Aerosol and Air Quality Research

Residential Indoor and Outdoor PM Measured Using Low-cost Monitors during the Heating Season in Monroe County, NY

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ABSTRACT

Continuous 1-minute indoor and outdoor PM concentrations (~$PM_{2.5}$) were measured from November through April of 2015/16 and 2016/17 at 50 single family residences in Monroe County, NY (25 per season) using Speck (Airviz Inc., Pittsburgh, PA) low-cost monitors (LCMs). While the accuracy of LCMs is inconsistent and source dependent, the LCMs provided reasonable precision for estimating indoor/outdoor (I/O) ratios based on laboratory and field testing, understanding the relationship between indoor sources and concentration, and comparing PM concentrations across residences for the detected size range (0.5–3 μm). The indoor $PM_{2.5}$ concentration pattern showed clear morning and evening peaks as well as higher indoor concentrations during the weekends when people are typically at home. The mean I/O $PM_{2.5}$ ratio was 1.1 for all homes and increased to 1.7 when a combustion source was in use as indicated by an elevated CO concentration whereas most prior studies have found this ratio to be < 1. Increases in wood-burning appliance temperature and indoor CO concentrations were found to be associated with an overall moderate (mean value of 2.1 μg m^{-3}) increase in indoor PM concentration averaged over the heating season. Short-term PM increases greater than 100 μg m^{-3} were periodically observed in homes with and without wood-burning appliances operating. This study provides an approach for exposure assessment in homes that can be utilized by employing appropriate calibration and quality assurance procedures for the LCMs.

Keywords: Residential PM, Indoor aerosol, Indoor-outdoor relationship, Low-cost monitors, Wood-burning appliance

1 INTRODUCTION

Indoor particulate pollution can arise from multiple sources including the penetration of the ambient aerosol into indoor spaces (Chen and Zhao, 2021) and a variety of indoor particle sources such as cooking, cleaning, and indoor combustion sources (Isaxon *et al.*, 2015). Ilacqua *et al.* (2022) reviewed the literature on indoor/outdoor ratios for various particulate matter (PM) sizes.
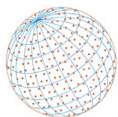
Liu and Zhang (2019) have extensively reviewed the literature on the relationships between outdoor and indoor PM$_{2.5}$ concentrations and constituents and report that outdoor PM$_{2.5}$ concentrations are generally higher than indoor values. Of the 40 studies they reviewed, only 14 had indoor/outdoor ratios $\geq$ 1.0. Thus, for most people, the indoor environment produces a lower exposure to PM$_{2.5}$ than does the ambient environment.

Since most people spend most of their time indoors (Klepeis et al., 2001; Duan et al., 2021), the building envelope provides mitigation from exposure to high ambient concentrations. Alternatively, intake fractions (the mass fraction of an emitted pollutant that is breathed by a person) is approximately 1000 times higher for indoor sources than for outdoor sources (Lai et al., 2000). Thus, depending on the source, indoor emissions may be more important than the penetration of ambient PM when assessing inhalation exposure.
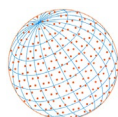
Residential solid fuel combustion is a major source of airborne PM and related pollutants during the heating season in many cold climate communities (Li et al., 2019a, 2019b). Even in the United States, residential wood combustion can be a significant PM$_{2.5}$ source. For example, Wang et al. (2012a, 2012b) found that emissions from wood combustion represent up to 30% of the ambient winter-time PM$_{2.5}$ concentrations in Rochester, NY, even though relatively few residents rely on wood combustion as their primary heat source (NYSERDA, 2016). Exposure to emissions from wood product burning appliances can arise in several ways depending on the source of the smoke. If people have wood burning appliances in their homes, there is likely to be leakage from the appliance, giving rise to direct exposure to the combustion emissions indoors. For all residents in a community where wood products are combusted, there will be exposures from the wood smoke in the ambient atmosphere both when the person is outdoors and through infiltration of ambient air into their homes.

Gardner et al. (2014) demonstrated that acute myocardial infarctions (MI) can be triggered by short-term increases in ambient PM$_{2.5}$ concentrations in the previous hour. However, Evans et al. (2017) did not observe increased odds of MIs associated with increases in markers of wood smoke. However, Assibey-Mensah et al. (2019) found that exposure or a marker of wood smoke during the 7th gestational month was associated with increased odds of hypertensive disorders of pregnancy. Subsequently, Assibey-Mensah et al. (2020) found wood smoke exposures during the first gestational month were also associated with increased odds of early-onset preeclampsia. However, to assess the effects of wood smoke on human health and inform policy on residential biomass burning, the patterns of exposure must be better understood. By improving analytical tools, we can obtain better estimates of short-term, spatially resolved exposure for health-related studies.

Low-cost monitors (LCMs) for air quality monitoring have proliferated on the market. These monitors allow inexpensive, simultaneous measurement of air pollutants at multiple locations. However, the accuracy of the sensors used in the LCMs is relatively low compared with scientific-grade air quality instrumentation. Thus, they require careful calibration and post-processing of the data (Manikonda et al., 2016; Zíková et al., 2017a). Recent studies have used LCMs to assess temporal and spatial PM levels for exposure analysis (Mousavi and Wu, 2021) as well as to separate contributions of indoor PM sources and infiltration of outdoor PM into occupied buildings (Bi et al., 2022; Krebs et al., 2021).

A landmark study conducted in 2005 and 2006 in Winsor, Canada reported PM$_{2.5}$, black carbon (BC) and ultrafine particles (UFP) measured indoors and outdoors at 48 homes, 5 days per home, in winter and summer using portable, but not low-cost, monitors (Wallace et al., 2011). Those investigators found that mean outdoor concentrations for all three pollutants were higher outdoors than indoors, and that indoor sources were relatively more important for UFP than for PM$_{2.5}$ and BC. They found that the concentrations for UFP and PM$_{2.5}$ increased significantly between 5 and 7 pm, consistent with times of peak cooking activities. Knowing the patterns of occupancy and activity and how they impact indoor pollutant levels can contribute to exposure assessment as well as mitigation strategies. With LCMs, this type of study can be conducted for lower cost and across more communities.

