Original Article

The Characterization of Polycyclic Aromatic Hydrocarbons in Northeastern US Trucking Terminals

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Abstract

In recent years, significant attention has been given to polycyclic aromatic hydrocarbons (PAHs) exposures given their mutagenic and carcinogenic properties. However, levels of exposure and the key determinants of exposure are not well defined for the trucking industry. We measured ultrafine particle characteristics at 10 trucking terminals of varying operating size and location in the Northeast region of the United States using particle concentration counter and a surface area analyzer. Multivariate mixed-effects linear regression models were used to assess determinants of the concentration of total bound PAHs (tPAH), the total aerosol active surface area (AS), and the ratio tPAH/AS overall and individually within docks, trucking cabs, and administrative offices. Associations between PAH measures with integrated measures of elemental carbon (EC), organic carbon (OC), and particulate matter (PM)_{2.5} were assessed by Spearman rank correlation. In adjusted models, tPAH, AS, and tPAH/AS average concentrations (95% confidence interval) were significantly higher in truck cabs compared to office locations (1.26 (ng m⁻³) (1.18, 1.35); 0.99 (mm² mm⁻³) (0.91, 1.08); 0.26 (ng mm⁻²) (0.18, 0.33), respectively). In the loading dock, AS concentrations were significantly higher than in the office (0.67 (0.61, 0.71), while the tPAH/AS was not (−0.63 (−0.67, −0.58)). In each location, average tPAH concentrations were moderately but significantly correlated with EC ($r = 0.47$ – 0.63) and with tPAH/AS

($r = 0.34\text{--}0.40$) in the truck cabs and loading docks. In conclusion, key predictors of tPAH, AS, and tPAH/AS within the trucking industry are work location (in particular truck cabs and terminal docks) and terminal characteristics (size). The association of tPAH and tPAH/AS with EC concentrations in dockworkers and pick-up and delivery drivers is consistent with occupational exposure attributable to vehicle exhaust. Therefore, measurement of tPAH, AS, and tPAH/AS to characterize ultrafine particles and bound PAH concentrations provide additional information regarding exposures in the trucking industry not captured by integrated measures by EC, OC, and $\text{PM}_{2.5}$.

Keywords: PAHs; Ecochem; trucking industry

Introduction

Vehicle exhaust from diesel, gasoline, and propane vehicles is a complex mixture of gases and particulate matter (PM). The main constituents of PM from exhaust consist of agglomerates of elemental carbon (EC) and organic carbon (OC) $<1.0\text{ }\mu\text{m}$ in diameter. Traditionally, EC and OC have been used as markers of exposure to vehicle particulate emissions (Song *et al.*, 2011). However, more recently, significant attention has been given to polycyclic aromatic hydrocarbons (PAHs), which are usually bound to particles, since they are major contributors to the mutagenic and carcinogenic properties of PM from vehicle exhaust (Polidori *et al.*, 2008).

These emissions—PAHs in particular—have been previously shown to be associated with adverse health effects resulting from both short-term and long-term exposure. In addition to dermal exposure, a potential PAH exposure pathway is via inhalation where PM size, the concentration of PM with bound PAHs, and the relationship between PM active surface area and bound PAHs determine the potential for adverse health effects (Polidori *et al.*, 2008). Previous research has suggested that potential long-term exposure to PAHs include disease manifestation such as an increased risk of skin, lung, bladder, and gastrointestinal cancer (Boffetta *et al.*, 1997; Armstrong *et al.*, 2004; Olsson *et al.*, 2010; Diggs *et al.*, 2011).

Of major concern are PAHs levels in trucking terminals as vehicle exhaust from diesel, gasoline, and propane vehicles is ubiquitous in the trucking industry. As a result, these workers, from those working in administrative offices to those working on the docks and driving trucks, may be exposed to harmful PAH levels. Unfortunately, little is known about the extent of their exposure. There have been only a handful of papers describing PAH exposures in the trucking industry—most notably Polidori *et al.* (2008), Ott and Seigman (2006), and Marr *et al.* (2004) who each sampled truck driver exposures by placing PAH monitors within trucking cabs. The magnitude, temporal patterns, and determinants of exposure intensity are not well defined for the trucking industry. Further, less is known about PAH exposures

experienced by non-drivers in the trucking industry, such as those who work on the terminal docks and in the administrative offices.

