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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