Perfluoroalkyl Acids Including Perfluorooctane Sulfonate and Perfluorohexane Sulfonate in Firefighters

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Objective: Firefighters were likely exposed to perfluorooctane sulfonate since it was a component of extinguishing foams and perfluorohexane sulfonate (PFHxS), a surfactant coating carpet and other building materials, during firefighting. The objective of the study is to evaluate serum concentrations of perfluoroalkyl acids (PFAAs) in firefighters. **Methods:** A total of 8826 male adults, including 37 firefighters, were analyzed. Multivariate analysis was conducted by using a general linear model. The least square mean of serum PFAAs was obtained after adjustment for demographic and socioeconomic variables. **Results:** Serum concentration of PFHxS was statistically higher in firefighters both before and after adjustment. Perfluorooctane sulfonate and perfluorononanoic acid were also found higher in firefighters, though not statistically significant. **Conclusions:** The study suggests that fighting fire can be a risk of exposure to PFAAs, specifically PFHxS.

The health effects of firefighting have long been a major interest of occupational health researchers. There are clear physical hazards and exposures to both toxic agents and toxic products of combustion. Toxins used by firefighters, as well as toxins present or created at fire scenes, may be a source of acute and chronic illness, including cancer[1–7] and heart disease or pulmonary disease.[3,8] In this article, we evaluate the intersection of firefighting and exposure to a class of potentially hazardous chemicals, perfluoroalkyl acids (PFAAs), with an emphasis on perfluorooctane sulfonate (PFOS) and perfluorohexane sulfonate (PFHxS) as likely firefighter exposures.

These PFAAs and related telomers are used to produce numerous products, including both industrial and household products that have provided a source of human contact. The telomers may rapidly break down to their related PFAA, adding to industrial exposures. There has been increasing recognition that some PFAAs are widespread in the environment, animal species, and humans.[9,10]

The manufacturing of PFOS began in the United States in 1948. In 2000, the major manufacturer, 3M Corporation, announced that it would stop making many of its well-known Scotchgard stain-repellent products after finding that PFOS persists in the environment and is found widely in numerous species, including humans. Perfluorooctane sulfonate has been in numerous products, including water-repellent coatings and fire-suppression foams. While PFOS is the most common PFAA in the human environment, it is not the only PFAA that could be inhaled or contact skin during fire suppression. Perfluorohexane sulfonate, which has been used for postmarket carpet-treatment applications, is also found in house dust, and it has been a component of fire-suppression foams.[11]

Demographic predictors of human PFAA exposures have emerged for perfluorooctanoate (PFOA) and PFOS. In addition to any local water contamination, known demographic predictors of PFOS human serum concentrations include increased dietary meat and animal fat consumption,[12] male gender, higher social class,[10] age greater than 60 years,[13,14] residence in the United States[15] or in contaminated parts of Japan,[16] and on-the-job exposure at manufacturing facilities.[17] Perfluorohexane sulfonate is found more in the serum of children compared with adults, and in males compared with females.[13,14]

Health outcomes of PFAAs are under increasing investigation. In humans, PFOS exposure is clearly associated with higher levels of cholesterol in adults[18] and children.[19] Concerns have been raised about bladder cancer in workers[20] on the basis of small numbers, and some types of human reproductive or developmental toxicity.[21–24] Even less is known about human outcomes of PFHxS exposure. In animal toxicology studies, a wide range of toxicity is exhibited. Cross-sectional investigation of the National Health and Nutrition Examination Survey data revealed a relationship between PFHxS exposure and parental report of attention-deficit disorder diagnoses.[25] Currently, no data address biomarkers of firefighter exposure to PFOS, PFHxS, or other PFAAs.

This study was conducted to examine data collected from a large, multiple-community study in a region with PFOA-contaminated drinking-water systems. Perfluorooctanoic acid contamination of six water districts, while the reason for the enrollment, was not hypothesized to be associated with firefighting. On the basis of reported components of firefighting foams, we hypothesized that firefighters were exposed to PFOS and PFHxS during firefighting and would have higher serum markers of exposure. The objective of the study is to evaluate serum concentrations of PFOA, PFOS, PFHxS, and related chemicals in firefighters and compare them with a contemporary, also-employed control group.