Our objectives were to characterize PAH concentrations in trucking terminal-based jobs in the Northeastern USA, describing temporal and spatial variations of PAHs. Specifically, we descriptively characterized the temporal and spatial variations of PAHs quantified by the tPAH on the surface of particles with a diameter below $1\text{ }\mu\text{m}$ (ng m^{-3}) and the AS of all PAH particles ($\text{mm}^2\text{ mm}^{-3}$); We leveraged these data to calculate the ratio (tPAH/AS), a measure of the amount of bound PAHs per unit of active surface of particles. This ratio has been proposed to be a measure of PAHs delivered to the lower respiratory tract (Ott and Seigmann, 2006).

Further, we assessed determinants of short-term PAH exposures, and determined the correlation of PAHs with traditional markers of air pollution and vehicle emissions such as EC, OC, and PM less than 2.5 microns in diameter ($\text{PM}_{2.5}$) to determine their added value to exposure assessment in the industry. This paper represents the first step towards estimating PAH exposure of trucking terminal workers in 10 work locations in the Northeastern USA. Our next step will be to additionally consider the relationship between PAH exposure and oxidative stress as indicated by biomarkers systemic inflammation, endothelial activation, and oxidative stress as was done previously for EC, OC, and $\text{PM}_{2.5}$ (Neophytou *et al.*, 2013).

Materials and Methods

Measurement sites

From May 2009 to September 2010, we measured ultrafine particle characteristics at 10 trucking terminals, each located within 450 miles of Boston, Massachusetts. The terminals included a mixture of urban and rural locations. Each terminal was sampled for 1 week (typically Sunday–Friday) and the order of terminals was determined randomly. Within each terminal, we identified three different work locations, representing a range of exposures to vehicle emissions: the cab of local pick-up

and delivery trucks, freight loading docks, and office areas. Area samples in the dock and office were placed with assistance of the shift supervisors to be close to areas of greatest activity for that shift while remaining out of the path of employees. Two 12-hour samples (typically 7 AM to 7 PM and 7 PM to 7 AM) were collected daily in the dock and the office. For samples within truck cabs, all samplers were placed directly in the cab with the driver, as close to the breathing zone as feasible.

Exposure measures

We measured ultrafine particle concentrations of PAHs and particle active surface area using two real-time, portable instruments, the EcoChem PAS 2000CE (PAS) and the EcoChem DC 2000CE (DC). The limits of detection were $\sim 1 \text{ ng m}^{-3}$ and $\sim 1 \text{ mm}^2 \text{ mm}^{-3}$ for tPAH and AS, respectively. Data were logged in 10- to 30-second intervals, and all data averaged over 30-second intervals to produce a uniform time series. Only samples that ran for longer than 2 hours were included.

Integrated samplers for $\text{PM}_{2.5}$ and both EC and OC in $\text{PM}_{1.0}$ (PM with a diameter $\leq 1.0 \mu\text{m}$) were co-located with the tPAH and AS measurements. Details of the integrated sample collection have been provided in detail previously (Smith *et al.*, 2006; Neophytou *et al.*, 2013). Briefly, EC and OC were measured by collecting $\text{PM}_{1.0}$ on a 22-mm quartz tissue filter, preceded by a precision-machined cyclone separator (SCC1.062 Triplex, BGI, Inc., Waltham, MA), which was then analyzed with a thermal-optical carbon analyzer using the NIOSH 5040 method (Smith *et al.*, 2006). $\text{PM}_{2.5}$ was collected on a 37-mm Teflon filter (with a pore diameter of $0.2 \mu\text{m}$) after passing through a precision-machined cyclone pre-selector (GK2.05 SH (KTL), BGI, Inc.) to remove particles greater than $2.5 \mu\text{m}$ in aerodynamic diameter. The mass was measured gravimetrically using a method consistent with the EPA PQ200 Federal Reference Method (Smith *et al.*, 2006).

