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Infiltration of Black Carbon Particles from Residential Woodsmoke into Nearby Homes

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Abstract

In many communities, residential wood burning is the source of a significant fraction of wintertime PM2.5 and produces exposures to nearby residents inside their homes. To evaluate the magnitude of this effect, black carbon particles were measured as a proxy for woodsmoke indoors and outdoors in a community where residential woodsmoke is the only significant particle source. Thirteen indoor/outdoor measurement pairs were obtained at 4 different residences and showed an average indoor/outdoor concentration ratio of 0.78 ± 0.21 for residences without indoor generation. In addition, a time dependent mass balance model was used in conjunction with aethalometer measurements taken over 16 nights at a single residence to estimate an average air exchange rate of 0.26 ± 0.08 h^{-1}, an average deposition loss rate of 0.08 ± 0.03 h^{-1}, and an average penetration factor of 0.97 ± 0.02. Using a mechanistic approach which utilizes these average values in a steady state model, the predicted average infiltration factor was 0.74 for the residence studied. The high values for both measured I/O ratio and modeled infiltration factor show that residential environments provide inhabitants with relatively little protection from recently generated wood smoke particles.

Keywords

I/O Ratio, Infiltration Factor, Black Carbon, Woodsmoke Particles, Indoor Exposure

1. Introduction

Residential wood burning is a significant source of wintertime particulate matter in many regions. In residential areas, the primary woodsmoke exposure is expected to occur indoors. Not only do many people spend most of their time indoors [1], but during peak periods of woodsmoke generation, evenings and nights, people are more

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likely to be inside their homes. An understanding of the relationship between outdoor woodsmoke concentrations and the corresponding indoor concentration is critical when assessing the impact of woodsmoke on populations. A question of particular importance is whether sheltering indoors effectively reduces a person's exposure to woodsmoke particles.

Although much research has been directed at better understanding the impact that outdoor particles have on concentrations in the indoor environment, the complexity of the processes involved complicates our understanding. The relationship between indoor and outdoor concentrations is influenced by a myriad of factors including home age, construction, and condition; occupant behavior and indoor sources; particle composition and size; reactions in the indoor environment; meteorology; and indoor loss rates. A common metric for describing this relationship is the indoor/outdoor ratio (I/O ratio). A more specific metric is the indoor concentration of particles of outdoor origin, often referred to as the infiltration factor. In the absence of indoor sources, the average I/O ratio is equal to the infiltration factor. Study of I/O ratios and infiltration factors typically take either a holistic approach where measured concentrations are compared under representative circumstances or a mechanistic approach where one or more of the influential factors are investigated. For some studies, the two approaches are combined.

Chen and Zhao [2] reviewed research regarding the migration of particles into the indoor environment. They reported I/O ratios that ranged from values significantly above 1 (indicating indoor sources) to values less than 0.1. Although some of the differences could be attributed to different particle size ranges, there were still large variations of values for particles of the same size range. For infiltration factor, they reported values for PM2.5 from 21 studies between about 0.3 to 0.8 and for PM10 from six studies between about 0.1 to 0.5. The variability exhibited between studies can be attributed to differences in building characteristics, meteorology, and particle size/composition.

Kearney *et al*. [3] reported infiltration factors for fine (<2.5 μm) and ultrafine (<100 nm) particles from 74 homes when indoor sources were removed from the concentration data using an algorithm. They found median infiltration factors were higher in the summer than the winter and also higher for fine particles than ultrafines. However the ranges for both seasons and particle sizes were much larger than the differences between them.

Residential wood burning within a neighborhood exposes residents in nearby homes to freshly combusted woodsmoke particles. The infiltration factor for these particles will be influenced by the specific properties of the woodsmoke particles. A principle component of woodsmoke particles is inorganic carbon. Several previous studies have investigated infiltration factors for either black carbon or elemental carbon. For instance, Lunden *et al*. [4] reported an indoor/outdoor ratio in the absence of indoor sources of 0.6, Geller *et al*. [5] reported 0.85, and Polidori *et al*. [6] reported 1.0. MacNeill *et al*. [7] found daily black carbon indoor/outdoor ratios that ranged from 0.28 to 0.59. Fuller *et al*. [8] investigated I/O ratios based on particle number counts (6 - 3000 nm) near highways, which may be similar in some ways to near field woodsmoke particles, and found a median ratio of 0.95. The wide range in ratios can be attributed to the diversity in the residences studied, differences in meteorological factors, and variations in particle characteristics due to the wide range of sources which produce inorganic carbon particles.

