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Indoor source apportionment in urban communities near industrial sites

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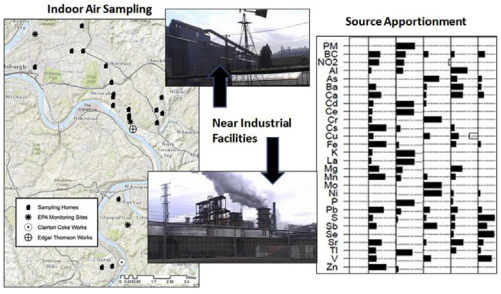
HIGHLIGHTS

- Indoor elemental constituents were assessed near industrial sources.
- Indoor sources (smoking) explained greater variability than outdoor sources.
- Outdoor infiltration was indicated by coal and motor vehicle markers.
- Source-specific indoor constituents should be further examined for health.

GRAPHICAL ABSTRACT



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ABSTRACT

Because fine particulate matter ($PM_{2.5}$) differs in chemical composition, source apportionment is frequently used for identification of relative contributions of multiple sources to outdoor concentrations. Indoor air pollution and source apportionment is often overlooked, though people in northern climates may spend up to 90% of their time inside. We selected 21 homes for a 1-week indoor sampling session during summer (July to September 2011), repeated in winter (January to March 2012). Elemental analysis was performed using inductively-coupled plasma mass spectrometry (ICP-MS), and factor analysis was used to determine constituent grouping. Multivariate modeling was run on factor scores to corroborate interpretations of source factors based on a literature review. For each season, a 5-factor solution

Abbreviations: $PM_{2.5}$, particulate matter with aerodynamic diameter less than 2.5 μm; BC, black carbon; PEM, Personal Exposure Monitor; NO_2, nitrogen dioxide; ICP-MS, inductively-coupled plasma mass spectrometry; Cd, cadmium; K, potassium; Ca, calcium; Fe, iron; Al, aluminum; Cu, copper; Sb, antimony; As, arsenic; Ba, barium; Ce, cerium; Cr, chromium; Cs, cesium; Mo, molybdenum; Sr, strontium; Tl, thallium; PMF, positive matrix factorization; GIS, geographic information system; AER, air exchange rate; S, sulfur; Zn, zinc; Se, selenium; Ni, nickel; V, vanadium; Mn, manganese; Pb, lead; La, lanthanum; P, phosphorus; r, correlation coefficient; R^2, coefficient of determination.

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explained 86–88% of variability in constituent concentrations. Indoor sources (i.e. cooking, smoking) explained greater variability than did outdoor sources in these industrial communities. A smoking factor was identified in each season, predicted by number of cigarettes smoked. Cooking factors were also identified in each season, explained by frequency of stove cooking and stovetop frying. Significant contributions from outdoor sources including coal and motor vehicles were also identified. Higher coal and secondary-related elemental concentrations were detected during summer than winter. Our findings suggest that source contributions to indoor concentrations can be identified and should be examined in relation to health effects.

1. Introduction

A substantial literature documents elemental composition of ambient fine particulate matter ($PM_{2.5}$), both for source identification and epidemiology (Stanek et al., 2011; Thurston et al., 2011). Fewer studies, however, have examined composition and source apportionment of indoor residential $PM_{2.5}$, although individuals in northern climates spend a majority of their time indoors (Koutrakis et al., 1992). In addition, indoor concentrations may be both higher and more complex in composition than are ambient concentrations, because indoor concentrations are influenced by indoor and outdoor sources, both modified by ventilation (EPA; Morawska et al., 2001; Adgate et al., 2002; Clougherty et al., 2011; Wallace, 1996a). As a result, there is a need for stronger methods to disentangle source contributions in the indoor environment – particularly in urban communities with complex source mixtures – to identify modifiable indoor sources, and to more clearly elucidate potential health effects (Stanek et al., 2011; Larson et al., 2004).