Meteorological variables

Meteorological variables such as average daily temperature ($^{\circ}\text{C}$), average humidity (%), average precipitation (cm), and wind speed (kph), were gathered from Weather Underground (wunderground.com/history) for the day the sampling occurred and were used as proxies for infiltration, window and door opening in the offices, number of open freight doors on the dock, and window opening in the truck cabs.

Statistical analysis

Time-series plots for tPAH, AS, and tPAH/AS over each sampling period were generated to visually examine exposure patterns. Descriptive statistics (mean, standard

deviation, geometric mean, geometric standard deviation, median, and range) were calculated overall and by work location.

Linear mixed-effects models that accounted for the correlation of repeated measures within each sample were used to determine the predictors of tPAH, AS, and tPAH/AS. The unique identifier for each sampling session and specific sampler served as our random intercept and an autoregressive covariance matrix was chosen to adjust for the correlation of 30-second measurements within each sampling session as it was shown to minimize Akaike's information criterion. We examined a number of potential predictors including: sampler location (office, dock, truck cab); time of day (morning rush (6 AM to 10 AM), work day (10 AM to 4 PM); evening rush (4 PM to 8 PM) and night (8 PM to 6 AM); day of the week; daily averages for temperature, humidity, precipitation, and wind speed; and season (winter, spring, summer, and fall). For location-specific models, Tuesday, Wednesday, and Thursday concentrations were collapsed due to small numbers of mid-week samples on the loading dock and truck cab. We also considered the impact of instrument placement within the office (near a fan or near a copier); and whether or not a truck driver smoked in his cab. Response variables (exposure concentrations) were log-transformed to ensure normally distributed residuals.

To determine the precision and accuracy of our models, we used a cross-validation procedure. We randomly selected 10% of the samples to hold out of the model development steps described above. After our final models were determined, we used the regression parameters to predict tPAH, AS, and tPAH/AS within the held out samples. We calculated correlations between the measured and predicted values and regressed the predicted values against the measured values to obtain intercepts and slopes.

To determine the associations of tPAH, AS, and tPAH/AS with integrated measures of EC, OC, and $\text{PM}_{2.5}$, matched to the same sampling run times, we calculated the Spearman rank correlation coefficient. We considered the average concentration, 95th percentile and maximum concentration of tPAH, AS, and tPAH/AS. An alpha level of 0.05 was used to determine statistical significance. All analyses were conducted using SAS (version 9.3; SAS Institute Inc., Cary, NC).

Results

We obtained a total of 99 unique real-time, continuous samples: 33 truck cab, 27 dock, and 39 office samples. In total, there were 127,063 30-second measurements.

Table 1 describes the distribution of these 30-second measurements and integrated measures of EC, OC, and PM_{2.5} both overall and across each sampler location by the covariates of interest. Overall, the most samples were collected in the office, mid-week, between 10 AM and 4 PM, and during the spring season. Throughout the course of sampling, average temperature and relative humidity were 13°C and 69%, respectively. Wind speed was on average 11 kilometers per hour and average precipitation was 0.47 centimeters. The majority of office samples were not obtained near a copier or fan, and only 14% of the truck cab samples were in the truck cabs of drivers who smoked. Due to study design, the majority of truck cab samples were collected on Mondays and

Fridays and the time of collection patterns varied across the locations. EC concentrations were higher in the truck cabs and loading dock compared to the office.

Representative time plots for each sampling location are shown in Fig. 1. Generally, for dock samples, log(AS) was higher than log(tPAH). Truck cab log(AS) and log(tPAH) levels were similar, and in the office samples, log(tPAH) levels were much higher than log(AS). Additionally, patterns of temporal variability suggested that time of day influenced both log(tPAH) and log(AS) concentrations. For example, in dock samples, peaks were correlated with shift changes. In office settings, lower levels during overnight sampling coincided with times when the area was likely less occupied.

