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Indoor air sampling for fine particulate matter and black carbon in industrial communities in Pittsburgh

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HIGHLIGHTS

- $PM_{2.5}$ and BC indoor concentrations were assessed near industrial sources.
- Indoor concentrations were consistently higher than outdoors during both seasons.
- We detected higher indoor $PM_{2.5}$ concentrations during summer than winter.
- Smoking explained greater variability in indoor $PM_{2.5}$ than outdoor estimates.

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ABSTRACT

Impacts of industrial emissions on outdoor air pollution in nearby communities are well-documented. Fewer studies, however, have explored impacts on indoor air quality in these communities. Because persons in northern climates spend a majority of their time indoors, understanding indoor exposures, and the role of outdoor air pollution in shaping such exposures, is a priority issue. Braddock and Clairton, Pennsylvania, industrial communities near Pittsburgh, are home to an active steel mill and coke works, respectively, and the population experiences elevated rates of childhood asthma. Twenty-one homes were selected for 1-week indoor sampling for fine particulate matter ($PM_{2.5}$) and black carbon (BC) during summer 2011 and winter 2012. Multivariate linear regression models were used to examine contributions from both outdoor concentrations and indoor sources. In the models, an outdoor infiltration component explained 10 to 39% of variability in indoor air pollution for $PM_{2.5}$, and 33 to 42% for BC. For both $PM_{2.5}$ models and the summer BC model, smoking was a stronger predictor than outdoor pollution, as greater pollutant concentration increases were identified. For winter BC, the model was explained by outdoor pollution and an open windows modifier. In both seasons, indoor concentrations for both $PM_{2.5}$ and BC were consistently higher than residence-specific outdoor concentration estimates. Mean indoor $PM_{2.5}$ was higher, on average, during summer (25.8 ± 22.7 μg/m^3) than winter (18.9 ± 13.2 μg/m^3). Contrary to the study's hypothesis, outdoor concentrations accounted for only little to moderate variability (10 to 42%) in indoor concentrations; a much greater proportion of $PM_{2.5}$ was explained by cigarette smoking. Outdoor infiltration was a stronger predictor for BC compared to $PM_{2.5}$, especially in winter. Our results suggest that, even in industrial communities of high outdoor pollution concentrations, indoor activities – particularly cigarette smoking – may play a larger role in shaping indoor exposures.

1. Introduction

Impacts of industrial emissions on outdoor air pollution in nearby communities are well-documented (Pope, 2007; Elliott et al., 1999; Curtis et al., 2006; Perlin et al., 1995) and, although outdoor concentrations explain a significant proportion of indoor pollution (Baxter et al., 2007a,b; Abt et al., 2000b; Levy et al., 2010), fewer studies have explored indoor air quality in industrial communities. Indoor pollution

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may have significant bearing on health, and better characterize personal exposures (Clougherty et al., 2011), because persons in Northern climates spend a majority of their time indoors (Wallace, 1996), and indoor pollutant concentrations have been shown higher than outdoor concentrations, even in developed countries (EPA, 2012a; Morawska et al., 2001; Adgate et al., 2002).

Although ambient air pollution has decreased over the past three decades in the U.S., systemic diseases associated with ambient pollution have increased (Lioy and Georgopoulos, 2011; Dominici et al., 2007), and this burden has not been equitably distributed (Clougherty et al., 2011; Gauderman et al., 2004; Samet et al., 2000; Self et al., 2005; Pope et al., 2009; Brunekreef et al., 1997). In low income communities, often located near industrial sites or alongside major roadways in western countries, both indoor and outdoor residential exposures may be highly elevated, and adversely impact health (Pope et al., 2009; Brunekreef et al., 1997).

Indoor concentrations are a composite of outdoor concentrations (which vary by residential location) and indoor sources, modified by ventilation characteristics (Baxter et al., 2007a; Abt et al., 2000a). Spatial variance in outdoor concentrations of fine particulate matter ($PM_{2.5}$) can vary by orders of magnitude across an urban area, attributable to proximity to industrial and traffic sources, and modifying factors such as elevation or meteorology (Clougherty et al., 2011; Adgate et al., 2002). While this variance in outdoor air pollution may result in substantial indoor concentration variability, indoor sources, such as cooking, smoking, and cleaning activities, can contribute significantly to indoor air pollution (Abt et al., 2000a; Semple et al., 2012).

The communities of Braddock and Clairton, Pennsylvania, located immediately east of Pittsburgh, are situated in river valleys along the Monongahela River, and are home to an active steel mill and coke works, annually producing 725.2 and 1048.8 tons of primary $PM_{2.5}$, respectively (USS, 2012; EPA, 2012b). These industrial sources represent two of the largest stationary sources of fine particles in Allegheny County, which has consistently exceeded National Ambient Air Quality (NAAQS) Standards for $PM_{2.5}$ (CDC, 2010; Kelly, 2007; EPA, 2009).

