subsets of dust and wipe samples were analyzed for 24 PFAS, including PFAAs and PFAA precursors. Targeted compounds and their respective carbon chain lengths (C) included: perfluoroalkyl carboxylates (PFCAs): perfluorobutanoate (PFBA; C-4), perfluoropentanoate (PFPeA; C-5), perfluorohexanoate (PFHxA; C-6), perfluoroheptanoate (PFHpA; C-7), PFOA (C-8), perfluorononanoate (PFNA; C-9), PFDA (C-10), perfluoroundecanoate (PFUnDA; C-11), perfluorododecanoate (PFDoDA; C-12), perfluorotridecanoate (PFTTrDA; C-13), and perfluorotetradecanoate (PFTeDA; C-14); perfluoroalkyl sulfonates (PFASAs): perfluorobutane sulfonate (PFBS; C-4), perfluoropentane sulfonate (PFPeS; C-5), PFHxS (C-6), perfluoroheptane sulfonate (PFHpS; C-7), PFOS (C-8), perfluorononane sulfonate (PFNS; C-9), and perfluorodecane sulfonate (PFDS; C-10); precursors: 4:2 fluorotelomer sulfonate (4:2 FtS; C-6), 6:2 fluorotelomer sulfonate (6:2 FtS; C-8), 8:2 fluorotelomer sulfonate (8:2 FtS; C-10), perfluorooctane sulfonamide (FOSA; C-8), N-methyl perfluorooctane sulfonamidoacetic acid (N-MeFOSAA; C-8), and N-ethyl perfluorooctane sulfonamidoacetic acid (N-EtFOSAA; C-8). Average MDLs are provided in Table S1.

Data Analysis

Fluorine and PFAS levels were blank-corrected by subtracting the mean of all field blanks. Non-detect values were substituted with the MDL divided by the square root of two before analyses. The MDLs were calculated as the sample concentrations at which the signal-to-noise ratio was three. To calculate the amount of measured fluorine explained by the 24 PFAS, the measured concentrations of each PFAS in dust were converted to expected fluorine levels following previously published methods.^{50,51}

Associations between potential predictors and fluorine, total PFAS, or individual PFAS in dust were modeled using multilevel regression models with a random intercept for the station to account for correlation between multiple samples within the same station. Only individual PFAS detected in over 50% of samples were modeled in order to minimize multiple testing issues. Concentrations were not normally distributed (based on a Shapiro-Wilk test and histogram) so the data were log transformed before modeling. Model estimates were transformed back to the linear scale for presentation of results as the percent change in outcome for each covariate. Two binary station-level covariates were included, regular washing of turnout gear after every fire and use of AFFF that contains PFAS, as well as a room-level covariate for floor cleaning frequency to account for differences in dust build-up. The model was defined by Equation (1).

$$\ln(C_{ij}) = \beta_0 + \beta_1 \text{RoomType}_{ij} + \beta_2 \text{AFFF}_j + \beta_3 \text{GearWashed}_j + \beta_4 \text{Cleaning}_{ij} + b_j + \epsilon_{ij} \quad \text{Equation (1)}$$

where C_{ij} is the concentration for the i^{th} room in the j^{th} station, b_j is the random station effect, and ϵ_{ij} is the within-room random error. The data were well-balanced, and an

unstructured covariance pattern was assumed. Statistical significance was evaluated at the $\alpha=0.05$ level. All analyses were conducted in R (version 3.3.1).

RESULTS

Fire Station Practices

The studied fire stations reported that firefighters spend an average 72% of their usually 24-hour shifts inside the station. Most stations (87%) have turnout gear washers and provide two sets of gear to every firefighter. The stations use a 10-year replacement cycle for gear, with varying ages of gear depending on timing of hires. Six unique gear manufacturers are used; 80% of stations purchase from multiple manufacturers. Most station chiefs (87%) reported that gear is washed after every fire, though usually the firefighter's responsibility. One station reported that washing all gear can take two days. Most stations have the gear professionally washed and inspected at least annually, but two stations do not have a consistent policy. Only one station had carpeting in the living room.

Six fire stations (40%) use AFFF or alcohol resistant AFFF (AR-AFFF, for fires involving polar solvents) reported on manufacturer websites to contain fluorosurfactants (assumedly PFAS). Nine stations use an extinguishing foam or suppression agent specified to not contain any fluorosurfactants. All the stations rarely use firefighting foam (at most a few times per year) for flammable liquids and vehicle fires. The foams are stored on the apparatus (truck). No stations train with PFAS-containing foam in-house.

Fluorine Concentrations in Dust

The median total fluorine concentration in sampled fire station rooms was 157 $\mu\text{g/g}$ (range: 6.17–952; $n=88$). By room, the median total fluorine concentrations were 108 $\mu\text{g/g}$ (range: 10.2–491; $n=15$) for living rooms, 296 $\mu\text{g/g}$ (range: 155–651; $n=15$) for the gear locker areas, 271 $\mu\text{g/g}$ (range: 18.2–952; $n=15$) for the apparatus bays, 195 $\mu\text{g/g}$ (range: 66.2–740; $n=13$) for the gyms, 80.2 $\mu\text{g/g}$ (range: 39.2–289; $n=15$) for the sleeping quarters, and 86.2 $\mu\text{g/g}$ (range: 6.17–256; $n=15$) for the kitchens (Table 2). The turnout gear locker areas had the highest median fluorine concentrations, followed by apparatus bays (Figure 1). Geometric means and standard deviations are provided in Table S2.

