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Predictors of concentrations of nitrogen dioxide, fine particulate matter, and particle constituents inside of lower socioeconomic status urban homes

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Air pollution exposure patterns may contribute to known spatial patterning of asthma morbidity within urban areas. While studies have evaluated the relationship between traffic and outdoor concentrations, few have considered indoor exposure patterns within low socioeconomic status (SES) urban communities. In this study, part of a prospective birth cohort study assessing asthma etiology in urban Boston, we collected indoor and outdoor 3–4 day samples of nitrogen dioxide (NO_2) and fine particulate matter ($PM_{2.5}$) in 43 residences across multiple seasons from 2003 to 2005. Homes were chosen to represent low SES households, including both cohort and non-cohort residences in similar neighborhoods, and consisted almost entirely of multiunit residences. Reflectance analysis and X-ray fluorescence spectroscopy were performed on the particle filters to determine elemental carbon (EC) and trace element concentrations, respectively. Additionally, information on home characteristics (e.g. type, age, stove fuel) and occupant behaviors (e.g. smoking, cooking, cleaning) were collected via a standardized questionnaire. The contributions of outdoor and indoor sources to indoor concentrations were quantified with regression analyses using mass balance principles. For NO_2 and most particle constituents (except outdoor-dominated constituents like sulfur and vanadium), the addition of selected indoor source terms improved the model's predictive power. Cooking time, gas stove usage, occupant density, and humidifiers were identified as important contributors to indoor levels of various pollutants. A comparison between cohort and non-cohort participants provided another means to determine the influence of occupant activity patterns on indoor–outdoor ratios. Although the groups had similar housing characteristics and were located in similar neighborhoods, cohort members had significantly higher indoor concentrations of $PM_{2.5}$ and NO_2, associated with indoor activities. We conclude that the effect of indoor sources may be more pronounced in high-density multiunit dwellings, and that future epidemiological studies in these populations should explicitly consider these sources in assigning exposures.
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Introduction

Given that people spend the majority of their time indoors, residential indoor air pollution levels may serve as a better, albeit still imperfect, surrogate for personal exposures in epidemiological studies than outdoor concentrations. Studies have consistently demonstrated that residential indoor concentrations are more strongly correlated with personal exposures than outdoor concentrations (Levy et al., 1998; Koistinen et al., 2001; Kousa et al., 2001). While numerous studies have identified important sources affecting longer-term indoor exposures (e.g. smoking and cooking) (Koutrakis and Briggs, 1992; Schwab et al., 1994; Ozkaynak et al., 1996; Lee et al., 2000), few have considered indoor exposure patterns among lower socioeconomic status (SES) populations.

For health outcomes such as asthma, which demonstrate significant socioeconomic gradients (The American Lung Association, 2001), accounting for indoor–outdoor (I/O) concentration patterns in low SES populations may take on an added importance. Low SES urban residents often live in smaller apartments, possibly resulting in greater contributions from indoor sources (given smaller volumes) and in different ventilation patterns (given adjoining units and a lack of central air conditioning). There may also be different activity patterns for this population such as air conditioning prevalence and use, which will influence the opening of windows and exposures to outdoor-generated pollutants; or increased prevalence of indoor sources such as cigarette smoking (Centers for Disease Control and Prevention, 2004) or gas stoves (which may be used for supplemental heating in the winter) (Centers for Disease Control and Prevention, 1997). In spite of these unique characteristics, and their disproportionate burden of air pollutant exposures (American Lung Association, 2001), few studies have examined I/O pollutant relationships in this population (O'Neill et al., 2003), and have focused only on asthmatic children (Wallace et al., 2003; Breysse et al., 2005), which may not inform understanding about asthma development, as relationships

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may differ for very young children. These local-scale intraurban indoor exposure patterns may contribute to differential health outcomes between and within urban neighborhoods, resulting in inequality of risk within a metropolitan area.

The current paper seeks to develop predictive models of residential indoor air pollutant concentrations for lower SES households in an urban area, which will ultimately inform exposure assessment for a birth cohort study. In a large cohort study, it is not feasible to monitor the entire study population, so models must be developed to assign exposures from questionnaires and other available data. Our objective is to determine the connection between these data and actual exposure measures, determining what information is needed to reduce exposure measurement error. We also aim to understand the relative importance of outdoor concentrations, building characteristics, and occupant activities in a low SES setting for a number of pollutants. Since it is unclear which air pollutant may be the causative agent for health outcomes, it is important to understand the exposure patterns for multiple pollutants. We hypothesize that the predictive power of outdoor concentrations, indoor source terms, and ventilation-related terms will differ significantly among pollutants, indicating that long-term personal exposure patterns would vary differentially from outdoor concentration patterns across pollutants.

Methods

Study Design

The air pollution measurements and home characteristics/occupant behavior data analyzed were collected as part of the Asthma Coalition for Community, Environment, and Social Stress (ACCESS), a prospective birth cohort study recruiting pregnant women throughout Boston with an overarching goal to assess asthma etiology related to social (maternal stress, exposure to violence), genetic, and environmental factors (traffic-related pollution and indoor allergens). For traffic-related pollution, sampling was conducted at a representative subset of homes allowing for model development and ultimate extrapolation to the remainder of the cohort. The target sample size was 40 sites based on both logistical considerations and the size of previous studies that demonstrated robust regression models linking outdoor concentrations of the pollutants of interest with traffic characteristics (Briggs et al., 2000; Brauer et al., 2003).

Measurements were taken in a variety of traffic settings by selecting sampling locations to capture the range of traffic densities present in urban neighborhoods. Geographic Information System (GIS)-based estimates of traffic density for all available participants were calculated, with the goal of capturing heterogeneity in traffic exposures both across all participants and within neighborhoods of interest. Using the Spatial Analyst Extension of ArcGIS 9.1, traffic scores were calculated for 50-m raster cells, using traffic volume data (provided by the Massachusetts Highway Department) for all road segments within 100 m of the home, and a kernel technique (quadratic inverse-distance weighing function) to more heavily weight segments near the cell's center. Homes were divided into tertiles by traffic density scores and selected to represent a range of traffic densities and neighborhoods, with an aim to oversample in neighborhoods where recruitment was occurring, but with representative sites across Boston to ensure generalizability. Participants were drawn in part from the ACCESS cohort, with additional non-cohort participants (42% of total participants) enrolled for geographic representativeness (i.e., to reflect neighborhoods not yet represented in the cohort but where future recruitment was planned).

