Elevated Exposures to Polycyclic Aromatic Hydrocarbons and Other Organic Mutagens in Ottawa Firefighters Participating in Emergency, On-shift Fire Suppression

Jennifer L.A. Keir¹, Umme S. Akhtar¹, Lt. David M.J. Matschke², Tracy L Kirkham³, Hing Man Chan¹, Pierre Ayotte⁵, Paul A. White^{1,4*}, Jules M. Blais^{1*}

 ¹ Department of Biology, University of Ottawa, 30 Marie Curie, Ottawa ON, K1N 6N5 Canada
 ² Ottawa Fire Service, 1445 Carling Ave, Ottawa, ON, K1Z 7L9 Canada;
 ³ Dalla Lana School of Public Health, University of Toronto, 155 College St, Toronto, ON, M5T 3M7 Canada;
 ⁴ Environmental Health Science and Research Bureau, Health Canada, 50 Colombine Driveway, Ottawa, ON, K1A 0K9 Canada
 ⁵ Centre de toxicologie du Québec, Institut national de santé publique du Québec, and Université Laval, 945 avenue Wolfe, Québec City, QC, G1V 5B3 Canada

*Corresponding Authors: Jules.Blais@uottawa.ca; Paul.White@hc-sc.gc.ca

Supporting information is nine pages containing one figure and seven tables.



Figure S1. Illustration showing sample collection procedures for firefighter participants. The first and last panels show the locations for wipe sample collections. For the personal air samples, the white arrows in the center panel indicate the location of the pump, the connecting tubing, and the sample collection tube. Particulate material was collected on quartz filters; volatiles and semi-volatiles were collected on polyurethane foam (PUF) plugs. This paper focused on evaluating urinary metrics and compared with dermal wipe and personal air sample results. Other metrics will be considered in a subsequent paper. Photos courtesy of D. Matschke and A. Wu, used with permission.

Table S1. PAHs measured in personal air and wipe samples, and their respective urinary

metabolite(s) measured in urine.

Parent PAHs measured	
in personal air & wipe	
samples	Urinary PAH Metabolite(s)
Naphthalene	1-Hydroxynaphthalene, 2-Hydroxynaphthalene
Fluorene	2-Hydroxyfluorene, 3-Hydroxyfluorene, 9-Hydroxyfluorene
Phenanthrene	1-Hydroxyphenanthrene, 2-Hydroxyphenanthrene,
	3-Hydroxyphenanthrene, 4-Hydroxyphenanthrene,
	9-Hydroxyphenanthrene
Fluoranthene	3-Hydroxyfluoranthene
Pyrene	1-Hydroxypyrene
Benz(<i>a</i>)anthracene	1-Hydroxybenz(<i>a</i>)anthracene, 3-Hydroxybenz(<i>a</i>)anthrancene
Chrysene	2-Hydroxychrysene, 3-Hydroxychrysene, 4-Hydroxychrysene,
	6-Hydroxychrysene
Benzo(a)pyrene	3-Hydroxybenzo(<i>a</i>)pyrene
Acenaphthylene	
Acenaphthene	
Anthracene	
Benzo(b)fluoranthene	
Benzo(k)fluoranthene	
Indeno(1,2,3-cd)pyrene	
Dibenz(ah)anthracene	
Benzo(ghi)perylene	

Table S2. Summary of creatinine-adjusted urinary PAH metabolite concentrations for office worker controls and firefighters (FF) (i.e., both pre- and post-fire suppression).

	Urinary PAH metabolites (µg/g creatinine)											
	1-hydroxypyrene			Σhydroxyphenanthrenes			Σhydroxyfluorenes			<i><u>Shydroxynaphthalenes</u></i>		
	Ν	Range	GM (SE)	Ν	Range	GM (SE)	Ν	Range	GM (SE)	Ν	Range	GM (SE)
Office workers	13 ^a	0.03-0.16	0.07 (0.01)	17 ^c	0.12-0.73	0.26 (0.02)	18	0.14-1.58	0.39 (0.02)	18	1.01-15.99	4.92 (0.37)
FF pre- fire	27 ^b	0.02-0.33	0.10 (0.01)	30 ^d	0.09-0.98	0.35 (0.02)	31	0.12-1.17	0.48 (0.02)	31	1.94-13.30	5.59 (0.21)
FF post- fire	31	0.06-1.81	0.27 (0.02)	31	0.20-6.56	0.89 (0.06)	31	0.32-7.09	1.31 (0.07)	31	2.83-75.79	12.52 (0.72)
Fold change ^e	27	NI-38.9	4.0	30	NI-63.4	5.3	31	NI-33.2	3.9	31	NI-12.2	2.9

SE, standard error; NI, no increase; GM, geometric mean.

Bold values indicate significant differences between post-event firefighters and both office workers and pre-event firefighters (p<0.0001).