Following on our prior studies on spatial variance in multiple ambient air pollutants across this area (Tunno et al., 2015; Shmool et al., 2014; Tunno et al., 2012), here we examined indoor $PM_{2.5}$ and black carbon (BC) concentrations in Braddock and Clairton households, during summer 2011 and winter 2012, to quantify the contribution of high outdoor concentrations in industrial communities to indoor concentrations, and to compare the contribution of outdoor concentrations vs. indoor sources. We hypothesize that the high outdoor air pollution concentrations in these communities should contribute significantly to indoor concentrations, and further hypothesize that pollutant concentrations would: (1) be higher indoors vs. outdoors, (2) vary by season, and (3) vary by indoor source activity, including cooking and smoking.

2. Methods

2.1. Study design

Families with at least one asthmatic child participating in a cohort recruited by the Pediatric Environmental Medicine Center (PEMC) at Pittsburgh Children's Hospital were invited to participate in the study. Twenty-one homes in and around the Braddock and Clairton communities were sampled for one week during both a summer (July 25th to September 13th, 2011) and winter (January 30th to March 5th, 2012) sampling session. For spatial contrast, six convenience sample homes were recruited from neighborhoods further from the industrial sites. The study area containing the homes was selected within the previously-sampled outdoor monitoring domain, enabling development of spatio-temporal home-specific outdoor estimates, detailed below.

2.2. Monitoring instrumentation and quality control

Indoor $PM_{2.5}$ samples were collected using a Harvard Personal Exposure Monitor (PEM) with a MEDO linear-piston vacuum pump. Teflon™ filters (37 mm) were pre- and post-weighed in a temperature and relative humidity (RH)-controlled (20.0 °C and 35% RH) glove box (PlasLabs Model 890 THC, Lansing, MI) on an ultramicrobalance (Mettler Toledo Model XP2U, Columbus, OH). $PM_{2.5}$ concentrations were calculated using the two PEMs from each home and averaged, for overall $PM_{2.5}$ concentration for the week-long (7-day) sampling duration. Reflectometry was performed on these $PM_{2.5}$ filters using an EEL43M Smokestain Reflectometer (Diffusion Systems Limited, London, England) to estimate black carbon (BC) absorbance units (ISO 9835:1993, 1993), prior to compositional analysis by inductively-coupled plasma mass spectrometry (ICP-MS) at Wisconsin State Hygiene Laboratories. A HOBO Data Logger (Onset devices, Pocasset, MA) recorded temperature and RH every five minutes. Temperature and RH measures from the HOBO device were averaged for the entire sampling period. All measures were corrected using full method blanks.

Samplers were placed in the main activity room, away from windows and combustion or heat sources. After three days, the PEM was replaced, to avoid particle overload on the impactor plate and perturbation of the particle size cut-point. To assess reproducibility, two homes were randomly selected each season for co-located sampling. A standardized log sheet was used to record sampling start and stop times, and questionnaire on indoor source activities was administered in-person to an adult resident of each home, on the final sampling day.

2.3. Indoor questionnaire

An adult over 18 years of age in each home completed an indoor air pollution questionnaire for both summer and winter sampling sessions (Baxter et al., 2007a; Dutta et al., 2007). Questions included items on household composition (i.e., number of adults and children, pets), details on smoking, cooking, cleaning and solvent use, use of pesticides or scented sprays, use of matches, burning of candles or similar, use of doormats, carpeting, and wearing shoes indoors, pests (incl. mice, roaches, insects), mold and mildew, and heating and ventilation characteristics (draftiness, percent of time windows open, air conditioning or humidifier use). Study data was managed using REDCap (Research Electronic Data Capture) hosted by the University of Pittsburgh (Harris et al., 2009). Questionnaire covariates were created, and correlations with $PM_{2.5}$ and BC examined, using SAS version 9.3 (SAS Institute Inc., Cary, NC).

2.4. Outdoor concentration estimates

Home- and week-specific outdoor concentration estimates were derived using our previously-published full-week LUR models for $PM_{2.5}$ and BC (Tunno et al., 2015). This outdoor sampling campaign was systematically designed to sample across 37 areas with contrasting gradients of traffic density, elevation, and industrial emissions (Shmool et al., 2014), in an attempt to seek out the effect of industry, terrain, and traffic congestion on outdoor $PM_{2.5}$ and BC. In these outdoor models, industrial emissions, traffic density, and elevation explained substantial spatial variance across our domain, after accounting for temporal variability using an upwind reference site (Tunno et al., 2015). For the present study, we calculated outdoor concentrations at each home using the mean value from the LUR surface for the area within 300 m of each home, as in Ross et al. (2013), and hourly EPA Air Quality System (AQS) data for $PM_{2.5}$ from the nearby Liberty and Lawrenceville monitoring locations (Fig. 1), averaged for the specific sampling hours at each home. These LUR-based outdoor estimates (Tunno et al., 2015) were also used to calculate indoor/outdoor ratios for $PM_{2.5}$ and BC at each home.

