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Modeling indoor particulate exposures in inner city school classrooms

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Abstract

Outdoor air pollution penetrates buildings and contributes to total indoor exposures. We investigated the relationship of indoor to outdoor particulate matter in inner-city school classrooms.

The School Inner City Asthma Study investigates the effect of classroom-based environmental exposures on students with asthma in the northeast United States. Mixed-effects linear models were used to determine the relationships between indoor $PM_{2.5}$ and BC and their corresponding outdoor concentrations, and to develop a model for predicting exposures to these pollutants. The indoor-outdoor sulfur ratio was used as an infiltration factor of outdoor fine particles.

Weeklong concentrations of $PM_{2.5}$ and BC in 199 samples from 136 classrooms (30 school buildings) were compared to those measured at a central monitoring site averaged over the same timeframe. Mixed effects regression models found significant random intercept and slope effects, which indicate that: 1) there are important $PM_{2.5}$ sources in classrooms; 2) the penetration of outdoor $PM_{2.5}$ particles varies by school, and 3) the site-specific outside $PM_{2.5}$ levels (inferred by the models) differ from those observed at the central monitor site. Similar results were found for BC except for lack of indoor sources. The fitted predictions from the sulfur-adjusted models were moderately predictive of observed indoor pollutant levels (Out of sample correlations: $PM_{2.5}$: r^2 = 0.68, BC; r^2 = 0.61).

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Conflict of interest

The authors declare no conflict of interest.

Our results suggest that $PM_{2.5}$ has important classroom sources, which vary by school. Furthermore, using these mixed effects models, classroom exposures can be accurately predicted for dates when central site measures are available but indoor measures are not available.

Keywords

air pollution; particulate matter; black carbon; school; asthma; exposure modeling; sulfur tracer method; $PM_{2.5}$

Introduction

Personal exposure to ambient and in-home air pollution is directly linked to adverse outcomes across a wide range of ages and health conditions (1, 2) including childhood asthma (3, 4). Outdoor pollutants penetrate the home environment and contribute to the overall personal exposure and asthma morbidity(5, 6); however, home characteristics, heating and cooking (7), and other personal activities(8) contribute to the overall burden of respirable pollutants.

Over the past 2 decades, attention has been directed to inhalant pollutants in primary schools across diverse locations, internationally(9-26). Particulate matter (measured as PM_{10} or $PM_{2.5}$, predominately) has consistently found to be elevated indoor compared to outdoor, with few exceptions(27). In many studies, levels greatly surpassed indoor air quality (IAQ) reference thresholds(28, 29). The differences in locations by urbanization, ambient climate, ventilation system, and school characteristics likely contribute to diverse findings in comparing indoor to outdoor pollutants across studies. However, there is a lack of U.S. based school studies.

The school classroom environment is an important source of exposures to pollutants for children. While workplace micro-environments have been extensively studied for adults, less is known about the personal exposures in schools and classrooms – the child's occupational environment. In part, this is due to the challenges of deploying sampling equipment in school classrooms while school is in session. For children in urban United States, the school classroom presents a unique set of circumstances and exposures to those found in homes. Exposure to traffic-related pollutants may be intensified as schools are frequently located centrally within the community for easy access to traffic routes and have specific periods of drop-off and pick-up during which large numbers of buses and cars may idle next to the building(30). The buildings tend to be older and may suffer from poor maintenance and ventilation (31, 32). Additionally, there are fewer indoor pollutant emissions, as most schools have no active cooking and prohibit tobacco smoke on the premises. Few studies have evaluated the extent of poor indoor air quality in schools (9, 31).

Because most school classrooms lack indoor sources of combustion, it is reasonable to hypothesize that indoor pollutant concentrations are highly correlated to the outdoor pollution measures. In this analysis, we aim to 1) describe the relationship between indoor and outdoor levels of particulate matter ≤2.5 μm ($PM_{2.5}$) and black carbon (BC) in inner city school classrooms attended by children with asthma in the School Inner City Asthma Study,

and 2) develop a model for predicting classroom $PM_{2.5}$ and BC utilizing the sulfur tracer method and measurements from the local central monitor. We further compared this method with satellite predictions for classroom measures.