For this project, we examined this spatial-temporal exposure relationships by measuring PM concentrations with LCMs indoors and outdoors at 50 residences during two heating seasons. Outdoor data from this project were previously used to assess the spatial and temporal variation of PM concentrations (Zíková et al., 2017a) and to estimate the hourly small-scale variability of

PM using land-use regression models (Masiol *et al.*, 2018). This study provides an assessment of the application of LCMs for human exposure assessment through evaluation of 1) patterns of indoor residential versus outdoor PM$_{2.5}$ concentrations, and 2) responses to both combustion and non-combustion sources indoors. Although the accuracy of the sensors is limited, with careful calibration, the relative concentration comparisons provide a means to assess health effects for indoor exposures for larger populations than has previously been possible.

2 MATERIALS AND METHODS

2.1 Study Location

This study was performed in Monroe County, New York, U.S., which lies on the southern shore of Lake Ontario and includes the City of Rochester (~210,600 inhabitants, 2020 Census). Monroe County is the center of the greater Rochester metropolitan area (~1.1 million inhabitants), which encompasses several surrounding counties and is the third largest metropolitan area in New York State. Local emissions are dominated by typical urban emissions including traffic and building space heating with few industrial sources. Road traffic across the metropolitan area include highways and secondary and local roads. Natural gas and bottled liquid petroleum gas (LPG) are largely used for domestic and commercial heating with some No. 2 oil (distillate oils, after July 1, 2013 must be ultralow sulfur, < 15 ppm) burned in the suburban areas. Residential wood combustion is used primarily for ambiance, but it provides primary space heating for about 2000 dwellings (NYSERDA, 2016). Off-road transport (diesel rail, shipping, airport) are other local PM sources.

A source apportionment study (Squizzato *et al.*, 2018) reported that in 2009/2016, Rochester was affected by 8 main PM$_{2.5}$ sources (in µg m^{-3}): secondary sulfate (2.1 ± 2.5), secondary nitrate (1.5 ± 2.3), spark-ignition vehicles (1.4 ± 1.6), diesel (0.8 ± 0.6), road dust (0.1 ± 0.1), biomass burning (0.6 ± 0.5), pyrolyzed organic-rich (0.6 ± 0.6), and road salt (0.1 ± 0.2). In addition, regional advection of polluted air masses from Toronto (ON), Buffalo (NY), the Ohio River Valley, and the eastern coast of the U.S. may also affect the air quality in this region (Emami *et al.*, 2018; Masiol *et al.*, 2019).

2.2 Indoor/Outdoor Monitoring with Low-Cost Sensors

Continuous 1-minute indoor and outdoor PM and indoor CO concentrations were measured from November through March of 2015/16 and 2016/17 at 50 residences across Monroe County (25 residential locations in each of the two heating seasons, Fig. 1) using LCMs. Inclusion criteria for the study were homes that had wood burning appliances or homes for which the residents frequently smelled wood smoke in the vicinity of their homes. Thirteen homes used the wood-burning appliance as their primary heating source. Table S1 provides the information on the homes included for this study.

LCMs available for measurement of PM are physically small and light, have low power demands, and require little or much less handling and maintenance compared to scientific-grade instruments. Consequently, a large number of sampling points can be monitored over wide areas to better elucidate the spatial variation of the air pollutants. For this study, the third generation Speck monitor (Airviz Inc., Carnegie Mellon University, Pittsburgh, PA) was selected after laboratory comparisons of several available PM LCMs using both generated tobacco smoke and aerosolized Arizona test dust (Manikonda *et al.*, 2016). The Speck uses an infra-red LED-based Samyoung (South Korea) DSM501A optical dust sensor (nominal size range 0.5 to 3 µm). Like other optical-based PM instruments, the response is dependent on the aerosol light-scattering characteristics. Because the intended target aerosol included wood combustion aerosol with a submicron size distribution (Chandrasekaran *et al.*, 2011, 2013; Wang *et al.*, 2019), the Speck LCMs were tested and calibrated using mainstream cigarette aerosol (Manikonda *et al.*, 2016) to ensure they could provide the required data. While PM$_{2.5}$ mass concentrations were not measured directly, the optical dust sensor size range (~0.5 to 3 µm) is roughly comparable with PM$_{2.5}$ given the calibration with research-grade instruments and are reported here as PM$_{2.5}$.