In addition to the LUR-based outdoor concentration estimates, we separately examined effects of "*reference site*" concentrations, and

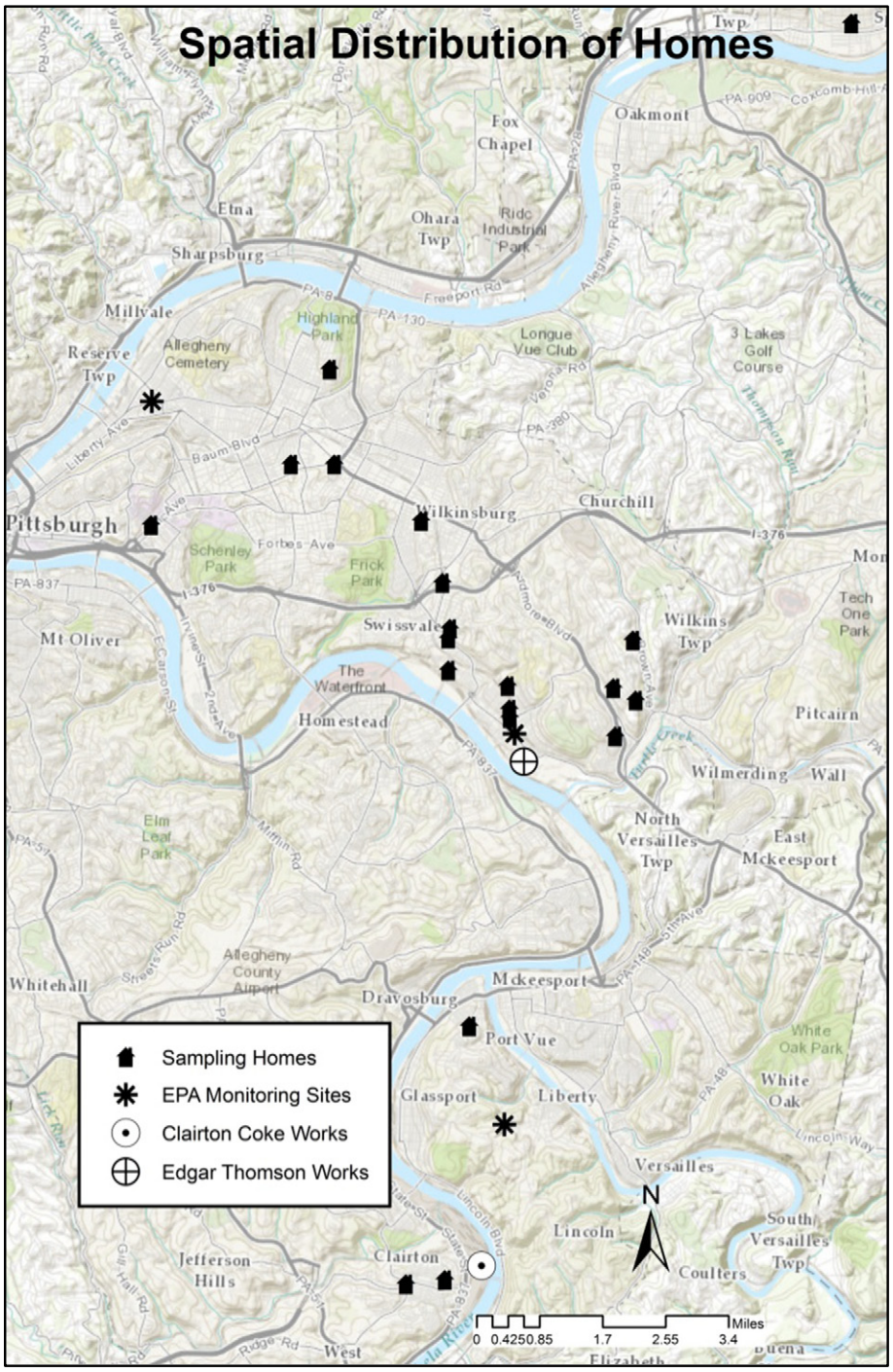

Fig. 1. Spatial distribution of sampling homes, EPA monitoring sites, and large industrial sources.

three additional indicators of spatial variance in outdoor contributions: *distance to industry* (Euclidean distance from each home to the nearer of the Clairton Coke Works or Braddock Edgar Thomson Steel Works), *IDW $PM_{2.5}$ emissions* (reported annual emissions based on 2011 EPA NEI data EPA, 2013), and *emissions/distance* (reported annual emissions from the nearer industrial facility, divided by the home's distance from the site).

2.5. Ventilation indicators

Lacking direct measures of air exchange rates (AER), we derived two proxy indicators: (1) an approximate I/O sulfur ratio, calculated using the indoor sulfur concentrations from ICP-MS analysis of our indoor $PM_{2.5}$ filters, divided by sulfur concentrations from our reference monitors for those sampling days, and (2) percentage of time when windows were open during the sampling period (Baxter et al., 2007a).

2.6. Statistical analyses

Descriptive statistics, scatterplots, and histograms were used to characterize distributions of $PM_{2.5}$ and BC concentrations, outdoor concentration indicators, and indoor source terms (Table 1). Prior to model-building, to assess the relative contribution of indoor and outdoor

sources on indoor concentrations, we compared bivariate Pearson correlations between indoor $PM_{2.5}$ and BC concentrations with each source term, by season (Table 2).