Materials and Methods

The School Inner City Asthma Study (**ClinicalTrials.gov:**NCT01756391) is a five-year prospective study evaluating the effects of the indoor environment of school classrooms on asthma morbidity for children with asthma attending public school. Detailed methods have been published previously (33). Briefly, children with asthma enrolled in the public school were recruited each summer prior to start of school and followed for the length of a school year with quarterly symptom assessments and biannual lung function testing. Informed consent/assent was obtained for all subjects. The enrolled subject's school classroom was assessed twice per year (fall and spring) for allergen, mold and endotoxin levels. Indoor levels of nitrogen dioxide and fine particulate matter were measured in a subset of these subjects' classrooms. Additionally, school/classroom inspections and other relevant data were collected, such as cafeteria cooking operations, fuel type, and relation of classroom location to street/driveway. This study was approved by the investigational review board at Boston Children's Hospital

Particle sampling and chemical analysis

Weeklong indoor $PM_{2.5}$ samples were collected using a personal exposure monitor (PEM) in school classrooms in 1 or 2 seasons during the academic school years between 2008 – 2013. PEM includes an inertial impactor designed specifically for personal or indoor sampling. PEM collects $PM_{2.5}$ on Teflon filters at a low flow rate of 1.8 L/min. In addition, concurrent daily $PM_{2.5}$ samples were collected at the central monitoring supersite, which is located within 10 km of the schools. Details on outdoor $PM_{2.5}$ measurement at the central site have been previously described by Kang et al. (34). While the central site is part of the urban community, it is located approximately 20 m above ground level and is not adjacent to local highways.

Teflon filters, including blank filters, were weighed on an electronic microbalance (MT-5 Mettler Toledo, Columbus, OH) prior to and after field measurements, after being equilibrated for a period of 48 hours in a particle-free room of controlled temperature (22±1.5°C) and relative humidity (40±5%). Following gravimetric measurement, Teflon filters were analyzed for indoor BC concentrations by measuring filter blackness using a smoke stain reflectometer (model EEL M43D, Diffusion Systems Ltd., United Kingdom). We assumed a factor of 1.0 for converting the absorption coefficient to BC mass, and then divided by each sampled volume to calculate indoor BC concentration. On the other hand, outdoor BC concentrations were measured using a single (λ=880 nm) channel aethalometer (model AE-16, Magee Scientific, Berkeley, CA). Two different BC methods were compared to establish a linear relationship (R^2=0.72, Slope= 0.95). Sulfur concentration in both the indoor and outdoor $PM_{2.5}$ samples was determined using X-Ray Fluorescence (XRF) spectroscopy (model Epsilon 5, PANalytical, The Netherlands) (35). Quality assurance and

control of XRF analysis was performed as previously described(36). All analyses were conducted at the Harvard T.H. Chan School of Public Health.

Sulfur Tracer Method for Determining Indoor $PM_{2.5}$ of Outdoor Origin

Particle concentrations depend on particle infiltration, exfiltration, emission and deposition. Assuming no indoor sources of sulfur, the indoor penetration of **$PM_{2.5}$** emitted from outdoor sources can be approximated by the sulfur tracer method (37, 38). Accordingly, the indoor to outdoor sulfur ratio (S_{IN}/S_{OUT}) was calculated for each school classroom and classroom sampling period to approximate the infiltration fraction of fine particles.

Satellite predictions

Satellite-based exposure assessment of $PM_{2.5}$ was assessed by a hybrid approach(39, 40). This hybrid approach combined simulation outputs from chemical transport model, land-use terms, satellite measurements and meteorological fields into a neural network. The neural network was trained with monitoring $PM_{2.5}$ data from EPA Air Quality System (AQS) and made prediction of $PM_{2.5}$ at 1 km×1 km grid cell on a daily basis. Ten-fold cross-validation demonstrated that predicted $PM_{2.5}$ correlated well with monitoring data. Individual exposure levels were ascertained by matching schools geocoded location to the nearest 1 km×1 km grid cell. Estimates of ambient Black Carbon (BC) concentrations were generated using a validated nu-SVR (Support Vector Regression) prediction model trained with 24,709 observations and 408 monitors throughout the region. Ten-fold cross validation showed good correlation between observed and predicted concentrations. Daily predictions at each school from 2000 to 2011 were made by extracting temporal and spatial predictors at each location and for each date.

Statistical Analysis

The indoor to outdoor ratio of sulfur concentrations in $PM_{2.5}$ was used as the infiltration ratio for each sampled classroom, as noted above.

