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Case Study

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Flame-retardant contamination of firefighter personal protective clothing – A potential health risk for firefighters

Reported By

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ABSTRACT

There is a high incidence of cardiovascular disease and certain cancers in firefighters that may be related to their occupational exposure to hazardous substances. Exposure may result from contaminated personal protective gear, as well as from direct exposure at fire scenes. This study characterized flame-retardant contamination on firefighter personal protective clothing to assess exposure of firefighters to these chemicals. Samples from used and unused firefighter protective clothing, including gloves, hoods and a coat wristlet, were extracted with methylene chloride and analyzed by EPA method 8270D Specific Ion Method (SIM) for polybrominated diphenyl ethers (PBDEs). Until recently PBDEs were some of the most common flame-retardant chemicals used in the US. Fifteen of the seventeen PBDEs for which analysis was performed were found on at least one clothing swatch. Every clothing sample, including an unused hood and all three layers of an unused glove, held a detectable concentration of at least one PBDE. These findings, along with previous research, suggest that firefighters are exposed to PBDE flame retardants at levels much higher than the general public. PBDEs are found widely dispersed in the environment and still persist in existing domestic materials such as clothing and furnishings. Firefighter exposure to flame retardants therefore merits further study.

KEYWORDS

Exposures; flame retardants; firefighters; PBDEs

Introduction

Firefighters are subject to an elevated risk of coronary heart disease^[1,2] and certain cancers.^[3,4] A recent NIOSH mortality study of firefighters found elevated risks of several types of cancers, including cancers of the digestive and respiratory systems, and malignant mesothelioma.^[5] A meta-analysis^[3] of 32 studies of cancer in firefighters ranked elevated risks of testicular and prostate cancer, non-Hodgkins lymphoma and multiple myeloma in firefighters as “probable.” They also reported a “possible” elevated risk of eight additional cancers, including malignant melanoma and brain cancer.^[3]

Exposures proposed to be responsible for the above adverse outcomes include toxic gases, particulates, and a wide variety of organic chemicals in the vapor state or adsorbed onto protective gear and particulate matter.^[6–8] In several studies, contaminants have been identified on firefighter personal protective gear, including heavy

metals, plasticizers and polycyclic aromatic hydrocarbons (PAHs).^[7,9,10]

In a study performed at Underwriters Laboratories (UL), it was found that gloves and protective hoods worn by firefighters at residential fires become contaminated with a wide range of organic and inorganic compounds, some of which are known to be carcinogens. The organic chemicals found in the largest quantities on firefighter gloves and hoods were phthalate diesters, especially di-(2-ethylhexyl) phthalate (DEHP), a plasticizer used primarily in polyvinyl chloride (PVC) plastics, and certain PAHs.^[7] In our previous research, DEHP contamination measured on firefighters’ personal protective gear was 18–215 times higher than the level of any other semi-volatile contaminant.^[10]

Exposure to these agents may result from inhalation, but deposition onto protective gear with subsequent skin contact creates a further possibility of exposure by

transdermal absorption. Polyhalogenated hydrocarbons such as PBDEs and phthalate diesters are highly lipophilic and would be expected to be readily absorbed through the skin, especially at the elevated skin temperatures experienced in firefighting situations.^[11] In the current study, used and unused firefighter protective clothing was analyzed for the presence of polybrominated diphenyl ether (PBDE) flame retardants.

In a study by Shaw et al., blood samples from 12 San Francisco (SF) firefighters who had responded to a fire within the previous 24 hours were found to contain a range of chlorinated, fluorinated, and brominated chemical species, including PBDE flame retardants. Measured levels of PBDEs were 2–3 times the levels detected in the general U.S. population.^[12] In a more recent study by Park et al., blood samples were collected from 101 Southern California firefighters. PBDE levels in the blood serum were very similar to those found for the SF firefighters.^[13]