2.7. Multivariate modeling

We built multivariate linear regression models, using a manual forward-stepwise procedure to determine outdoor infiltration and indoor sources of pollution. Outliers outside of the mean ± 3× standard deviations were removed prior to model building; one outlier home in the summer was removed where $PM_{2.5}$ was greater than 121 μg/m^3, where residents reported smoking. Covariates significant at $p <= 0.20$ in the bivariate analysis were considered candidate covariates, and individually incorporated into each model. Given our interest in assessing the impact of outdoor concentrations in industrial communities on indoor exposures, we first incorporated the location- and week-specific (LUR-based) outdoor concentration estimate into each model, and examined effect modification by the ventilation proxies (I/O sulfur ratio, and percent of time windows were open). We then tested each of the additional candidate outdoor source term, ordered by descending strength of the bivariate correlation, then tested significant source terms for effect modification by ventilation. Finally, we tested indoor source terms, ordered by descending strength of the bivariate correlation, and tested for effect modification by ventilation on each term (Baxter et al., 2007a).

Model fit was assessed at each stage, using the coefficient of determination (R^2) and root mean square error (RMSE). For a covariate to be retained at each stage, we required p-value < 0.10, an increase in R^2 of at least 0.01, a decrease in RMSE, and VIF < 2.0 for all model terms. At each stage, non-significant covariates were individually removed by descending p-value, and the model re-fit. In Tables 3 and 4, we report the pollutant concentration increases associated with an interquartile range (IQR) increase in each source indicator ($\beta \times$ IQR) from each multivariate model.

2.8. Sensitivity testing

Scatterplots were examined to assess fits between each significant predictor and pollutant concentrations, to ensure that covariate selection was not reliant on outliers. We rank ordered I/O sulfur ratios for the 21 homes as an attempt to reveal indoor sources of sulfur effects of air conditioning and the frequency of opening windows. We tested rank ordered I/O sulfur ratios instead of the outdoor LUR concentration estimates for all four multivariate models, but these did not improve model fit. Final model residuals were examined to ensure normality. Each model term was examined against the residual of the model without that term, to ensure that each term explained unique variance. Indoor concentration predictions were compared to observed concentrations using scatter plots. Finally, we tested the sensitivity of averaging outdoor pollutant concentrations within a 300 m buffer by comparing it to predicted concentrations at the point of the home location, finding correlations greater than 0.86. Statistical analyses were conducted using Proc Reg and Proc GLM in SAS version 9.3 (SAS Institute Inc., Cary, NC).

Table 1
Descriptive statistics for pollutant and meteorological data.

	Summer 2011	Winter 2012
Mean $PM_{2.5}$ (μg/m^3) (SD)	25.8 (±22.7)	18.9 (±13.2)
Mean BC (abs) (SD)	2.8 (±1.2)	2.3 (±1.4)
Temperature (°F) (mean, min–max)	77.4 (68–83)	68.7 (63–80)
Relative humidity (%) (mean, min–max)	55.6 (42–65)	33.1 (17–48)

3. Results

3.1. Indoor summer and winter $PM_{2.5}$ and BC concentrations

A higher mean indoor $PM_{2.5}$ concentration of 25.8 μg/m^3 (SD 22.7 μg/m^3) was detected during summer, vs. 18.9 μg/m^3 (SD 13.2 μg/m^3) during winter. For BC, a slightly higher mean of 2.8 absorbance units (SD 1.2 abs) was found during summer, vs. 2.3 abs (SD 1.4 abs) during winter.

3.2. Indoor/outdoor pollutant ratios

Pittsburgh-based outdoor $PM_{2.5}$ and BC LUR estimates were determined for each home. We found consistently higher concentrations indoors than outdoors for $PM_{2.5}$ and BC. In the summer, median I/O ratios were 1.30 for $PM_{2.5}$ and 1.47 for BC. In the winter, median I/O ratios were 1.21 for $PM_{2.5}$ and 1.38 for BC. Correlations between indoor and outdoor predicted concentrations were fairly low, as correlations ranged from 0.11 to 0.20 for $PM_{2.5}$ and 0.14–0.28 for BC across both seasons (Table 2).

Using EPA ambient data from Lawrenceville, PA, an urban Pittsburgh neighborhood, the median summer I/O ratio was 1.29 for $PM_{2.5}$. For the winter, the ratio was 1.08 for $PM_{2.5}$. Using ambient data from Liberty, PA, an industrial Pittsburgh neighborhood, the median $PM_{2.5}$ ratio was 1.26 for summer and 1.46 for winter.

3.3. Pollutant correlation to proximity to industry and outdoor LUR estimates

For summer, Pearson's correlation coefficients between both indoor $PM_{2.5}$ and BC concentrations with increasing distance from industry were −0.35 ($p = 0.13$) for BC and −0.30 ($p = 0.19$) for $PM_{2.5}$. For the winter, correlations between $PM_{2.5}$ and BC concentrations with increasing distance from industry were −0.31 ($p = 0.17$) for BC and −0.10 ($p = 0.66$) for $PM_{2.5}$. Outdoor estimates from LUR models were correlated most strongly with BC during winter ($r = 0.34$), suggesting industrial influence consistent with previous work (Table 2) (Fig. 2).

