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Biomonitoring in California Firefighters

Metals and Perfluorinated Chemicals

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Objective: To assess California firefighters' blood concentrations of selected chemicals and compare with a representative US population. **Methods:** We report laboratory methods and analytic results for cadmium, lead, mercury, and manganese in whole blood and 12 serum perfluorinated chemicals in a sample of 101 Southern California firefighters. **Results:** Firefighters' blood metal concentrations were all similar to or lower than the National Health and Nutrition Examination Survey (NHANES) values, except for six participants whose mercury concentrations (range: 9.79 to 13.42 μg/L) were close to or higher than the NHANES reporting threshold of 10 μg/L. Perfluorodecanoic acid concentrations were elevated compared with NHANES and other firefighter studies. **Conclusions:** Perfluorodecanoic acid concentrations were three times higher in this firefighter group than in NHANES adult males. Firefighters may have unidentified sources of occupational exposure to perfluorinated chemicals.

The California Environmental Contaminant Biomonitoring Program (Biomonitoring California), the first legislatively mandated ongoing state biomonitoring program in the United States, is a collaborative effort involving the California Department of Public Health, the Office of Environmental Health Hazard Assessment, and the Department of Toxic Substances Control.[1] Chemicals measured in Biomonitoring California studies are selected on the basis of potential for exposure to the general public or sensitive populations, known or suspected health effects because of chemical exposure, the availability of valid laboratory analytical methods, and the testing laboratory's capacity to analyze these chemicals.[2]

Biomonitoring California collaborated with the University of California, Irvine (UC Irvine), Center for Occupational and Environmental Health staff to measure selected environmental chemicals in a study of firefighters. Firefighters were identified as an important group to study because there are very few biomonitoring studies of this population and they may have greater exposures to many hazardous chemicals. Several studies have demonstrated that firefighters are at increased risk for some types of cancer, and some authors have postulated that this may be a result of occupational chemical exposures.[3–5] During routine fire response activities, firefighters may inhale toxic gases, vapors, or particles and may also ingest particles released during a structural or vehicle fire, building collapse, or hazardous materials spill.[6] Predicting firefighters' exposures to specific environmental chemicals or combustion products is difficult because of the variability of fuels (eg, plastics, wood, and petroleum products) and fire characteristics (eg, temperature, duration, and availability of oxygen in the fire environment).[7] Firefighters may also be exposed to hazardous chemicals during the overhaul process, when searching debris for embers that may reignite. During overhaul, some firefighters may remove their self-contained breathing apparatus (SCBA), even though volatile organic compounds and particulate matter are still likely to be present in the environment.[8] In addition, Fent and Evans[9] reported that firefighters may not routinely use SCBA when suppressing vehicular fires. Inconsistent use or improper handling of personal protective equipment may increase firefighters' chemical exposure.

Firefighter exposure to heavy metals has been documented in smoke and on turnout gloves.[8,10] Studies have documented the adverse neuropsychological and renal function effects of heavy metal exposure.[11,12]

Perfluorinated chemicals (PFCs) are widely used in homes and offices as stain repellent fabric and carpet coatings.[13] Firefighters may also be exposed to PFCs through the use of some firefighting foams.[14,15] Although foams designed to suppress Class A fires (eg, involving burning buildings or vegetation) are not reported to contain PFCs,[16,17] those designed to suppress Class B fires (eg, involving flammable liquids) routinely contain fluorinated surfactants.[18] Animal toxicology and epidemiologic studies on some PFCs indicate that this class of chemicals can affect the human endocrine, nervous, and immune systems.[19,20] Possible adverse health outcomes include decreased fertility, neurodevelopmental toxicity, and cancer.[21–26]

Biomonitoring has been conducted in only a few investigations of firefighter exposure to environmental chemicals.[6,14,15,27–31] Notably, community studies have shown elevated levels of PFCs among firefighters,[14,32] and occupational studies have shown elevated levels of metals and PFCs after responding to an incident.[6,15,28]

Because we considered firefighters to be a potentially sensitive subpopulation at risk for exposure to environmental chemicals, we conducted a biomonitoring study in Southern California firefighters. This paper, on analysis of selected heavy metals and PFCs, is the first publication from this population. This study also measured levels of polybrominated diphenyl ethers, polycyclic aromatic hydrocarbons, phthalates, and other analytes identified as priority chemicals for Biomonitoring California. Biomonitoring results of other chemicals analyzed in this study will be reported in separate publications.

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METHODS

Study Design

A convenience sample of firefighters in a southern California county scheduled for their annual or biannual voluntary, nonpunitive wellness examination between mid-October 2010 and February 2011, were invited to take part in the Firefighter Occupational Exposures (FOX) study. Eligibility was limited to those who had been employed as full-duty active firefighters for at least 12 months. The study was approved by the California Committee for the Protection of Human Subjects and UC Irvine's Institutional Review Board.

After obtaining informed consent, we administered an exposure assessment questionnaire that could be completed in 15 minutes during the examination. Data collected included firefighting experience, recent incident response, occupational duties, personal protective equipment (PPE) use and maintenance, exposure to firefighting foam, selected dietary intake, and demographic information. The following data were abstracted from the UC Irvine, Center for Occupational and Environmental Health occupational records: current job title, annual incident response, information about a second job, self-reported current health status, tobacco use, and medical history.

A brief standardized environmental survey was used to collect data on potential chemical exposure sources at each fire station with a participant. Data collected included building age, the number of fire department vehicles, presence of carpeting, and the number and condition of nonstick cookware.