PBDEs are a class of brominated aromatic organic chemicals which may contain from 1–10 bromine atoms arranged in different configurations, with each chemical being assigned a congener number from 1–209 (e.g., PBDE-209). Adverse health effects of PBDE exposure are not well characterized, but include neurotoxicity and endocrine disruption.^[14] PBDEs are also suspected of thyroid toxicity and carcinogenicity.^[15] In 2004, 21% of flame retardants produced globally were PBDEs and other brominated flame-retardant chemicals.^[16] Three commercial mixtures of PBDE congeners were commonly used in consumer products in the last half of the 20th century: pentaBDE, octaBDE, and decaBDE. Approximately 75% of the commercial flame-retardant mixture known as pentaBDE consists of the congeners PBDE-47 and PBDE-99. It was used primarily in polyurethane foams.^[17] PentaBDE was banned in California in 2003 due to its environmental persistence, bioaccumulation, and toxicity. Since that time, eight other states and the European Union also banned its use, and the sole U.S. producer voluntarily stopped production. It is still found commonly in polyurethane foam used in furniture and other items made before 2004.^[18]

The second commercial mixture, octaBDE, is a complex combination of different congeners, and it was used primarily in plastic housings and office equipment.^[17] The sole U.S. manufacturer of octaBDE also voluntarily ceased production in 2004. In addition, the US EPA released a Significant New Use Rule (SNUR) in 2006. The SNUR ensured that no new production or import of pentaBDE or octaBDE could start without EPA review.^[19]

PBDE-209 is deca-brominated diphenyl ether. PBDE-209 makes up over 97% of the commercial flame-retardant mixture known as decaBDE. DecaBDE was used primarily in electrical and electronic equipment and

in textiles.^[17] Until recently, it was manufactured and used in large quantities.^[20] In late 2009, the two major U.S. producers of decaBDE made agreements with the US EPA to phase out production, import, and sales. All U.S. uses of decaBDE were to end after 2013.^[21] As part of the effort to end the use of decaBDE in consumer goods, the US EPA has issued a report on the evaluation of alternative flame-retardant chemicals that could be used in its place.^[22] No recommended alternative was selected; rather, the alternatives must be weighed according to how well characterized they are, as well as their known toxicity, environmental persistence, carcinogenicity, efficacy and cost, when selecting a substitute.^[22]

Levels of several PBDE congeners on firefighter personal protective clothing were determined in this study for comparison with those previously found for PAHs and phthalates.^[10] As in our previous research, contaminant concentrations were measured separately on each layer of a used and an unused firefighter glove. This helps to answer the question of whether dermal exposure to contaminants could occur not only by handling of contaminated gear, but also by direct exposure by skin contact even when gloves are being worn. Used and unused firefighter hoods and a coat cuff were analyzed concurrently, to further assess the contribution to firefighter risk of dermal absorption. The same brands of both used and unused gear were sampled. Due to the expense of firefighter turnout coats, a cuff from an unused coat was not sampled.

Materials and methods

Sample collection

Samples of unused and occupationally soiled firefighter protective clothing, were analyzed for the presence of PBDEs. This clothing included gloves, hoods and a coat, and was donated to the University of Cincinnati by several Cincinnati metropolitan fire units. Used clothing had been utilized for firefighting for an unspecified period of time, and was not laundered before being donated for analysis. Detailed descriptions of the gear sampled are given in [Table 1](#).

Samples from different types of personal protective clothing were obtained using similar procedures. Samples approximately 10 cm square were cut from the clothing and one half was weighed and then sent to the analytical laboratory (ALS Environmental, Kelso, WA) for analysis. The other halves of the samples were stored in a freezer at -10°C until needed for any potential future analysis. In this study, some of the samples analyzed were the second half of samples that had been analyzed in the previous study.^[10] (See the Discussion section below.)

Table 1. Firefighter gear sampled.