Sampling was conducted in two seasons, the non-heating (May–October) and heating (December–March), to capture potential seasonal effects on concentrations. A standardized questionnaire was administered at the end of each sampling period to gather housing characteristics/occupant behavior data. Our questionnaire was derived from a questionnaire used in the Inner-City Air Pollution sub-study within the Inner-City Asthma Study (Wallace et al., 2003), and the US Environmental Protection Agency's Residence Survey and Daily Follow-up Questionnaire (Williams et al., 2003). In addition to home location, other home information included home type, built year of house and type of heating/cooking fuel. Some occupant activities considered were environmental tobacco smoke (ETS) exposure, opening windows, time spent cooking, use of candles or air fresheners, cleaning activities, and use of air conditioning. These factors could affect air exchange rates (AER), penetration efficiencies, and resuspension, as well as identify possible indoor sources.

Field Monitoring

Indoor and outdoor 3- to 4-day samples of nitrogen dioxide (NO_2) and particulate matter ($PM_{2.5}$) were collected simultaneously at each home in both seasons. This sampling period was selected empirically to avoid limit of detection issues and sampler overload, and to represent longer-term rather than acute exposures. When possible, two consecutive measurements were collected at each home, providing 1-week average concentrations and minimizing weekday/weekend effects. Indoor samples were taken in the main living space of the home, away from windows, stoves, and heat sources. Outdoor samples were taken on a free-standing tripod whenever possible; else samplers were extended from a window on a three-foot sampling arm made of polyvinyl chloride (PVC) piping to avoid the building envelope. A four-inch deep stainless-steel rain dish was placed over both indoor and outdoor samplers to reduce interference by rain, snow, wind, and curious children living in the homes. Additional data not reported here include: indoor/outdoor

temperature and humidity monitored with a HOBO device (Onset Computer Corporation, Pocasset, MA, USA), and continuous traffic counts recorded directly on the largest road within 100 m of the home with a Trax I Plus traffic counter (JAMAR Technologies, Horsham, PA, USA).

Analytical Methods

$PM_{2.5}$ samples were collected using a Harvard Personal Environmental Monitor (PEM), a size-selective inertial impactor attached to a Medo linear-piston vacuum pump running at 4 l/min. Particles were captured on 37-mm Teflon filters and an elutriator (10 cm long, 5 cm in diameter) was attached to each PEM. Sampling, preparation, and analysis procedures have been described previously (Marple et al., 1987). In addition, given our interest in addressing traffic-related air pollution, elemental carbon (EC) concentrations were estimated using reflectance analysis on the particle filters, a non-destructive process that provides measurements highly correlated with concentrations measured using thermal-optical methods (Kinney et al., 2000). To allow for more detailed source apportionment and better capture indoor source factors, elemental analysis was conducted by X-ray fluorescence spectroscopy (XRF) on the particle filters. The analyses were conducted according to standard operating procedures at the Desert Research Institute laboratories (DRI; Reno, NV, USA) including their quality control and assurance (Chow and Watson, 1998; Watson et al., 1999). Finally, NO_2 samples were collected using Yanagisawa passive filter badges, which absorb NO_2 on a triethanolamine solution on a cellulose fiber filter (Yanagisawa and Nishimura, 1982).

Quality Control and Quality Assurance

Field blanks were collected totaling approximately 10% of the number of samples. Field blanks were transported and handled like regular samples, but the filters were not attached to pumps. These samples were used to determine background contamination and for the calculation of method limits of detection (LODs). Concentrations were blank corrected using the mean blank value from the field blanks, when this value was significantly different from zero at the 95% level. The LODs were calculated as the standard deviation of the field blank concentrations multiplied by three. The exception to this was for the elemental concentrations determined by XRF. For each element and sample, concentration uncertainties were given that equaled one standard deviation of error estimates based on analytical precision. As in previous studies reporting aerosol elemental data, three times the uncertainty was considered to be the LOD for each element and sample (Long and Sarnat, 2004). Random number generation was used to assign values to those samples below the LOD, constrained to values between 0 and the LOD and assuming a uniform distribution. Precision of the method was determined by duplicate samples (10% of number of samples) and was equal to the mean relative difference (RD), the average of the absolute difference of a pair of duplicates divided by the mean of the pair. Finally, responses to the questionnaire were evaluated for completeness and, when applicable, validated with data available through the City of Boston, Brookline, Cambridge, and Somerville property tax records.

Data Analysis

In this analysis, we utilize the principles of the mass-balance model to determine the relationship between indoor and outdoor concentrations, indoor sources, and ventilation. However, adequate data for some parameters are lacking; in particular, air exchange rates (AER) measurements were not possible in this study, so we relied on other methods to capture the effect of ventilation, as described below.

A sequential model-building approach was taken. First, with information gathered from the questionnaire, potential indoor sources and source-related activities were identified. The list of potential covariates was narrowed down by only considering those factors significant at the 80% level for several pollutants in univariate regression analyses. From these, only variables with a logical causal connection to indoor concentrations of the investigated pollutants were considered, including those related to the generation and/or resuspension of the considered pollutants. For example, gas stoves have been shown to be a source of NO_2 (Lee et al., 1998), with the time spent cooking per day as an effect modifier. Crustal elements (i.e., Ca, Fe, and K) and elements associated with sea salt (i.e., Na and Cl) tend to be associated with resuspension activities such as cleaning and foot traffic (Wallace, 1996), so we created an occupant density variable equal to the number of occupants divided by the number of rooms. Humidifier use has been shown to lead to increasing levels of $PM_{2.5,}$ especially those elements (Ca, Cl, K, Si, and S) characteristic of the "tap water fingerprint" (Highsmith et al., 1992). Cooking has been associated with $PM_{2.5}$ as well as numerous elements (Ozkaynak et al., 1996). Candle use is a potential source of EC.

Home-specific outdoor concentrations and exposure-related activities that occurred during the sampling period were regressed against the individual indoor concentrations, forcing outdoor concentrations into the model and using the predetermined indoor source terms. This initial model can be expressed as:

$$C_{\mathrm{in}_{ij}} = \beta_{oj} + \beta_{1j} * C_{\mathrm{out}_{ij}} + \beta_{xj} * Q_{ij} \qquad (1)$$

where $C_{\mathrm{in}_{ij}}$ (ppb, μg or ng/m^3, or m$^{-1}\times10^{-5}$) is the indoor concentration of pollutant j for participant i, $C_{\mathrm{out}_{ij}}$ (ppb, μg or ng/m^3, or m$^{-1}\times10^{-5}$) is the outdoor concentration, and Q_{ij} is a vector of the various indoor source terms.