The multilevel model results indicated that fluorine concentrations were 273% significantly higher (95% confidence interval [CI]: 104–579%; $p<0.0001$) in gear locker areas and 191% higher (95% CI: 59.1–429%; $p=0.0012$) in apparatus bays compared to living rooms on average, after adjusting for covariates (Table 3). Six of the 15 studied stations had AFFF products that reportedly contain fluorosurfactants (PFAS), but these stations did not have significantly different levels of fluorine in dust ($p=0.26$). Regular gear washing ($p=0.46$) and floor cleaning frequency ($p=0.35$) were also not significant predictors of fluorine. Only 3.97% of the variance in log fluorine concentrations was attributable to differences between fire stations, so the vast majority of the variability was explained by room type.

PFAS Concentrations in Dust

PFAS were detected in 92.3% of the subset of dust samples from living rooms, apparatus bays, and turnout gear locker areas (Table 2). The most frequently detected compounds were 6:2 FtS (89.7% of samples), N-EtFOSAA (82.1%), 8:2 FtS (79.5%), PFOA (76.9%), and PFOS (71.8%) (Table 2). The maximum concentration detected for an individual PFAS was 1800 ng/g for N-EtFOSAA, followed by 316 (6:2 FtS), 185 (PFDS), 131 (8:2 FtS), and 91.5 ng/g (PFOS).

Overall, the sum of 24 PFAS (Σ_{24} PFAS) had a median dust concentration of 98.7 ng/g (range: 16.8–2170; $n=39$). The bays had substantially lower Σ_{24} PFAS concentrations (median: 60.1 ng/g; range: 29.9–259; $n=14$) compared to living rooms (median: 170 ng/g; range: 16.8–2170; $n=11$) and gear locker areas (median: 189 ng/g; range: 47.8–723; $n=14$) (Figure 1). The multilevel model results indicated that Σ_{24} PFAS concentrations were 63.5% (95% CI: 24.5–82.0%) significantly lower in apparatus bays compared to living rooms on average ($p=0.015$) but did not significantly differ between locker areas and living rooms ($p=0.84$), after adjusting for covariates (Table 3). Use of AFFF containing PFAS ($p=0.16$), frequent gear washing after fires ($p=0.68$), and cleaning frequency ($p=0.50$) were not statistically significant predictors of Σ_{24} PFAS levels. Similar to fluorine concentrations, only 2.27% of the variance in log Σ_{24} PFAS was attributable to differences between stations as opposed to differences in room type.

The 24 PFAS analytes accounted for up to 1.2% of measured total fluorine concentrations. The bays had the lowest median percent of fluorine accounted for by measured PFAS (median: 0.013%; range: 0.0059–0.50%), compared to living rooms (median: 0.11%; range: 0.0043–1.2%) and gear locker areas (median: 0.038%; range: 0.0095–0.13%).

PFAS Profiles in Dust

Figure 2 shows the median concentrations of each PFAS in dust samples across fire stations by room. The living room dust samples had a substantially higher absolute median concentration and proportion of N-EtFOSAA than the gear locker areas and apparatus bays, while the gear locker area dust samples had a higher absolute median concentration and proportion of 6:2 FtS, 8:2 FtS, PFHxA, PFOA, PFHpA, PFDA, and PFNA.

N-EtFOSAA was the dominant compound in 64% of living rooms ($n=11$), comprising 23–83% of Σ_{24} PFAS concentrations; the other four living room dust samples were each dominated by a different PFAA. In half of samples from turnout gear locker areas ($n=14$), 6:2 FtS or in two cases 8:2 FtS were the highest measured compounds, constituting 18–21% of Σ_{24} PFAS concentrations; another five samples were dominated by N-EtFOSAA and another two by PFNS. In half of samples from apparatus bays ($n=14$), 6:2 FtS was the main compound comprising 14–57% of Σ_{24} PFAS; the remaining samples were mostly comprised of N-EtFOSAA in three cases, PFNS in three, and PFHxA in one.

Results from multilevel models of the ten PFAS detected in over half of samples showed that turnout gear locker areas had significantly higher levels of PFHxA (168%; 95% CI: 35.8–449%; $p=0.012$), PFHpA (104%; 95% CI: 14.5–293%; $p=0.033$), and PFDA (135%; 95% CI: 37.8–310%; $p=0.0060$) in dust compared to living rooms, and apparatus bays had

88.5% significantly lower levels of N-EtFOSAA than living rooms (95% CI: 40.7–97.2%; $p=0.010$), after adjusting for AFFF use, regular gear washing, and cleaning frequency.