CO was measured using data loggers with electrochemical sensors (EL-USB-CO, Lascar Electronics, Erie, PA). During the second heating season, a thermocouple with a datalogger was attached to the wood burning appliance to record when the appliance was in use.
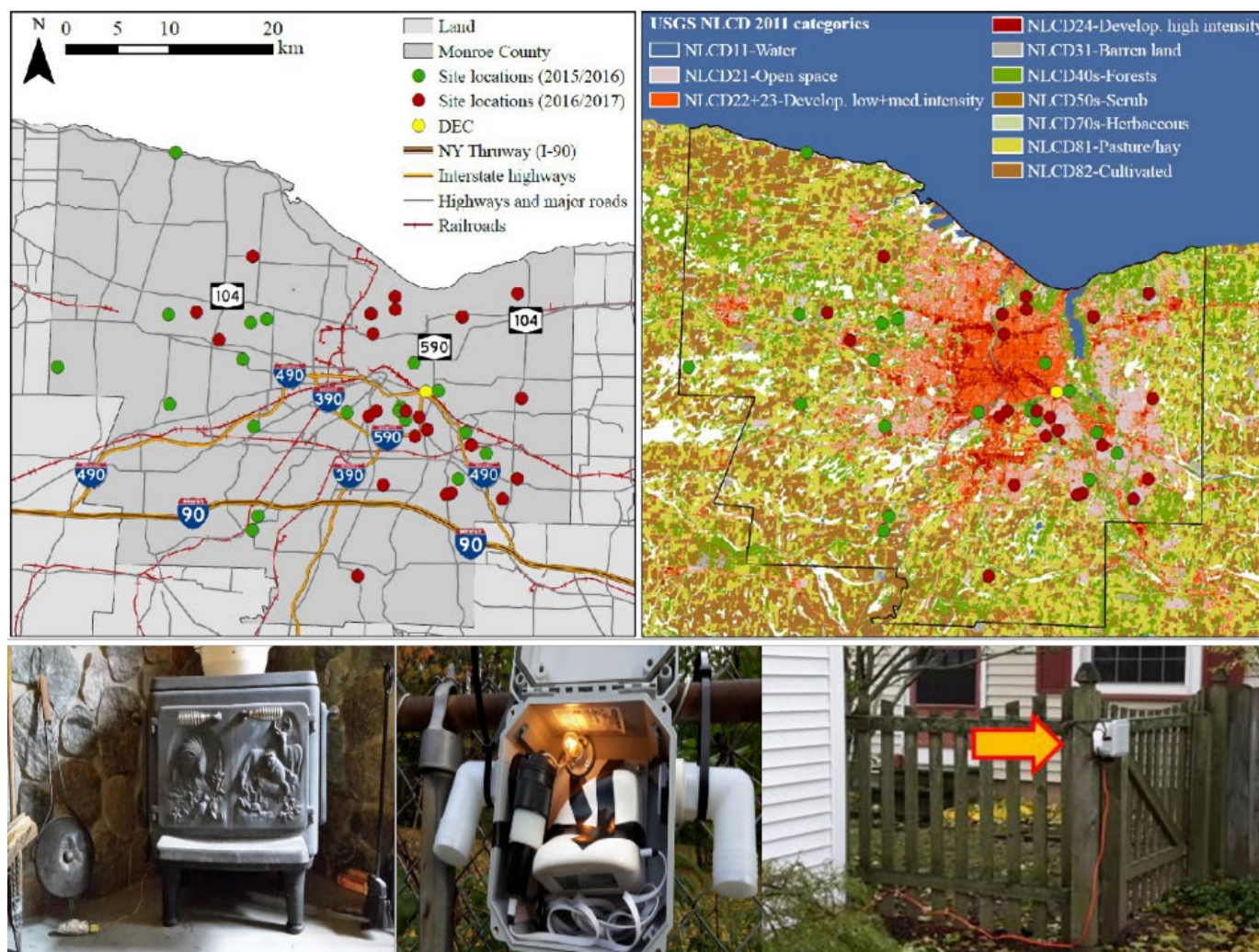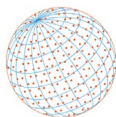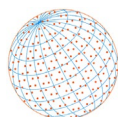
Fig. 1. Locations of Monroe County residences used for indoor/outdoor monitoring for 2015/16 (green) and 2016/17 (red) heating seasons (top). Example of thermocouple placement on woodstove (bottom left), Speck LCM in weatherproof enclosure (bottom middle), and example outdoor location of LCM in weatherproof enclosure (bottom right).

The indoor PM and CO LCMs were placed in the main living area of the home. The outdoor LCMs were placed outside the homes, generally in their backyards as described by Zíková *et al.* (2017a). The outdoor LCMs were mounted in waterproof plastic-fiberglass boxes with two 90° bent inlets (2 cm in diameter) for air exchange and a 6 W bulb to prevent freezing. An additional LCM was placed at the New York State Department of Environmental Conservation monitoring site in Rochester, NY (U.S. EPA site code 36-055-1007; 43°08'46"N, 77°32'52"W). PM concentrations were recorded every minute. However, hourly averages were used for all of the results presented below.

2.3 Quality Assurance for Low-Cost PM Monitor Data

Co-locations experiments were conducted to characterize the LCMs as well as to provide correction factors for the field study data (Zíková *et al.*, 2017a). The quality of the data was also assessed after the LCMs were deployed for the monitoring study (Zíková *et al.*, 2017a) using periodic colocation studies. Preliminary tests reported stable responses over wide ranges of particle concentrations (0–1000 µg m⁻³) and high repeatability of results (Manikonda *et al.*, 2016). Lab and field tests showed these units had a nominal limit of detection of 10 µg m⁻³ and large biases. For individual monitors, bias values varied from 40% to 400% for indoor data with a mean value of 190 ± 120% for hourly averages while outdoor bias values ranged from 150–1100% and an average bias for hourly data of 440 ± 260% (see Fig. 4 in Zíková *et al.* (2017b) for individual

performance values). However, they showed good reproducibility and overall precisions of $8.8 \pm 1.7\%$ indoors and $12.2 \pm 6.2\%$ outdoors (Zíková *et al.*, 2017b). Thus, the Speck values were corrected using the ratio of the average of the ratio of the $PM_{2.5}$ concentration measured by a GRIMM 1.109 aerosol spectrometer (Douglasville, GA, USA) to the concentration for each Speck LCM during the multi-day collocation periods (Zíková *et al.*, 2017a). The GRIMM 1.109 aerosol spectrometer is an optical-based detects aerosols in a size range of 0.25–32 μm in 31 size channels. It has strong correlation with the GRIMM EDM-180 spectrometer (Wang *et al.*, 2020), which is a United States Environmental Protection Administration designated Federal Equivalent Method (FEM) for measuring $PM_{2.5}$ (https://www.epa.gov/amtic/air-monitoring-methods-criteria-pollutants).

2.4 Participant Surveys and Activity Diaries

A participant survey tool was developed to provide information regarding the activity patterns and sources of air pollutants in the residences. After assessing the 2015/16 survey data, additional topics were introduced or expanded upon for the 2016–17 participant survey tool, including information on ventilation, the presence of pets, air fresheners, and humidifiers, the frequency of vacuuming, and the typical cooking methods used in the home. A full version of the survey with the additional questions for the 2016–17 heating season (highlighted) can be found in the supplemental material. A one-week diary was also included for 15 participants during the 2016–17 heating season. The purpose was to establish and better define weekly habits of different participants, e.g., times in or out of the home, when the stove may be on, or when cooking occurs.

To assess the indoor personal exposure to PM due to wood smoke, different contributing sources were identified within the data time series and separated from the background indoor concentration during low activity times (e.g., early morning), when other indoor sources were limited.

To complete time comparisons, all outdoor and indoor data points provided by the Speck monitors (in one-minute intervals) were examined for missing points. Data completeness varied between monitors and location. To reduce sensor noise, hourly averages were calculated for all monitors. Data for the CO monitors and thermocouples were also averaged to hourly values. Comparisons were between background levels when homeowners are not present compared to high activity times (evenings when homeowners are present) to determine the contribution of indoor sources. The low activity times denoting a background concentration were selected as 11 am to 1 pm during the weekdays. High activity times were selected as 6 pm to 8 pm, also during the weekdays.