This model has the benefit of being simple and easy to interpret; however, it does not account for variations in homes in AER, penetration efficiency, removal rate, and

other factors. These effects are theoretically described under steady-state conditions with a single compartment mass balance model seen in Equation (2):

$$C_{\mathrm{in}_{ij}} = \frac{P_j a_i}{a_i + k_j} C_{\mathrm{out}_{ij}} + \frac{Q_{ij}/V_i}{a_i + k_j} = F_{\mathrm{INF}_{ij}} C_{\mathrm{out}_{ij}} + \frac{Q_{ij}/V_i}{a_i + k_j} \quad (2)$$

where P_j is the penetration efficiency (dimensionless), a_i is the AER (h^{-1}), k_j is the decay rate (h^{-1}), Q_{ij} is the indoor source strength (ppb/h, μg or ng/h or $m^{-1} \times 10^{-5}$/h), and V_i is the house volume (m^3). The quantity $P_j a_i/(a_i + k_j)$ is also called the infiltration factor ($F_{\mathrm{INF}_{ij}}$) and represents the fraction of $C_{\mathrm{out}_{ij}}$ that penetrates indoors in each home. Therefore $F_{\mathrm{INF}_{ij}} C_{\mathrm{out}_{ij}}$ describes the ambient contribution to the indoor concentration, and $Q_{ij}/V_i(a_i + k_j)$ represents the contribution of indoor sources to the indoor concentration.

In this study, no direct measurements of AERs were taken so a reasonable proxy must be used. As shown in Equation (3), sulfur I/O ratios represent the infiltration factor if there are no indoor sources of sulfur as hypothesized (Sarnat et al., 2002).

$$C_{\mathrm{in}_{iS}} = F_{\mathrm{INF}_{iS}} C_{\mathrm{out}_{iS}} + \frac{Q_{iS}/V_i}{a_i + k_S}, \quad Q_{iS} = 0 \Rightarrow \frac{F_{\mathrm{INF}_{iS}} C_{\mathrm{in}_{iS}}}{C_{\mathrm{out}_{iS}}} \quad (3)$$

where $F_{\mathrm{INF}_{iS}}$, $C_{\mathrm{in}_{iS}}$, $C_{\mathrm{out}_{iS}}$ are the individual infiltration factor, indoor, and outdoor concentrations of sulfur, respectively. The assumption of no indoor sources of sulfur was tested empirically by regressing the indoor sulfur concentrations against outdoor sulfur concentrations and examining if the intercept was significantly different from zero or if any hypothetical sources of sulfur significantly predicted indoor concentrations.

F_{INF} is directly associated with AER, especially if P and k are relatively less variable across homes, so dichotomizing on high/low infiltration factor will be equivalent to dichotomizing on AER. This has been corroborated in previous studies, which observed a strong relationship between F_{INF} and AER in Boston area homes (Long and Sarnat, 2004). We chose to dichotomize instead of using the actual values because of instability of the estimated AER at higher values, uncertainty about P and k (both mean values and the degree to which they vary by home), and because the effective penetration efficiency varies across particle constituents. Backwards elimination is performed in the models developed for Equation (1) with only terms significant at $P < 0.2$ remaining in the final model. The $F_{\mathrm{INF}iS}$ variable can be incorporated into these pollutant specific models as an interaction term as seen in Equation (4):

$$C_{\mathrm{in}_{ij}} = \beta_{oj} + \beta_{1j} * C_{\mathrm{out}ij} * AERDummy_i + \beta_{xj} * Q_{ij} * AERDummy_i \quad (4)$$

where $AERDummy_i$ is the dichotomized variable from the $F_{\mathrm{INF}iS}$s. This allows us to determine if this proxy variable contributed more information as to the influence of outdoor concentrations and the identified indoor sources from Equation (1), although due to statistical power considerations, we present models only incorporating the $AERDummy_i$ term for outdoor concentrations. All regression analyses were done using SAS version 8.

Results

Participant Characteristics

A total of 66 sampling sessions were conducted, with 23 homes monitored in both seasons, 15 in the non-heating season only, and five in the heating season only. The 43 sites were distributed among 39 households with four of the participants moving and allowing us to sample in their new home. The locations of all of the sampling sites are shown in Figure 1 and are distributed throughout the metropolitan Boston area. Of the 43 sites, 25 (58%) were members of the ACCESS cohort while 18 (42%) were selected outside the cohort. The distributions of basic home characteristics for all sites, those in the cohort, and those outside the cohort are

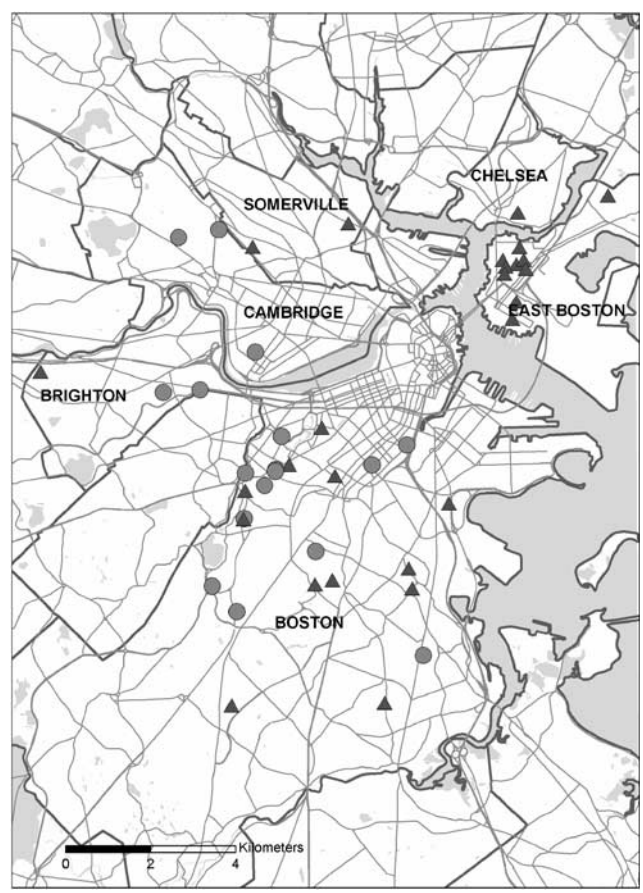

Figure 1. Locations of 43 air pollution sampling sites. ▲, Cohort; ●, Non-cohort; ——, Major roads.

summarized in Table 1. The age and type of home are similar in both cohort and non-cohort members. The non-cohort members have slightly larger apartments, but the mean size (less than five rooms) remains relatively small.