PFAS Profiles in Gear Wipes

In the pilot testing of wipes of turnout gear taken from six fire stations, the five highest detected PFAS masses were 30,000 (PFOA), 20,800 (PFNA), 18,200 (PFBS), 12,400 (PFDA), and 5,600 ng/wipe (PFUnDA). The maximum Σ_{24} PFAS mass detected on a gear wipe was 84,500 ng/wipe. PFCAs (including PFOA, PFHxA, PFDA, PFNA, and PFHpA) and 8:2 FtS were consistently detected in all gear wipes (Figure 3). PFCAs made up over half of the Σ_{24} PFAS mass for most of the gear wipes. The 6:2 FtS contributions were 44% and 4.4% in two gear wipes but lower in the others. PFOS was a dominant compound (at 18%) in one gear wipe. Gear was reported to vary in age, washing frequency, and manufacturer, and there were wide ranges in masses of PFAS on gear wipes.

DISCUSSION

We evaluated the occurrence of PFAS inside fire stations and measured fluorine in dust to determine potential unidentified PFAS contamination. The results showed that dust in turnout gear locker areas and adjoining apparatus bays had significantly higher fluorine concentrations compared to living rooms in fire stations. There were also differences in PFAS profiles in dust between the living rooms and the turnout gear locker areas and apparatus bays, indicating different potential sources of PFAS contamination from consumer products versus firefighting gear. The living room dust samples were most often dominated by N-EtFOSAA, and at higher concentrations than in the apparatus bays. N-EtFOSAA has been associated with consumer products such as carpet, non-stick cookware, and food packaging,^{52–54} which would all be expected as products in/near the living areas but not bays. N-EtFOSAA is a precursor compound that degrades to PFAAs such as PFOS and PFOA; all three have largely been phased out of production.^{55,56} Thus, legacy PFAS may persist in indoor environments even after phase-outs due to the continued use of PFAS-containing products with long life spans.

By contrast, the main compound in dust from both turnout gear locker areas and apparatus bays was usually 6:2 or 8:2 FtS. The median concentrations of 6:2 and 8:2 FtS were lower in the bays than locker areas, so bays may experience less cross-contamination from gear stored in the usually adjoining locker areas. Compared to station living rooms, concentrations of PFHxA, PFHpA, and PFDA were significantly higher in dust from turnout gear locker areas based on multilevel models. These five chemicals were also detected at relatively high levels in at least some of the six wipe samples of turnout gear in the stations, whereas N-EtFOSAA was only found at relatively very low levels in the wipes. In fact, every PFCA was consistently detected in all the turnout gear wipes. Although PFAS have not been previously reported in wipes of clothing, the levels of PFHxA, PFHpA, PFDA, and other PFAAs in the gear wipes in this pilot study (ranges: 110–3500 ng, 4.1–2600 ng, and 82–12000 ng, respectively) were typically orders of magnitude higher than the levels on wipes of hands of 60 people in a recent study using the same size gauze and same solvent (ranges: <MDL–0.61 ng, <MDL–5.7 ng, and <MDL–0.61 ng; detection frequencies: 7%, 2%, and 20%, respectively). That study did not measure 6:2 or 8:2 FtS.

The differences suggest that PFAS occurred on the wiped turnout gear from more than just trace contamination.⁵⁷ The PFAA precursors 6:2 FtS and 8:2 FtS are newer replacement chemicals with understudied sources, although they have been recently detected in AFFF and textiles.^{58,59} PFDA, a long-chain legacy chemical, and PFHxA and PFHpA, two short-chain replacements, have been detected in AFFF, carpet, clothing and furniture textiles, food packaging, and building materials.^{13,15,16,59–61} The different PFAS loadings in gear locker area dust may be introduced from storage of turnout gear that is contaminated by: 1) intentional additives of old and/or new PFAS depending on the age of the gear (up to 10 years at the stations), and 2) exposure to old and/or new PFAS in AFFF of varying ages and in smoke from combustion of consumer products of varying ages.

These findings are consistent with a recent study that found detectable levels of 6:2 FtS, 8:2 FtS, PFHxA, PFHpA, PFDA, and several other PFAAs in various layers of four turnout gear garments.³³ Furthermore, a California biomonitoring study reported that firefighters whose turnout gear was not professionally cleaned within the previous year had significantly higher blood levels of two PFCAs (PFOA and PFNA).²⁸ Another firefighter study in Finland reported a potentially unidentified source of exposure, because the two most elevated PFAS (PFNA and PFHxS) in blood samples after AFFF training sessions were not detected above the limit of quantification in the actual AFFF used.²⁵ In comparison, this study found higher median and maximum levels of PFOA and PFNA in dust from turnout gear locker areas than living rooms, although differences did not reach statistical significance. PFOA and PFNA were also found at the two highest levels in turnout gear wipes in the studied fire stations. This study's results corroborate contaminated turnout gear as a potential source of some PFAS, although we need further research on routes of exposure and implications for firefighters.