Table 1. Distribution of covariates overall and by sampler location—average (SD)

	Overall	Dock	Truck cab	Office
N				
Samples	99	27	33	39
30 sec averages	127,063	34,407	39,910	52,746
30 sec averages, N (%)				
Day of the week				
Monday	25,269 (19.9)	2,646 (7.7)	19,846 (49.7)	2,777 (5.3)
Tuesday	21,093 (16.6)	7,965 (23.1)	1,305 (3.3)	11,823 (22.4)
Wednesday	30,023 (23.6)	12,127 (35.2)	0 (0.0)	17,896 (33.9)
Thursday	30,339 (23.9)	10,296 (29.9)	2,719 (6.8)	17,324 (32.8)
Friday	20,339 (16.0)	1,373 (4.0)	16,040 (40.2)	2,926 (5.5)
Time of Day				
Morning Rush (6–10 am)	20,042 (15.8)	4,432 (12.9)	7,750 (19.4)	7,860 (14.9)
Work day (10 am–4 pm)	46,578 (36.7)	9,631 (28.0)	22,037 (55.2)	14,910 (28.3)
Evening Rush (4–8 pm)	24,758 (19.5)	6,487 (18.9)	8,804 (22.1)	9,467 (17.9)
Night (8 pm–6 am)	35,685 (28.1)	13,857 (40.3)	1,319 (3.3)	20,509 (38.9)
Season				
Spring	49,958 (39.3)	13,854 (40.3)	16,154 (40.5)	19,950 (37.8)
Summer	30,538 (24.0)	10,527 (30.6)	4,401 (11.0)	15,610 (29.6)
Fall	34,535 (27.2)	7,513 (21.8)	16,759 (42.0)	10,263 (19.5)
Winter	12,032 (9.5)	2,513 (7.3)	2,596 (6.5)	6,923 (13.1)
Location				
Near copier	11,761 (9.3)	0 (0.0)	0 (0.0)	11,761 (22.3)
Near fan	10,673 (8.4)	0 (0.0)	0 (0.0)	10,673 (20.2)
Smoker	5,428 (4.3)	0 (0.0)	5,428 (13.6)	0 (0.0)
Mean (SD)				
Meteorological variables				
Temperature (°C)	13.1 (8.1)	14.3 (9.7)	11.4 (5.1)	13.5 (8.5)
Humidity (%)	69.4 (12.8)	68.7 (10.5)	66.8 (12.3)	71.9 (14.1)
Precipitation (cm)	0.47 (0.85)	0.34 (0.46)	0.29 (0.67)	0.70 (1.08)
Wind speed (kph)	10.9 (7.4)	10.6 (7.4)	8.8 (6.0)	12.7 (7.8)
Integrated samples				
EC (µg m ⁻³)	0.59 (0.72)	0.78 (1.00)	0.92 (0.64)	0.19 (0.18)
OC (µg m ⁻³)	8.51 (3.94)	5.32 (2.61)	10.49 (5.08)	9.05 (1.89)
PM _{2.5} (µg m ⁻³)	10.47 (6.91)	13.27 (7.46)	12.55 (7.99)	6.92 (3.26)

Figure 1. Representative time-series plots of active surface (AS) (ng/m^3) and total PAH (tPAH) (mm^2/mm^3) concentration by sampler location (from top to bottom: dock, truck cab, and office). The blue lines represent tPAH concentrations and the red lines are AS concentrations.

In truck cab samples, there were marked rises and falls likely corresponding to variability in in-traffic exposures and freight pickups and drop offs. Summary measures of the exposures are presented in [Table 2](#). The highest

average concentrations were consistently observed in the truck cabs, while the lowest were in the office.

[Table 3](#) presents the multivariable models for $\log(\text{tPAH})$, $\log(\text{AS})$, and $\log(\text{tPAH}/\text{AS})$. Work day and

Table 2. Summary statistics by sampler location: across all terminals

Work location	Variable	Mean	Stdev	Median	Minimum	Maximum	Geometric mean	Geometric stdev
Dock (<i>n</i> = 34,407)	tPAH (ng m ⁻³)	17.0	25.8	7.67	1.0	1,088.7	9.0	2.9
	AS (mm ² mm ⁻³)	61.4	89.2	31.3	1.0	1,388.0	33.1	2.9
	tPAH/AS (ng mm ⁻²)	0.51	0.81	0.28	0.00	49.3	0.3	3.4
Truck cab (<i>n</i> = 39,910)	tPAH (ng m ⁻³)	79.8	112.9	28.7	1.0	1,556.0	30.6	4.4
	AS (mm ² mm ⁻³)	73.4	103.1	41.0	1.0	2,367.3	41.3	3.0
	tPAH/AS (ng mm ⁻²)	1.73	5.04	0.71	0.00	468.0	0.7	3.6
Office (<i>n</i> = 52,746)	tPAH (ng m ⁻³)	14.7	24.0	8.0	1.0	294.0	8.0	2.9
	AS (mm ² mm ⁻³)	25.0	27.4	18.0	1.0	467.7	16.4	2.6
	tPAH/AS (ng mm ⁻²)	1.41	4.21	0.42	0.01	168.3	0.5	3.4