MATERIALS AND METHODS

Study Population/Survey Design

The C8 Health Project was a court-directed result of a class-action settlement, attendant to the discovery of PFOA contamination in six water districts within Ohio and West Virginia, in the mid-Ohio Valley. Project enrollment was conducted between August 2005 and August 2006. An independent company, Brookmar, was created solely for the purpose of developing, testing, and conducting a health survey of eligible participants within the six water districts. An extensive health survey included demographic, socioeconomic, employment, and personal health information. A total of 18 diagnoses were independently verified. A description of the survey, survey history, design, enrollment, population, laboratory methods, and laboratory quality assurance has been published.[26] The survey itself is available on-line at http://www.hsc.wvu.edu/som/cmed/c8/.

Briefly, 69,030 eligible participants, including an estimated 81% of current adult residents, enrolled.[27] Blood samples were submitted for PFAA analysis by 66,899 participants. In this study, the subset of male adult participants with self-reported employment information was included. To study occupations, we excluded

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those who reported more than one occupation, since the current dominant job could not be determined from information available in the database. Women were excluded because there was one female firefighter.

Study Variables and Outcomes

The study design was cross-sectional analysis. The primary independent variable was participants' employment, with a specific focus on firefighters. Those reporting a single other trade or no job composed the comparison populations. The main study outcomes were the serum mean concentrations of PFAAs that were detected in more than 97% of the study population. For these four PFAAs, including PFOA, PFOS, PFHxS, and perfluorononanoic acid (PFNA), the serum mean, geometric mean, and median were calculated for three employment categories: (1) firefighters, (2) people with self-reported employment in 20 industrial job categories, and (3) those who did not report an employment.

The other PFAAs, detectable in a variable but much lower percentage of the population, were examined as to presence/absence of nonzero values because they were detectable only in a portion of the study population and mean serum concentrations were low. These PFAAs were perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnA), and perfluorododecanoic acid (PFDoA).

For these chemicals, we modeled as zeros and explored presence/absence of the chemical. In contrast, PFOA, PFOS, PFHxS, and PFNA were common exposures, and nondetects were recorded as half the detection limit.

Data were analyzed using SAS (version 9.1.3; SAS Institute Inc, Cary, NC). A general linear model was used to perform multivariate analysis with the natural log-transformed serum concentrations of PFAAs. The log-transformed data better fitted the model because PFAA levels were not normally distributed. The least squares mean of serum PFAAs analyzed in the study was used after adjustment for demographic and socioeconomic variables previously found to be associated with PFAA serum concentrations. Age, water district, and average household incomes were considered as covariables and in a multivariable model.

Statistical analysis was performed to examine whether differences in detection of PFPeA, PFHxA, PFHpA, PFDA, PFUnA, and PFDoA were statistically significant for the studied employment categories, by using a logistic model.

RESULTS

A total of 26,944 male adults, including 42 firefighters, were identified. Among 8826 men who identified a single employment category, 41 firefighters reported work activities that included line firefighting and saving lives. Four reported cessation of fighting fire more than 8 years before the survey date; these were excluded given consideration of half-life of PFAAs. One firefighter did not have serum PFAA laboratory tests and was excluded from the study. A total of 36 firefighters were included in the study.

All but one firefighter (97.2%) self-identified as white, consistent with central Appalachian demographics and the overall study population. Data are presented without regard to race or ethnicity. Average time of employment was 14 years, with a median of 13 years. The mean age of the firefighters was 43.2 years (median age of 40 to 41 years). The characteristics of occupational employment are summarized in Table 1.

The detection ranges for these and other PFAAs varied in the study cohort, from 0.7% for PFDoA to 99.9% for PFOA. Perfluorohexane sulfonate was found in 98.1% of participants; PFOS was detected in 99.4%; and PFNA was found in 97.6% of the study population. Perfluoropentanoic acid, PFHxA, PFHpA, PFDA, and PFUnA were found in intermediate, varying concentrations.