Descriptive Model

To examine the relationship between the indoor and outdoor concentrations of $PM_{2.5}$ and BC, linear mixed-effect models were deployed, which used indoor concentration as the dependent variable, the product outdoor concentration *sulfur ratio as the independent variable, and the random effects at the school level (intercept and slope with unstructured covariance). A random component was dropped from the model if its variance estimate was less than twice its standard error. Additional covariates to reflect time were considered such as year of study and a restricted cubic spline of days since October 1st.

$$\mathrm{indoor}_{ij}=\beta_0+\beta_1\mathrm{outdoor}_{ij}+u_{oi}+u_{1i}\mathrm{outdoor}_{ij}*\mathrm{S\_ratio}_{ij}+\varepsilon_{ij}$$

for i^{th} school and j^{th} classroom measurement

Predictions of indoor concentrations

To determine an infiltration factor for a given day, a model was developed to predict the sulfur ratio. The following parameters were considered as predictors: ambient temperature, cubic spline of days, school year sampled, random intercept for school, random slope for school, condition of the classroom ceiling, walls, and windows, the number of windows in the classroom, whether the windows open, whether the windows face the drop-off/pick-up area, and floor level of the classroom. The final model included ambient temperature, spline of days, and school random intercept. The predicted sulfur ratio for a given classroom sample was estimated with this final model using all data points except for the classroom sample being predicted. This predicted sulfur ratio multiplied by the outdoor concentration equals the concentration of indoor particles that are of outdoor origin, the outdoor source contribution. Subsequently, indoor concentration was predicted for a given classroom sample using the mixed effects model described above using all data points except for the classroom sample being predicted.

Step 1—Calculate sulfur ratio for j^{th} classroom in i^{th} school

$$\mathrm{S\_ratio}_{ij}=\mathrm{indoor\_S}_{ij}/\mathrm{outdoor\_S}_{i}$$

Step 2—Model the sulfur ratio at i^{th} school and j^{th} classroom measurement from cubic spline of days since school started, temperature, and school specific intercepts.

$$\mathrm{S\_ratio}_{ij}=\beta_0+\beta_1\mathrm{time}_1+\beta_2\mathrm{time}_2+\beta_3\mathrm{time}_3+\beta_4\mathrm{time}_4+\beta_5\mathrm{temp}+u_{oi}+\varepsilon_{ij}$$

Step 3—Estimate outdoor contribution of indoor on given day

$$\mathrm{out\_in}_{ij}=\widehat{\mathrm{S\_ratio}}_{ij}*\mathrm{outdoor}_{i}$$

Step 4—Predict indoor concentration from estimate of outdoor contribution of indoor and school specific intercepts and coefficients.

Satellite-based estimates of $PM_{2.5}$ and BC were evaluated in the models described above by interchanging the term for central monitor site measurement with the temporally related satellite-based value at the school. Satellite-based model results were compared with the central site sulfur-adjusted model.

Results

In total, 199 samples of $PM_{2.5}$ mass and elements including sulfur and 198 samples of BC obtained from 136 classrooms across 30 school buildings were compared with $PM_{2.5,}$ sulfur, and BC measured at the nearby central monitoring site. Figure 1 depicts the geographic relationship between schools and the central monitoring site. Notably, the central site is on the roof of a 6 story building and median distance from schools was 4974 m (range 1065 m – 11592 m). Table 1 displays selected key characteristics of the schools and classrooms

sampled in this analysis. The majority of schools included were built in the early part on the 20th century. None of the schools have gas cooking in the building and all had recently serviced furnaces. The classrooms largely had windows that were able to be opened and about 1/3 faced the pick-up and drop-off area.

Table 2 demonstrates the distribution of indoor and outdoor concentrations measured. The sulfur indoor/outdoor ratio mean was 0.72 (standard deviation=0.14) and was normally distributed. Evaluation of the sulfur ratio variability over time demonstrated that it was relatively stable across schools and classrooms with a slight increase at the end of the school year (Figure 2). This reflects local seasonal trends in which there may have been increased air exchange rates due to opening of windows and doors in warmer weather, leading to greater infiltration of outside particles.