Household activities identified as possible indoor sources are presented in Table 2, along with their distributions and our determination of which were considered as a possible predictor for NO_2, $PM_{2.5}$ and selected particle constituents. For the hypothesized sources of interest, such as cooking and cleaning activities, there is adequate heterogeneity within the study population in the frequency of these activities.

Table 1. Distributions of basic home characteristics for all participants, cohort members, and non-cohort members.

Categorical variables	Total ($n=43$) (%)	Cohort members ($n=25$) (%)	Non-cohort members ($n=18$) (%)
Type of housing			
Single-family home	5	4	6
Multi-family home	56	60	50
Apartment building	39	36	44
Year built			
Before 1900	20	16	27
1900–1949	56	52	61
1950–1969	12	16	6
1970–later	12	16	6
Continuous variable	Mean (SD)	Mean (SD)	Mean (SD)
Number of rooms	4.2 (1.7)	3.7 (1.2)	4.8 (2.1)

Indoor and Outdoor Concentrations

The percentage of samples above the LOD for each pollutant is shown in Figure 2, with only those with at least 70% of the

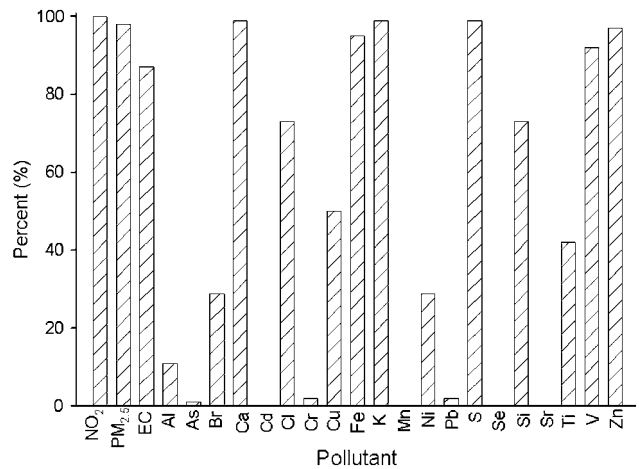

Figure 2. Percentage of samples above the LOD. (LOD's for NO_2, $PM_{2.5}$, and EC based on method blank values, all others are based on uncertainty limits).

Table 2. Distributions of selected household activities for all sampling sessions.

Categorical variables	Percent (%)	Potential source term for
Cleaning activities[a] *($n=61$)*		$PM_{2.5}$, Ca, Fe, K , Si, Na, Cl, Zn
0–1 activity/week	33	
2–3 activities/week	67	
Humidifier use ($n=61$)		$PM_{2.5}$, Ca, K, Si, Cl
No	82	
Yes	18	
Candle use ($n=61$)		$PM_{2.5}$, EC
No	74	
Yes	26	
Cooking time ($n=61$)		$PM_{2.5}$, EC, Ca, Fe, K, Si, Na, Cl, Zn
≤ 1 h/day	66	
>1 h/day	34	
Gas stove usage ($n=59$) (stove type∗cooking time/day)		NO_2
Electric∗≤ 1 h/day, Electric∗>1 h/day, and Gas∗≤ 1 h/day	75	
Gas∗>1 h/day	25	
Continuous variables	Mean (SD)	
Occupant density ($n=61$) (number of occupants/rooms)	0.99 (0.58)	$PM_{2.5}$, Ca, Fe, K, Si, Na, Cl, Zn

[a]Cleaning, sweeping, and/or vacuuming.

Table 3. Indoor and outdoor residential pollutant concentrations and I/O ratios.

	Outdoor concentrations				Indoor concentrations				Indoor/outdoor ratios	
Pollutant	*N*	Median	Mean (SD)	Range	*N*	Median	Mean (SD)	Range	*N*	Median (CV)
NO_2 (ppb)	52	16.8	17.2 (5.67)	5.21–33.3	54	17.1	19.6 (11.0)	5.67–61.1	51	0.99 (0.63)
$PM_{2.5}$ ($\mu g/m^3$)	60	12.6	14.2 (5.43)	6.75–31.3	64	16.7	20.3 (12.5)	6.77–74.9	58	1.14 (0.71)
EC ($m^{-1} \times 10^{-5}$)	58	0.55	0.63 (0.49)	0.08–3.8	62	0.49	0.57 (0.35)	0.12–2.2	56	0.89 (0.64)
Ca (ng/m^3)	58	24.4	29.4 (17.2)	9.12–113	62	32.1	48.0 (84.8)	12.8–676	56	1.16 (1.90)
Fe (ng/m^3)	58	61.7	66.5 (26.4)	15.4–162	62	44.5	46.1 (20.0)	3.50–101	56	0.69 (0.40)
K (ng/m^3)	58	46.8	57.5 (56.7)	19.1–446	62	82.2	78.6 (8.22)	15.2–477	56	1.10 (0.95)
Si (ng/m^3)	58	34.9	55.9 (71.9)	4.81–467	62	38.3	83.4 (200)	5.53–1480	56	1.04 (1.31)
Na (ng/m^3)	58	119	144 (160)	29.2–1260	62	128	185 (320)	2.27–2520	56	1.05 (1.84)
Cl (ng/m^3)	58	4.25	15.2 (41.4)	0.194–3050	62	20.8	87.2 (3680)	0.528–2920	56	3.18 (3.79)
Zn (ng/m^3)	58	10.9	14.7 (19.3)	5.23–1520	62	8.86	14.7 (18.0)	3.40–1050	56	0.83 (1.13)
S (ng/m^3)	58	1280	1540 (1450)	413–11300	62	928	1120 (699)	310–3910	56	0.76 (0.32)
V (ng/m^3)	58	3.52	4.79 (4.00)	1.08–22.5	62	2.84	3.54 (2.66)	0.392–15.5	56	0.76 (0.46)

samples above the LOD included in formal regression analyses. With the exception of NO_2 (32%) and Cl (40%), the mean RD was less than 25% for all pollutants, indicating reasonable method precision.