Figure 1 represents a flowchart of sample collection and transport. Whole blood was collected in 3 mL ethylenediaminetetraacetic acid-coated tubes, stored at 4°C at the collection site, and periodically shipped on refrigerated gel packs to the California Department of Public Health Environmental Health Laboratory (Richmond, California) for metals analysis. For serum preparation, approximately 40 mL of blood was collected in tubes without additives or anticoagulants. Serum was separated by allowing blood to clot at room temperature, then centrifuging the sample twice at 2000 rpm—first for 15 minutes and then for 10 minutes. Serum was frozen and stored at −20°C, then shipped on dry ice to the Department of Toxic Substances Control Environmental Chemistry Laboratory (Berkeley, California) for PFC analysis.

Laboratory Methods

Whole blood specimens were analyzed for total mercury, manganese, cadmium, and lead. We used an Agilent 7500cx with a helium collision cell (Agilent Technologies, Inc, Folsom, CA) inductively coupled plasma-mass spectrometry system.[33,34] Quality control reference materials and intermediate calibration standards were prepared from stock standard solutions traceable to the National Institute of Standards and Technology (Gaithersburg, MD). Blood specimens were diluted 1:50 to minimize blood matrix effects.

Whole blood specimens were stored at −20°C until analyzed. About 80% of blood specimens were analyzed within 72 hours of sample receipt, with the remainder completed within 10 days. Each specimen was analyzed in duplicate, and the final result was calculated by averaging the 2. Acceptance criteria for the analytical results were based on the relative percentage difference (RPD%) between the 2 specimens. The RPD% rejection threshold for manganese, lead, and mercury was 10%, and, for cadmium, it was 20%. Samples that did not meet these criteria were reanalyzed. The average RPD% for manganese, lead, mercury, and cadmium was 2.9%, 2.2%, 2.9%, and 11.6%, respectively.

Quality control reference materials were prepared by spiking defibrinated sheep blood obtained from the Hemostat Laboratories (Dixon, CA) with stock standard solutions at three concentrations (low, medium, and high). All reference materials were analyzed at both the beginning and end of each batch analysis. Four concentrations of the National Institute of Standards and Technology Standard Reference Material 955c were periodically analyzed throughout the study to assure independent confirmation.

Serum specimens were analyzed for perfluorooctane sulfonate (PFOS), perfluorohexane sulfonate (PFHxS), perfluorobutane sulfonate, perfluorooctanoic acid (PFOA), perfluorononanoic

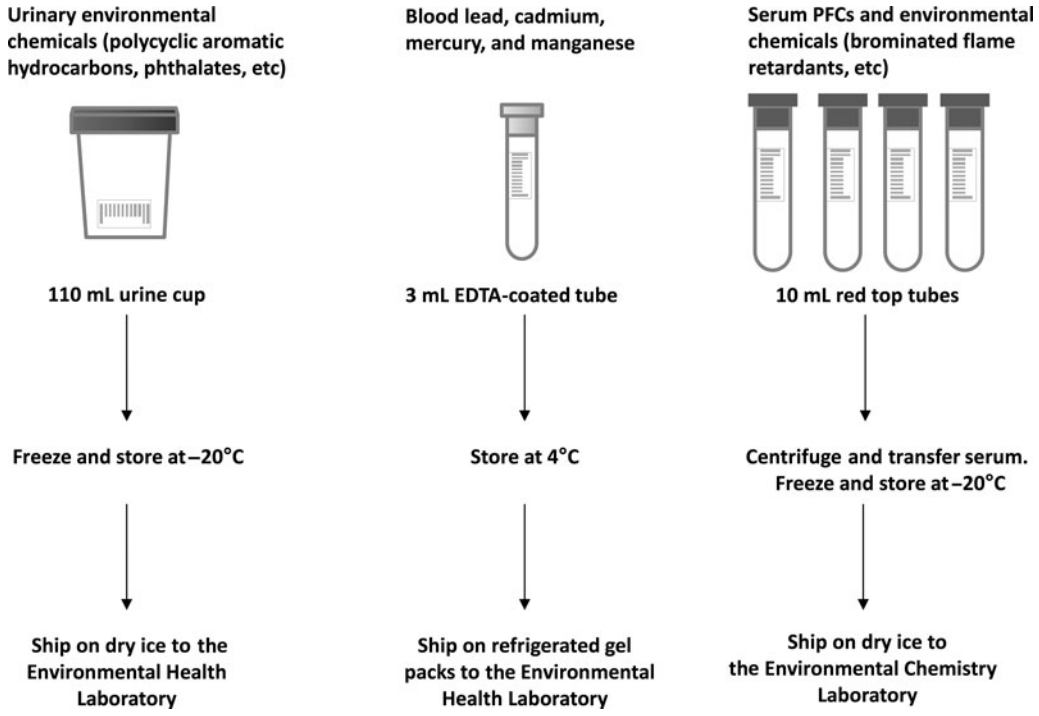

FIGURE 1. Specimen collection and transport flow chart for the FOX study, 2010 to 2011. EDTA, ethylenediaminetetraacetic acid; PFCs, perfluorinated chemicals.

acid (PFNA), perfluoroheptanoic acid (PFHpA), perfluorodecanoic acid (PFDeA), perfluoroundecanoic acid, perfluorododecanoic acid, perfluorooctane sulfonamide (PFOSA), 2-(*N*-ethyl-perfluorooctane sulfonamido) acetic acid, and 2-(*N*-methyl-perfluorooctane sulfonamido) acetic acid (N-MeFOSAA). We used an on-line solid phase extraction high-performance liquid chromatography tandem mass spectrometry method. Details of this method were described previously.[35,36] For quantification, seven-point external calibration curves were processed together with each batch of samples. Calibration curves were constructed on the basis of ratios of target analyte to internal standard, plotting the peak area ratio versus the concentration ratio. Linear regression was conducted and regression coefficients were 0.98 to 0.99.