Station use of PFAS-containing AFFF while fighting fires did not significantly predict either fluorine or Σ_{24} PFAS concentrations in dust, perhaps because 60% of sampled stations only use products specified as PFAS-free. In addition, any AFFF use by these stations is limited to at most a few times per year, which minimizes exposures and cross contamination from gear carried back to the station. Firefighter training with PFAS-containing AFFF always occurs off-site at most twice per year for the studied stations. Because our study was limited to stations in one geographical area with similar policies, the results could underestimate indoor PFAS burdens for other stations across the country that may use AFFF regularly to fight fires or train, may not wash turnout gear, or may store turnout gear in living spaces. In fact, the dust samples from fire station living areas in our study all had lower detected median levels, and order(s) of magnitude lower maximum levels, of PFAAs compared to dust collected in living areas of 49 fire stations in the US and Canada (the authors did not measure the same PFAA precursors as we did) (Table S3).³⁰ Thus, the nonexistent or otherwise rare use of PFAS-containing AFFF by the fire stations in our study may be one reason for the lower PFAA dust levels than the fire stations in that other study, which further motivates shifts towards firefighting products that do not contain any PFAS and that are only used sparingly.

Regular gear washing after fires was associated with non-statistically significant declines in fluorine. However, most stations (87%) reported regularly washing gear, and thus this model

may not have had sufficient statistical power, and individual firefighter washing practices in reality may have differed from the station-wide policies. The impact of gear washing on chemical levels may also depend on the situation. For example, prior work has reported that washing turnout gear reduced levels of PAHs and some flame retardants^{34,36} but increased flame retardants (not PAHs) through cross-contamination when other used gear was washed in the machine.³⁴ Daily cleaning of floors was associated with lower levels of fluorine and PFAS in dust, but the association did not reach statistical significance. This result may be due to the limited statistical power and/or the potential over-reporting of cleaning frequencies based on policy not practice.

Finally, the results showed that the 24 targeted PFAS only accounted for at most 1.2% of total fluorine detected in the dust samples, suggesting the potential presence of unknown non-polymeric and polymeric PFAS.^{42,51} The amount of unexplained total fluorine in the dust samples aligns with previous studies. In a study of several samples of firefighting turnout gear, 17 measured PFAS typically explained only about 1% of the levels of total fluorine.³³ The similar proportion in our fire station dust samples suggests that inorganic fluoride from soil tracked in on the ground⁶² likely did not significantly interfere with our results. In Swedish cosmetics, 39 quantified PFAS represented less than 1.3% of EOF in 28 cosmetics and 11–28% in three other cosmetics with the highest concentrations. The EOF only accounted for an average 9% of total fluorine, which, unlike EOF, would include inorganic fluoride and any PFAS or other organofluorine compounds that were not extractable.^{42,50} In a small sample of Swedish disposable food packaging, 44 PFAS only explained up to 0.28% of EOF and 0.011% of total fluorine.⁵¹ In papers and textiles, concentrations of 73 ionic PFAS accounted for up to 0.41% of total fluorine and four volatile PFAS up to 2.2%; the authors also found that unknown precursor compounds could at least explain up to 14% of the fluorine by using the total oxidizable precursor (TOP) assay,⁴² which measures changes in known PFAA concentrations before and after forced oxidation of PFAA precursors.⁶³ These studies suggest that large portions of unexplained total fluorine and organic fluorine levels are likely due to unknown PFAS.

For PFAAs, concentrations in the fire station living room dust samples were generally lower than previous studies of dust in U.S. homes or offices, although these environments are not directly comparable (Table S3).^{30,47,64–69} Only one living room in this study had carpet, which is an important source of PFAAs indoors.^{13,47,67} In addition, those studies did not measure the three most frequently detected chemicals in this study's samples—N-EtFOSAA, 6:2 FtS, and 8:2 FtS—which were found at higher concentrations than in previous European studies. In the fire station living rooms (chosen to be the most comparable reference point), N-EtFOSAA had a median concentration of 87.5 ng/g (maximum: 1800), whereas a study of Swedish preschools measured a median of 18.4 ng/g (95th percentile: 283),⁷⁰ and a study of Finnish bedrooms reported a median 3.00 ng/g for linear N-EtFOSAA (maximum: 422).⁷¹ The Swedish preschools did not have detectable levels of 6:2 FtS,⁷⁰ while another study of Norwegian households detected 6:2 FtS in dust at a median of 4.8 ng/g (maximum: 53).⁷² In comparison, our fire station living rooms had higher concentrations of 6:2 FtS at a median of 9.85 ng/g (maximum: 316). For 8:2 FtS, this study's median 6.56 ng/g (maximum: 66.1) was similar to levels in Norwegian homes (median: 5.3 ng/g, maximum: 99).⁷² In summary, the fire station dust samples generally had lower levels of PFAAs than prior studies, except

for two PFAA precursors (N-EtFOSAA and 6:2 FtS) that were higher. However, dust contact may only be one pathway of exposure to PFAS in fire stations, so other exposure routes such as dermal contact with contaminated products or inhalation should also be researched.