By taking a mechanistic approach, the factors which influence the indoor/outdoor concentration ratio can be used to explain the ratios found. Since black carbon is fairly inert in the indoor environment, the most important processes which influence the indoor/outdoor ratio are air infiltration rate, penetration factor, deposition losses, indoor generation, and resuspension [9] [10]. For this study, the small size of the particles and the limited activity within the residences served to limit the impact of resuspension. Both the penetration factor and deposition rate can be highly influenced by site characteristics. The penetration factor describes the fraction of infiltrating particles which enter the indoor environment with the infiltrating air, as opposed to depositing with the building walls. A wide range of penetration factors have been reported in the literature [2]. Stephens and Siegel [11] measured penetration factors for 18 homes based on particle number concentrations (20 - 1000 nm) and found that they ranged between 0.17 and 0.78 with a mean of 0.47. For the same homes, deposition loss rate ranged from 0.31 to 3.24 h^{-1} with a geometric mean of 1.01 h^{-1}. Monte Carlo simulations used to determine infiltration factor for a wide range of particle sizes agreed with the experimental data which show a large variation in infiltration factor across the building stock [12].

By sampling in a region, ambient particles are dominantly from residential combustion; this study adds important information to our understanding of transport of woodsmoke particles into homes and, ultimately, to our understanding of human health impacts of residential wood burning.

2. Experimental Methods

To investigate the migration of recently generated woodsmoke particles into residences, field sampling was performed at 4 different homes within a 1 square kilometer residential area encompassing approximately 400 homes. During field sampling events, referred to as Intensive Operation Periods (IOPs), integrated filter and aethalometer samples were collected both inside and outside the homes from 6 pm to 6 am. In addition, the study area was surveyed using an infrared camera to determine which homes were actively using their chimneys. This study was performed in conjunction with a study of variability of outdoor woodsmoke concentrations within a residential neighborhood [13].

The homes selected were within the town of Cambria, California, located on the central coast of California (35.554030, −121.087394). Cambria is located between the Pacific Ocean and the Santa Lucia Range. It has a population of approximately 6500 and lists tourism, light industry, and agriculture as the main industrial activities [14]. One of the most significant advantages of the chosen town was the nearly complete lack of non-woodsmoke particle sources, such as industry or vehicular traffic. There are no major freeways in the area and little or no traffic during the evenings and nighttime. The largest road, Highway 1, is one lane each way and serves only local truck traffic. There are no major PM2.5 producing local industries. The nearest major cities are Monterey (population 410,000) approximately 80 miles north-west, San Luis Obispo (population 270,000) approximately 30 miles south-east, and Paso Robles (population 28,000) approximately 20 miles east. Due to the distance, topography, prevailing meteorology, and relatively good air quality in these nearby regions, transport of pollutants does not significantly impact air quality in Cambria. Although Cambria does not have other significant pollutant sources, residential wood burning is common. Because of the age of the neighborhoods and the characteristics of the homes, a large percentage of the homes in Cambria have fireplaces and/or woodstoves. In addition, the close proximity to available wood sources results in many homes using wood for all or a significant portion of their heating needs.

During each IOP, integrated 12 hours PM2.5 filter samples were collected between 6 pm and 6 am using Model 200 Personal Environmental Monitors (PEMs, SKC, Inc) using flow-controlled pumps (Leland Legacy, SKC Inc.) A mask containing a 2 cm opening was used to decrease the deposit area and thus increase measurement sensitivity. The twelve hours sampling period was chosen to coincide with the hours where wood burning is most prevalent and exclude daytime hours where burning is not prevalent in this region and other aerosol sources are more dominant. Sampling locations were restricted to volunteers' homes and locations where the PEMs would be less likely disturbed. Outdoor samplers were placed near the exterior of the home and indoor samplers were generally located in the main living area of the home, with the exception of one home (*J*) where the resident requested that the sampler be placed in a different room with less usage. Access and logistical constraints did not allow for all homes to be sampled on all nights.