The method was validated by repeatedly analyzing blank bovine serum spiked with unlabeled PFC standards at two different concentrations (low and high). Experimental values and the respective target values were summarized and verified.[36] Blank samples (bovine serum) were also processed with each batch of participant samples, and no PFCs were detected above the respective limits of detection (LODs) (defined as three times the standard deviation of the blank).

Early Reporting of Results

The Centers for Disease Control and Prevention (CDC) has established early reporting thresholds for use in the National Health and Nutrition Examination Survey (NHANES) for blood mercury (10 μg/L or more in adult males or 5.8 μg/L or more in females aged 18 to 49 years, based on Sue[12] and Rice et al,[37] respectively). Any study participant whose blood mercury result exceeded the CDC early reporting threshold was notified.

Data Analysis

The FOX questionnaire allowed respondents to report one or more ethnicities. Participants were defined as Hispanic if they indicated "Hispanic or Latino," regardless of other ethnicities reported, or white if they indicated "Caucasian or White" and did not also report another ethnicity. Participants who indicated "Asian," "Native Hawaiian or Other Pacific Islander," or "African American or Black" were categorized as "other" for data analysis.

Analyte concentrations were nonnormally distributed, so geometric means (GMs) were calculated for analytes detected in more than 60% of samples. Measurements below the LOD were imputed using LOD/$\sqrt{2}$.[38] We calculated GMs and 95% confidence intervals (CIs) of 2009 to 2010 NHANES men aged 20 years or older for comparison.[39] We assessed correlations among covariates and analytes using the Spearman method. Analytes detected in 75% or more of samples were included in regression analyses.

Covariates considered for inclusion in modeling included demographics (age, educational level, race/ethnicity, body mass index, body fat percentage, current tobacco use, years as firefighter, and job title); incident response activities (ie, the number of hazardous material spills, industrial fires, commercial fire, house fires, car fires, or brush/vegetation fires attended in the last year); fire station characteristics (age, floor coverings, upholstered furniture number and condition), intake of selected dietary items, and the use of firefighting foam; and work practices (PPE storage location and decontamination within the last year, SCBA usage patterns, and handwashing frequency).

Unadjusted regression models were used to assess the relative importance of independent variables as predictors for each chemical (see Supplemental Digital Content Table S1, available at http://links.lww.com/JOM/A172). A priori variables for all regression models included age and race, with job title added for metals models only. We examined firefighter blood metal and PFC concentrations in relation to participants' frequency of responding to fires or hazardous materials incidents during the last year for this analysis because participants indicated infrequent responses within the preceding month. For incident response type and PPE usage categories, results of age- and race-adjusted PFC models were ranked by the Akaike information criterion to determine order of entry for predictors in the final model. Predictors that improved the previous model's Akaike information criterion were included. Factors that were significantly associated with metals concentrations in single predictor models, improved model fit, and showed a consistent response trend across categories were included in the metals regression models. We repeated all significant analyses excluding the two females. Regression model coefficients were exponentiated to represent the proportional change in the GM associated with each level of predictor, compared with a referent level and adjusted for other predictors. Statistical analysis was conducted using SAS (version 9.3; SAS Institute Inc, Cary, NC).

RESULTS

Of the 137 invited firefighters, 101 participated in this study (74% response rate). Because 98% of participants were male, biomonitoring data are presented without regard to sex. Demographic and occupational information are summarized in Tables 1 and 2, respectively. None of the 10 firefighters who reported using tobacco smoked cigarettes; all smoked cigars or used chewing

TABLE 1. Demographic Characteristics of 101 Participants in the Firefighter Occupational Exposures Study, 2010 to 2011

Characteristic	*n*
Sex	
Male	99
Female	2
Age (mean ± SE), yrs	42.8 ± 0.9
Age, yrs	
20–29	9
30–39	29
40–49	36
≥50	27
Education at hire	
High school/General Education Development	39
Some college/technical school	29
College graduate	33
Race/ethnicity (n = 100)	
White non-Hispanic	78
Hispanic	15
Other	7
Body mass index (kg/m^2)†	
Normal (18.5–24.9)	18
Overweight (25–29.9)	57
Obese (≥30)	26
Body fat percentage‡	
≤17.5	36
>17.5–24	32
>24	33
Current tobacco use	
Yes	10
No	91

†Calculated from height and weight measurements at clinical examination.
‡Caliper skinfold measurements at clinical examination.