Table 3. Mixed model results of log(tPAH), log(AS), and log(tPAH/AS) for all locations

	Log(tPAH)	Log(AS)	Log(tPAH/AS)
	β (95% CI)	β (95% CI)	β (95% CI)
Time of day			
Morning rush (6–10 am)	-0.008 (-0.071, 0.055)	0.013 (-0.047, 0.074)	-0.039 (-0.094, 0.017)
Work day (10 am–4 pm)	-0.235 (-0.29, -0.181)	-0.365 (-0.418, -0.312)	0.155 (0.108, 0.202)
Evening rush (4–8 pm)	-0.194 (-0.254, -0.134)	-0.220 (-0.278, -0.163)	0.032 (-0.020, 0.085)
Night (8 pm–6 am)	Reference	Reference	Reference
Day of week			
Monday	0.208 (0.051, 0.365)	-0.237 (-0.396, -0.077)	0.442 (0.307, 0.576)
Tuesday	-0.046 (-0.26, 0.169)	-0.409 (-0.626, -0.192)	0.358 (0.174, 0.541)
Wednesday	0.300 (0.094, 0.505)	0.016 (-0.192, 0.225)	0.280 (0.104, 0.456)
Thursday	0.327 (0.140, 0.513)	0.081 (-0.108, 0.271)	0.243 (0.084, 0.403)
Friday	Reference	Reference	Reference
Season			
Spring	1.048 (0.949, 1.148)	0.619 (0.518, 0.720)	0.435 (0.350, 0.520)
Summer	1.491 (1.315, 1.666)	1.333 (1.155, 1.512)	0.165 (0.015, 0.315)
Fall	1.381 (1.268, 1.495)	0.270 (0.155, 0.386)	1.118 (1.021, 1.216)
Winter	Reference	Reference	Reference
Sampler location			
Dock	0.036 (-0.016, 0.089)	0.661 (0.607, 0.714)	-0.625 (-0.670, -0.580)
Truck cab	1.263 (1.176, 1.349)	0.993 (0.906, 1.081)	0.258 (0.184, 0.332)
Office	Reference	Reference	Reference
Meteorological variables			
Temperature (°C)	-0.037 (-0.043, -0.030)	-0.052 (-0.058, -0.045)	0.015 (0.009, 0.020)
Humidity (%)	-0.019 (-0.021, -0.017)	0.010 (0.008, 0.012)	-0.029 (-0.031, -0.027)
Precipitation (cm)	0.105 (0.071, 0.139)	-0.129 (-0.163, -0.094)	0.234 (0.206, 0.263)
Wind speed (kph)	-0.010 (-0.015, -0.006)	-0.035 (-0.040, -0.031)	0.025 (0.021, 0.029)

CI = confidence interval.

evening rush hours were the only time periods where statistically significant changes in pollutant concentrations were observed for log(tPAH) and log(AS). Specifically, during the work day and evening rush hours, log(tPAH) and log(AS) concentrations were lower com-

pared to night time concentrations, while for the ratio, they were higher. Log(tPAH) concentrations and the ratio were generally higher on all days compared to Friday. While concentrations were higher in all seasons as compared to winter, the greatest seasonal impact on

log(tPAH) and log(AS) was during the summer while for log(tPAH/AS) it was during the fall. Location tended to play an important role in variations in concentrations. All exposures were statistically significantly higher in the truck cabs compared to the office. There was no statistically significant difference between dock and office log(tPAH) levels; however, log(AS) levels were significantly higher on the dock and the log(tPAH/AS) ratio was significantly lower as compared to the office.