Statistical analyses were applied to evaluate the data distributions and significance in subset differences. Using the Kolmogorov Smirnov test, normality was rejected at the 0.05 significance level for all datasets. Accordingly, Wilcoxon rank sum tests (Wilcoxon, 1945) were used to determine whether there are significant differences between the indoor and outdoor values at the 0.05 level for each measured home and between data with or without burning activities pooled for all homes.

3 RESULTS

3.1 Indoor/Outdoor Ratio

The ratio of the hourly averaged indoor $PM_{2.5}$ concentration to the hourly averaged outdoor $PM_{2.5}$ concentration (I/O ratio) for all the homes as measured by the LCMs are presented in Fig. 2. For the 2015/16 heating season, the I/O ratio was greater than 1.0 for 38 % of homes (overall mean I/O ratio = 1.3), and increased substantially (overall mean I/O ratio = 1.6) when a combustion source was present indoors as indicated by measured CO > 0 ppm. The results were similar for the 2016/17 heating season, with the overall mean I/O ratio of 1.1, increasing to 1.7 when a combustion source was present. The mean indoor and outdoor concentrations and I/O ratios and their associated standard deviations for each subject house are provided in Table S2. Average $PM_{2.5}$ concentrations were ~70% higher than the outdoor $PM_{2.5}$ concentrations due to the presence of indoor PM sources. Concentration peaks were associated with both combustion sources (e.g., wood burning appliances, cooking, candles) and non-combustion sources (e.g., particles resuspended from human activity like cleaning) as shown for one representative participant (Fig. 3).
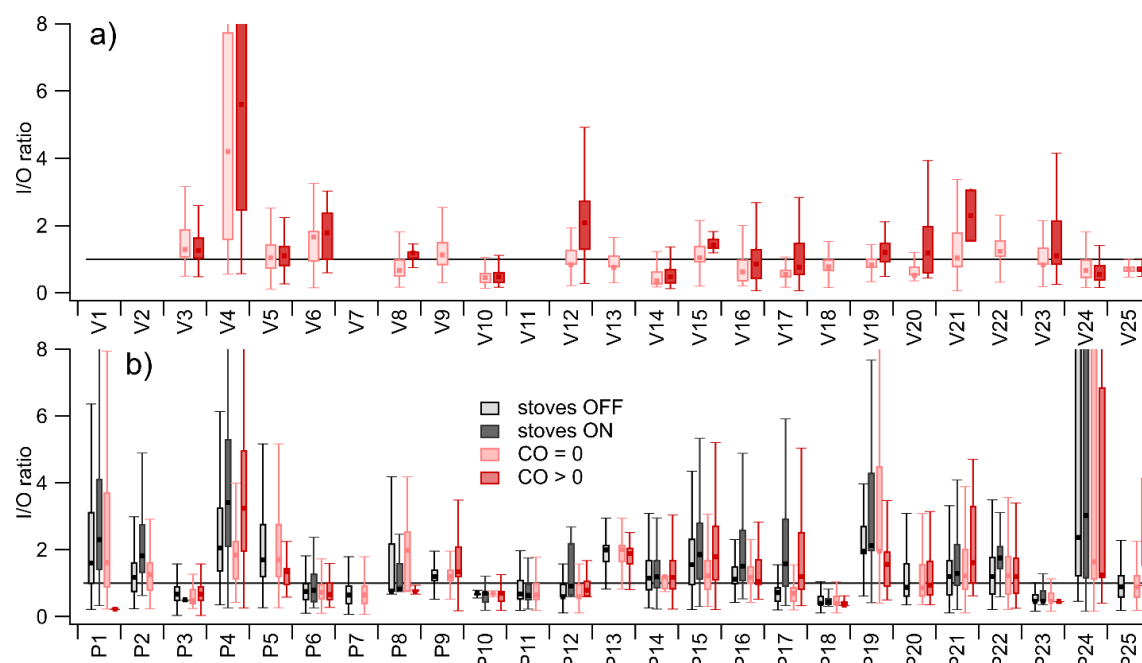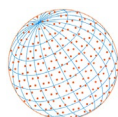
Fig. 2. Mean indoor/outdoor (I/O) PM$_{2.5}$ concentration ratio with no combustion sources present (CO = 0 ppm) and during combustion periods (CO > 0 ppm) during the a) 2015/16 heating season and b) 2016/17 heating season. The 2016/17 heating season also includes the I/O ratio for when the wood appliances (stoves) were ON versus OFF, as indicated by the thermocouple attached to the appliance.
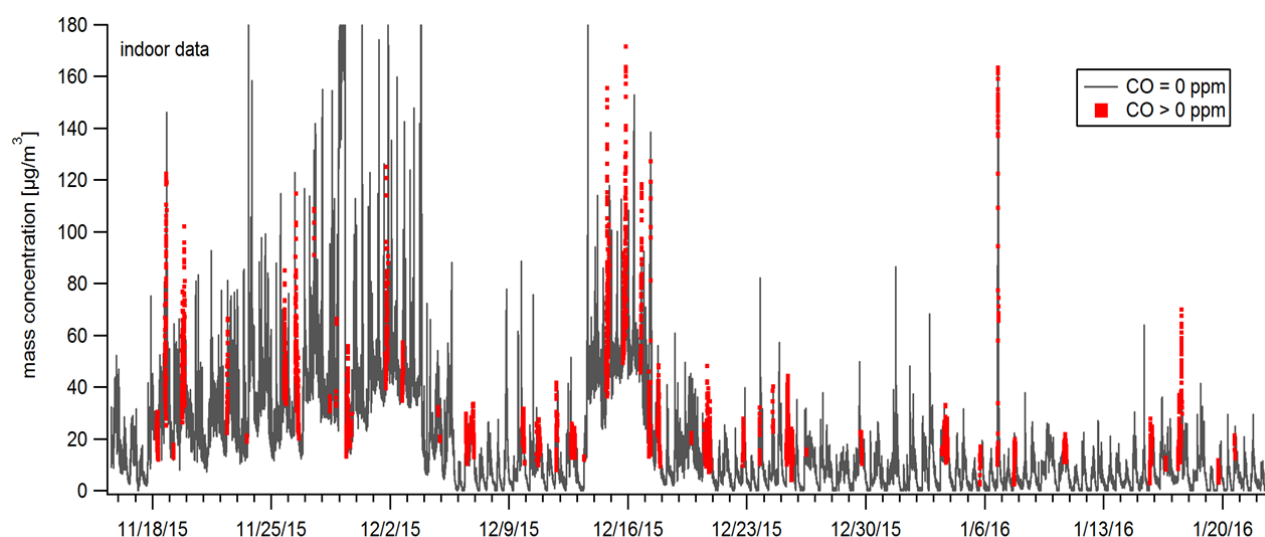