The final descriptive models predicting indoor $PM_{2.5}$ and BC (table 3) included an outdoor source fixed effect and school-level random effects. Sampling year and a term for time from start of school did not strengthen the model and so they were not included in the final mixed effects model for $PM_{2.5}$ or BC. There was significant variation in both school-level intercepts and outdoor $PM_{2.5}$ slopes, and these random effects were negatively correlated. The mixed effect model for BC showed little variance in school-level intercepts (σ^2=0.01, s.e.=0.01), so this random effect was removed from the model. The BC model showed significant variation in school-level outdoor BC slopes, and the fixed effect of outdoor BC showed a stronger average school effect compared to that seen in the $PM_{2.5}$ model (coefficient=0.80 vs. 0.54). The predictions from these models were highly associated with the observed indoor pollutant levels ($PM_{2.5}$: r^2=0.80; BC: r^2=0.75).

Figure 3 illustrates the random effects of school in the $PM_{2.5}$ and BC models. The random intercept for each school reflects the pollutant levels generated within the school. The random slopes of each school represent variation in the relationship between the central site and the indoor environment concentrations, independent of what is accounted for by indoor generation, penetration of the school envelope, or variability in the regional pollutant measured at the central site. Therefore, the slopes capture the variation in the immediate outside pollutant levels influencing the indoor environment.

These analyses suggest the presence of $PM_{2.5}$ sources within the schools, but not for BC. The variability in school-level slopes in the BC model indicates that the site-specific outside BC levels (not measured but inferred from the model) differ from those observed at the central monitor site.

The predicted sulfur ratio was modestly correlated to the observed one (r^2=0.31). However, when it was included in models predicting indoor $PM_{2.5}$ and BC, estimates were well correlated with the observed ones ($PM_{2.5}$: out of sample r^2=0.68; BC: out of sample r^2 =0.62). The relationship between the predictive model for $PM_{2.5}$ and BC is shown in figure 4.

Since satellite-based $PM_{2.5}$ models were only available through 2012 and BC models through 2011, we analyzed156 and 102 matched classroom samples, respectively. Satellite

models produced similar estimates to the sulfur-adjusted central monitor site (table 3), though the out of sample predictions were less accurate ($PM_{2.5}$ r^2=0.57; BC r^2=0.40).

Discussion

School and classroom exposures to particulate air pollution are likely to have significant health effects in children, since they spend the majority of their time in this environment. However, particle monitoring inside schools is challenging. Understanding the relationship between indoor and outdoor particulate matter is important to assess the origin of exposures. Both indoor generated and ambient PM elicit an inflammatory response in the respiratory system(41). Additionally, given the constraints of conducting large population studies in school classrooms, accurate predictions of indoor pollutants is critical to understanding their health effects.

Here we demonstrate the strong relationship between the measured indoor classroom levels of $PM_{2.5}$ and BC with the corresponding outdoor levels, even with overall low levels of particulate concentrations compared with other studies of similar findings (9, 13, 16, 18). Moreover, mixed effects modeling allows for understanding the relative contributions of indoor sources, local outdoor sources near the schools and regional sources of particle pollution to classroom air pollution. Our study suggests that indoor sources of $PM_{2.5}$ significantly contribute to indoor levels in many schools despite the absence of smoking and cooking on premises. Previous studies suggest that re-suspension of particles already deposited on the floor by classroom activities (8, 9, 21, 42) is an important source, particularly in temperate climates where windows and doors are frequently closed(15). Others have shown that while the majority of indoor $PM_{2.5}$ is generated outdoors, a significant portion may be due to organic compounds and calcium-rich particles from chalk and building deterioration (9, 43, 44), as well as textiles, and outdoor soil components(13). Moreover, the composition of the classroom $PM_{2.5}$ has been found to be significantly different from that of outdoor particles (9) and may contain more toxic (17, 45) and biologically active products(41). Despite this, $PM_{2.5}$ of outdoor origin has been shown to have greater association with health effects in children with asthma (46).

While levels of $PM_{2.5}$ associated with indoor sources varied across schools, this variability was not explained by school characteristics such as building age, furnace, or structural conditions, similar to schools studies from Munich, Germany(16), Belgium(9), and France(23, 24) in which classroom and school characteristics were not found to associate with indoor/outdoor particle concentration ratios. In the current study, samples were collected when school was in session to investigate the effects of normal classroom activity; therefore, resuspension of fine particles is likely to be an important contributor to indoor fine particle levels, a dominant feature noted in the Primequal study of French schools(23, 24).