Table 2 summarizes descriptive statistical features, least squares means before adjustment, and also a general linear model of the serum concentrations of the commonly found analytes, PFHxS, PFOA, PFOS, and PFNA. Serum concentration of PFHxS (least square mean) was higher in firefighters both before and after adjustment (5.19 and 4.79 ng/mL, compared with 3.78 and 3.80 ng/mL) when compared with other employment category. The least squares means for PFOS and PFNA were also higher in the firefighter group than in other employment categories both before and after adjustment for age, district, and income (27.77 and 26.32 ng/mL for PFOS; and 1.74 and 1.63 ng/mL for PFNA). Mean serum concentration of PFOA was incidentally found to be high in people with self-reported industrial jobs, both before and after adjustment of socioeconomic covariants (37.87 ng/mL vs 39.28 ng/mL). This is expected, as some survey participants were employed by the industry that was the initial source of community water pollution. After adjustment, serum mean concentration of PFOA was lower in the firefighters, compared with other employment or the unemployed.

Statistical analysis of the serum concentrations for PFHxS, PFOA, PFOS, and PFNA in different employment categories was further conducted by using a general linear model both with and without adjustment for age, water district, and average household income (Table 3). The serum concentration of PFHxS was statistically significantly higher in firefighters in comparison with other job categories before and after adjustment of demographic and socioeconomic factors compared with no job reported.

Serum mean concentration of PFOS was higher in firefighters than in participants who reported other employment and those who reported no employment. Nevertheless, the difference reached statistical significance only between firefighters and people who reported no employment (Table 3).

The difference in detection of PFPeA, PFHxA, PFHpA, PFUnA, and PFDoA in firefighters and in the study groups was examined by using logistic methods. The detection rate of PFPeA, PFHxA, PFHpA, PFDA, PFUnA, and PFDoA was 0%, 51.35%, 45.95%, 59.46%, 18.92%, and 2.7% in firefighters. Detection rates of these six PFAAs in firefighters were not statistically significantly different in comparison with the other two employment categorical groups.

TABLE 1. Demographic Characteristics of Study Adult Males by Employment Category

Employment	Number*	%	Age Mean, y	Age Median, y	Employment Duration Mean, y	Employment Duration Median, y
Firefight	36	0.5	40.1	40	13.3	12.5
Other employment	5373	67.4	42.6	43	12	7
No job reported	2563	32.1	40.5	34		

*Number of subjects with detectable serum PFAAs.

TABLE 2. Serum Least Square Mean, Geometric Mean, Arithmetic Mean, Median, and Standard Deviation in Study Groups by Employment Category

Employment	N	LS Mean*	LS Mean†	GeoMean	Mean	Median	SD	Min	Max
Perfluorohexane sulfonate									
Firefight	36	5.19	4.79	4.77	5.87	4.60	3.62	0.25	14.60
Other employment	5373	3.78	3.80	3.62	5.11	3.60	20.42	0.25	1447.60
No job reported	2563	3.77	3.75	3.52	5.13	3.50	6.41	0.25	100.30
Perfluorooctanoic acid									
Firefight	36	29.70	27.06	37.69	87.47	31.50	240.11	0.25	7534.60
Other employment	5373	37.87	39.28	31.59	72.96	26.90	148.54	0.60	1925.20
No job reported	2563	31.59	36.35	29.70	43.60	36.30	31.93	1.50	104.50
Perfluorooctane sulfonate									
Firefight	36	27.77	26.32	24.37	29.18	27.85	12.84	0.25	67.50
Other employment	5373	22.61	22.86	22.14	26.12	23.00	16.62	0.25	564.30
No job reported	2563	19.88	20.48	19.37	23.94	20.90	23.16	0.25	759.20
Perfluorononanoic acid									
Firefight	36	1.74	1.63	1.56	1.77	1.60	0.81	0.25	4.40
Other employment	5373	1.53	1.50	1.48	1.64	1.50	0.83	0.25	11.60
No job reported	2563	1.39	1.38	1.30	1.48	1.40	0.82	0.25	14.70

*Least square mean by GLM model unadjusted.
†Least square mean by GLM model adjusted with age, water district, and average household income.