^a Five samples omitted due to technical difficulties and/or chromatographic interferences (i.e., overlapping peaks).

^b Four samples omitted due to chromatographic interferences.

^cOne sample omitted due to technical difficulties.

^dOne sample omitted due to technical difficulties.

^eBold values indicate significant changes in average post- to pre-event fold changes across all subjects (p<0.05).

Table S3. Summary of creatinine-adjusted mutagenic potency values for office worker controls and firefighters (FF) (i.e., both pre- and post-fire suppression).

	(re	Urinary Mutagenicity (revertants/µmol creatinine)					
	N Range GM						
Office workers	18	0.17-10.35	0.87 (0.08)				
FF pre-fire	31	0.19-5.76	1.01 (0.07)				
FF post-fire	31	0.51-22.68	1.90 (0.12)				
Fold change ^a	31	NI-74.7	4.32				

SE, standard error; NI, no increase; GM, geometric mean.

Bold values indicate significant differences between post-event firefighters and both office workers and pre-event firefighters (p<0.0001).

^a Bold values indicate significant changes in average post- to preevent fold changes across all subjects (p<0.05). **Table S4.** Summary of creatinine-adjusted urinary biomarker concentrationsfor office worker controls and firefighter (FF) subjects (i.e., both pre- and post-fire event).

	Urinary Biomarkers (ng/mg creatinine)									
		8-iso-P	GF _{2α}	Clara Cell 16						
	N	Range	ge GM (SE)		Range	GM (SE)				
Office workers	18	0.2-9.8	2.1 (0.2)	18	0.2-50.6	8.5 (1.1)				
FF pre-fire	31	0.2-5.9	1.0 (0.4)	31	0.4-17.8	3.8 (0.2)				
FF post-fire	31	0.2-8.0	1.1 (0.1)	31	0.6-15.8	3.2 (1.0)				
Fold change	31	NI-13.2	1.8	31	NI-3.8	1.2				

SE, standard error; NI, no increase; GM, geometric mean.

Bold values indicate significant differences between office workers and firefighters both pre- and post-fire event (p<0.05).

Table S5. Effects of Personal Air Total PAH Level, or the Product of Personal Air PAH Level

and Fire Suppression Time, on Urinary Levels of Various PAH Metabolites.

		Log Person PAH Leve	al Air Total el (ng/m ³) ¹	Log Product of Personal Air Total PAH Level (ng/m ³) and Duration of Fire Suppression (min) ¹			
Dependent Variable (post versus pre fold change)	N	r ²	p value ²	r ²	p value ²		
Total PAH Metabolites ³	24	0.27	0.0097	0.34	0.0025		
Σ OH-Naphthalenes	29	0.23	0.0090	0.32	0.0018		

¹Log of difference between post-event level and pre-event level. ²p value associated with F ratio.

³One highly influential observation removed (see text); Four samples were omitted due to incomplete sums of urinary metabolites (i.e., concentration of one or more metabolite unavailable due to technical difficulties)

		Log In Derm Levels PAHs	crease in al Wipe of LMW (ng/cm ²) ¹	Log In Dermal V of HM (ng/	crease in Vipe Levels W PAHs ′cm ²) ¹	Log Increase in Dermal Wipe Level of Total PAHs (ng/cm ²) ¹		
Dependent Variable (post versus pre fold change)	N ⁴	r ²	p value ²	r ²	p value ²	r ²	p value ²	
Total PAH Metabolites ³	25	0.43	0.0022	0.44	0.0026	0.43	0.0018	
OH-Pyrene ³	25	0.25	0.028	0.23	0.045	0.28	0.017	
ΣOH -Phenanthrenes ³	28	0.26	0.019	0.34	0.0052	0.31	0.0067	
ΣOH -Fluorenes ³	29	0.19	0.043	0.35	0.0036	0.36	0.0027	
Σ OH-Naphthalenes	29	0.44	0.0005	0.38	0.0020	0.43	0.0005	

Table S6. Effect of Dermal PAH Contamination on Urinary Levels of Various PAH Metabolites.

¹Log of difference between post- and pre-event level. ²p value associated with F ratio. ³One outlier removed (see text for explanation). ⁴Four individual(s) lacked 1-OHP values (i.e., one or more metabolite was not quantified due to technical difficulties), and one individual lacked Σ OH-Phen data.

Table S7. General linear model describing event-related increases in total urinary PAH

 metabolites (fold-change) as a function of PAH concentrations in air and differences in PAHs on

 dermal wipes between post and pre fire events (N=25).

Dependent Variable	Independent Variable	t value	p value					
Fold-change of Total Urinary	Log Difference in Total Dermal PAH	3.03	0.0075					
PAH Metabolites	Log Personal Air Total PAH	2.09	0.052					
Model $r^2 = 0.54$, F ratio = 10.11, p=0.0013								