All meteorological variables were shown to have statistically significant effects on the means of each pollutant. Increases in average temperature and wind speed were associated with small reductions in log(tPAH) and log(AS) and small increases in the ratio. As average humidity increased, log(tPAH) and log(tPAH/AS) concentrations decreased slightly, while log(AS) concentrations increased, all effects were statistically significant. Precipitation was associated with increases in log(tPAH) and log(tPAH/AS) and decreases with log(AS).

Results from location-specific mixed models are presented in Supplementary Tables S1–S3 (available at *Annals of Work Exposures and Health*). Similar to the overall models, season and day of the week were important predictors of exposure, and instrument placement was important in the office. Within truck cab samples, smoking status was only a statistically significant predictor of log(tPAH/AS).

Cross-validation results demonstrated that the multivariable model had limited predictability. The correlations between predicted and observed tPAH and tPAH/AS were 0.53 and 0.41, respectively. The linear regression for predicted versus observed tPAH had an R^2 of

0.28, an intercept of 0.81, and a slope of 0.75. The equivalent regression parameters for AS were 0.02, 2.2, and 0.34, and those for the ratio were 0.22, –0.21, and 0.97.

Table 4 shows the Spearman correlations between summary measures of tPAH, AS, and tPAH/AS with co-located integrated EC, OC, and PM_{2.5} measurements—both overall and by each sampler location. When all samples were considered together, the correlation coefficients were largest (0.61–0.68) and most statistically significant for tPAH correlated with EC. This relationship held across the different sampling locations. Statistically significant relationships between tPAH and PM_{2.5} were mainly observed in the full set of samples that was driven by the large differences between the loading dock and the office. AS was correlated with both EC and PM_{2.5} in the full set of samples (0.38–0.46); however, these associations appear to be driven by the office samples (see online Supplementary Material, available at *Annals of Work Exposures and Health*). The tPAH/AS ratio was correlated more strongly with EC than with OC, and was stronger and statistically significant in the dock and truck locations in contrast to the office.

Discussion

In this study, we have demonstrated that concentrations of tPAH, AS, and their ratio vary between and within work locations in the US trucking industry and that features related to characteristics predictive of terminal operations, such as season, time of day, and day of the week were determinants of exposure. In exam-

Table 4. Spearman correlation coefficients for different parameters of tPAH, AS, and tPAH/AS with EC, OC, and PM_{2.5}

	Overall			Dock			Truck Cab			Office		
	EC	OC	PM _{2.5}	EC	OC	PM _{2.5}	EC	OC	PM _{2.5}	EC	OC	PM _{2.5}
tPAH												
Average	0.68***	0.20	0.32**	0.61**	0.26	0.47*	0.51**	0.08	–0.27	0.63***	–0.08	0.15
95 th %ile	0.67***	0.20	0.33**	0.66**	0.12	0.39	0.45*	0.09	–0.21	0.59***	–0.08	0.13
Max	0.61***	0.06	0.37**	0.43*	–0.04	0.09	0.33	0.03	–0.05	0.27	–0.10	0.05
AS												
Average	0.46***	–0.03	0.40***	0.08	0.11	0.05	–0.20	–0.02	0.09	0.52**	0.04	0.32*
95 th %ile	0.44***	–0.04	0.38**	–0.09	0.10	–0.04	–0.16	–0.02	0.12	0.44**	0.12	0.26
Max	0.41***	–0.15	0.41***	–0.19	–0.09	–0.18	–0.29	0.12	0.32	0.20	–0.10	0.04
tPAH/AS												
Average	0.34**	0.27**	0.07	0.48*	0.15	0.34	0.40*	0.15	–0.19	–0.07	–0.11	–0.22
95 th %ile	0.32**	0.25*	0.07	0.36	0.18	0.36	0.45*	0.09	–0.26	–0.15	–0.11	–0.28
Max	0.22*	0.21*	0.14	0.11	0.01	0.19	–0.34	0.18	0.10	–0.26	–0.17	–0.25

* $P < 0.05$; ** $P < 0.01$; *** $P < 0.0001$.