Gear	Manufacturer	Description and Material
Protective Hoods (n = 4)	American Firewear (Dayton, OH)	Style HD 300 22887FNF, material - Nomex
Coat (n = 1)	Honeywell First Responder (Dayton, OH)	Morning Pride, Nomex wristlets
Structural Gloves (n = 3)	American Firewear (Ohatchee, AL)	Model 6500, outer layer – kangaroo leather, middle layer - Pyroprotect, inner layer - Kevlar/Nomex, cuff - Nomex

Hoods were composed of a double layer of a single type of material. A 10 cm square sample was removed from the front area immediately below the face, keeping both layers of fabric together. Samples were cut in half, from the chest edge to the chin edge. The Hood 1 sample analyzed in this study was the second half of a sample analyzed in the previous study.^[10] Figure 1 shows an unused and a used hood, with the area sampled being removed.

The coat was fitted with double wristlets at the end of each sleeve. The entire right inner wristlet, which included a thumb loop, was removed from the coat for the previous study,^[10] and the wristlet was cut in half from the hand side to the arm side. The half of the wristlet that did not include the thumb loop was analyzed for this study. Figure 2 shows the coat sampled for this study, with the inner wristlet removed.

The gloves were made up of three layers of different fabrics. The materials in the three layers are listed in Table 1. A sample of each layer was cut from the palm area of the glove. Samples were cut in half, from the wrist edge to the finger edge. The three layers were extracted and analyzed separately. Used and unused



Figure 2. Turnout coat sampled for this study, with right inner wristlet removed.

gloves, with the three layers removed, are shown in Figure 3.

Analytical methods

Samples were extracted with methylene chloride and the extracts analyzed for 17 PBDEs by EPA method 8270D Specific Ion Method (SIM). The limit of detection for each analyte was 0.005 µg/g.

Results

When extracts of the clothing samples were analyzed, 15 of the 17 PBDEs of interest were detected for at least one clothing sample. No detectable levels of PBDE-128 or -190 were found. Every clothing sample, including an unused hood and all three layers of an unused glove, held a detectable quantity of at least one PBDE. Results of the



Figure 1. Unused hood (left) and used hood (right), showing area of sample removal.



Figure 3. Unused glove (left) and used glove (right) sampled for this study, showing all three layers sampled. Color differences can be noted, especially on middle and inner layers.

analysis of gloves are shown in Table 2; results of the analysis of hoods are shown in Table 3; results of the analysis of a coat cuff are shown in Table 4, along with results for the hood that was worn together with the coat.

At least one PBDE congener was detected in each sample of used and unused firefighter personal protective clothing that was sent for analysis. The most abundant congeners were PBDE-47, PBDE-99, and PBDE-209, which were tetra-, penta-, and deca-brominated diphenyl ethers. PBDE-47 and PBDE-99 are the major constituents of the commercial flame-retardant mixture known as

pentaBDE.^[17] PBDE-47 was detected in every sample of used and unused personal protective clothing analyzed except the inner layer of Glove 2 (used) and the unused hood. PBDE-99 was detected in every sample except the inner layer of the unused glove and the unused hood.

The commercial flame-retardant mixture known as decabDE is made up almost entirely of PBDE 209.^[17] PBDE-209 was detected in every sample of used and unused protective clothing analyzed. In every sample, the PBDE found in the highest concentration was either PBDE-209 or PBDE-47.

Table 2. PBDE congeners detected on firefighter glove samples (µg/g).

PBDE	Unused Glove			Glove 1			Glove 2
	Inner	Middle	Outer	Inner	Middle	Outer	Inner
17	<LOD	<LOD	<LOD	<LOD	0.060	<LOD	<LOD
28	<LOD	0.017	<LOD	<LOD	0.18	0.067	<LOD
71	<LOD	A	<LOD	0.15	E	0.37	0.13
47	0.009	0.10	0.30	0.89	12.0	4.9	<LOD
66	<LOD	A	<LOD	<LOD	0.21	0.067	<LOD
100	<LOD	0.006	0.029	0.54	1.9	1.0	0.037
99	<LOD	0.019	0.097	1.5	6.2	3.9	0.11
85	<LOD	A	<LOD	0.11	0.42	0.23	<LOD
154	<LOD	A	<LOD	0.15	0.36	0.28	0.015
153	<LOD	A	0.007	0.24	0.41	0.37	0.012
138	<LOD	A	<LOD	<LOD	0.067	0.062	<LOD
128	<LOD	A	<LOD	<LOD	<LOD	<LOD	<LOD
183	<LOD	A	<LOD	<LOD	0.028	0.013	<LOD
190	<LOD	A	<LOD	<LOD	<LOD	<LOD	<LOD
203	<LOD	A	<LOD	0.043	0.028	0.012	0.081
206	B	C	B	0.51	0.43	0.23	1.1
209	0.87	0.45	0.38	15.0	2.8	3.0	7.6
Weight (g)	1.57	0.93	6.51	1.74	1.12	6.99	2.00