TABLE 3. Statistical Analysis of the Serum Concentration of Perfluoroalkyl Acids With General Linear Model by Employment Category in Comparison With Firefighters

Job	Adjust	Estimate	P	95% CI	
Perfluorohexane sulfonate					
Other employment	Not adjusted	−0.32	0.01	−0.55	−0.08
	Adjusted*	−0.23	0.05	−0.46	0.00
No job reported	Not adjusted	−0.32	0.01	−0.55	−0.08
	Adjusted*	−0.24	0.04	−0.48	−0.01
Perfluorooctanoic acid					
Other employment	Not adjusted	0.24	0.21	−0.14	0.63
	Adjusted*	0.37	0.01	0.09	0.65
No job reported	Not adjusted	0.06	0.75	−0.32	0.45
	Adjusted*	0.30	0.04	0.01	0.58
Perfluorooctane sulfonate					
Other employment	Not adjusted	−0.21	0.04	−0.40	−0.01
	Adjusted*	−0.14	0.16	−0.34	0.06
No job reported	Not adjusted	−0.33	0.00	−0.53	−0.14
	Adjusted*	−0.25	0.01	−0.45	−0.05
Perfluorononanoic acid					
Other employment	Not adjusted	−0.13	0.07	−0.27	0.01
	Adjusted*	−0.08	0.26	−0.22	0.06
No job reported	Not adjusted	−0.23	0.00	−0.37	−0.09
	Adjusted*	−0.16	0.02	−0.30	−0.02

*Adjusted with age, water district, average household income, and smoking status by using general linear model.

DISCUSSION

The results of our biomarker study confirmed firefighter exposure to PFHxS but did not confirm our hypothesis that firefighters from the mid-Ohio Valley were exposed to PFOS (compared with other worker populations). For PFOS, there was a difference, but it did not reach statistical significance. It is unclear whether a similar study in a larger cohort might find a difference. In contrast, the positive association between firefighting and PFHxS persisted after adjustment for several covariants that were previously found to be associated with serum PFAA levels in this population.[26,27]

Perfluorohexane sulfonate is a surfactant, used to make fluoropolymers, fire-suppression foam, and stain-protective coatings for carpets, papers, and textiles. Along with other PFAAs, PFHxS serum levels have been declining in the serum of the US population; the decline is probably due to specific changes made by a major manufacturer of PFOS and related compounds in 2002.[10] Nevertheless, PFAAs in nondisposable household products and in house dust are known to persist, and broad population exposures are anticipated for many years,[28,29] particularly associated with carpeting.

The finding that firefighters had higher PFHxS serum concentrations suggests an exposure source different from the community exposure via contaminated drinking water sources previously reported for this region[27,30] and elsewhere.[31,32] Exposure for firefighters is most likely through a respiratory mechanism and, possibly to a lesser extent, via skin exposure. We infer that the likely source is fire-suppression foam, and fire conditions in households with stain-resistant carpet applications may contribute.

The health effects of PFHxS have not been studied in animals to the same extent as PFOS or PFOA. It does not appear to have the same paradoxical positive associations with serum lipids that have been seen with PFOA and PFOS.[33] Mouse and human peroxisome proliferator-activated receptor alpha were found to be activated by PFAAs including PFHxS.[34] The half-life of PFAAs is generally related to the length of their carbon chain; compounds containing fewer carbons have shorter half-lives. Nevertheless, PFHxS, which is eliminated in 8.5 years,[35] is an exception. Because of its long half-life, PFHxS can persist in the body long after cessation of exposure or employment.

The critical weakness for this study is the small firefighter cohort. Fire suppression in rural areas such as the mid-Ohio Valley is typically volunteer work outside of the few small cities. Only 36 firefighters were analyzed on the basis of the self-reported employment table. The small sample significantly restricts analysis. The presence of a statistically significant effect in this small number may understate or otherwise fail to represent the findings in busier, urban settings. Because of the small sample, the exposure population noted here is insufficient for the future detection of health effects. The finding highlights a need to determine whether the hypothesized primary cause of exposure, use of fire-suppression foam, is the cause.

CONCLUSIONS

Perfluorohexane sulfonate concentrations measured in the study population were found to be highest in firefighters, and the finding was statistically significantly different from that of other employment categories. We hypothesize that the use of firefighting foams is the source. Perfluorooctane sulfonate and PFNA were higher in firefighters, although the difference was not statistically significant. The small sample of firefighters is an important limitation. Further study with a large sample is needed to retest and refine the hypothesis that fighting fire can be a risk of exposure to PFAAs, especially PFHxS.

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