Summary statistics for the measured residential indoor and outdoor concentrations and I/O ratios are presented in Table 3. All pollutant concentrations appear to be lognormally distributed, as shown by Shapiro–Wilks tests on the log-transformed data ($\alpha = 0.05$), with concentrations both indoors and outdoors spanning an order of magnitude. Median I/O ratios are significantly greater than 1 for $PM_{2.5}$, Ca, and Cl as determined by a one sample median test ($P < 0.05$). I/O ratios for Fe, Zn, S, and V are significantly less than 1 ($P < 0.05$), with the I/O ratio for EC being marginally significantly less than 1 ($P < 0.1$). For NO_2, K, and Si, I/O ratios are not significantly different than 1, although they varied significantly across sites.

Results from the regression analyses of outdoor concentrations against indoor concentrations are shown in Table 4. Outliers were removed that unduly influenced regression results, defined as having an absolute studentized residual greater than four. The β_1s are the coefficients of the outdoor term in Equation (1), without consideration of indoor source terms. Outdoor pollutant concentrations explained between 78% (S) and 7% (NO_2) of the variability seen in indoor concentrations (excluding Si), and the outdoor term was significant ($P < 0.1$) for all pollutants except Si. As anticipated, the coefficients (β_1s) are generally higher for combustion pollutants and lower for crustal elements, which are larger and have lower penetration efficiencies.

Identification of Indoor Source Terms

Regression analyses were performed according to the model described by Equation (1), with the indoor concentrations as the dependent variables and outdoor concentrations and selected source terms as the independent variables. Variables

Table 4. Univariate regression analysis of outdoor concentrations on indoor concentrations.

Pollutant	β_1 (SE)	*P*-value	R^2
NO_2	0.48 (0.26)	0.07	0.07
$PM_{2.5}$	0.91 (0.23)	0.01	0.23
EC	0.72 (0.10)	<0.0001	0.49
Ca	0.56 (0.12)	0.01	0.30
Fe	0.38 (0.09)	<0.0001	0.26
K	0.83 (0.11)	<0.0001	0.52
Si	0.02 (0.07)	0.78	0.00
Na	0.46 (0.07)	<0.0001	0.43
Cl	0.40 (0.15)	0.01	0.12
Zn	0.85 (0.19)	<0.0001	0.28
S	0.95 (0.07)	<0.0001	0.78
V	0.60 (0.04)	<0.0001	0.77

and regression coefficients are shown in Table 5. Cooking for more than an hour per day (as compared to less than an hour) was associated with a significant increase in $PM_{2.5}$ and Zn. Occupant density significantly predicted indoor $PM_{2.5}$, Na, Cl, and Si concentrations. Additionally, performing more than one cleaning activity was negatively associated with indoor Fe levels, and humidifier use was associated with an increase in indoor Ca. Cooking on a gas stove for more than an hour significantly increased indoor NO_2 levels, as compared to cooking for less than an hour or on an electric stove. Of note, no source terms were associated with indoor V, but humidifier use was associated with indoor S. However, this term was actually associated with a decrease in indoor levels, and before the addition of any source terms, the intercept was statistically insignificantly different from zero for S. In addition, few homes demonstrated indoor/outdoor ratios of sulfur significantly greater than 1, with only four of 56 ratios above 1.05. Thus, this does not provide significant evidence of indoor sources of S in this cohort. Outdoor

Table 5. Identification of indoor source terms contributing to indoor concentrations after adjusting for outdoor concentrations.[a]

Pollutant	R^2	Outdoor concentration Estimate (SE)	Cooking time ($\leq$1h/day = 0, > 1h/day = 1) Estimate (SE)	Candle use (No = 0, Yes = 1) Estimate (SE)	Humidifier use (No = 0, Yes = 1) Estimate (SE)	Cleaning activities ($\leq$1/week = 0, > 1/week = 1) Estimate (SE)	Occupant density Estimate (SE)	Gas stove usage (other[b] = 0, gas* > 1 h/day = 1) Estimate (SE)
NO_2 (ppb)	0.16	0.531 (0.224)**	N/A[c]	N/A	N/A	N/A	N/A	5.70 (3.06)*
$PM_{2.5}$ (μg/m^3)	0.37	0.878 (0.225)**	5.71 (2.92)**	−2.46 (3.17)	−1.34 (3.17)	−0.364 (2.78)	4.11 (2.55)*	N/A
EC (m^{-1} × 10^{-5})	0.52	0.738 (0.111)**	−0.054 (0.056)	0.012 (0.068)	N/A	N/A	N/A	N/A
Ca (ng/m^3)	0.48	0.446 (0.117)**	−4.27 (4.67)	N/A	15.2 (5.72)**	3.28 (4.34)	4.19 (3.91)	N/A
Fe (ng/m^3)	0.30	0.391 (0.093)**	0.950 (5.53)	N/A	N/A	−9.95 (5.15)**	8.14 (4.38)	N/A
K (ng/m^3)	0.54	0.817 (0.117)**	10.3 (15.7)	N/A	19.1 (19.0)	11.8 (14.8)	−14.7 (13.2)	N/A
Si (ng/m^3)	0.05	0.020 (0.070)	−5.38 (12.3)	N/A	−13.3 (15.2)	−3.62 (11.0)	15.4 (10.1)*	N/A
Na (ng/m^3)	0.47	0.452 (0.075)**	17.1 (28.9)	N/A	N/A	−16.0 (26.4)	30.5 (22.5)*	N/A
Cl (ng/m^3)	0.24	0.227 (0.168)*	5.46 (15.4)	N/A	−3.17 (19.30)	12.3 (13.9)	27.0 (12.9)**	N/A
Zn (ng/m^3)	0.36	0.813 (0.211)**	5.82 (2.61)**	N/A	N/A	−1.18 (2.44)	0.694 (2.22)	N/A
S (ng/m^3)	0.80	0.951 (0.070)**	N/A	N/A	−214.5 (114.3)**	N/A	N/A	N/A
V (ng/m^3)	0.77	0.596 (0.044)**	N/A	N/A	N/A	N/A	N/A	N/A

[a]**$P<0.1$ and *$P<0.2$.
[b]Includes all homes with electric stoves and those with gas stoves and cooking time of $\leq$1 h per day.
[c]N/A: variable not considered a potential covariate for the pollutants.