Indoor $PM_{2.5}$ levels were influenced by both indoor-school and outdoor particle sources. Whereas, the $PM_{2.5}$ in the urban area originates from different sources, BC is more specific to traffic-related particles and thus exhibits a greater spatial variability in the city landscape (34, 47, 48). Indoor BC had negligible indoor source emissions and its concentrations were correlated with those measured at the central monitor supersite. This finding is consistent with our assumptions that, in the absence of cooking and tobacco smoke on school grounds,

BC concentrations would be a direct reflection of traffic pollutants infiltrating the classroom. Black carbon, along with gaseous byproducts of traffic, has been shown to easily penetrate schools and classrooms in urban areas (43).

We found little variability in the infiltration of outdoor particles across the school year as measured by sulfur tracer method. The sulfur ratio was consistently less than unity across classrooms with few exceptions, indicative of the lack or absence of indoor sources emitting sulfur, which is consistent with findings from previous studies (37, 49). Building conditions, ventilation, and seasonal influences have been implicated in the penetration of sulfur to the indoor environment (50, 51). We found ambient temperature, time, and the school random intercept to be significant predictors of the sulfur ratio, confirming the importance of seasonal and school building envelope attributes in $PM_{2.5}$. Specific classroom characteristics, including the physical condition, total number of windows, and number of windows facing a drop-off area, were not predictors of sulfur indoor/outdoor ratio.

Moreover, using the sulfur tracer method, we were able to capitalize on this close relationship of indoor to outdoor levels of $PM_{2.5}$ and BC to predict $PM_{2.5}$ and BC classroom levels on days when no particle samples were available. Our cross-validation model provided good estimates of predicted indoor concentrations (PM : out of sample $r^2_{2.5}$ =0.68; BC: out of sample r^2=0.62). These correspond to correlations between held out and modeled values of 0.79-0.82. Model performance was superior to those of indoor-home models due to the low contribution of the indoor sources (52). In a limited subgroup of schools based on available data, we found that satellite-based $PM_{2.5}$ and BC estimates to be similar, though overall less accurate in predicting indoor levels than the central monitor-based approach. The similarities are not surprising as the satellite estimates rely, in part, on the central monitor measures for calibration. The limited number of samples in the satellite-based analysis may account for part of the increased variance found in these models.

Our data analysis and interpretation of the findings presented here may be limited because of the relatively small number of samples and classroom time-activity data collected for each indoor sampling period. Additionally, weekday and weekend periods may have been included in the same sampling period. While our findings clearly show strong indoor-outdoor correlations for $PM_{2.5}$ and BC, the predictive model would likely be enhanced by greater quantity of indoor measures and discrimination of classroom activities. This belies the difficulty in population-based sampling in the inner-city school classroom where considerations of space, interference with educational activities, and sampling cost limit access. Despite these difficulties, our models were able to predict indoor $PM_{2.5}$ and BC with good accuracy. Finally, the lack of cooking, geographic characteristics, and local weather patterns affecting this school district may limit the generalizability of these findings to other school classroom based cohorts; however, the purpose here was to predict exposures within the classrooms measured at least once. The methodology may serve as a framework to determine the relative outdoor contributions to indoor pollution more generally.

Despite extensive study of the indoor and ambient pollution health effects at the home(53), few published studies have explored the health effects of classroom/school specific pollution in children with asthma(54-56). We demonstrate that outdoor $PM_{2.5}$ and BC concentrations

can be used to predict indoor concentrations in the unique microenvironment of the school classroom. These models may contribute to more robust health effects analyses in future studies.