TABLE 2. Occupational Characteristics of FOX Participants, 2010 to 2011 ($n = 101$)

Characteristic	*n*
Job title	
Firefighter	45
Engineer	20
Captain or battalion chief	36
Years as firefighter ($n = 100$) (mean ± SE), yr	18.8 ± 1.0
Years as firefighter ($n = 100$), yr	
1–10	27
11–20	24
21–30	39
31–40	10
Turnout gear decontaminated within the last year ($n = 100$)	
Yes	46
No	54
Hazardous materials responses in the last year	
None	34
<1/mo	54
≥1/mo	13
Commercial fire responses in the last year	
None	33
<1/mo	58
≥1/mo	10
Brush fire responses in the last year	
None	14
≥1	87
Firefighting foam use in the last year ($n = 100$)	
Class A only	64
Class B or both Class A & Class B	23
Neither	13
Handwashing frequency per 12-h duty shift ($n = 100$)	
≤8	31
9–11	33
12–19	21
≥20	15
Year built—fire station where FOX participant was assigned ($n = 89$)	
before 1980	46
1980–1999	28
2000 and later	15

FOX, Firefighter Occupational Exposures.

tobacco. Years as a firefighter was strongly correlated with age ($r = 0.88$; $P < 0.0001$), whereas job title was moderately correlated with age ($r = 0.54$; $P < 0.0001$) and years as a firefighter ($r = 0.65$; $P < 0.0001$). Some variables could not be included in final models because of missing data, including fire station characteristics.

We compared participants' blood metal and PFC concentrations with adult men from the 2009 to 2010 NHANES cycle (Tables 3 and 4). All blood lead and cadmium concentrations were below the CDC early reporting thresholds. In six male firefighters, total blood mercury concentrations (when rounded to a whole number) equaled or exceeded (range: 9.79 to 13.42 μg/L) the early reporting threshold. Urine metals analysis found low inorganic mercury levels in FOX firefighters,[40] indicating that the modestly elevated blood concentrations are predominantly organic mercury. Elevated blood mercury concentrations are often related to consumption of high-mercury fish. All FOX participants had blood manganese concentrations consistent with the usual range of about 4 to 15 μg/L.[11]

Seven PFCs were detected in all 101 participants (Table 4). Perfluorododecanoic acid was not detected in any participant. Of the PFCs measured in this study, PFOS concentrations were the highest, similar to results reported in the NHANES.[39] Geometric means for most PFCs in FOX participants were similar to or lower than corresponding NHANES GMs. Nevertheless, PFDeA concentrations were approximately three times higher in these firefighters (GM = 0.90 μg/L; 95% CI, 0.78 to 1.03) than in NHANES (GM = 0.30 μg/L; 95% CI, 0.28 to 0.34).

Multivariate models identified higher blood cadmium, lead, and mercury in participants aged 50 years or older, although only cadmium concentrations were significantly elevated. Significantly higher blood cadmium was also found in firefighters who washed their hands less frequently during a work shift (Table 5). Manganese was higher in firefighters who responded to commercial fire incidents at least once in the last year than those who did not, and in firefighters assigned to fire stations built after 2000. Mercury was significantly higher in firefighters who responded to brush fires at least once in the last year than those who did not. Multivariate models accounted for 14% to 30% of the variability in blood metal values.

Perfluorinated chemical multivariate models (Tables 6 to 8) identified significantly higher (PFOSA) concentrations in firefighters aged 50 years or older. Monthly or more frequent responses to commercial fires were associated with higher PFHpA concentrations. Those who responded to hazardous materials incidents at least monthly had higher concentrations of 2-(*N*-methyl-perfluorooctane sulfonamido) (N-MeFOSAA) than those who did not, whereas any hazardous materials response was associated with significantly higher PFNA values. PFNA and PFOA were also significantly higher in firefighters whose turnout gear had not been professionally decontaminated within the last year. Participants who used Class A firefighting foam had significantly higher PFHpA concentrations than those who did not use any class of foam.

DISCUSSION

Firefighting is recognized as a hazardous occupation, and firefighters have a presumed medical predisposition to developing injuries or disease related to their work.[41] Studies have documented firefighters' increased risk of coronary heart disease and some types of cancer.[3–5,42] There are few studies that have examined chemical exposures as factors that may affect this population. This study adds to the biomonitoring literature on firefighters and illustrates that both occupational and environmental sources may contribute to chemical exposure.

Perfluorinated chemicals were of particular interest in this study because of their use in Class B firefighting foam.[14] In a recent study of 8826 adult men in the Ohio River Valley, Jin et al[14] found significantly increased PFHxS concentrations in 36 firefighters compared with other employed adults. The authors hypothesize that this finding may be associated with firefighting foam exposure. Nevertheless, the chemical composition of the foam was not determined. Because manufacturers consider PFCs used in Class B firefighting foams as proprietary information, we were not able to identify the PFCs in foams used by FOX participants. Firefighter Occupational Exposures participants' PFHxS concentrations (GM = 2.26 μg/L; 95% CI, 2.00 to 2.54) were not significantly different from NHANES (GM = 2.15 μg/L; 95% CI, 1.93 to 2.40) and were half that of the Jin et al firefighters (GM = 4.77 μg/L). Nevertheless, we identified an unexpected positive association between Class A firefighting foam use and PFHpA concentrations (Table 8). Class A foam, used to suppress structural and vegetation fires more effectively than water alone, is not reported to contain PFCs.[43] We were unable to chemically analyze the Class A foam used by firefighters in this study.