Filter samples were analyzed using a light transmission method to characterize the spectral light absorption by aerosol samples collected on quartz fiber filters. Transmission was measured using a custom built optical spectrometer that allowed placement of filter samples between the light source and detector. The light source was a lamp with an emission spectrum extending from the near ultraviolet to the near infrared and the spectrometer was equipped with a fixed diffraction grating and a linear CCD-array detector. Light attenuation (ATN) was calculated from measured sample light transmission (T): ATN = $100\ln(1/T)$. In this study, T was defined as $(I_s/I_{s,o}) \times (I_{r,o}/I_r)$, where I_s and $I_{s,o}$ are the measured intensities of light transmitted through a quartz filter sample prior to and after removal of carbonaceous material by heating to 800˚C in oxygen, I_r and $I_{r,o}$ are the intensities of light transmitted through a reference quartz filter measured at the same time as I_s and $I_{s,o}$, respectively. Measurements of I_s and $I_{s,o}$ were made using the same quartz filter.

Spectral analysis was used to determine the value of the absorption Ångström exponent (AAE) which is indicative of the source of the particulate matter. For example, the AAE of motor vehicle exhaust is ~1, whereas biomass burning smoke exhibits stronger absorption selectivity with an AAE ~2 [15]. The average AAE for the indoor filters was 2.6 ± 0.7 and the average for the outdoor samples was 2.2 ± 0.8 indicating that woodsmoke was the dominant particle source for both the indoor and outdoor samples. In addition, analysis showed that black carbon concentration performed as well as any of the other surrogate measurements of woodsmoke particle concentration [13]. As a result, the analysis of the indoor and outdoor woodsmoke concentrations within the study area was performed using black carbon (BC) concentrations based on attenuation at 880 nm to represent the amount of woodsmoke at the sampling locations.

During each IOP, aethalometers (Magee Scientific) were also placed at selected indoor and outdoor locations, co-located with the filter samplers. In addition, indoor and outdoor aethalometers sampled for an extended period at one of the residences to better characterize particle behavior for that home. Five aethalometers were used during the course of this research, four were dual-wavelength (370 nm and 880 nm) and one was a multi-wavelength aethalometer (370, 470, 520, 590, 660, 880 and 950 nm). The attenuation at 880 nm wavelength was used to determine the black carbon concentration for all aethalometers, which was consistent with the black carbon analysis used for the filters. To ensure adequate precision between the instruments, the aethalometers were placed side by side and the specific attenuation values were adjusted to match black carbon concentrations (880 nm). Before the adjustment the F statistic was 63.84 corresponding to a *P*-value of 0. After the adjustment, the F statistic was 1.78 with a corresponding *P*-value of 0.131, statistically equivalent using a minimum *P*-value level of 0.05.

This study was designed to minimize some of the inherent complexity of the relationship between indoor and outdoor concentrations. The results assume that the black carbon from woodsmoke particles is inert and that resuspension was negligible, based on the small particle size and the relative inactivity of the residents during the sampling period. Indoor sources were considered to be negligible for all samples where the fireplace was not used during the sampling period. The primary metric for analysis of the data was the indoor/outdoor ratio, which for those homes without indoor sources will equal the infiltration factor. Although the average indoor/outdoor ratio is a traditional metric for describing the relative concentration of a pollutant in the indoor environment, care must be taken in determining this value. Since changes in outdoor concentration are transmitted slowly through the building shell (often described as an apparent time lag in the concentration series), the indoor/outdoor ratio at any particular time is more dependent on recent changes in outdoor concentration than on characteristics of the home or pollutant. Therefore, to obtain useful information about the relationship between indoor and outdoor exposure, concentrations must be averaged across longer periods; ideally time periods which encompass an entire "event" from the introduction of the pollutant outdoors until it is completely removed from the indoor environment. The sampling interval of 6 pm to 6 am was chosen to most effectively encompass the entire evening woodsmoke generation event based on observations of wood burning patterns for the area.