<LOD - Below the limit of detection of 0.005 µg /g.

A - Below the limit of detection of 0.0054 µg/g.

B - Below the limit of detection of 0.05 µg /g.

C - Below the limit of detection of 0.054 µg/g.

D - Below the limit of detection of 0.34 µg/g.

E - Below the limit of detection of 0.5 µg /g.

F - Below the limit of detection of 1 µg/g.

Table 3. PBDE congeners detected on firefighter hoods ($\mu\text{g/g}$).

PBDE	Unused Hood	Hood 1*	Hood 2	Hood 3
17	G	0.29	0.079	D
28	G	0.35	0.071	0.04
71	G	0.35	0.74	0.28
47	G	18.0	22.0	3.0
66	G	0.24	0.31	0.025
100	G	3.6	3.9	0.55
99	H	13.0	18.0	2.2
85	G	0.63	0.64	0.12
154	G	0.42	0.54	0.16
153	G	1.0	0.92	0.23
138	G	<LOD	0.035	0.036
128	G	<LOD	<LOD	<LOD
183	G	<LOD	<LOD	<LOD
190	G	<LOD	<LOD	<LOD
203	G	<LOD	B	<LOD
206	G	0.83	0.68	F
209	0.7	16.0	7.8	22.0
Weight (g)	4.68	3.24	3.23	3.04

*Another sample of the same clothing previously analyzed for phthalates and PAHs.

<LOD - Below the limit of detection, 0.005 $\mu\text{g}/\text{gram}$.

A - Below the limit of detection of 0.0054 $\mu\text{g}/\text{gram}$.

B - Below the limit of detection of 0.05 $\mu\text{g}/\text{gram}$.

C - Below the limit of detection of 0.054 $\mu\text{g}/\text{gram}$.

D - Below the limit of detection of 0.34 $\mu\text{g}/\text{gram}$.

E - Below the limit of detection of 0.5 $\mu\text{g}/\text{gram}$.

F - Below the limit of detection of 1 $\mu\text{g}/\text{gram}$.

G - Below the limit of detection of 0.0042 $\mu\text{g}/\text{gram}$.

H - Below the limit of detection of 0.023 $\mu\text{g}/\text{gram}$.

All layers of the unused glove tested contained detectable quantities of PBDE-209 and -47. The middle layer also contained PBDE-28, -99, and -100. In addition, the outer layer contained PBDE-99, -100, and -153. Of all PBDE congeners found in the layers of the unused glove, PBDE-209 was present in the highest quantities. It was also the only PBDE found in the unused hood.

Discussion

From the results of this study, it is apparent that flame retardants, especially DecaBDE, were being added to firefighter glove and hood materials during manufacture. Additional PBDE contamination accumulated during fire response. With only one exception, all detectable levels of PBDEs on items of used firefighter personal protective clothing (82 concentrations of 15 different PBDEs) were higher than those on the unused clothing. The only exception was that the level of PBDE-47 on the inner layer of Glove 2 was non-detectable ($<0.005 \mu\text{g/g}$), while that on the inner layer of the unused glove was measured as $0.009 \mu\text{g/g}$.

In the analysis of SF firefighter blood samples by Shaw et al., the most abundant PBDE congeners found were PBDE-47, -153, and -209.^[12] In the study reported here, higher levels of PBDE-99 and -100 were detected

than -153. This could reflect differences in the compositions of flame retardants used in California and in Ohio, or differences in the biological uptake and metabolism of the different congeners. It has been suggested that debromination of PBDE-209 to lower-brominated congeners such as PBDE-153 may occur.^[17] This is a subject for further study.