This study has a few limitations. First, only one gear wipe from each of six stations were analyzed as a pilot analysis due to the lack of studies on PFAS in wipes at the time, but gear varies widely in age, washing frequency, and manufacturer. Wipe samples did not differentiate between chemicals contaminating the surface of the gear versus originating from the gear. This study was also limited in determining the exact fraction of total fluorine due to PFAS or distinguishing potential contributions from unknown non-polymeric versus polymeric PFAS. Inorganic fluoride or other non-PFAS organic fluorinated compounds like chlorofluorocarbon refrigerants⁷³ should not significantly impact differences in living rooms and gear locker rooms, and this study's low proportions of explained total fluorine align with previous research on turnout gear and studies of consumer products that were able to compare PFAS to both total fluorine and organic fluorine separately. Finally, this study was limited to stations in one state with little variation in policies such as gear washing, which makes assessments of impacts of station practices difficult. There could have been some over-reporting of the frequency of cleaning and gear washing in the stations. As strengths, this study evaluated PFAS as a previously understudied contaminant in fire stations, analyzed a novel elemental indicator of potentially unknown PFAS in dust, measured more PFAS than other U.S. studies of dust, and compared different rooms within stations to characterize potential sources of PFAS.

CONCLUSIONS

This study identifies turnout gear as a potential source of PFAS inside fire stations, either due to the addition of PFAS in the gear itself or contamination of the gear from firefighting activities involving AFFF or combustion of consumer products in fires. Dust in turnout gear locker areas had elevated within-station levels of total fluorine and certain PFAS that were also found in wipe samples of gear. On the other hand, the use of PFAS-containing AFFF by the studied stations did not statistically explain differences in PFAS levels, likely because most stations in this study do not use these products at all or only use them very rarely. Further research should evaluate firefighter exposures to PFAS from turnout gear through dermal contact or inhalation, as dust ingestion is likely not the primary route of exposure. Finally, this study's analysis of fluorine in dust points to potential emerging uses of unknown PFAS and the usefulness of an indicator that captures total PFAS content. More research is needed on the relative importance of unknown PFAS, polymeric PFAS, and fluoride on total fluorine concentrations in dust.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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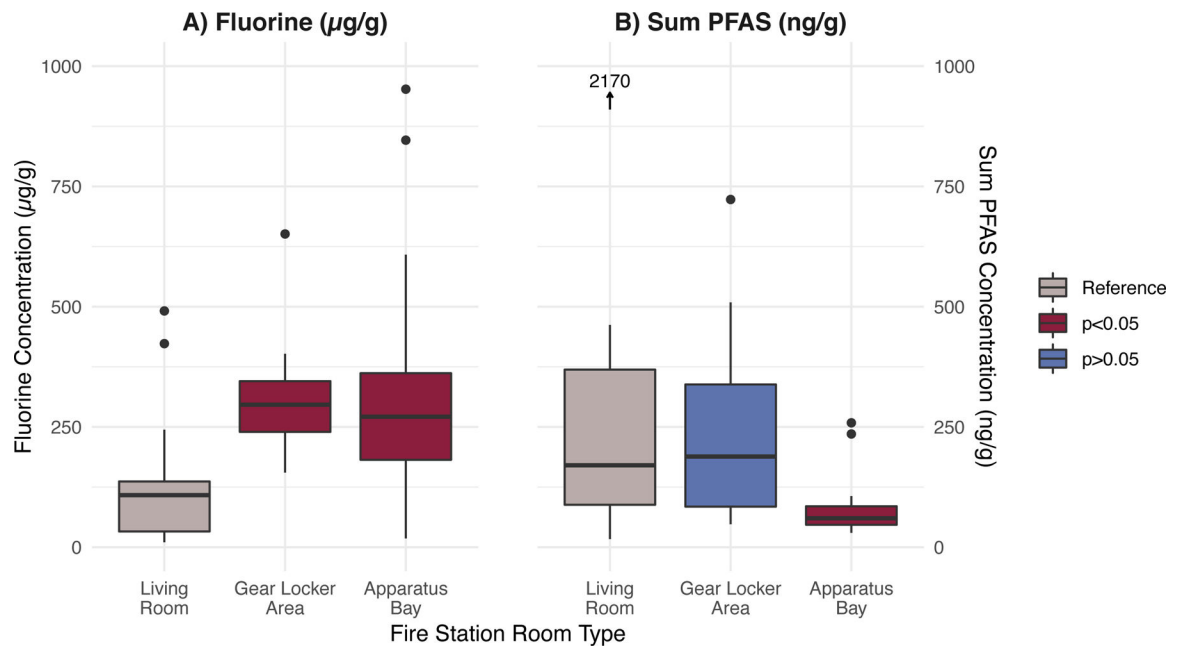


Figure 1. Boxplot of concentrations of A) total fluorine (µg/g) and B) Σ_{24} PFAS (ng/g) in dust by room type in 15 fire stations in Massachusetts.