3.4. Indoor questionnaire covariates

During summer, $PM_{2.5}$ concentrations positively correlated with stovetop frying ($r = 0.47$) and cooking activity ($r = 0.41$). Higher concentrations were found in homes reporting more cigarettes smoking. BC correlated with time spent cooking ($r = 0.41$), and higher absorbance values were found in homes reporting more cigarettes smoking. Potential modifiers, such as frequency of open windows ($r = 0.30$) and the I/O sulfur ratio ($r = 0.56$) positively correlated with BC.

During winter indoor sampling periods, $PM_{2.5}$ concentrations were positively correlated with frequency of cleaning the kitchen ($r = 0.36$) and stovetop frying ($r = 0.32$). Higher concentrations were found in homes reporting more cigarettes smoking and more children in the home. Potential modifiers, such as open windows ($r = 0.37$) and the I/O sulfur ratio ($r = 0.58$) positively correlated with $PM_{2.5}$. BC correlated with stovetop frying ($r = 0.40$), as well as a potential modifier such as I/O sulfur ratio ($r = 0.53$) (Table 2).

3.5. Multivariate linear regression model building

Candidate covariates were selected from the correlations shown in Table 2 using the criteria of $p < 0.20$. All models contained a predicted outdoor concentration estimate multiplied by the I/O sulfur ratio, as an outdoor infiltration term. This term explained 10 to 42% of $PM_{2.5}$ and BC variability across models. For the summer, the final multivariate model for $PM_{2.5}$ ($R^2 = 0.52$) included number of cigarettes smoked in the home and stovetop frying. The final model for BC ($R^2 = 0.50$) included the number of cigarettes smoked (Table 3).

Table 2
Univariate analysis of outdoor concentration indicators and indoor source indicators correlated with pollutants. Correlations ≥ 0.30 are in bold ($p < 0.20$).

Covariate	Summer 2011*		Winter 2012	
	$PM_{2.5}$	BC	$PM_{2.5}$	BC
	r (p)	r (p)	r (p)	r (p)
Outdoor concentration indicators:				
LUR predicted $PM_{2.5}$ estimate	0.11 (0.65)	0.13 (0.58)	0.20 (0.38)	0.05 (0.83)
LUR predicted BC estimate	0.10 (0.68)	0.14 (0.54)	**0.34 (0.13)**	0.28 (0.21)
Reference site $PM_{2.5}$ concentration	0.07 (0.78)	0.17 (0.45)	0.05 (0.83)	0.18 (0.44)
Reference site BC absorbance	0.15 (0.53)	0.07 (0.78)	0.28 (0.22)	0.20 (0.38)
Distance to steel mill	**−0.30 (0.19)**	**−0.35 (0.13)**	−0.10 (0.66)	**−0.31 (0.17)**
$PM_{2.5}$ emissions IDW	0.10 (0.68)	0.02 (0.93)	0.27 (0.23)	0.16 (0.49)
$PM_{2.5}$ emissions/distance to steel mill	0.22 (0.35)	**0.35 (0.12)**	**0.47 (0.03)**	0.25 (0.27)
Indoor source indicators:				
Time spent cooking	**0.41 (0.07)**	**0.41 (0.07)**	−0.04 (0.85)	0.15 (0.51)
Frequency of stove frying	**0.47 (0.04)**	0.29 (0.20)	**0.32 (0.15)**	**0.40 (0.07)**
Frequency of cleaning the kitchen	0.27 (0.24)	0.08 (0.72)	**0.36 (0.10)**	0.29 (0.20)
Potential modifiers:				
Frequency of open windows	−0.19 (0.42)	**0.30 (0.20)**	**0.37 (0.10)**	0.19 (0.40)
Sulfur I/O ratio	0.08 (0.74)	**0.56 (0.01)**	**0.58 (0.01)**	**0.53 (0.01)**
Categorical indoor source indicators:				
Covariate	$PM_{2.5}$	BC	$PM_{2.5}$	BC
	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)
# Cigarettes smoked	–	–	–	–
No cigarettes	19.19 (4.55)	2.58 (0.99)	15.96 (8.48)	3.61 (1.69)
1–4 cigarettes per day	31.32 (4.32)	3.21 (1.67)	27.00 (−)	2.20 (−)
>4 cigarettes per day	–	4.99 (−)	36.75 (3.45)	3.32 (1.28)
# Children in home	–	–	–	–
0–1	17.41 (5.41)	2.48 (1.07)	12.21 (4.77)	1.54 (1.00)
2–3	22.33 (7.60)	2.94 (1.10)	20.70 (11.17)	2.74 (1.35)
4–5	31.28 (13.37)	3.44 (2.31)	33.83 (14.31)	3.16 (1.56)

* An outlier ($PM_{2.5} = 121\ \mu g/m^3$) was removed for summer 2011 $PM_{2.5}$ analyses.