Table 6. Effects of outdoor concentrations on indoor concentrations modified by I/O sulfur category.[a]

Pollutant	R^2	High I/O S Estimate (SE)	High I/O S *P*-value	Low I/O S Estimate (SE)	Low I/O S *P*-value	Significance of interaction term[b]
Pollutants without indoor sources						
EC (m^{-1} × 10^{-5})	0.50	0.791 (0.121)	<0.0001	0.705 (0.103)	<0.0001	NS
K (ng/m^3)	0.52	0.834 (0.111)	<0.0001	0.963 (0.287)	<0.01	NS
S (ng/m^3)	0.93	1.00 (0.040)	<0.0001	0.641 (0.050)	<0.0001	**
V(ng/m^3)	0.79	0.754 (0.085)	<0.0001	0.596 (0.043)	<0.0001	**
Pollutants with indoor sources						
NO_2 (ppb)	0.16	0.557 (0.235)	0.03	0.473 (0.250)	0.07	NS
$PM_{2.5}$ (μg/m^3)	0.35	0.898 (0.230)	<0.001	0.781 (0.254)	<0.01	NS
Ca (ng/m^3)	0.46	0.557 (0.133)	<0.001	0.439 (0.125)	0.001	NS
Fe (ng/m^3)	0.39	0.543 (0.100)	<0.0001	0.347 (0.086)	<0.001	**
Si (ng/m^3)	0.06	0.118 (0.104)	0.26	−0.010 (0.077)	0.90	NS
Na (ng/m^3)	0.46	0.493 (0.184)	0.01	0.442 (0.074)	<0.0001	NS
Cl (ng/m^3)	0.29	1.16 (0.460)	0.02	0.139 (0.157)	0.38	**
Zn (ng/m^3)	0.38	0.659 (0.223)	<0.001	0.877 (0.190)	<0.0001	*

[a]Final models after backwards elimination model, effect estimates of indoor sources not shown
[b]NS = not significant, **$P<0.1$ and *$P<0.2$.

concentrations remained significant for all pollutants but Si. To note, all regression analyses were repeated using a stepwise selection approach and the results were unchanged.

Effect Modification by Air Exchange Rates

The I/O ratio of sulfur was dichotomized at the median (0.76) to serve as a proxy for 'high' and 'low' AERs and was only included as an effect modifier of outdoor concentrations without modifying the effect of indoor sources (Table 6), due to the limited statistical power and resulting statistical instability when effect modification of indoor sources was included. The effect estimates for the indoor source terms remained similar in magnitude and significance after the addition of the interaction terms. The exception was in the indoor Fe model where the effect estimate for the cleaning activities term was still negative, but smaller in magnitude (−3 ng/m^3) and no longer significant. For the pollutants without identified indoor sources, with the exception of K, there was generally a larger effect of outdoor concentrations in homes in the high AER category compared with homes in the low AER category, as anticipated. However, the interaction term was only significant for S and V. For most

of the pollutants with identified indoor sources, a larger effect of outdoor concentrations was observed in homes with high AERs compared with homes with low AERs, with the difference in effects significant for Fe and Cl. The exception is for Zn where the effect appeared significantly larger in homes with low AERs (albeit in a model excluding an interaction term on indoor sources). For Si outdoor concentrations did not have a significant effect on indoor concentrations for homes in either AER category.

Table 7. Distributions of selected household activities among cohort and non-cohort members for all sampling sessions.

Categorical variables	Cohort members ($n=38$) (%)	Non-cohort members ($n=23$) (%)
Cleaning activities[a]		
0–1 activity/week	18	57
2–3 activities/week	82	43
Humidifier use		
No	76	91
Yes	24	9
Candle use		
No	76	70
Yes	24	30
Cooking time		
≤ 1 h/day	53	87
>1 h/day	47	13
Opening of windows		
No	47	22
Yes	53	78
Air conditioning use		
No	63	86
Yes	37	13
Continuous variables	Mean (SD)	Mean (SD)
Number of occupants spending >4 h in the home	4.6 (1.8)	2.2 (1.2)
Occupant density (people/room)	1.3 (0.46)	0.43 (0.14)

[a]Dusting, cleaning, and vacuuming.

Influence of Home Vs. Participant Characteristics

Although basic home characteristics are similar between cohort and non-cohort members (Table 1), multiple activity patterns differed significantly (Table 7). Cohort members tended to cook and clean more, had more frequent air conditioning use, less opening of windows, and greater occupant density. Thus, a comparison between these groups can provide another means to determine the influence of occupant activity patterns on I/O ratios.

After the removal of outliers (following identical criteria as above), the median I/O ratios of $PM_{2.5}$, EC, and NO_2 were significantly ($P<0.05$) different between the two groups as determined by a Wilcoxon rank-sum test (Figure 3). The median I/O ratios of $PM_{2.5}$ and NO_2 were greater in the cohort group's homes than in the non-cohort group's homes, indicating a greater influence of indoor sources. However for EC, the opposite was true, indicative of higher outdoor EC and/or lower AERs for cohort members. This is supported by Table 8, where outdoor EC concentrations are higher for cohort members yet indoor concentrations are the same for the two groups, and by Table 7 illustrating higher infiltration of outdoor EC due to the more opening of windows in non-cohort homes. Also shown in Figure 3, the median I/O ratio

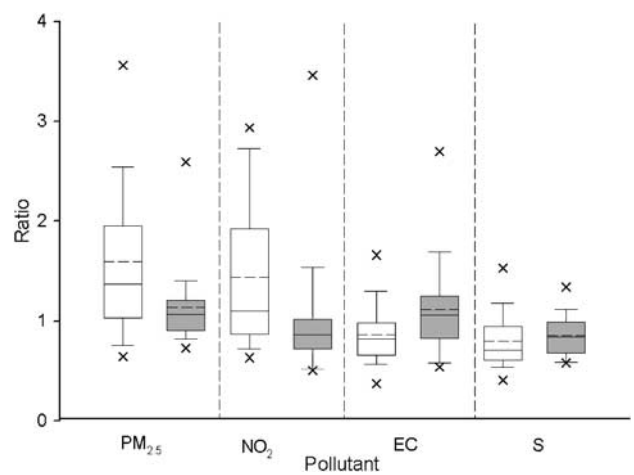

Figure 3. Indoor/outdoor pollutant ratios among cohort and non-cohort members (solid line = median; dotted line = mean; boxes = interquartile range; whiskers = 10th and 90th percentiles; X's = 5th and 95th percentiles). ▭, Cohort; ▭, Non-cohort.

Table 8. Indoor and outdoor concentrations among cohort and non-cohort members for selected pollutants.