Studies of urban ambient $PM_{2.5}$ composition have identified elemental tracers, and characteristic source signatures, associated with key urban sources (e.g., vehicular emissions, brake/tire wear, diesel, industry, etc.) (Gunawardana et al., 2012; Schauer, 2003; Sternbeck et al., 2002; Lall and Thurston, 2006). In addition, source characterizations and studies focused in industrial communities have validated tracers for industrial sources including coal burning and steel production (Thurston et al., 2011; Pekney et al., 2006; Hammond et al., 2008; Salvador et al., 2008; Rizzo and Scheff, 2007; Irvine et al., 2009). Studies of indoor residential environments have examined additional indicators specific to indoor sources, such as smoking and cooking (Rizzo and Scheff, 2007; Zhao et al., 2006; Semple et al., 2012; Ozkaynak et al., 1996a).

The communities of Braddock and Clairton, Pennsylvania, located east of Pittsburgh along the Monongahela River, are home to an active steel mill and coke works, respectively. Among the largest sources of their types in the county, the Edgar Thomson Steel Works and Clairton Coke Works produce approximately 725 and 1049 tons of primary $PM_{2.5}$ per year (USS; EPA, 2008). In multivariate models for $PM_{2.5}$ and black carbon (BC) concentrations in these homes, we identified an important impact of indoor smoking, and a significant contribution from nearby industry (Tunno et al., 2015a).

In this study, we examined elemental constituents in indoor $PM_{2.5}$ sampled from homes in varying proximities to these industrial sources, to assess the relative contributions of indoor and outdoor sources to indoor concentrations. We hypothesized that these industrial sources would contribute to indoor concentrations – potentially with varying influence by season and ventilation – and that indoor sources such as smoking and cooking would contribute to indoor concentration year-round.

2. Methods

2.1. Study design

Families with at least one child participating in an asthmatic cohort at the Pediatric Environmental Medicine Center at Children's Hospital, Pittsburgh, PA, were invited to participate. A total of 21 homes from the communities of Braddock and Clairton 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 comparability, to capture concentrations in homes further from these industrial sources, convenience samples of six additional homes were included in the dataset. Samplers were placed in the main activity room, away from windows and combustion or heat sources. Study design, sample data collection, and monitoring instrumentation are detailed elsewhere (Tunno et al., 2015a).

2.2. Monitoring instrumentation and constituent measurements

A total of 42 week-long measurements were collected using a Harvard Personal Exposure Monitor (PEM) on a MEDO linear-piston vacuum pump. $PM_{2.5}$ was measured using 37 mm Teflon™ filters (Pall Life Sciences) that were pre- and post-weighed in a temperature and relative humidity controlled glove box (PlasLabs Model 890 THC, Lansing, MI), using an ultramicrobalance (Mettler Toledo Model XP2U, Columbus, OH). The PEM was switched mid-week in each home due to concern of filter overloading. $PM_{2.5}$ 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. Black carbon was measured (in absorbance units) from each filter using an EEL43M Smokestain Reflectometer (Diffusion Systems Limited, London, England). Passive Ogawa badges were used to measure nitrogen dioxide (NO_2) (Ogawa USA, Pompano Beach, FL) for a continuous 24-h, 7-day sample. Ogawa badges were stored in the refrigerator at 4 °C, then analyzed using Ultraviolet–Visible (UV–VIS) spectrophotometry to determine NO_2 concentrations (Thermo Scientific Evolution 60S UV–Visible Spectrophotometer). All measures were blank-corrected using full method blanks. For determination of elemental concentrations, inductively-coupled plasma mass spectrometry (ICP-MS) analyses were conducted by the Wisconsin State Laboratory of Hygiene following documented protocols (ESS INO Method 400.4; EPA Method 1638) (Sutton and Caruso, 1999).

2.3. Indoor questionnaire

An adult over 18 years of age in each home completed an indoor sources questionnaire adapted from prior studies (Baxter et al., 2007; Dutta et al., 2007). Questions included items on smoking, cooking, stove frying, ventilation, heating system, air conditioning,