TABLE 3. Blood Metal Concentrations in FOX Firefighters, 2010 to 2011, Compared With NHANES*

Blood Metal	Population	*n*	LOD	DF (%)	Percentiles				Maximum	Geometric Mean (95% CI)
					25th	50th	75th	95th		
Cadmium (ug/L)	FOX	101	0.15	78.2	0.15	0.20	0.24	0.35	0.77	0.19 (0.18, 0.21)
	NHANES	2,784	0.20	71.4	<LOD	0.28	0.54	1.60	6.88	0.33 (0.31, 0.34)
Lead (ug/dL)	FOX	101	0.02	100	0.69	0.95	1.22	2.01	5.92	0.96 (0.87, 1.05)
	NHANES	2,784	0.25	100	0.96	1.42	2.17	4.26	43.52	1.47 (1.41, 1.53)
Mercury (ug/L)	FOX	101	0.06	100	1.82	2.90	5.48	9.79	13.42	2.79 (2.36, 3.30)
	NHANES	2,784	0.33	88.1	0.53	0.99	2.16	6.43	85.70	1.09 (0.99, 1.21)
Manganese (ug/L)	FOX	101	0.54	100	6.50	7.70	8.79	11.40	15.81	7.61 (7.26, 7.98)

*2009 to 2010 NHANES males aged 20 years or older, manganese not measured.

CI, confidence intervals; DF, detection frequency; FOX, Firefighter Occupational Exposures; LOD, limit of detection; NHANES, National Health and Nutrition Examination Survey.

TABLE 4. Serum PFC Concentrations (μg/L) in FOX Firefighters, 2010 to 2011, Compared With NHANES*

Serum PFCs	Population	*n*	LOD	DF (%)	Percentiles				Maximum	Geometric Mean‡ (95% CI)
					25th	50th	75th	95th		
PFOS	FOX	101	0.083	100	10.10	12.70	16.80	24.70	46.60	12.50 (11.34, 13.78)
Perfluorooctane sulfonic acid	NHANES	876	0.2	99.8	8.30	12.30	17.60	40.40	281.0	12.13 (10.43, 14.10)
PFOA	FOX	101	0.301	100	2.96	3.86	4.89	9.54	18.10	3.75 (3.37, 4.17)
Perfluorooctanoic acid	NHANES	876	0.1	99.7	2.70	3.70	5.10	8.20	24.00	3.61 (3.28, 3.98)
PFHxS	FOX	101	0.012	100	1.61	2.27	3.13	4.64	13.20	2.26 (2.00, 2.54)
Perfluorohexane sulfonic acid	NHANES	876	0.1	99.6	1.40	2.20	3.40	6.90	44.80	2.15 (1.93, 2.40)
PFNA	FOX	101	0.075	100	0.89	1.13	1.49	2.21	4.23	1.15 (1.06, 1.25)
Perfluorononanoic acid	NHANES	876	0.082	99.8	0.98	1.31	1.89	4.18	17.95	1.40 (1.20, 1.63)
PFDeA	FOX	101	0.032	100	0.51	0.72	1.72	2.63	4.60	0.90 (0.78, 1.03)
Perfluorodecanoic acid	NHANES	876	0.1	96.4	0.20	0.30	0.40	0.90	20.70	0.30 (0.28, 0.34)
PFHpA	FOX	101	0.059	75.2	0.06	0.12	0.22	0.63	0.98	0.13 (0.11, 0.15)
Perfluoroheptanoic acid	NHANES	876	0.1	16.3	<LOD	<LOD	<LOD	0.20	1.00	—
PFOSA	FOX	101	0.009	95.0	0.019	0.029	0.050	0.151	0.396	0.032 (0.027, 0.037)
Perfluorooctane sulfonamide	NHANES	876	0.1	0.1	<LOD	<LOD	<LOD	<LOD	0.10	—
N-MeFOSAA	FOX	101	0.013	100	0.09	0.14	0.24	0.61	1.86	0.16 (0.13, 0.18)
2-(*N*-methyl-PFOSA) acetic acid	NHANES	876	0.1	75.9	0.10	0.20	0.30	1.00	3.80	0.19 (0.18, 0.21)
N-EtFOSAA	FOX	101	0.011	65.3	<LOD	0.016	0.023	0.060	0.464	0.016 (0.014, 0.018)
2-(*N*-ethyl-PFOSA) acetic acid	NHANES	876	0.1	6.0	<LOD	<LOD	<LOD	0.10	1.00	—
PFUA	FOX	101	0.010	100	0.17	0.26	0.37	0.53	0.73	0.24 (0.21, 0.27)
Perfluoroundecanoic acid	NHANES	876	0.1	75.4	0.10	0.20	0.30	0.90	28.50	0.18 (0.16, 0.21)
PFDoA	FOX	101	0.040	0.0	<LOD	<LOD	<LOD	<LOD	<LOD	—
Perfluorododecanoic acid	NHANES	876	0.1	4.6	<LOD	<LOD	<LOD	<LOD	2.80	—
PFBuS	FOX	101	0.020	6.9	<LOD	<LOD	<LOD	0.02	0.04	—
Perfluorobutane sulfonic acid	NHANES	876	0.1	0.7	<LOD	<LOD	<LOD	<LOD	0.40	—

‡Geometric means were calculated for analytes detected in more than 60% of samples.

*2009 to 2010 NHANES males aged 20 years or older.

CI, confidence intervals; DF, detection frequency; FOX, Firefighter Occupational Exposures; LOD, limit of detection; NHANES, National Health and Nutrition Examination Survey; PFC, perfluorinated chemical.