ining correlations between these measures and more traditional integrated measures of EC, OC, and $PM_{2.5}$ in filter samples, we found that tPAH, AS, and tPAH/AS provide that describe the air concentrations of PAH compounds in relation to ultrafine particles that are not otherwise captured. Adjusting for season, time of day, day of the week, and meteorological variables, tPAH, AS, and tPAH/AS average concentrations were significantly higher in a truck cab than in an office location. In the loading dock, AS concentrations were significantly higher than in the office and the tPAH concentrations were also higher (although not statistically so). In each location, the EC in $PM_{1.0}$ was significantly correlated with tPAH, AS, and tPAH/AS. Although tPAH/AS ratios were greater in the office compared to the dock, unlike the office, tPAH/AS in the truck cab and loading dock was also significantly correlated with EC. These results indicate that amount of bound PAHs per unit of active surface of particles is significantly associated with EC exposures in dockworker and pick-up and delivery drivers, jobs with regular exposure to vehicle exhaust in the trucking industry, as compared to office workers.

Temporally, we observed that tPAH levels were lower during the work day as compared to night time and lower on Friday as compared to the other days of the week. Given that these terminals were busiest at night and were closed during the weekend (Fridays were days when activity tapered down in response to the start of the weekend), we are not surprised by these findings. We also hypothesize that lower tPAH and AS concentrations during the winter than other seasons were due to window/door opening patterns.

The results presented in the tables in the online Supplementary Material (available at *Annals of Work Exposures and Health*) indicate that placement of the sampler near the copier significantly increased AS concentrations. In the office, unlike on the loading dock or truck cabs, AS was also correlated with $PM_{2.5}$, suggesting a contribution from indoor exposures that (including the copier) that can also contribute to both small and larger particles. EC exposures on the loading dock and truck cab were higher than in the office. EC in these jobs is a measure of exposure to the carbon core of particles from vehicle emissions, including from diesel exhaust (Attfield *et al.*, 2012; Garshick *et al.*, 2012).

In cross-validation analyses, we demonstrated that our models under-predicted the observed exposures. This was most evident for AS where the slope between predicted and observed was 0.34. Our models were better at predicting the ratio with an intercept of -0.21 and a slope of 0.97. This under-prediction is not surprising given that our models lacked fine temporal scale predictors.

The PAS and DC EcoChem instruments have been used individually and simultaneously in a number of studies to describe differences in PAH and/or AS concentrations across time spans and locations, to categorize particulate type, and allow for comparison to other integrated measures of air pollution (Agarwal *et al.*, 2013; Banks *et al.*, 2013; Hudda *et al.*, 2013; Adar *et al.*, 2015; Kim *et al.*, 2015; Niu *et al.*, 2015). Our results are most appropriately compared to those measuring tPAH and AS within trucking terminals or facilities with diesel-operated vehicles and to other studies in on-road vehicles.

Our findings are similar to our previous research in Mexico City conducted in 2002, where we used both EcoChem instruments to describe ambient and occupational tPAH and AS concentrations in cargo terminals, distribution centers, and trucking cabs (Marr *et al.*, 2004). In that study, we also observed much higher tPAH averages in trucking cabs (596 versus 79.8 $ng\ m^{-3}$ observed here) associated with the substantial air pollution in Mexico City. Assessment of correlations between the EcoChem and integrated filter measures showed strongest correlations between tPAH and EC and weakest between tPAH and OC (Marr *et al.*, 2004). Compared to Mexico City, however, our study was conducted after the introduction of particulate emission controls of heavy duty diesel trucks, a major contributor to on-road EC, and we found a substantially lower tPAH concentration. Our study also found weak or no meaningful associations between OC and PAH, AS, and tPAH/AS, suggesting that total OC on $PM_{1.0}$ is a weak surrogate for total and particle-bound PAH. A number of studies have sampled tPAHs within vehicles to obtain information on in-vehicle commuting exposures. Hudda *et al.* (2013) examined tPAH concentrations in six vehicles of varying age, mileage, volume, and manufacturer while in operation on the Los Angeles Freeway. Average tPAH concentrations inside vehicle cabs were 13 $ng\ m^{-3}$, comparable to the means found on our dock samples (17 $ng\ m^{-3}$) but much lower than our observed levels in truck cabs (79.8 $ng\ m^{-3}$; Hudda *et al.*, 2013). In a study of commuters in Atlanta, Sarnat *et al.* (2014) observed higher average tPAH levels of 118.8 $ng\ m^{-3}$, with a range of exposures from 50 to 207 $ng\ m^{-3}$ (Sarnat *et al.*, 2014). The tPAH exposures in that study had similar patterns of correlation (tPAH correlation with $PM_{2.5}$ = 0.44, EC = 0.76, and OC = 0.30) to what we observed in our full sample. Adar *et al.* (2015) reported levels of tPAHs on diesel school buses with a number of different emission reduction technologies and in electric cars on the same roadways. Average tPAH concentrations in the buses were 101 $ng\ m^{-3}$, with the lowest levels observed in buses using biodiesel and higher levels in