In addition to the need to collect samples over an entire peak event, there are also inherent uncertainties introduced when extrapolating a given indoor/outdoor ratio to represent indoor concentrations under differing circumstances. The ratio is dependent on factors such as the infiltration, deposition, and penetration rates which will depend not only upon characteristics of the building and the particles of concern, but also upon meteorological and mixing factors which will change with time. Therefore, a given indoor/outdoor ratio is most accurate when it is applied to homes with similar infiltration characteristics and under similar meteorological conditions. This means that not only can two homes in the same region have different indoor/outdoor ratios for the same time period, but also that the ratio for the same home will typically vary with meteorology. Nevertheless, indoor/outdoor ratios are a useful construct to provide a basic understanding of indoor contaminant levels.

Using the extended aethalometer data from one residence, measured indoor concentrations were compared with modeled indoor concentrations to estimate air exchange rate, penetration factor, and deposition rate. A mass balance model was developed and parameters were fit using a minimization of the sum of squared differences between the modeled and measured indoor concentrations. The model assumed resuspension, chemical reactions, and source generation inside the home were negligible. Minimal foot traffic during the night hours allowed the dismissal of resuspension. Because black carbon is relatively non-reactive, chemical reactions were assumed to be insignificant. Finally, for the home chosen the residents did not burn firewood, eliminating source generation. Cooking is a possible generation source of black carbon, however no anomalous increases in black carbon concentrations were observed indoors during the study period. Based on these assumptions, the following time dependent forward marching mass balance equation was used to model indoor concentrations.

$$C_{in,t} = C_{in,t-1} + \left[C_{out,t-1} P \lambda_{v,t} - C_{in,t-1} \left(\lambda_{v,t} + k_{dep} \right) \right] \Delta t \quad (1)$$

where $C_{in,t}$ = modeled indoor concentration at time t (μg/m^3), $C_{in,t-1}$ = modeled indoor concentration 1 time step before time t (μg/m^3), $C_{out,t-1}$ = measured outdoor concentration 1 time step before time t (μg/m^3), P = dimensionless penetration factor, $\lambda_{v,t}$ = air exchange rate at time t (h^{-1}), k_{dep} = deposition loss rate (h^{-1}), and Δt = time between samples (h).

It was assumed that penetration and deposition loss rate were constant throughout the event. The infiltration rate, however, is unlikely to remain constant throughout the entire period, since it is dependent on meteorological conditions and indoor/outdoor temperature difference. Consequently, the value for infiltration rate was allowed to vary every 2 hours. For minimization, the Solver program for Microsoft Excel, developed by Frontline Systems was used. The infiltration model used the minimization of the sum of squared differences between the measured and modeled indoor concentrations normalized by dividing the difference term by the measured concentration. Normalization deemphasizes high concentrations, which may otherwise shift the parameters to minimize small regions of high concentrations.

$$SSD = \sum \left(\frac{C_{\text{measured}} - C_{\text{modeled}}}{C_{\text{measured}}} \right)^2 \quad (2)$$

where C_{measured} (ng/m^3) = indoor concentration of black carbon determined by the aethalometer, and C_{modeled} (ng/m^3) = concentration of black carbon determined from the indoor infiltration model.

One issue that arises when using this method is that the equation is over parameterized, that is, there can be more than one value for each parameter that minimizes the sum of squared differences. Depending on the starting values for each parameter, Solver may find different solutions for minimization. To provide a more robust solution, deposition rates and penetration factors were calculated based on periods of relative dominance for each term. The deposition rate has a greater influence on the concentration of indoor BC when indoor concentrations are larger than outdoor concentrations (decreasing indoor concentrations) while the penetration factor has a greater influence when outdoor concentrations are higher than indoor concentrations (increasing indoor concentrations). Consequently, the penetration factor was fit only during periods when indoor concentrations were increasing and the deposition rate was fit only during periods when indoor concentrations were decreasing. This method is based on the analytical method developed by Thatcher *et al*. [16].