Fig. 3. Example PM$_{2.5}$ concentration time series from one home for which red indicates measured CO > 0 ppm, indicating the presence of a combustion source. Many peaks are not associated with combustion sources.

Short-term PM concentration increases to greater than 100 μg m^{-3} were observed in homes with and without wood-burning appliances operating. For a few participants, CO concentrations were substantially elevated indicating the presence of a combustion source that was not operating properly or was not properly vented. These participants were provided guidance to address this issue.

A comparison of mean indoor and outdoor concentrations as measured by LCMs when the combustion appliance was operating is shown in Fig. 4. Increases in wood-burning appliance temperature and indoor carbon monoxide (CO) concentrations were found to be associated with an overall moderate (0.8 μg m^{-3}) increase in indoor PM$_{2.5}$ concentration averaged over the
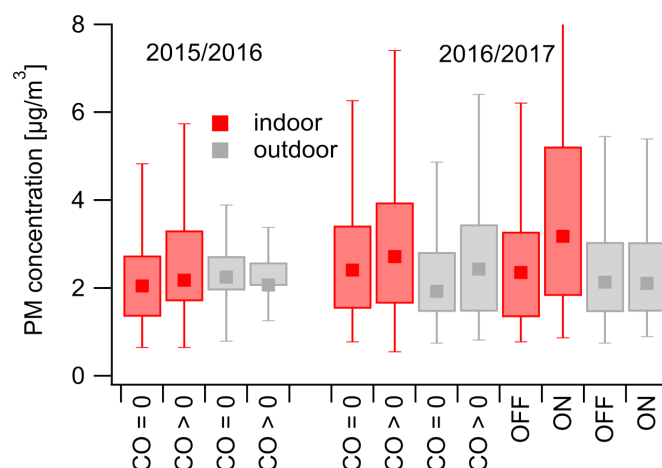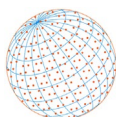
Fig. 4. Indoor and outdoor concentrations, as measured by LCM. Concentrations are statistically greater during times of combustion.

heating season. The largest difference (1.8 μg m^{-3}) occurred when the wood burning appliance was on versus off. Based on the rank sum test, there were significant differences between the groups at the 0.05 level for all datasets.

3.2 Association of PM$_{2.5}$ with Human Activity Patterns

The daily and weekly profiles of indoor PM$_{2.5}$ concentrations were shaped by human activities. The daily PM$_{2.5}$ concentration pattern (Fig. 5, top left) shows a minimum of PM$_{2.5}$ concentrations during late night/early morning coinciding with an expected minimal indoor and outdoor PM source emissions. In the early morning, an increase of PM$_{2.5}$ concentrations was observed, consistent with the human activities in the morning (e.g., cooking breakfast; resuspending dust via walking). The concentrations remain reasonably stable until 3 pm. In the evening, a second concentration peak (maximum around 6 pm) occurred and coincides with expected increases in indoor sources (cooking dinner, possibly burning wood products).

The links between the human activities and the indoor PM$_{2.5}$ concentrations is also evident by looking at the weekly cycle (Fig. 5, top right) and weekday-hour pattern (Fig. 5, bottom). The higher weekly concentrations were recorded during the weekends, when people were at home. The hourly pattern during the weekdays shows a clear difference between the morning and evening peaks, which was attributed to the absence of most participants during the weekday lunch period. Alternatively, the hourly patterns during Saturdays and Sundays were different from the midweek patterns and show a unique peak that coincides with increased human activities in the home. The PM concentrations start to increase 1–3 hours later in the day during the weekend days compared to the weekdays, consistent with a later wakeup time on the weekends.

The daily and weekly profiles of outdoor (ambient) PM concentrations are shaped by both human activities and meteorological conditions. The daily cycle (Fig. 6, top left) shows a minimum for PM concentration during late night/early morning, coinciding with the expected minimal traffic and wood smoke sources overnight. In the morning, some increase in PM concentration is evident, consistent with expected increases in human related sources, such as traffic and heating. The concentrations peak in the evening, again coinciding with expected increases in traffic emissions during the afternoon rush hour. Given that most homes are heated with natural gas, local residential heating would not be expected to be a significant PM source throughout the day. Thus, vehicle traffic was the likely major driver of the observed diel variations. Most of the homes were distant from major highways, so the PM daily pattern (Fig. 6) was less clearly defined than the main road traffic pattern (Fig. 7)