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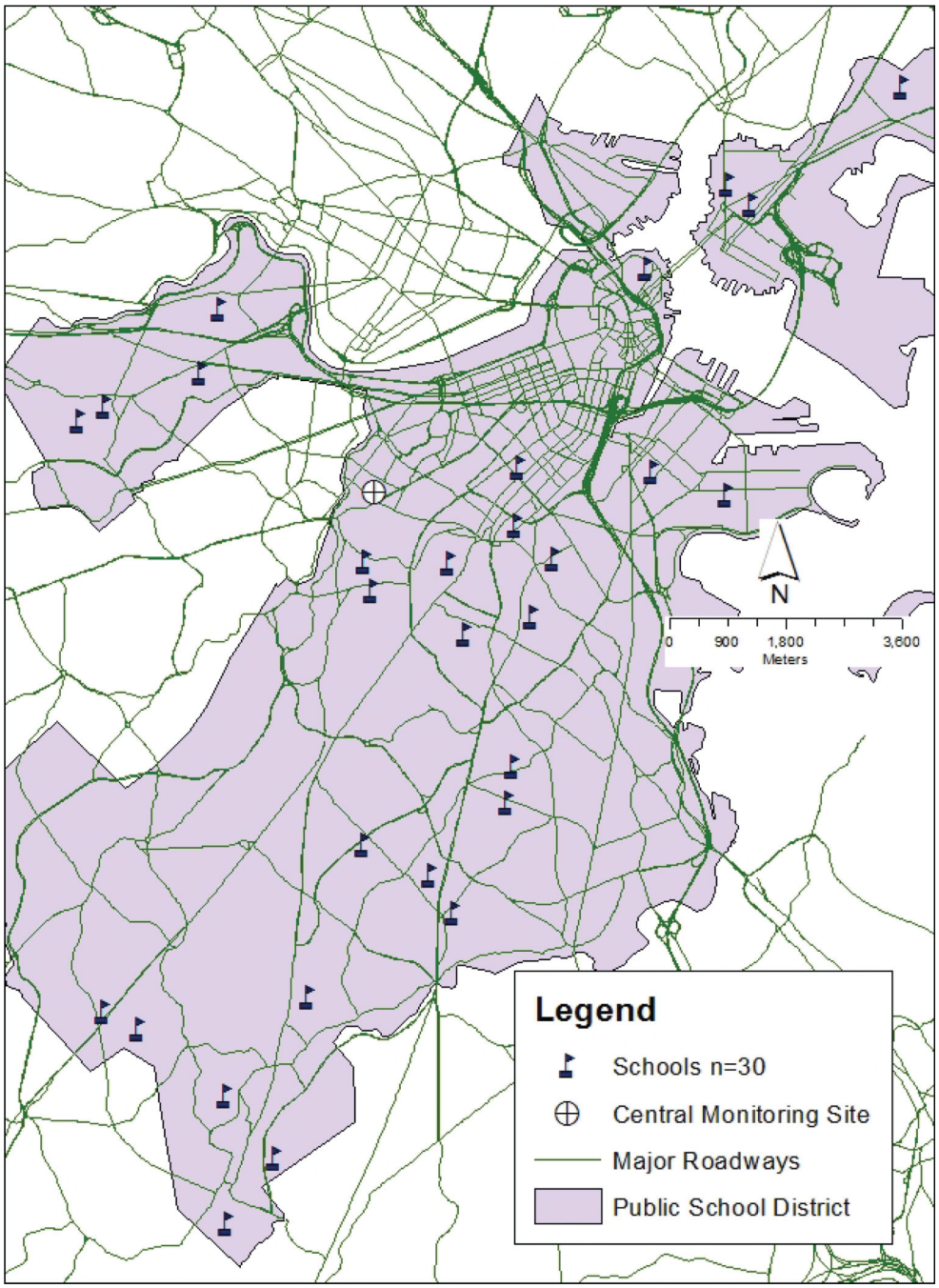