3. Results and Discussion

For this study, 12 hours integrated filter samples were taken simultaneously indoors and directly outdoors at four different residences on 6 nights during the winter. This sampling occurred in conjunction with an outdoor study of residential woodsmoke described by Thatcher *et al*. [13]. The four homes were all located within the 1 km^2 study region and were detached single family residences which were constructed in the 1980s and 90s. For each home, a filter sampler was placed indoors and a second sampler was placed outdoors near the home's exterior. **Table 1** gives basic details of the residences where sampling occurred.

Filters were analyzed for black carbon using optical absorbance. Since outdoor black carbon concentrations typically peaked in the early evening and decreased around midnight, this timing had a higher probability of capturing an entire peak event. A total of 14 pairs of filter samples were collected, however 4 of the pairs had at least one filter below the detection limit and 2 other filters had sampling errors. By supplementing the filter data with aethalometer data, 13 indoor/outdoor pairs taken were complete as shown in **Table 2**. During a given IOP, the variability between outdoor sample concentrations taken at the different residences varied by as little as 7% (IOP 1a) and as much as 80% (IOP 7). There was no consistent pattern of concentrations for the homes studied. A home could have the lowest outdoor concentration for one IOP and the highest for another.

Table 1. Description of homes used in study.

Residence	# Stories	Year built	Floor area (ft^2)	# Residents	Surrounding foliage	Indoor sampling location
E	2	1976	1800	2	Moderate	Living room
G	1	1998	2900	2	Heavy	Living room
J	2	1988	2500	1	Moderate	1st floor room
M	1	1982	2900	2	Heavy	Living room

Table 2. Indoor/outdoor ratios for 12 hours integrated black carbon concentrations by residence and intensive operation period (IOP).

IOP	Residence	I/O ratio
1a	E	0.99[a]
2a	E	0.86[a]
3a	E	0.42[a]
4a	E	0.87
7a	E	1.06[a]
8a	E	0.87[a]
1a	G	0.69
3a	G	2.97[b]
4a	J	1.96[c]
3a	M	0.86
4a	M	0.69
7a	M	0.45
8a	M	2.25[b]
Average		**0.78[d]**
Standard deviation		**0.21**

[a]Ratio includes concentrations integrated from an aethalometer; [b]Resident burned wood in fireplace during test; [c]Resident burned mail in fireplace during test; [d]Ratios from 3a-G, 4a-J and 8a-M were excluded from the average due to fireplace usage.

During sampling, two of the homes reported burning wood in their fireplaces during one sampling period. Both of these homes experienced indoor concentrations that were over 2 times higher than the measured concentration outside the home. In addition, Home J reported burning "mail" in the fireplace resulting in an indoor/outdoor ratio of about 2. Since the I/O ratio is being used to represent the level of particles of outdoor origin (infiltration factor), these homes were excluded from the I/O ratio average due to the presence of the indoor sources.

Although there was a large amount of variability in the indoor/outdoor ratios, the average value indicates that the indoor environment was not highly effective at reducing exposures to black carbon from residential woodsmoke generated in the near-field. As can be seen in Equation (1), a value for I/O close to one can result either from high air exchange rates, low loss rates, or a combination of theses factors. If the relatively high ratio resulted from a higher than normal air exchange rate, then this conclusion would have limited applicability to other residences. However, if the high I/O ratio is due to low loss rates then a similar ratio might be expected in other residences. Abnormally high air exchange rates would not be expected for this study, since wind speeds were low, indoor/outdoor temperature differences were moderate, windows were closed, and homes were of standard construction and well maintained. On the other hand, relatively fresh woodsmoke particles would be expected to be small, trending toward the accumulation mode. Particles in this size range don't settle or diffuse quickly, leading to an expectation of low loss rates and high penetration rates.

To help confirm the findings of the filter-based measurements, additional tests were performed at residence E. A total of 16 burning events were recorded between November 30, 2008 and March 20, 2009. A burning event was defined as a substantial increasing in outdoor black carbon concentration followed by a return to pre-event concentrations. These events usually began around 6 to 8 pm and the time period for the outdoor concentration increase ranged from a few hours to less than an hour. **Figure 1** shows the indoor and outdoor BC concentrations for a single burning event. The length of the time lag between the start of elevated outdoor concentrations and the indoor response is an indication of a relatively low air exchange rate.