For the winter, the final multivariate model for $PM_{2.5}$ ($R^2 = 0.61$) included number of cigarettes smoked. The final model for BC ($R^2 = 0.70$) was driven by outdoor infiltration and included the predicted outdoor estimates modified by the frequency of open windows (Table 4).

4. Discussion

Our results highlight the importance of both infiltration of outdoor pollutant concentrations and indoor sources, especially smoking, on indoor exposures in industrial communities. We hypothesized that outdoor concentrations would be strong predictors for our indoor models. For both the summer and winter indoor BC models and the winter $PM_{2.5}$ model, outdoor estimates modified by the I/O sulfur ratios explained at least 33% of the variability found in indoor concentrations. We hypothesized smoking to be the most predictive indoor pollution source, which was corroborated in both summer $PM_{2.5}$ and BC models, as well as the winter $PM_{2.5}$ model. Outdoor infiltration explained moderate variability in indoor $PM_{2.5}$ and BC concentrations, yet smoking contributed to higher concentration increases. In both seasons, indoor concentrations for both $PM_{2.5}$ and BC were consistently higher than residence-specific outdoor concentration estimates.

For seasonal differences in $PM_{2.5}$, other studies found lower PM concentrations during summer months compared to winter (Leaderer et al., 1999; Hazenkamp-Von Arx et al., 2004). We found slightly higher indoor $PM_{2.5}$ concentrations during the summer, though the difference was not statistically significant. BC absorbance levels did not differ by season.

Smoking is an important source of fine and coarse particulate matter indoors (Wallace, 1996), and was the strongest predictor for both summer and winter $PM_{2.5}$ models. Wallace (1996) had found estimated

Table 3
Summer 2011 multivariate model covariates and model fits for pollutants.

		Multivariate model						
	Covariates	β	p-Value	RMSE	VIF	IQR of source indicator	Conc. increase per source indicator	Seq R^{2a}
Summer $PM_{2.5}$ ($\mu g/m^3$)[b]	Intercept	−11.96	0.67	–	–	–	–	–
	Outdoor $PM_{2.5}$ estimate * I/O sulfur ratio	2.29	0.18	7.93	0.61	2.03	4.65	0.10
	Cigarettes smoked	–	–	–	–	1	–	–
	1–4 cigarettes	8.40	0.05	–	–	–	8.40	–
	No cigarettes	0	–	6.73	0.82	–	–	0.33
	Frequency of stove frying	0.99	0.02	5.86	0.62	4.5	4.46	**0.52**
Summer BC (abs)	Intercept	3.97	0.001	–	–	–	–	–
	Outdoor BC estimate * I/O sulfur ratio	0.22	0.01	0.99	0.99	2.51	0.55	0.33
	Cigarettes smoked	–	–	–	–	1	–	–
	>4 cigarettes	2.11	0.04	–	–	–	2.11	–
	1–4 cigarettes	1.51	0.16	–	–	–	1.51	–
	No cigarettes	0	–	0.90	0.28	–	–	**0.50**

[a] Seq R^2 is the sequential model fit for each additional term incorporated into model.
[b] One outlier was removed for PM modeling.

Table 4
Winter 2012 multivariate model covariates and model fits for pollutants.

		Multivariate model						
	Covariates	β	p-value	RMSE	VIF	IQR of source indicator	Conc. increase per source indicator	Seq R^2
Winter $PM_{2.5}$ (μg/m^3)	Intercept	24.21	0.004	–	–	–	–	–
	Outdoor $PM_{2.5}$ estimate * I/O sulfur	0.76	0.002	10.51	0.97	10.33	7.85	0.39
	Cigarettes smoked	–	–	–	–	1	–	–
	>4 cigarettes	17.12	0.02	–	–	–	17.12	–
	1–4 cigarettes	1.00	0.93	–	–	–	1.00	–
	No cigarettes	0	–	8.87	0.68	–	–	**0.61**
Winter BC (abs)	Intercept	1.89	0.02	–	–	–	–	–
	Outdoor BC estimate * I/O sulfur	0.34	0.24	1.06	0.38	1.34	1.11	0.42
	Windows open	–	–	–	–	1	–	–
	1–4 h	1.77	0.05	–	–	–	1.77	–
	<1 h	0	–	1.04	0.22	–	–	0.46
	Outdoor BC estimate * windows open	–	–	–	–	1	–	–
	1–4 h	1.50	0.002		–	–	1.50	–
	<1 h	0	–	0.81	0.32	–	–	**0.70**

increases in $PM_{2.5}$ in homes with smokers ranging from 25 to 45 μg/m^3 (Wallace, 1996). Most residents who smoked reported 1–4 cigarettes per day in the home, which conferred significant increases in indoor $PM_{2.5}$ concentrations. For summer $PM_{2.5}$, stovetop frying was also an important contributor to indoor pollution. We identified little outdoor contributions in the summer and moderate contributions in the winter,