Figure 1.
Location of schools and central monitor

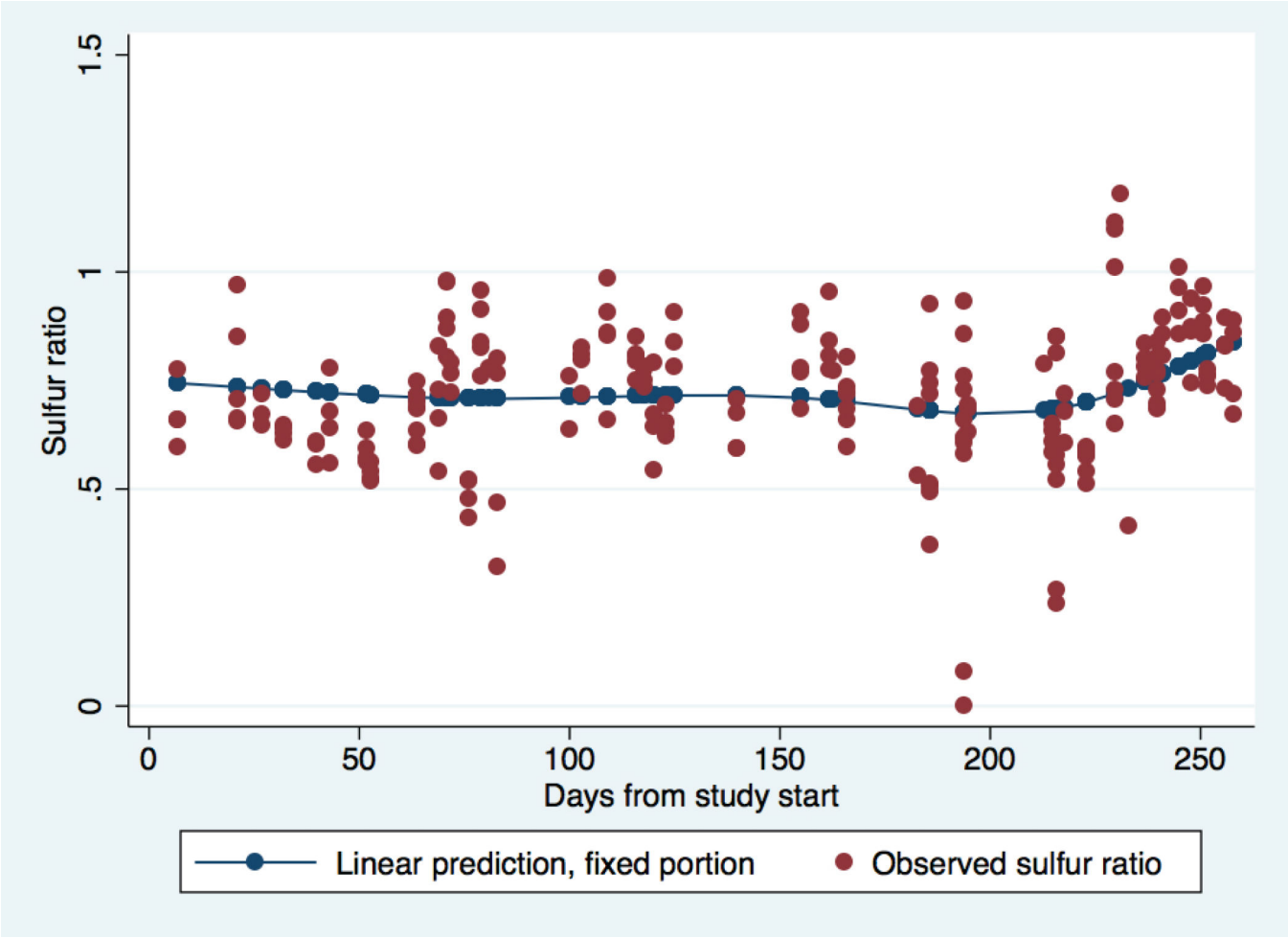

Figure 2. Variation in sulfur ratio over time

Sulfur ratio for each classroom sampled plotted by days starting October 1st. Each point represents the observed ratio of classroom sulfur/outdoor sulfur.