Comparison of PM$_{2.5}$ ranges in low activity versus high activity times shows that high activity times (6 pm to 8 pm weekdays) typically see higher median, mean, upper bound values than low activity times (11 am to 1 pm, weekdays) (Fig. S1). The differences in concentration indicate that important consideration must be given to additional indoor PM sources present during the high
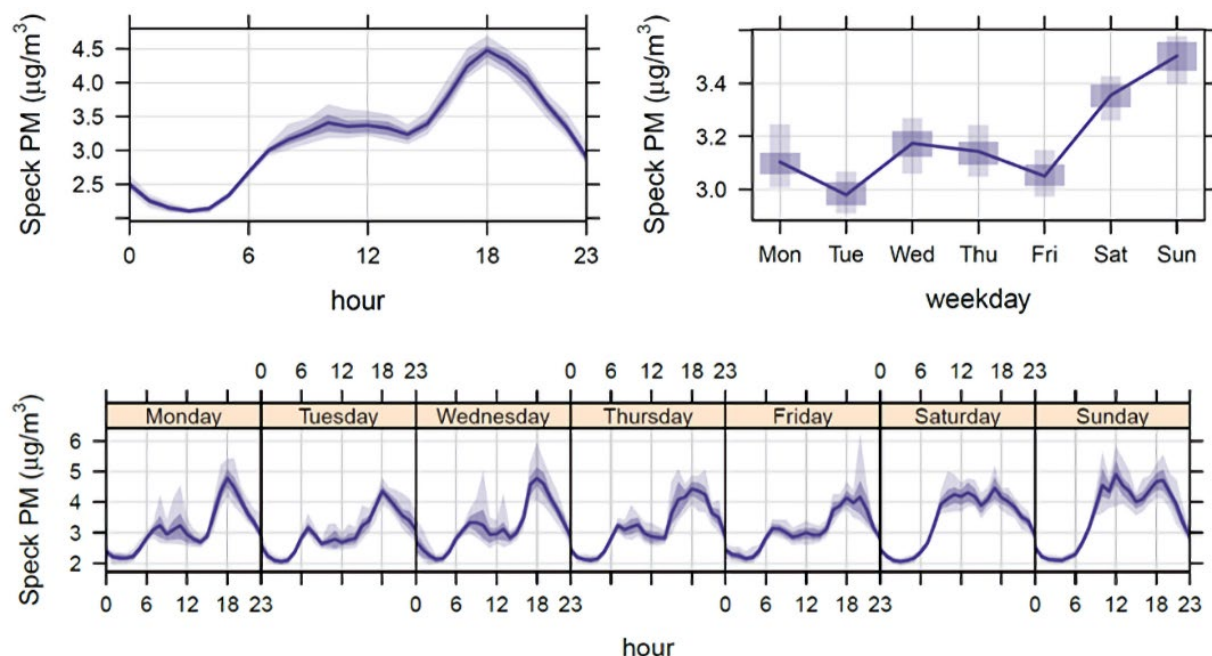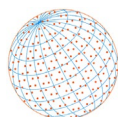
Fig. 5. Average daily (top left) and weekly (top right) indoor PM concentrations at individual locations included in the 2016/17 heating season. The combined pattern is reported in the bottom plot. The blue line represents the average concentration (all sites); the shaded areas represent the 75th and 99th confidence intervals calculated by bootstrapping the data (n = 2000).
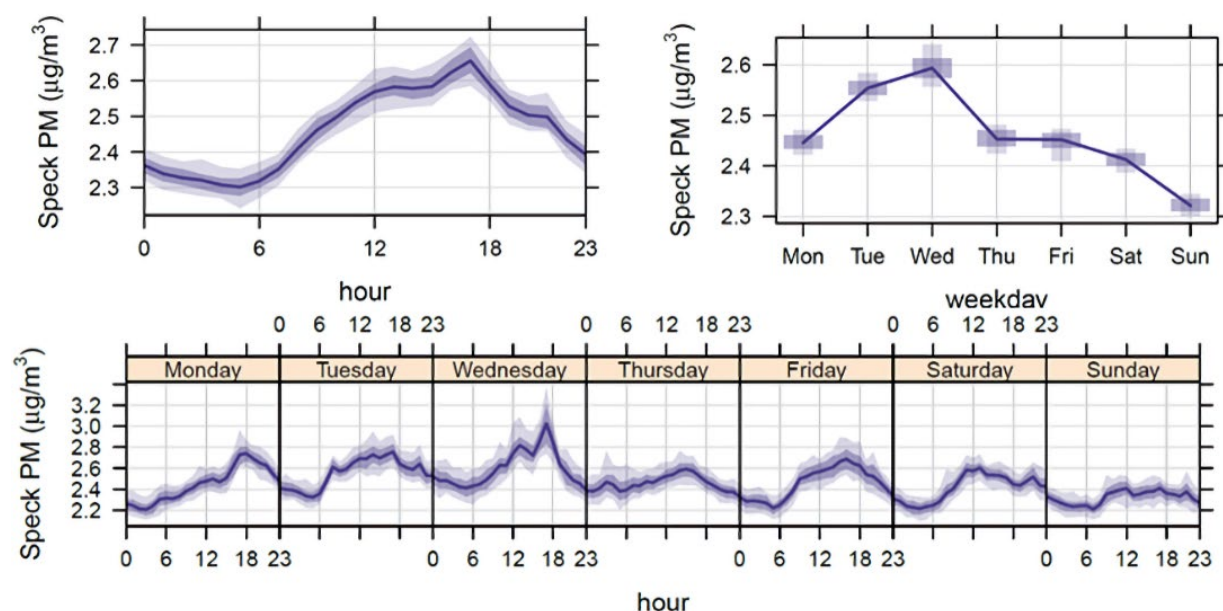

Fig. 6. Average daily (top left) and weekly (top right) outdoor concentrations at individual locations included in the 2016/17 heating season. The combined (daily + weekly) pattern is provided in the bottom plot. The lines and shaded areas are as described in Fig. 5.

activity times. However, these values do not help distinguish among the contributing sources during these periods.