In the SF firefighter study, exposures were attributed to inhalation, but dermal absorption is also a possibility. The firefighter with the highest serum levels of PBDEs did not wear respiratory protection.^[12] However, higher serum PBDE levels were not consistently observed for those firefighters who reported wearing no respiratory protection. Therefore, the possibility of PBDE dermal absorption cannot be ruled out.

In the current study, two samples of used firefighter personal protective clothing were taken from a coat and a hood that had previously been analyzed for the presence of PAHs and phthalate diesters.^[10] Table 4 shows the previous results for PAHs and phthalate diesters as well

Table 4. Hood and Wristlet 1 - Total contaminants ($\mu\text{g/g}$).

Chemical	Hood 1	Coat wristlet 1
1-Methylnaphthalene	1.4	<LOD
2-Methylnaphthalene	1.4	<LOD
Acenaphthylene	3.6	0.7
Anthracene	0.9	<LOD
Benzo(a)anthracene	1.4	<LOD
Benzo(a)pyrene	<LOD	0.4
Benzo(b)fluoranthene	<LOD	0.4
Benzo(g,h,i)perylene	<LOD	0.3
Benzo(k)fluoranthene	<LOD	0.8
Chrysene	1.3	<LOD
Dibenzo(a,h)anthracene	0.8	<LOD
Fluoranthene	<LOD	0.5
Fluorene	0.8	<LOD
Indeno(1,2,3-cd)pyrene	<LOD	0.2
Naphthalene	3.1	0.2
Phenanthrene	<LOD	3.8
Pyrene	<LOD	<LOD
Butyl benzyl phthalate	<LOD	8.0
Di(2-ethylhexyl)phthalate	340.0	220.0
Diethyl phthalate	<LOD	<LOD
Di-n-butyl phthalate	<LOD	<LOD
Di-n-octyl phthalate	<LOD	1.8
PBDE 17	0.29	0.010
PBDE 28	0.35	0.045
PBDE 71	0.35	0.54
PBDE 47	18.0	6.6
PBDE 66	0.24	0.094
PBDE 100	3.6	1.5
PBDE 99	13.0	5.7
PBDE 85	0.63	0.28
PBDE 154	0.42	0.38
PBDE 153	1.0	0.43
PBDE 138	<LOD	0.064
PBDE 128	<LOD	<LOD
PBDE 183	<LOD	0.015
PBDE 190	<LOD	<LOD
PBDE 203	<LOD	0.054
PBDE 206	0.83	0.8
PBDE 209	16.0	14.0

<LOD - Below the limit of detection

as PBDE concentrations from the current study. The relative proportions of PBDE congeners on the two clothing items, which were worn together during fire response, were similar. This is in contrast to the levels of PAHs and phthalates measured previously, which differed greatly between the two clothing items.

In this investigation, the three layers of the one unused and one of the two used gloves (Glove 1) were separated and analyzed. The outer layer of the glove provides cut resistance; the middle layer is a moisture barrier; and the inner layer provides thermal insulation. As the outer layer of the glove comes in closest contact with burned materials during use, one might expect it to hold the highest concentrations of contaminants. However, in our previous study that measured PAH and phthalate contamination, the highest concentration of many of the PAHs was found on the middle layer of the used glove.^[10]

Similarly, in this investigation, the middle layer of the used glove contained the highest concentration of many of the PBDEs, even though the levels on the middle layer in the unused glove were lower in almost every case than those on the outer layer. This could indicate that flame retardants are penetrating the other layers of the glove during use and accumulating on this water-impermeable layer, or that the fabric of this layer releases flame retardants during use. The innermost glove layers, worn in contact with the skin, also held detectable levels of at least 8 PBDEs. Contaminants might be present on this layer as a result of transfer from firefighters' hands, transfer from the middle layer, release from the inner-layer fabric, or a combination of all three.