The aethalometer raw data was reintegrated over the course of the entire burning period to obtain an average

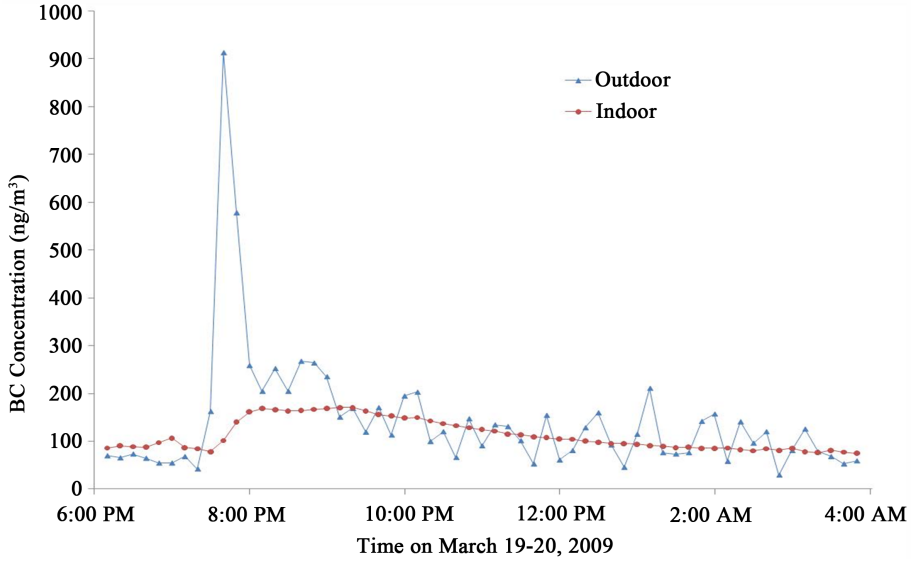

Figure 1. Indoor and outdoor aethalometer black carbon concentrations (10 minute averages) for Home E in Cambria, CA.

indoor and an average outdoor concentration. For black carbon the average indoor/outdoor ratio was 0.74 ± 0.05 for all 16 burning events using a 95% confidence interval. This means that over the course of the burning period, an individual indoors will be exposed to approximately three quarters of the exposure a person standing just outside the house would receive. This value is similar to the average of 0.78 obtained from the filter measurements.

Figure 2 shows the modeled indoor concentrations and measured concentrations over time for the burning event on March 16-17, 2009. **Figure 3** compares modeled and measured concentrations over the same burning event. Across all 16 burning events, the slope of the model fitting line varied between 0.88 and 1.02 with an average of 0.98. The lowest R2 for the model fit was 0.93 and the average for all events was 0.97.

For the 16 burning periods, the average air exchange rate for the home was 0.26 ± 0.08 h^{-1}. This value is lower than the average residential air exchange rate reported in many studies. However, wind speeds were low during most of the study and average outdoor temperatures, while cool, were not as cold as winters in many regions. Consequently, the lower driving forces would be expected to lead to lower air exchange rates.

Based on the mass balance model, the average deposition rate was 0.08 ± 0.03 h^{-1} and the average penetration factor was 0.97 ± 0.02. These values are consistent with the expectation that many recently generated BC particles will be accumulation mode or smaller. Although most studies investigate penetration factors and deposition rates based on particle size and not aerosol source, similar findings have been suggested by various authors. For woodsmoke, particle size distributions have been reported by different studies as between 30 - 100 nm in diameter [17], 38 ± 11 nm in diameter [18] and 100 - 200 nm in diameter [19] Some coagulation could be expected as these studies examined particles from freshly burned sources, not sizes at nearby buildings. Differences in burning conditions, type of wood, and time since combustion have the most predominate effects on the size distribution. Wallace [20] reviewed indoor particles studies for a large number of homes and reported an average deposition loss rate for ambient particles of 0.39 ± 0.16 h^{-1} for PM2.5, which is expected to be larger than the median woodsmoke particle in this study. For median optical diameter particles less than 0.225 µm, Thatcher *et al.* [16] reported average deposition rates ranged from 0.12 - 0.3 h^{-1}, while penetration factors ranged from 0.8 - 1. These previous results are consistent with our findings that deposition and penetration losses have only a slight mitigating effect on indoor concentrations.