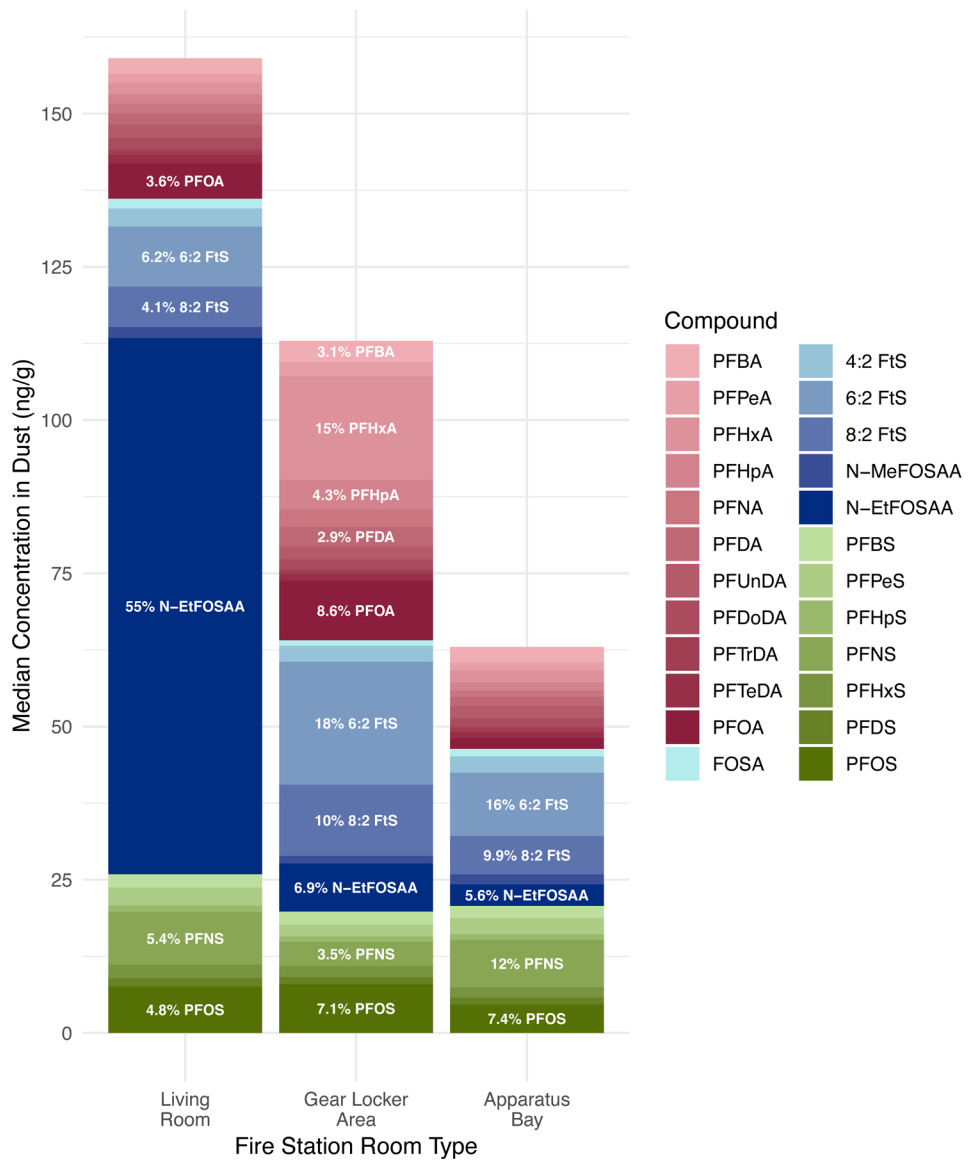


Figure 2. Stacked bar chart of median speciated concentrations of each PFAS (ng/g) in dust by room type in 15 fire stations in Massachusetts.