The PM concentration peak in the evening hours was fairly consistent for all participants, but it is difficult to separate specific source contributions during this time. The presence of carbon monoxide is associated with biomass combustion, but other combustion sources may coincide in time. For example, the wood burning device is most likely turned on while other sources of CO are present within the home such as cooking on a gas stove.
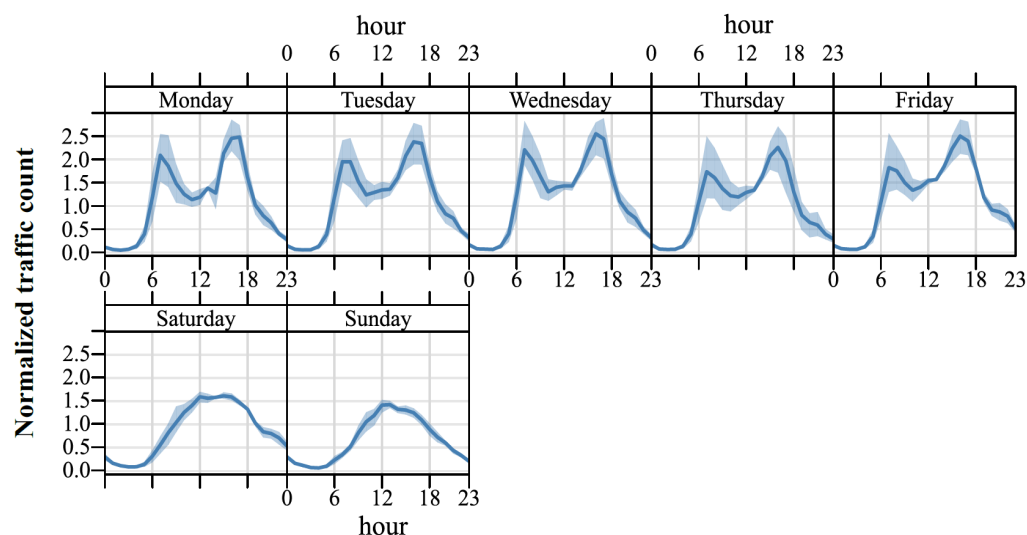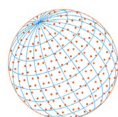
Fig. 7. The normalize traffic count per hour by day of the week based on annual average daily traffic (AADT) provided by the New York State Department of Transportation for a major roadway in Rochester.

To address this potential for simultaneous contributions, additional comparisons were made for the 2016/2017 heating season (when stove temperature data available). Examining the PM mass distributions for high and low activity times, the three additional comparison groups include:

1. High Activity without combustion – determined by the absence of CO and whether or not the stove was on.
2. CO > 0 with stove – determined by appliance temperature.
3. CO > 0 without stove – determined by temperature.

By comparing high activity PM with that of high activity without combustion, the contribution of combustion sources can be determined. Also, the comparison of PM due to CO with and without the stove allows the wood combustion source to be distinguished from other sources of combustion within the home. For example, PM concentrations for participant P01, who used wood as a primary heat source, are shown for these conditions in Fig. 8.

Box and whisker plots for other participants are provided in the supplemental information (Figs. S1 and S2). These comparisons show that there were variations among homes, but that combustion and the presence of a wood burning device (stove on) had an impact on the median PM concentrations. There may be other variables specific to each participant that further affects these differences.
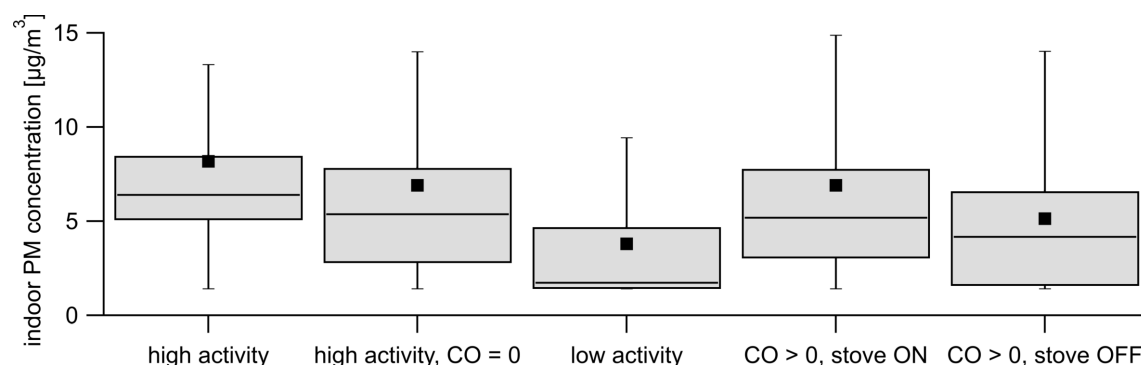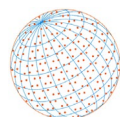

Fig. 8. $PM_{2.5}$ comparison for P01 for high activity (N = 336), high activity no combustion (N = 22), and low activity (N = 336) using the hourly average PM concentration during those periods. The PM concentration for CO > 0 with stove on (N = 2258) and CO > 0 no stove (N = 1267) are for all data when said criteria are met.
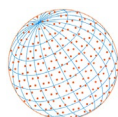
The rank sum test was used to evaluate the differences between two sets of data as to whether or not the difference was statistically significant. The difference between median values for comparable sets are presented and designated as significant if the calculated p-value was less than 0.05 (Table 1). Essentially, the "High activity with combustion vs. high activity without combustion" column in Table 1 show the contribution made by combustion within the home. For some of the participants, there were small differences, or even a negative change. These results indicate that there may be other significant sources of PM within the homes. However, the Sum1 value is positive (0.7 µg m^{-3} increase) and Sum2* is even higher (2.1 µg m^{-3} increase). The "Presence of CO or not" column also provides an estimate for the contribution of combustion sources, but for all times during the monitoring period. "CO with combustion" highlights the potential contribution by the wood burning device within that home. The Sum2* values for these two columns are a 1.0 and 1.1 µg m^{-3} increase, respectively. For only the homes with wood as a primary heat source, the CO presence values increase to 1.3 µg m^{-3}, while the stove contribution decreases slightly to 0.8 µg m^{-3}. Individual increases in PM$_{2.5}$ concentrations up to 5 µg m^{-3} increase can have serious implications regarding personal exposure and the health of individual exposed occupants.

Table 1. Median differences in PM$_{2.5}$ (µg m^{-3}) between subset groups during the 2016–17 heating season. There are two summary values for the median difference presented; "Sum1" represents the average difference in median values for all participants while "Sum2*" represents the average difference in median values only for participants with significantly different sets of data.