Firefighter hoods are intended to prevent dermal exposure to flames and heat; however, as noted previously, they are worn next to some of the most permeable skin in the human body.^[10] The permeability of the skin of the jaw angle, forehead and scalp is high and exceeded only by that of the scrotum.^[23] The used hoods examined in this study were all contaminated with at least 11 PBDEs, at levels up to 22 $\mu\text{g/g}$. If hoods are not washed frequently, they can continue to expose firefighters whenever they are worn, donned or doffed, even during training.

A large variety of PBDEs were detected on the soiled protective clothing samples. Even as they are eliminated from newly manufactured furnishings and electronics, PBDEs will continue to be found in older items at fire scenes. As shown in this study, even unused protective clothing items can contain flame retardants. PBDE-209 was detected in every sample of unused protective clothing at a minimum of 76 times the LOD, a level not likely to be caused by incidental contamination.

A study by Shen et al. found high levels of PBDEs, PAHs, and polychlorinated biphenyls (PCBs) in vacuum cleaner dust from 20 California (CA) fire stations. In fact, median PBDE-209 concentration exceeded the median

concentration found in a sampling of California residences by a factor of 20.^[24] The researchers attributed some of the PBDE and PAH contamination in fire station dust to ash carried back from fire events. The results of our study would support that conclusion.

Shen et al. could not attribute all of the notably high concentration of PBDE-209 in CA fire station dust to ash from fire events. They hypothesized that some of the PBDE-209 may stem from turnout gear and vehicles that had been treated with flame retardants.^[24] The significant concentrations of PBDE-209 found in an unused firefighter glove and hood in this study would also support that theory.

Stevenson et al.^[25] studied the endocrine-disrupting effect of deposits on firefighting gear and found significant differences in estrogenic and anti-estrogenic activity between extracts of different layers of a new firefighter glove, and between extracts of new and used gear. They attributed these differences to the potential presence of different flame retardants. This is also consistent with the findings of this study.

Conclusions

In multiple studies, contaminants including metals, PAHs and plasticizers have been shown to accumulate in firefighter personal protective clothing with use.^[7,9,10] This study is the first to show that PBDEs also accumulate in firefighter personal protective clothing. Because PBDEs are distributed so widely and are so environmentally persistent, firefighters are continuing to be exposed to them, even as they are being eliminated from newly manufactured items. This exposure may contribute to the high rates of certain cancers that have been observed in firefighters, as noted in the meta-analysis by LeMasters et al.^[3] and in the mortality and cancer incidence study of Daniels et al.^[5]

Contaminants found in the bloodstream of firefighters are often attributed to inhalation exposure, but the contribution of dermal exposure has not been quantified. The current findings show that significant concentrations of PBDEs are found in personal protective clothing worn in contact with the skin. The contribution of dermal exposure to the uptake of flame retardants remains unquantified.

Recommendations

Results of this study suggest that flame retardants such as PBDEs accumulate in firefighter protective clothing over time. As has often been noted, administrative controls (prompt and frequent changing and washing of protective clothing) and personal hygiene practices (hand-washing and showering), can reduce potential exposure to flame

retardants and other toxic agents. Following decontamination principles of removing contaminants at the scene of structural fires, much like at a hazmat incident, would therefore be a potentially important step in improving health protection. These practices could be performed at fire scenes, as well as during and after training sessions. This could help reduce PBDE levels in dust such as that found at CA fire stations.^[24]

In this study, significant levels of PBDEs were found on personal protective clothing worn in contact with the skin, including the inner layer of gloves, the cuff of a coat, and firefighter hoods. The contribution of dermal absorption to PBDEs should therefore be investigated in addition to inhalation exposure. We report significant levels of PBDE contamination on the clothing items examined, although the amount of di-(2-ethylhexyl) phthalate (DEHP) found previously was far higher, compared to levels of any other contaminant. There was over 15 times as much DEHP on the coat wristlet and Hood 1 as the most abundant PBDE congener. High levels of PBDEs have been detected in blood serum from California firefighters.^[12] Information on levels of DEHP metabolites in firefighters' bodies is lacking, however, and clearly a needed subject for further research.

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