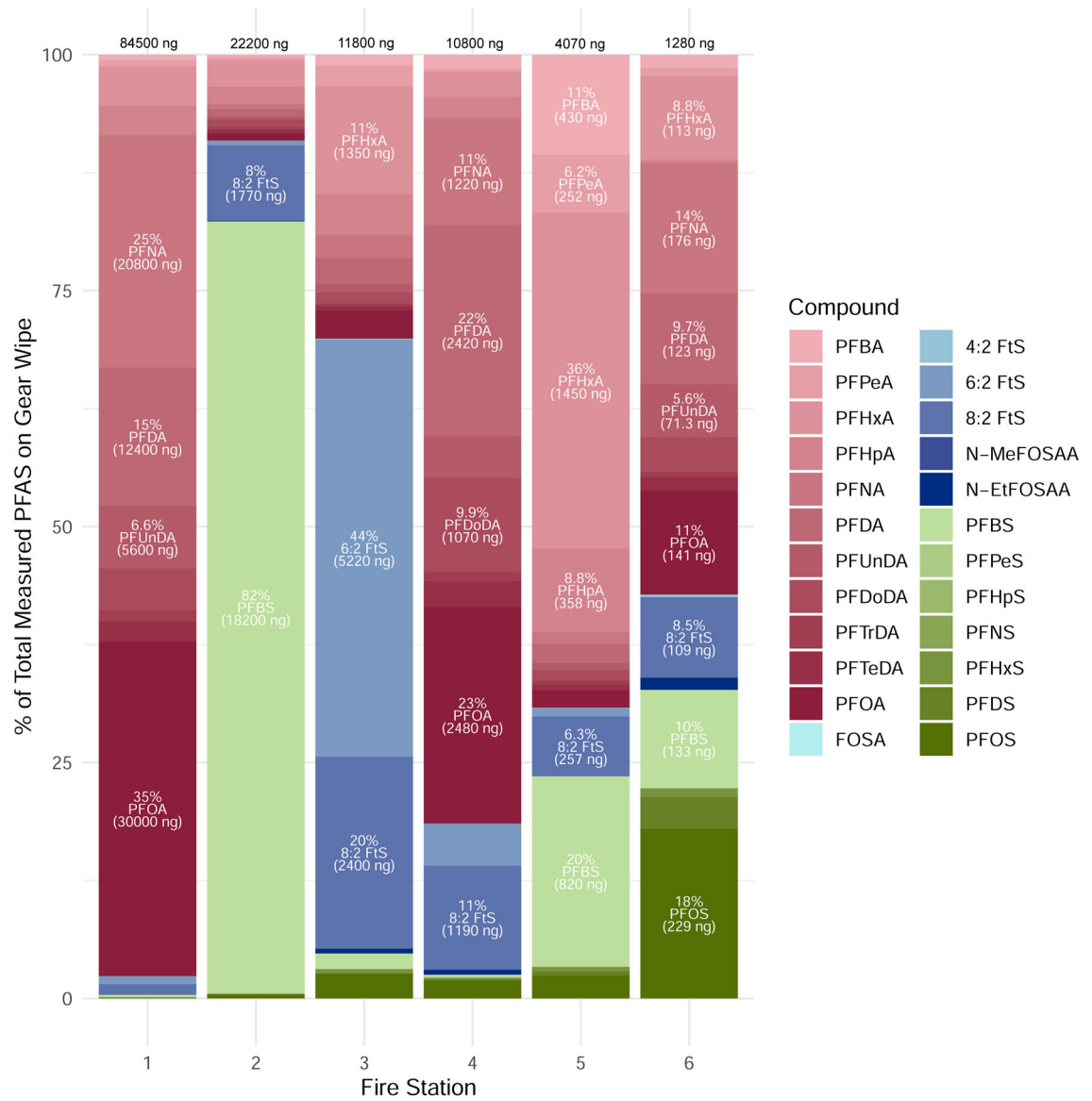


Figure 3. Profiles of PFAS on wipes of turnout gear in a subset of six fire stations in Massachusetts. *Note:* The total Σ₂₄PFAS concentration on each gear wipe (ng/wipe) is listed on top of the bar.

Table 1.

Characteristics of the 15 fire stations in Massachusetts sampled in this study.

Characteristic	<i>n</i> (%)	Median [Range]
Neighborhood Type		
Urban	10 (67)	
Suburban	5 (33)	
Estimated Annual No. of Calls		4000 [1800–7000]
No. of Firefighters on Duty		10 [5–14]
% of Shift Spent in Station		75 [50–90]
Gear Washer Present		
Yes	13 (87)	
No	2 (13)	
Gear Washed After Every Fire		
Yes	13 (87)	
No	2 (13)	
Foam Reported to be PFAS-Free		
Yes	9 (60)	
No	6 (40)	
Training with Firefighting Foam		
Annually off-site	4 (27)	
Once or twice yearly in-house	8 (53)	
None	3 (20)	
Number of Floors in Station		
One	3 (20)	
Two or three	12 (80)	

Table 2.

Summary statistics for concentrations of total fluorine ($\mu\text{g/g}$; $n=88$) and PFAS (ng/g ; $n=39$) in dust samples from different rooms in 15 fire stations in Massachusetts.