Firefighter Occupational Exposures participants' PFDeA concentrations were up to three times higher than the NHANES comparison group for the 25th to 95th percentiles and GM. Although unadjusted analysis demonstrated higher PFDeA concentrations in firefighters who used Class A foam or responded to hazardous materials incidents within the last year (see Supplemental Digital Content Table S1–1, available at http://links.lww.com/JOM/A172), these two variables did not meet inclusion criteria for the multivariate models. Shaw et al[28] also found elevated PFDeA concentrations in 12 San Francisco firefighters (median = 1 μg/L) compared with NHANES (median = 0.25 μg/L). Previous studies involving firefighters have demonstrated elevated concentrations of other PFCs, including PFOS,[14,15] PFOA,[15,28] and PFHxS[14,15] compared with NHANES.[39] Although not definitive, our findings combined with the results of these other studies suggest that an occupational factor, such as working with firefighting foam, may increase firefighters' exposures to PFCs. We recommend evaluating firefighting foam exposures and foam delivery methods, as well as other potential

TABLE 5. Adjusted Proportional Change in Geometric Mean (95% CI) Blood Metal Concentrations From Multivariate Regression Models[a]

Predictor	n[b]	Lead ($n = 99$)	Mercury ($n = 97$)	Cadmium ($n = 99$)	Manganese ($n = 96$)
Age, yrs					
≥50	26	1.22 (0.81, 1.84)	1.08 (0.48, 2.43)	1.78** (1.26, 2.51)	1.02 (0.82, 1.26)
40–49	35	0.86 (0.58, 1.26)	0.92 (0.42, 1.97)	1.22 (0.88, 1.69)	1.03 (0.84, 1.27)
30–39	29	0.80 (0.56, 1.14)	0.91 (0.45, 1.82)	1.10 (0.81, 1.48)	1.04 (0.86, 1.27)
20–29	9	1.0 (reference)	1.0 (reference)	1.0 (reference)	1.0 (reference)
Decontaminated PPE in last year					
No	53	1.18 (0.98, 1.43)			
Yes	46	1.0 (reference)			
Brush fire responses in last year					
≥1/mo	11		2.21* (1.02, 4.75)		
<1/mo	72		1.77* (1.02, 3.06)		
None	14		1.0 (reference)		
Hazardous materials responses in last year					
≥1/mo	13			1.25 (0.96, 1.63)	
<1/mo	53			0.90 (0.75, 1.08)	
None	33			1.0 (reference)	
Handwashing frequency per 12-h work shift					
≤8	31			1.29* (1.00, 1.66)	
9–11	32			1.11 (0.88, 1.41)	
12–19	21			0.99 (0.76, 1.29)	
≥20	15			1.0 (reference)	
Commercial fire responses in last year					
≥1/mo	10				1.16 (0.96, 1.40)
<1/mo	56				1.14* (1.02, 1.28)
None	30				1.0 (reference)
Fire station construction year					
<1980	54				0.82** (0.72, 0.94)
1980–1999	27				0.82* (0.71, 0.96)
≥2000	15				1.0 (reference)
Adjusted model R^2		0.21	0.14	0.30	0.26

*$P < 0.05$; **$P < 0.01$.
[a]Exponentiated b coefficient from each metal's multiple regression model adjusted for race, body mass index, and job title.
[b]Totals do not all equal 101 because of missing data.
CI, confidence interval; PPE, personal protective equipment.

occupational sources to PFCs, in larger firefighter studies designed to include personal exposure measurements.

Data on the potential adverse health effects of PFDeA exposure are limited. On the basis of what we know about other PFCs, PFDeA may affect endocrine activity and the immune system.[23,24] Because this study was not designed to assess the health effects of environmental chemical exposure, we did not collect health endpoint data. Thus, we were unable to investigate a potential association between elevated PFDeA levels and specific health outcomes in our firefighter group.

Firefighter exposure to heavy metals has been documented in both structural and wildland fires. Bolstad-Johnson et al[8] identified lead in air samples taken during overhaul, whereas Fabian et al[10] found metals (including lead, manganese, and mercury) on firefighter gloves from smoke and soot deposition, indicating the potential for additional exposure when gloves are removed. In wildland fires, combustion can release metals from soil organic material and vegetation, increasing their bioavailability.[44,45] Despite these potential occupational exposures, GMs of blood lead, manganese, and cadmium were low in this study group. Firefighter Occupational Exposures participants' lead exposure may be lower than those firefighters described above in part because many housing developments in the southern California county were built after lead was banned from house paint. Manganese differs from other metals in this study because it is an essential nutrient.[46] Although manganese concentrations in FOX participants were consistent with the usual range reported for the US population,[46] manganese concentrations were significantly lower in firefighters assigned to stations built prior to 2000.

Firefighter Occupational Exposures participants' blood cadmium concentrations (GM = 0.19 μg/L; 95% CI, 0.18 to 0.21) were also low compared with NHANES (GM = 0.33 μg/L; 95% CI, 0.31 to 0.34), firefighters in New York City (GM = 0.377 μg/L),[6] and adult males in the New York City Health and Nutrition Examination Survey (GM = 0.76 μg/L; 95% CI, 0.73 to 0.79).[47] The major source of cadmium exposure in the United States is cigarette or cigar smoking. Using smokeless tobacco products has not been found to increase blood cadmium concentrations.[48] There was no difference in the mean blood cadmium values for FOX cigar smokers and chewing tobacco users compared with nontobacco users (data not shown).