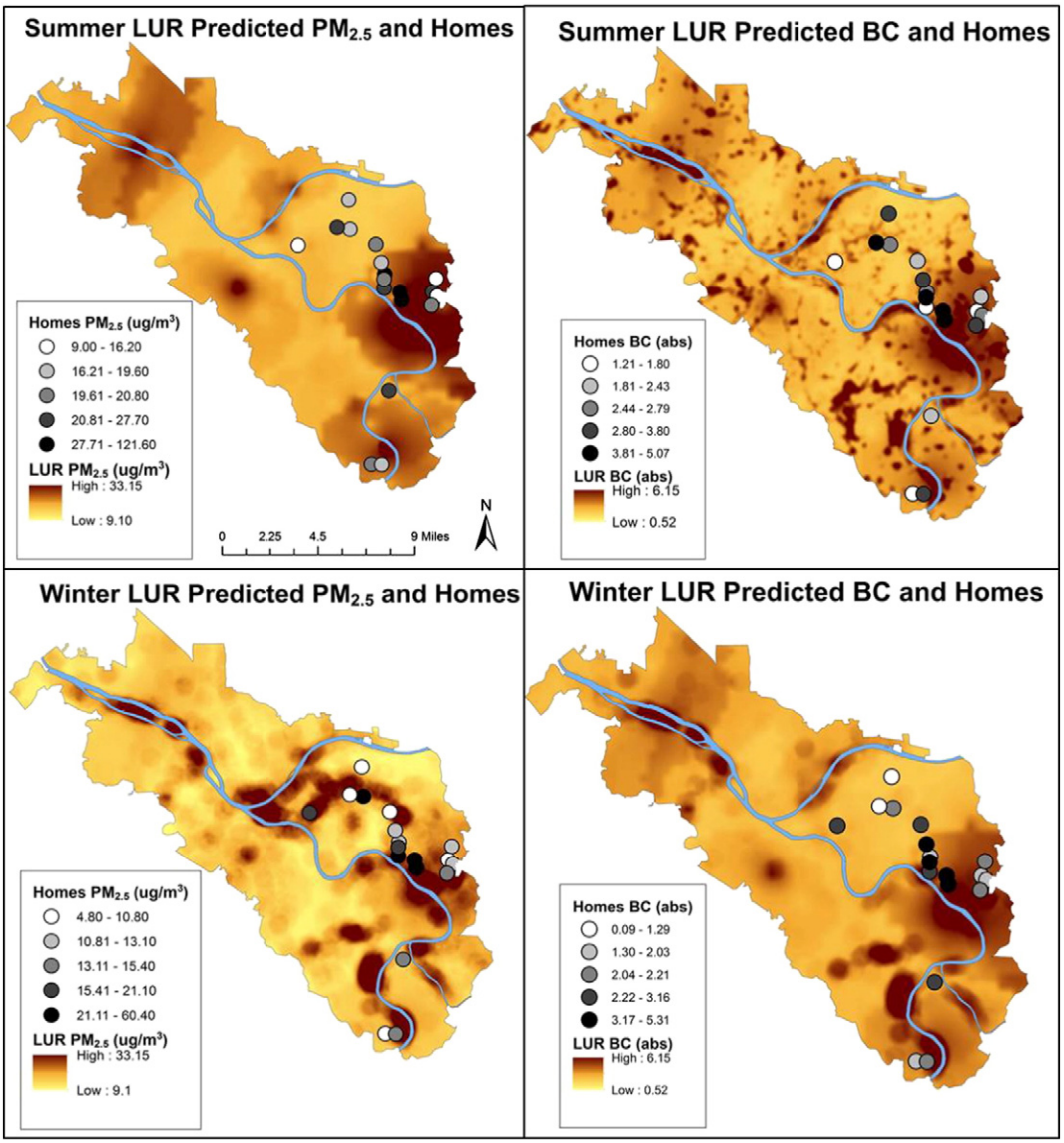

Fig. 2. Summer and winter $PM_{2.5}$ and BC indoor concentrations overlaid with LUR predicted estimates.

as our outdoor estimates modified by I/O sulfur ratios explained 10 to 39% of variability in indoor concentrations.

For both seasons in BC models, we were able to identify an outdoor contribution to indoor pollution based on outdoor BC estimates modified by I/O sulfur concentrations. Smoking was a significant predictor for summer BC variability, but not winter. For the summer, variability in BC was explained by outdoor BC and sulfur I/O ratios, as well as number of cigarettes smoked. Smoking and frying food both emit particulates, including black carbon, however, cooking predictors were not significant in either BC seasonal model (Wallace, 1996). A study in Boston found that indoor BC was associated with an outdoor measure of local traffic (Baxter et al., 2007a); we may be finding a similar association between indoor BC and outdoor measures based on industrial activity. Reinforcing the significance of outdoor BC and sulfur ratios as a strong predictor for BC, indoor BC was higher when windows were opened for greater than 12 h per day (mean BC = 3.15 (SD = 0.76 abs)), compared to windows open for less than 1 h per day (mean BC = 2.24 (SD = 1.22 abs)). In the final summer model, our outdoor predicted estimates and sulfur ratios were stronger predictors compared to open windows. The importance of outdoor infiltration in industrial communities was highlighted during winter BC. For the winter, variability in BC was solely explained by outdoor estimates and modifiers, including I/O ratios and frequency of open windows.