Analyte	% Detected >MDL ^A	Median [Range]			
		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	100	157 [6.17–952]	108 [10.2–491]	296 [155–651]	271 [18.2–952]
<i>In ng/g:</i>		$n=39$ ^C	$n=11$	$n=14$	$n=14$
Σ_{24} PFAS	92.3	98.7 [16.8–2170]	170 [16.8–2170]	189 [47.8–723]	60.1 [29.9–259]
Σ_6 Precursors ^D	92.3	42.0 [5.59–1830]	123 [5.59–1830]	94.1 [<MDL–446]	25.2 [8.03–183]
Σ_{11} PFCAs ^E	84.6	27.1 [7.57–251]	27.0 [7.57–129]	56.4 [<MDL–251]	17.6 [<MDL–48.9]
Σ_7 PFSAs ^F	56.4	22.7 [3.67–274]	28.4 [3.67–274]	23.5 [7.27–112]	16.7 [3.80–26.8]
6:2 FtS	89.7	11.1 [<MDL–316]	9.85 [1.61–316]	20.1 [<MDL–310]	10.3 [<MDL–133]
N-EtFOSAA	82.1	5.90 [.748–1800]	87.5 [.748–1800]	7.84 [<MDL–299]	3.51 [<MDL–159]
8:2 FtS	79.5	6.96 [<MDL–131]	6.56 [1.62–66.1]	11.6 [<MDL–131]	6.24 [<MDL–28.1]
PFOA	76.9	4.85 [<MDL–60.0]	5.77 [<MDL–30.6]	9.69 [<MDL–60.0]	1.74 [.735–27.5]
PFOS	71.8	4.95 [1.26–91.5]	7.57 [1.26–78.9]	7.97 [4.00–91.5]	4.64 [1.56–24.6]
PFNA	64.1	1.59 [.446–29.8]	1.59 [.543–15.1]	2.79 [<MDL–29.8]	1.12 [.446–5.57]
PFHxA	61.5	3.06 [<MDL–76.9]	1.82 [<MDL–76.9]	16.9 [<MDL–40.9]	1.98 [<MDL–23.6]
PFHpA	53.8	1.53 [<MDL–22.6]	1.53 [<MDL–11.0]	4.88 [<MDL–22.6]	1.24 [.577–5.53]
PFDA	53.8	1.78 [.276–42.0]	1.87 [.276–4.54]	3.24 [<MDL–42.0]	1.46 [1.19–2.90]
PFUnDA	51.3	2.04 [.281–17.5]	2.11 [.616–3.17]	2.10 [.554–17.5]	2.00 [.281–4.95]
PFDoDA	46.2	1.62 [.270–19.7]	1.92 [.270–2.83]	1.67 [1.15–19.7]	1.50 [.619–1.80]
PFTriDA	46.2	.699 [.136–7.03]	.849 [.177–2.68]	.688 [.214–7.03]	.686 [.136–3.05]
PFTeDA	43.6	1.13 [.173–7.83]	1.38 [.173–1.77]	1.13 [.51–7.83]	1.07 [.291–1.29]
PFBA	41	2.57 [<MDL–27.5]	2.57 [<MDL–18.4]	3.50 [<MDL–27.5]	2.57 [<MDL–6.94]
PFDS	38.5	1.15 [<MDL–185]	1.41 [.291–185]	1.13 [.332–12.5]	1.13 [<MDL–1.51]
PFPeA	35.9	1.32 [<MDL–16.6]	1.51 [<MDL–14.2]	2.27 [<MDL–16.6]	1.26 [<MDL–4.19]
FOSA	35.9	1.26 [<MDL–2.27]	1.58 [<MDL–2.27]	.865 [<MDL–1.74]	1.23 [<MDL–1.48]
N-MeFOSAA	30.8	1.67 [<MDL–2.70]	1.80 [<MDL–2.70]	1.24 [<MDL–2.32]	1.63 [<MDL–1.97]
PFHxS	25.6	1.77 [<MDL–12.2]	2.17 [<MDL–6.24]	1.79 [<MDL–12.2]	1.68 [<MDL–2.02]
PFBS	23.1	2.08 [<MDL–7.48]	2.17 [<MDL–3.25]	2.15 [<MDL–7.48]	1.97 [<MDL–2.37]
PFPeS	7.69	2.71 [<MDL–4.38]	2.93 [<MDL–4.38]	1.91 [<MDL–3.76]	2.66 [<MDL–3.20]
PFHpS	7.69	.940 [<MDL–1.52]	1.09 [<MDL–1.52]	.884 [<MDL–1.30]	.919 [<MDL–1.11]
4:2 FtS	7.69	2.73 [<MDL–4.42]	2.95 [<MDL–4.42]	2.64 [<MDL–3.79]	2.68 [<MDL–3.22]
PFNS	2.56	7.92 [<MDL–12.8]	8.56 [<MDL–12.8]	3.97 [<MDL–11.0]	7.74 [<MDL–9.33]

^AMDL = method detection limit. The average MDL for total fluorine was 25 $\mu\text{g/g}$. The PFAS MDLs calculated for each sample ranged from 0.0242 (PFTeDA) to 18.1 ng/g (PFNS).

^BSamples 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, PFTriDA, and PFTeDA.

^F PFSAAs included PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, and PFDS.

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Table 3.

Results from multilevel models of the association between concentrations of total fluorine ($\mu\text{g/g}$) and the sum of 24 PFAS^A (ng/g) in dust based on room type, use of AFFF that contains PFAS, regular gear washing after a fire, and frequency of room floor cleaning.

Covariate	% Difference [95% Confidence Interval] ^B	
	Fluorine	$\Sigma_{24}\text{PFAS}$
<i>Room Type</i>		
Living Room	Ref	Ref
Gear Locker Area	273% [104, 579%] ***	-7.78% [-54.7, 90.7%]
Apparatus Bay	191% [59.1, 429%] **	-63.5% [-82.0, -24.5%] *
Gym	130% [24.0, 332%] *	
Sleeping Quarters	24.5% [-31.7, 127%]	
Kitchen	-9.66% [-50.9, 64.7%]	
<i>Use of AFFF with PFAS</i>		
No	Ref	Ref
Yes	-26.4% [-52.8, 16.0%]	91.8% [-12.0, 330%]
<i>Gear Washed After Fires</i>		
No	Ref	Ref
Yes	-23.1% [-57.3, 37.9%]	24.9% [-51.6, 226%]
<i>Room Floor Cleaning</i>		
Weekly–Monthly	Ref	Ref
Daily	-9.1% [-44.5, 24.0%]	-21.9% [-59.4, 55.4%]

*
 $p < 0.05$

**
 $p < 0.01$

 $p < 0.0001$

^A Only samples from living rooms, gear locker areas, and apparatus bays were analyzed for PFAS.

^B Concentrations were log-transformed in the models, but the estimates were transformed back to linear scale for presentation.

Note: PFAS = per- and polyfluoroalkyl substances; AFFF = aqueous film-forming foams.