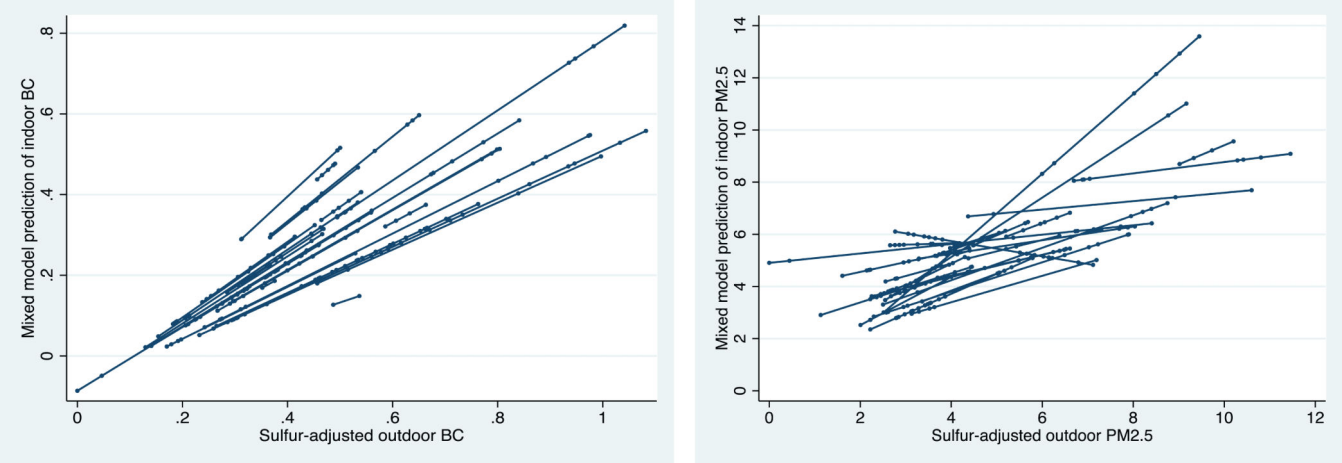

Figure 3. Random Effects of School on $PM_{2.5}$ and Black Carbon

Left Panel: Plot of the predicted indoor PM2.5 levels by the outdoor source contribution. Each point represents a classroom measure; each line represents a school's linear prediction that incorporates its school-specific random intercept and random slope. Right Panel: Plot of the predicted indoor BC levels by the outdoor source contribution. Each point represents a classroom measure; each line represents a school's linear prediction that incorporates its school-specific random slope (random intercept not included due to little school-level variation).

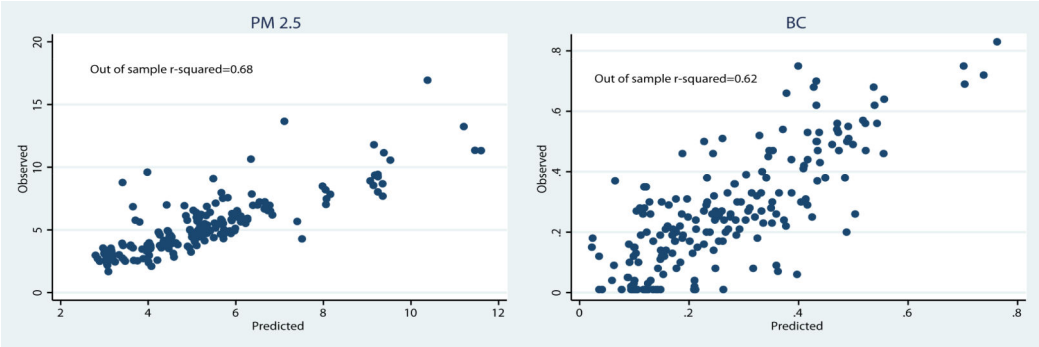

Figure 4. Out-of-sample correlations of predicted and measured $PM_{2.5}$ and BC by classroom
Left panel: Plot of predicted versus out of sample observed classroom $PM_{2.5}$. Right panel: Plot of predicted versus out of sample observed classroom BC.

Table 1

Selected Characteristics of Schools/Classrooms

SCHOOLS		**CLASSROOMS**	
Characteristic	**No. (%)**	**Characteristic**	**No. (%)**
Year Built		Flooring	
1900-1949	15 (50)	Carpet	3 (2)
1950-1969	12 (40)	Linoleum/Tile	93 (68)
Gas Cooking	1 (3)	Rugs	109 (80)
Furnace age		Classroom Level	
≤ 10 years	13 (43)	Basement	3(2)
>10 years	9 (30)	1st floor	3 (2)
Service/12 month	27 (90)	2nd floor	59 (43)
		≥ 3rd floor	71 (52)
		Windows open	127 (96)
		Faces drop-off	40 (31)

Table 2

Description of Particulate Exposures, by Site

Pollutant	Classroom[a]	Central Site[a]
$PM_{2.5}$, $\mu g/m^3$	5.3 (2.3)	6.5 (2.7)
BC, $\mu g/m^3$	0.27 (0.19)	0.64 (0.25)
Sulfur, $\mu g/m^3$	0.36 (0.18)	0.50 (0.22)
S_{in}/S_{out} [b]	0.72 (0.14)	

[a] Values expressed as mean (standard deviation).

[b] S_{in}/S_{out}: ratio of indoor to outdoor sulfur levels

Table 3

Results obtained from the mixed-effects models used to predict indoor $PM_{2.5}$ and BC concentrations

	$PM_{2.5}$				BC			
	Central site		Satellite		Central site		Satellite	
Fixed effects	**Coefficient**	**Standard error**	**Coefficient**	**Standard error**	**Coefficient**	**Standard error**	**Coefficient**	**Standard error**
Intercept	2.78	0.56	2.42	0.69	−0.09	0.02	0.09	0.04
outdoor source contribution	0.54	0.11	0.49	0.12	0.80	0.06	0.73	0.22

Random effects	**Variance estimate**	**Standard error**			**Variance estimate**	**Standard error**		
Intercept	5.39	2.20	5.70	3.31	--	--	--	--
outdoor source contribution	0.20	0.08	0.20	0.10	0.04	0.01	0.29	0.13
Covariance	−0.94	0.39	−0.97	0.56	--	--	--	--