Using residence-specific indicators and outdoor concentrations at EPA sites, median pollutant I/O ratios were greater than 1.0, indicating higher concentrations inside the home than outside. Many studies have had contrasting results, finding low or high correlations between indoor and outdoor pollutant concentrations when sampling inside and directly outside of homes (Adgate et al., 2002; Ramachandran et al., 2000; ALA, 2013). Levy et al. (2002) and Morawska et al. (2001) identified moderate to high (I/O) pollutant ratios (Morawska et al., 2001; Levy et al., 2002), whereas using residence-specific outdoor concentration estimates, our correlations between indoor and outdoor pollutant concentrations were low ($r < 0.28$), resulting in similar findings reported in Riverside, California (Ozkaynak et al., 1996).

This study builds upon other literature (Lioy, 2010), showing that daily activity patterns, such as smoking or opening windows, are important to adequately characterize indoor residential exposures. A strength of this indoor air sampling study is that it allowed us to determine indoor weekly pollutant concentrations across homes near active industrial sites using a simple air sampling device; little maintenance of the units was required (the PEM only needed to be changed to avoid filter overload). One-hundred percent retention of sampling homes was achieved, so direct seasonal comparisons could be made. The questionnaire allowed for assessment of indoor pollutant concentrations across a multitude of covariates that could be used in multivariate linear regression modeling.

One limitation was the sample size of only 21 homes; 56% of participants in the small asthmatic cohort from the industrial communities of Braddock and Clairton elected to take part in the indoor air sampling. Another limitation was the inability to directly collect outdoor concentrations at the homes, though we believe our seasonally-specific LUR estimates are reasonably sufficient, as LUR models predicted 64 to 86% of outdoor variability of pollutant concentrations across Pittsburgh (Tunno et al., 2015). Outdoor measurements were taken using portable air samples across randomly distributed locations during both the summer and winter months while indoor sampling was occurring, so there isn't a difference in time between our indoor measurements and outdoor-derived estimates. Unfortunately, we were unable to collect information on the house, such as the year built. Data primarily allowed us to gain an understanding of the complexity of indoor pollution exposures, and to estimate the outdoor contribution to indoor pollution through using outdoor concentration estimates from our published models (Tunno et al., 2015).

Better understanding exposures is an important public health need, as lower income communities are often clustered near industrial sites, potentially resulting in highly elevated residential exposures. Contrary to the initial hypothesis, outdoor concentrations accounted for low to moderate variability in indoor concentrations; a much greater proportion was explained by cigarette smoking in three of the four models (Tables 3 and 4). Our results showed that, even in industrial communities of high outdoor pollution concentrations, indoor activities – particularly cigarette smoking – may play a larger role in shaping indoor $PM_{2.5}$ exposures. Outdoor infiltration from nearby industrial facilities, combined with indoor smoking, is an important public health issue. The results of this study were reported to residents with the aim of reducing daily activities that lead to increased indoor exposures.

5. Conclusions

Smoking was the most significant identified contributor to indoor air pollution, and outdoor-derived estimates and sulfur I/O ratios explained a portion of the outdoor contribution to indoor pollution. Median I/O ratios indicated higher $PM_{2.5}$ and BC concentrations indoors than outdoors, even in these homes located near large active industrial sources. Slightly higher indoor $PM_{2.5}$ concentrations were found during the summer compared to the winter. This study was informative to parents, indicating that daily activities in the home, such as smoking, can result in higher concentrations of $PM_{2.5}$. Though outdoor pollution cannot be ignored and certainly contributes to indoor air pollution levels, indoor activities like smoking appear to be stronger sources of $PM_{2.5}$ concentrations compared to outdoor concentrations, even in these industrial communities. The combination of outdoor infiltration from nearby industry and indoor smoking is important to public health.

Abbreviations

$PM_{2.5}$	particulate matter with aerodynamic diameter less than 2.5 μm
BC	black carbon
I/O	indoor/outdoor
NAAQS	National Ambient Air Quality
PEMC	Pediatric Environmental Medicine Center
PEM	Personal Exposure Monitor
RH	relative humidity
ICP-MS	inductively-coupled plasma mass spectrometry
EPA	Environmental Protection Agency
AQS	Air Quality System
LUR	land use regression
IDW	inverse-distance weighted
NEI	National Emissions Inventory
AER	air exchange rate
r	correlation coefficient
R^2	coefficient of determination
β	parameter estimate
RMSE	root mean square error
VIF	variance inflation factor

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

BT and JEC were primarily responsible for the manuscript design, drafting, and development. KNS, LC, and ST contributed to acquisition of data. KNS, LC, ST, FH, and PL contributed to critical revision of this manuscript, as well as important intellectual content included in this review. All authors read and approved the final manuscript.

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