Blood mercury concentrations in FOX firefighters were higher than corresponding NHANES values across all percentile distributions (Table 3). Six male firefighters in this study had total blood mercury concentrations between 9.79 and 13.42 μg/L, which are

TABLE 6. Adjusted Proportional Change in Geometric Mean (95% CI) Serum PFC Concentrations From Multivariate Regression Models[a]

Predictor	n[b]	PFOS ($n = 100$)	PFOA ($n = 99$)	PFHxS ($n = 96$)
Age, yrs				
≥50	26	1.22 (0.82, 1.81)	0.66 (0.42, 1.03)	1.15 (0.69, 1.91)
40–49	35	1.19 (0.81, 1.75)	0.69 (0.45, 1.07)	1.21 (0.74, 1.96)
30–39	29	0.93 (0.63, 1.35)	0.66 (0.43, 1.00)	1.12 (0.70, 1.81)
20–29	9	1.0 (reference)	1.0 (reference)	1.0 (reference)
Early SCBA removal during overhaul				
Might remove	35			0.74* (0.57, 0.98)
Does not remove	61			1.0 (reference)
Decontaminated PPE in last year				
No	33		1.33* (1.06, 1.67)	
Yes	60		1.0 (reference)	
Adjusted model R^2		0.07	0.1	0.07

*$P < 0.05$.
[a]Exponentiated b coefficient from each PFCs multiple regression model adjusted for race.
[b]Totals do not all equal 101 because of missing data.
CI, confidence interval; PFHxS, perfluorohexane sulfonate; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonate; PPE, personal protective equipment; SCBA, self-contained breathing apparatus.

TABLE 7. Adjusted Proportional Change in Geometric Mean (95% CI) Serum PFC Concentrations From Multivariate Regression Models[a]

Predictor	n[b]	PFNA ($n = 99$)	PFUA ($n = 93$)	*N*-MeFOSAA ($n = 96$)
Age, yrs				
≥50	26	0.88 (0.62, 1.24)	0.76 (0.45, 1.27)	1.29 (0.69, 2.39)
40–49	35	0.90 (0.65, 1.26)	0.78 (0.48, 1.28)	1.21 (0.67, 2.18)
30–39	29	0.83 (0.60, 1.14)	0.70 (0.43, 1.12)	1.05 (0.59, 1.86)
20–29	9	1.0 (reference)	1.0 (reference)	1.0 (reference)
Hazardous materials responses in last year				
≥1/mo	13	1.49** (1.12, 1.99)		2.24** (1.35, 3.70)
<1/mo	53	1.32** (1.08, 1.61)		1.28 (0.89, 1.83)
None	33	1.0 (reference)		1.0 (reference)
Brush fire responses in last year				
≥1/mo	10		1.37 (0.81, 2.31)	
<1/mo	69		1.38 (0.95, 2.00)	
None	14		1.0 (reference)	
SCBA use during overhaul				
Sometimes	71			1.28 (0.89, 1.83)
Always	25			1.0 (reference)
Early SCBA removal during overhaul				
May remove	33		1.06 (0.80, 1.41)	
Does not remove	60		1.0 (reference)	
Decontaminated PPE in last year				
No	53	1.24* (1.04, 1.49)		
Yes	46	1.0 (reference)		
Adjusted model R^2		0.14	0.13	0.18

*$P < 0.05$; **$P < 0.01$.
[a]Exponentiated b coefficient from each PFCs multiple regression model adjusted for race.
[b]Totals do not all equal 101 because of missing data.
CI, confidence interval; *N*-MeFOSAA, 2-(*N*-methyl-perfluorooctane sulfonamido) acetic acid; PFNA, perfluorononanoic acid; PFUA, Perfluoroundecanoic acid; PPE, personal protective equipment; SCBA, self-contained breathing apparatus.

TABLE 8. Adjusted Proportional Change in Geometric Mean (95% CI) Serum PFC Concentrations From Multivariate Regression Models[a]

Predictor	n[b]	PFDeA (n = 99)	PFHpA (n = 94)	PFOSA (n = 99)
Age, yrs				
≥50	26	0.91 (0.53, 1.58)	1.46 (0.73, 2.91)	1.91* (1.02, 3.57)
40–49	35	1.11 (0.65, 1.91)	2.12* (1.09, 4.14)	1.44 (0.78, 2.64)
30–39	29	0.97 (0.58, 1.63)	1.60 (0.82, 3.10)	1.02 (0.56, 1.85)
20–29	9	1.0 (reference)	1.0 (reference)	1.0 (reference)
Hazardous materials responses in last year				
≥1/mo	13	1.10 (0.70, 1.75)		
<1/mo	53	1.43* (1.04, 1.97)		
none	33	1.0 (reference)		
Brush fire responses in last year				
≥1/mo	63		0.27** (0.12, 0.59)	
<1/mo	23		0.53* (0.31, 0.89)	
None	13		1.0 (reference)	
House fire responses in last year				
≥1/mo	27		1.16 (0.73, 1.83)	
<1/mo	67		1.0 (reference)	
Commercial fire responses in last year				
≥1/mo	10		2.46* (1.14, 5.34)	
<1/mo	54		1.22 (0.81, 1.84)	
None	30		1.0 (reference)	
Firefighting foam use in last year				
Class A only	63	1.35 (0.88, 2.08)	1.82* (1.07, 3.11)	1.29 (0.80, 2.10)
Class B or both Class A & Class B	23	0.92 (0.56, 1.50)	0.78 (0.42, 1.42)	0.78 (0.45, 1.35)
Neither	13	1.0 (reference)	1.0 (reference)	1.0 (reference)
Adjusted model R^2		0.17	0.32	0.18