		Outdoor concentration				Indoor concentration			
Pollutant	Group	*N*	Median	Mean (SD)	Range	*N*	Median	Mean (SD)	Range
NO_2 (ppb)	Cohort	24	16.1	16.9 (6.03)	5.21–33.3	26	20.2	21.7 (10.5)	5.67–49.8
	Non-Cohort	28	17.1	17.5 (6.0)	7.43–29.7	28	15.1	17.6 (11.3)	6.42–61.1
$PM_{2.5}$ ($\mu g/m^3$)	Cohort	31	12.7	15.0 (5.49)	7.70–31.3	36	20.7	22.6 (10.8)	6.77–46.4
	Non-Cohort	28	12.2	13.4 (5.38)	6.75–28.6	27	12.5	15.2 (8.56)	7.17–48.3
EC ($m^{-1} \times 10^{-5}$)	Cohort	31	0.58	0.61 (0.24)	0.30–1.4	35	0.48	0.53 (0.27)	0.12–1.3
	Non-Cohort	25	0.49	0.54 (0.24)	0.08–1.3	24	0.48	0.54 (0.24)	0.25–1.2

of S appears to follow a similar pattern as EC with a higher median for non-cohort members compared to cohort members. Although not statistically significant ($P = 0.2$), this suggests that infiltration rates and therefore AERs are higher in homes of non-cohort members than of cohort members, and that there is a greater influence of indoor sources of EC (not identified in the regression analyses) in non-cohort homes.

Discussion

Ambient and Non-ambient Contributions to Indoor Concentrations

In our study, predicting 1-week average indoor concentrations based solely on outdoor concentrations did not explain the majority of the variability in many cases, reflecting the influence of home-to-home variations in indoor sources, occupant behaviors, and AERs. For NO_2 and most particle constituents (with the exception of outdoor-dominated constituents like sulfur and vanadium), the addition of selected indoor source terms improved the model's predictive power, significantly so in many cases (i.e., NO_2, $PM_{2.5}$, Ca, Cl). Cooking time, gas stove usage, occupant density, and humidifiers were identified as important contributors to indoor levels. We also found additional information value in a dummy variable created from indoor/outdoor ratios of sulfur ($AERDummy_i$), which theoretically captured AERs and allowed us to better incorporate some of the principles of the mass balance model.

Our measured residential indoor and outdoor concentrations, and I/O relationships are largely comparable to those seen in other studies (Zipprich et al., 2002; Brunekreef et al., 2005; Meng et al., 2005). Our findings are also in general agreement with present day models for predicting the impact of indoor sources based on integrated measurements, which identified gas appliances and cooking as important sources of NO_2 (Linaker et al., 1996; Levy et al., 1998; Rotko et al., 2001; Garcia-Algar et al., 2004) and PM (Ozkaynak et al., 1994; Brunekreef et al., 2005), respectively. The current study also identified resuspension activities as affecting PM exposure. These activities, often treated as episodic events increasing short-term indoor particle concentrations, (Thatcher and Layton, 1995; Abt et al., 2000; Long et al., 2000) in our study seemed to also affect longer-term indoor levels. The additional significance of resuspension factors in our study may be associated with the smaller volumes and greater crowding (higher occupant densities) among our participants, as opposed to the single-family homes generally sampled in other studies.

PTEAM estimated that cooking caused an average increase of 9 $\mu g/m^3$ (Ozkaynak et al., 1996) in indoor $PM_{2.5}$, with EXPOLIS observing a 11.65 $\mu g/m^3$ (Amsterdam) cooking effect on $PM_{2.5}$ (Brunekreef et al., 2005). Although the differences in source covariates and averaging times impair direct interpretability, our cooking time covariate in Table 5 contributed a similar magnitude concentration increase to $PM_{2.5}$ as in the studies above (with a 5.7 $\mu g/m^3$ increment associated with cooking more than 1 h/day). Furthermore, our gas stove usage covariate in Table 5 contributed a similar magnitude increase (5.7 ppb) of NO_2 as in previous studies, where gas stove usage contributed approximately 10–20 ppb to indoor concentrations (Lee et al., 1998) and personal NO_2 exposure were found to be 14 ppb higher in homes with gas stoves compared to those without them (Levy et al., 1998). Of note, our study did not examine ETS, as smoking (1–4 cigarettes/day) inside the home was reported only during four sampling sessions. The results did not change with the exclusion of these participants; therefore they remained in the final models.

Two exceptions to our agreement with the literature are for NO_2 and Si, where outdoor concentrations explain indoor concentrations to a lesser extent than expected. For NO_2, there is evidence to suggest that inadequate statistical power and measurement error may be a cause. Due to an error in the laboratory, some samples taken during the heating season were lost, resulting in a smaller sample size and an unbalanced data set by season. Moreover, the mean RD was 32% (as compared with 9% for $PM_{2.5}$ and 12% for EC), indicating poorer precision potentially affecting the observed relationships. For Si, outdoor concentration was not a statistically significant contributor to indoor concentrations. One reason could be that the Si particles found outdoors are too large to readily penetrate the building envelope and/or have a short residence time so that they deposit quickly once entering the indoor environment. This rationale explains the weaker association for many other crustal elements as well.

While we were able to capture some significant indoor source terms, it is possible that they do not reflect direct causal influences on concentrations, and some covariates are somewhat difficult to interpret. For example, there are numerous types of cooking activities that may have differential effects on concentrations, and we were unable to incorporate all potential covariates due to limited statistical power. Additionally, some of the source terms are correlated with one another and may be proxies for multiple factors. For example, occupant density may not only represent resuspension activities (more people moving around causing more resuspension), but may also be associated with SES and related occupant activity patterns. In univariate regressions (results not shown), higher occupant densities were significantly ($P < 0.05$) associated with increased cooking and cleaning, indicating the difficulty in separating these source terms. Due to substantial differences in occupant density by cohort status, occupant density was also correlated with ventilation-related behaviors, further complicating interpretation of this term. While factor

analytic methods on questionnaire data could have resolved some of the collinearity issues, this would have impaired general interpretability and not resolved correlations between source terms and non-source factors.

As we did not find evidence of significant indoor sources of sulfur (all homes using humidifiers had lower AERs, potentially explaining the negative relationship in Table 5) and found a strong relationship between indoor and outdoor concentrations ($R^2 = 0.78$), this dummy variable should appropriately capture categories of AERs. Although not statistically significant for some of the outdoor dominated pollutants, there appears to be a trend with the coefficient for outdoor concentrations increasing with increasing AER category, indicating as expected that outdoor concentrations are greater contributors to indoor concentrations at higher AERs. The exception is for K where a larger effect estimate is seen in homes with low AERs, although this difference is insignificant. The reason for this result is unclear; one possibility is that this model did not take into account the effect on indoor sources. Although none were identified in the primary analysis, this does not ensure that there are no indoor sources of K.