buses with oxidative catalysts or ultra-low sulfur diesel. Average levels in the electric car were 125 ng m^{-3} (Adar *et al.*, 2015). Ott and Seigman (2006) measured tPAH/AS ratios in truck cabs as they drove through traffic and observed trip averages ranging from 0.42 to 0.58 ng mm^{-2} —levels comparable to our dock samples but roughly three times lower than averages observed in both trucking cabs during driving and office settings (Ott and Seigman, 2006). In a mobile monitoring campaign in New Mexico, Banks *et al.* (2013) found mean concentrations of 130 ng m^{-3} during the evening rush hour (Banks, 2013). Northcross *et al.* (2014) investigated tPAH concentrations in stationary vehicles with smoking occupants. The mean tPAH concentrations were 1325.1 ng m^{-3} , which is within the range of values observed in our trucking cabs (Northcross *et al.*, 2014). Taken together, these studies suggest a wide range of exposures exist in on-road vehicles. Therefore, vehicle characteristics, driver window usage and road types, all of which we were unable to assess in our study, may be key determinants of the intensity of PAH exposures, including particle-bound PAH compounds experienced during on-road driving.

Our mixed model results for both dock and truck cab specific locations showed an increase in log tPAH levels during the morning hours and lower levels during mid-day. This is similar to the pattern observed in a study located near the Los Angeles port, where ambient tPAH levels peaked during morning rush hour traffic, tapered off during the day and slightly increased in the evening (Polidori *et al.*, 2008). However, the average tPAH concentrations were 0.64 ng m^{-3} , much lower than even our office samples (Polidori *et al.*, 2008).

There are several potential limitations in our study. As noted above, we did not have time varying information on several types of potential predictors, such as forklift and/or truck activity on and around the docks or window opening behavior and the influence of specific driving activities in truck cabs. We also were unable to include information on specifics about the vehicles being used at any given time point (age, after-treatment system), the distance of all in-use vehicles from each of the sampling locations, or details regarding changes in ventilation in office and dock areas. This lack of information on key variables likely explains our cross-validation results, as we were limited to predicting average levels, as opposed to peaks, in an area. Our models also assume a common source of particles in all models, which is not true in the administrative offices. Further, while we placed area samples in central locations in the dock and office, these levels may not be fully representative of the full range of exposures experienced in these areas. Additionally, the Echochem

only provides information on total particle-bound PAHs in submicron particles. Therefore, we were not able to evaluate PAHs specific to each work location and in particular further evaluate factors associated with tPAH/AS concentrations in the office that included other indoor sources such as the copier. We are also unable to distinguish differences in mixtures of PAHs across or within terminals, as different mixtures cannot be detected from the Ecochem instruments. On the other hand, this study has a number of strengths. It is one of the largest to date to provide information on tPAH, AS, and their corresponding ratio in real world situations that may be experienced by the general public. Our results also indicate a significant correlation between relative amounts of particle-bound PAH per unit of surface area (tPAH/AS) and EC in vehicle exhaust-exposed occupations. Furthermore, it provides insight into predictors of these measures and indicates that they provide information over and above that provided by traditional integrated measures. Finally, we were able to better quantify levels of PAH exposures in trucking terminal workers—in particular, truck drivers and terminal dock workers. Given the potential for the adverse health effects, our results will serve to further inform the levels and nature of exposure to PAHs in the trucking industry.

Supplementary Data

Supplementary data are available at *Annals of Work Exposures and Health* online.

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Conflicts of Interest

The authors declare no conflict of interests.

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