*$P < 0.05$; **$P < 0.01$.
[a]Exponentiated b coefficient from each PFCs multiple regression model adjusted for race.
[b]Totals do not all equal 101 because of missing data.
CI, confidence interval; PFDeA, perfluorodecanoic acid; PFHpA, perfluoroheptanoic acid; PFOSA, perfluooctane sulfonamide.

close to or higher than the early reporting threshold for adult men (10 μg/L). No common factors were identified in these six participants, who varied in age, job title, years employed as a firefighter, on-duty activities, and fire station assignment. None reported contact with elemental mercury on- or off-duty. Our questionnaire did not include questions on fish consumption; however, the blood mercury concentrations for these six firefighters are consistent with those measured in individuals living in coastal regions,[47,49–52] where people tend to eat more fish. McKelvey et al,[47] in their representative sample of New York City adult men, reported blood mercury concentrations (GM = 2.67 μg/L; 95% CI, 2.48 to 2.87) similar to the FOX study (GM = 2.79 μg/L; 95% CI, 2.36 to 3.30). Mercury concentrations from both these studies are approximately 2.5 times higher than NHANES (GM = 1.09 μg/L; 95% CI, 1.00 to 1.21). Urine analyses identified very low inorganic mercury concentrations in these firefighters[40]; thus, it is likely that consumption of fish containing elevated mercury was responsible for the higher blood mercury concentrations in FOX participants. Consistent with previous general population studies, participants aged 50 years or older had higher blood cadmium, lead, and mercury concentrations than younger firefighters, even after adjusting for potential sources of exposure.[47,53,54]

We also examined whether self-reported SCBA use during overhaul operations was associated with chemicals measured in this study. Firefighters may remove their SCBA during overhaul when smoke is no longer visible but while they are disturbing debris searching for embers that may reignite. We found no association between heavy metal or PFC concentrations and self-reported use of respiratory protection during overhaul.

In summary, our results demonstrate that some workplace practices impact chemical exposures in firefighters. Specifically, handwashing was associated with lower cadmium levels, and professionally cleaning turnout gear was associated with lower levels of PFNA and PFOA. Handwashing has been reported to be an effective method of reducing potential chemical exposure among other occupation groups.[55,56]

There are several limitations to this study. The small population limited the power to find associations between exposure factors and blood chemical concentrations. Our best fit multivariate models explained 14% to 30% of the variation in firefighter metal values and 7% to 32% of PFC variation, consistent with the common presence of these chemicals in nonoccupational environments. Because of the exam scheduling, data and biosamples were collected from some FOX participants when they returned from being off duty. Due to varying frequency of incidents, participants may not have responded to incidents recently (in the previous 7 to 10 days or longer). Participants were on duty during study data collection, and limited time was available to complete a questionnaire during their examination. Thus, we were unable to collect potentially relevant data, such as a detailed dietary history for fish consumption or information on potential residential chemical exposures. Behaviors such as PPE use and maintenance, dietary intake, and handwashing were self-reported and relied on recall. Moreover, this study was not designed to identify specific exposure sources and, therefore, did not conduct

systematic environmental sampling for metals and PFCs (eg, in dust) at the workplace or participant's home. Finally, this study was not designed to assess health outcomes or their association with biomonitored chemical levels.

CONCLUSIONS

This biomonitoring study was designed to shed new light on chemical exposures in firefighters. This publication adds new information on heavy metal and PFC concentrations in this population, using very sensitive chemical analytical methods.

Lead and cadmium concentrations were lower than those in the US general population; although older study participants tended to have higher concentrations of lead, mercury, and cadmium, no values were high enough to be clinically significant. Modestly elevated blood mercury levels reported here are consistent with findings in other studies of coastal populations in California that are likely to eat more fish with mercury contamination.[47] Our finding that cadmium levels were lower in firefighters who reported more frequent handwashing provides support for the standard recommendation of thoroughly washing hands to reduce chemical exposure in workers.

This study population had PFC concentrations similar to those of the general US population, except for higher PFDeA concentrations. Although we were unable in this study to identify the factors leading to higher PFDeA exposure in this group of firefighters, our finding of elevated PFHpA concentrations in participants who used Class A firefighting foam is consistent with increased PFCs levels found among firefighters in other studies.[14,28] Larger studies of firefighters or other first responders with more detailed investigation should be conducted to determine whether firefighting foam or other occupational factors contribute to increased PFC levels in firefighters.

This study was not designed specifically to assess exposure reduction offered by PPE, so we were not able to directly evaluate the effect of its use on biomonitored chemicals. Nevertheless, on the basis of finding an association between professional cleaning of turnout gear and levels of some PFCs, we recommend that turnout gear be professionally cleaned to reduce exposures. We also support recommendations made by other authors that firefighters use PPE (eg, turnout gear and SCBA) during all phases of firefighting, including overhaul.[30]

Biomonitoring is a powerful tool that can provide information for an occupational exposure assessment of firefighters who may be frequently exposed to higher levels of harmful chemicals than the general population. Moreover, biomonitoring reflects chemical exposure regardless of the source or route of exposure. This study points to the possibility of specific PFC exposures related to firefighting. Nevertheless, as this study demonstrates, this is also a limitation because we were not able to distinguish occupational from residential or other sources of exposure. Future studies incorporating ambient exposure assessment in both home and workplace settings would help differentiate various chemical exposure sources and enable primary prevention efforts through targeted exposure reduction.

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