ID	Wood burning appliance on or off	High activity vs. all other times	High activity with combustion vs. high activity without combustion	Presence of CO vs. no CO	CO presence with combustion vs. CO presence with no combustion
P01***	0.97*	4.66*	0.06	0.56*	1.01*
P02	0.96*	1.65*	4.42	–	–
P03	0.7	0.63*	–	0.47*	–0.71
P04***	2.25*	0.71*	1.09*	2.7*	1.37*
P05[a]	–	0	–	0.07*	–
P06	0.51*	0.83*	0.13	–0.35*	0.56
P07[b,c]	–	0.7*	–	–	–
P08	0.4*	0	3.01	0	0.4*
P09[b]	–	0.61*	–	0.53*	–
P10***	0	0.14*	0.65	0	0
P11***[c]	0.01	1.86*	0.58	–	0.01
P12	0.98*	0.49*	–0.12	–0.33	–
P13[a]	–	0.27*	–	0.01	–
P14***	–0.03*	0.13	0.81*	0.7*	–0.03*
P15***	–0.71*	2.45*	–0.01	0.04	–0.7
P16	0.15*	0.31*	4.38*	0	5.06*
P17	2.07*	1.42*	0.82	1.33*	1.79*
P18	0.27	0.39*	0.40	0.04	–
P19	0.01	5.6*	–2.9	0	0*
P20[a]	–	1.1*	–	0.33*	–
P21	0.88*	2.78*	0.29	2.3*	–39.3**
P22	0.75*	0.99*	0.23	0.06	0.78*
P23***	0	0.60*	–0.72	0	0
P24	1.74*	–6.7	0.1	1.57*	1.74*
P25[a]	–	1.4*	–	1.86*	–
Sum1	0.7	1.0	0.7	0.7	0.9
Sum2*	0.8	1.4	2.1	1.0	1.3

[a] no episodes with stoves ON, [b] no T data, [c] no zero CO data.
*p value < 0.05, **data omitted, ***uses wood as primary heat source.

4 DISCUSSION

The LCMs used for this study, and most LCMs on the market, have limitations that prevent accurate comparison of the reported concentrations to health-based standards, especially for combustion sources. The concentrations measured by the LCMs at the 50 homes in this study were often lower than the nominal limit of detection of 10 µg m^{-3}, and the outdoor bias-corrected LCM measurements were ~40% of the mean PM$_{2.5}$ concentrations measured by the FEM located within the study geographic area (Zíková *et al.*, 2017a). Also, most combustion particles are in the size range below the nominal 0.5 µm sensor lower limit of detection. A laboratory comparison of the Speck monitors used for this study with a TSI (Shoreview, MN) Aerodynamic Particle Sizer (APS) found the Speck monitors were well correlated (R^2 = 0.96) with the APS but overestimated PM$_{2.5}$ mass for both cigarette smoke and Arizona test dust. However, both the Speck and APS only detect particles greater than ~0.5 µm in diameter. Thus, the PM concentrations produced by combustion sources were likely underestimated in this study, but the concentrations produced by resuspension may be overestimated.

The average (± standard deviation) PM$_{2.5}$ concentrations in 2015/16 and 2016/17 measured by the TEOM-FDMS (FEM method) at the DEC site were 8.0 ± 5.6 and 6.0 ± 4.7 µg m^{-3}, respectively. Thus, the second period mean value was only 74% of the first period average concentration. After the correction of the LCMs using the results of the co-location with the GRIMM spectrometer, a similar decrease in concentrations for the second year was also detected by the LCMs (71%). However, the corrected concentrations measured by the LCMs (average 3.5 ± 0.9 and 2.4 ± 0.4 µg m^{-3} during the first and second campaign, respectively) were consistently lower than the FEM values. The DEC site is located near the intersection of two interstate highways (I490 and I590), a major state highway (NY99), and the mainline of the CSX railroad that runs about 100 m SW of the monitoring site. Thus, it may have been possible to observe lower values in residential neighborhoods away from these major local sources.

This underestimation suggests that there could also be a suboptimal (too strong) bias correction that may be due in part to differences between TEOM-FDMS and the GRIMM. Zíková *et al.* (2017a) found a 55% overestimation of GRIMM PM$_{2.5}$ mass concentration compared with TEOM values at concentrations typically observed in Rochester. This result reconciles the TEOM-LCM differences. A further correction based on the TEOM-FDMS data was possible. Only the empirical bias correction with the GRIMM spectrometer was used to adjust the LCM concentrations because: (i) the analytical method used by the PM sensors (light scattering) is similar to the GRIMM. The TEOM-FDMS uses a different measurement principle (microbalance plus a diffusion drying system); and (ii) the purpose of this work was to identify the intraurban variations for the assessment of health outcomes, i.e., it aims to represent variations of indoor PM pollution at hourly basis rather than estimating "FRM-like" concentrations for regulatory purposes.

Subsequent to this study, improved LCMs have been introduced to the market that report concentrations closer to measured particle mass concentrations for various sources (Singer and Delp, 2018). However, any monitor that uses optical properties of particles as a measurement principle will be limited for measuring aerosol from combustion sources. Promising technologies for LCMs that measured ultrafine particles should remove this limitation in the near future (Ranjan and Dhaniyala, 2009).

5 CONCLUSIONS

The LCMs was able to provide valuable information on the contributions of wood burning devices to PM exposures within typical single-family homes in a northeastern United States metropolitan area. PM$_{2.5}$ concentrations varied throughout the day and week for each home, likely affected by a variety of factors including the type of wood burning device, frequency of device use, and the number of people within the home. Using carbon monoxide detectors as well as thermocouples, the periods of combustion were effectively separated from other times of indoor PM source contribution. Results show that a wood burning device on average contributed approximately 2.1 µg m^{-3} (p < 0.05), with a particle number contribution of approximately 67,000 cm^{-3}. This increase is relevant to investigations on human health impacts due to wood
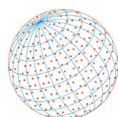
combustion within the home, as there is no threshold to the negative effects of PM exposure. Also, because the Speck does not measure most of the smoke particles (they are outside the optical range of the instrument), the true concentrations may be much larger. Traditional monitoring methods are expensive, and require infrastructure providing constant temperature and humidity for effective mass sampling. Thus, it is imperative to continue to improve the accuracy and sensitivity of low cost semi-continuous $PM_{2.5}$ monitors in defining the impacts of particulate matter on human health.

The results of this study confirm that contemporary wood-burning appliances increase indoor exposures to airborne particles in homes, particularly in the room housing the appliance. This study also provides improved estimated wood smoke exposures at any given time during the heating season for these individuals. Future studies may use this approach for measuring wood smoke exposures in epidemiologic studies.

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