For those pollutants with identified indoor sources, the AER proxy demonstrated similar results to those of the outdoor-dominated pollutants, with the exception of Zn where there was a larger effect of outdoor concentrations in the low AER category, although this difference was only marginally significant ($P = 0.19$), This unexpected results may possibly be due to the exclusion of an interaction between AER and indoor source terms. Ideally, regression models would incorporate this factor, but statistical power was limited in this investigation and the resulting models displayed significant instability. In addition to inadequate statistical power, there were correlations between some of the indoor source terms and F_{INF}, such as occupant density, which was negatively associated with the dichotomized I/O sulfur ratios. More generally, significant measurement error would also be anticipated using the effect modification on our questionnaire responses. These data are proxies rather than direct measurements of the indoor source strength, and in accordance with the mass balance model, other factors such as home volume may also have affected the relationships.

Even with the incorporation of ventilation characteristics, the predictive power for most of the pollutants with indoor sources was still relatively low, with less than half of the variability explained in many cases. This raises questions about the measurement error associated with using these models in order to estimate long-term exposures for a cohort study. However, in spite of the limitations, our models do however identify which indoor sources appear to be important, which both informs epidemiological investigations and offers guidance for future design of questionnaires in an epidemiological context. For example, more resolved information on the frequency and duration of cooking (including specific types of cooking) or humidifier use could improve the effectiveness of the models. While these issues were incorporated into our questionnaires, the categories may have been too coarse to capture relatively small gradients in exposure. In addition, many epidemiological studies wish to classify people into exposure categories as opposed to predicting the exact concentrations, and our models may prove effective in this regard. More generally, imperfect models addressing indoor exposures may reduce exposure misclassification more than models that only consider outdoor concentrations. Future analysis will focus on evaluating the implication of any exposure misclassification with our models or alternative approaches on epidemiological studies.

Building Vs. Occupant Characteristics

One of the important dimensions of our study is the fact that we recruited two distinct populations with similar basic housing characteristics in similar neighborhoods, allowing us to understand the influence of building vs. occupant characteristics on indoor exposures. Their different activity patterns resulted in more heterogeneity in their exposures due solely to indoor sources. The cohort, consisting of pregnant or recently pregnant women, spent much more time at home (mean = 19.5 h/day) than the non-cohort members (mean-10 h/day). Therefore, as suggested by Table 7, cohort members may perform more activities generating indoor air pollution, making their levels higher than those seen outdoors or in non-cohort participant homes. There may also be a difference in AERs, with homes in the cohort having lower AERs than non-cohort homes. This suggests a difference in housing characteristics or activity patterns that is not captured by Table 1.

As we used convenience sampling methods to recruit additional participants, largely focused on increased representation of undersampled neighborhoods, it could be argued that our findings may be less representative of the larger cohort population by the inclusion of participants outside the ACCESS cohort. Yet, these non-cohort homes were located in the target neighborhoods and had similar basic housing characteristics. More generally, in well-developed urban areas there may be limited new residential construction, so basic housing characteristics may be more homogeneous.

Limitations

As in any monitoring study, this study was potentially limited by errors in exposure measurements and methods of data collection. The measurements collected during the sampling period may not accurately represent typical conditions, limiting our ability to draw broad conclusions about long-term exposure patterns. In addition, in order to limit the effect of our study on the subject's activities, the sampling equipment was placed out of the way and did not require any maintenance by the participant. Thus, the time–activity patterns of the

individual should not have been altered due to the burden of the sampling equipment, although this meant our sampling captured a region of the house instead of the participant's (or their child's) personal exposures. However, the time–activity data indicate that residential concentrations may be a reasonable proxy for personal exposures of cohort members.

The housing characteristics and occupant activities used depended on questionnaire data, and although a standardized questionnaire was used, there may still be a lack of accuracy and reliability in the data. Further, although sampling was conducted in two different seasons (a heating and non-heating season), these were broadly defined and covered a period up to 6 months. Under ideal circumstances, data collection would have occurred simultaneously across all homes in each season to minimize seasonal variability, but this was not logistically feasible. Therefore, each sampling session was treated as an independent measurement.

In addition, the sample size limited our ability to explore a larger range of potential indoor source terms. More variations in indoor concentrations may have been accounted for by including continuous variables for AERs or in more closely adhering to a mass-balance model. Since AER was estimated via a proxy (I/O of sulfur), using a continuous AER would have required explicit assumptions about P and k, and we had less confidence in these estimates than in creating broad categories representing 'high' and 'low' AERs.

Conclusions

In conclusion, the current paper identified important predictors of indoor concentrations for multiple air pollutants in a low SES urban population, including outdoor concentrations, indoor source terms, and proxies for AERs. This allows us to determine the information necessary to assess long-term indoor exposures. The important indoor sources include average cooking time per day, humidifier use, occupant density, and gas stove usage, with different sources important for different pollutants, indicating that the questionnaire data needed will be dictated by the pollutant being studied. The crustal and sea salt elements were mainly associated with occupant density, with aggregate $PM_{2.5}$ also associated with cooking time, and indoor NO_2 increasing with increasing gas stove usage. We also found it useful to capture AERs in a context where AERs could not be measured by dichotomizing the I/O ratios of sulfur, although this covariate was more informative for outdoor source dominated pollutants. Additionally, our cohort vs. non-cohort analysis illustrated that it was the occupant activity patterns that were driving the indoor exposure patterns rather than the basic housing characteristics.

In general, our study provides some direction regarding how exposure-related questionnaires should be refined in population studies, in order to predict indoor exposures in the absence of measurements, which are often not possible for a large cohort. We have demonstrated that, in lower-SES urban dwellings (largely multi-unit), resuspension activities along with cooking and stove usage appear to contribute significantly to longer-term indoor exposures to some pollutants. The importance of resuspension activities has not been observed in studies focusing on single-family homes, indicating that more research is needed in urban areas where more people reside in multi-unit dwellings with higher occupant densities. Incorporating this information will lead to more accurate predictions of indoor pollutant levels for lower SES populations, improving our ability to detect health effects in large cohort studies. Future studies will consider the information value of GIS and other publicly available data in predicting indoor exposure patterns in the absence of outdoor monitoring and detailed activity information, which are often difficult to obtain in these types of studies.

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