Using the air exchange rate, deposition rate, and penetration factor from the mass balance model in the steady state indoor/outdoor equation (Equation (1)) yielded an estimated indoor/outdoor ratio of 0.74, which is 12% lower than the average ratio for the filter samples from House E (0.85).

4. Conclusion

Field experiments were performed within a residential neighborhood of Cambria, CA. Cambria was chosen due

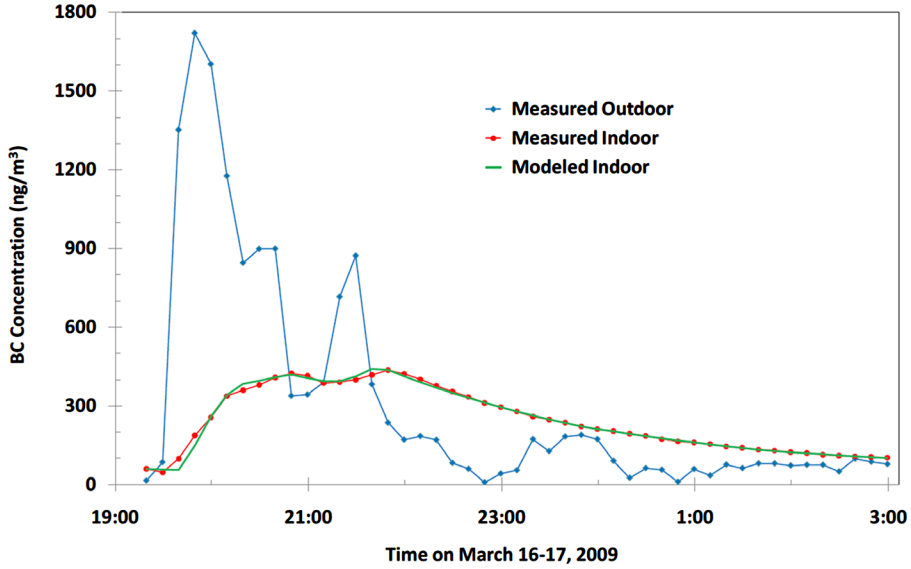

Figure 2. Modeled versus measured indoor black carbon aethalometer concentrations for Home E on March 16-17, 2009.

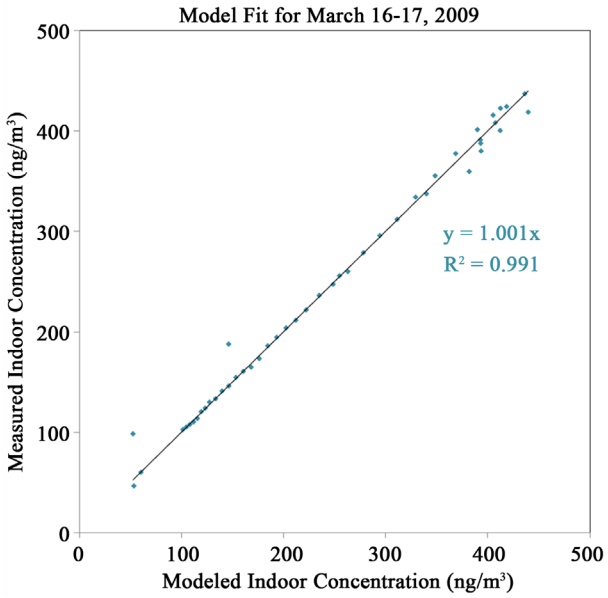

Figure 3. Model fit for the burning event on March 16-17, 2009 shown in **Figure 2**.

to the favorable meteorology, prevalence of wood burning, and lack of non-woodsmoke PM sources. To determine the impact of residential woodsmoke on residents within their homes, integrated filter samplers and aethalometers were placed indoors and outdoors at four residences within a 1 km^2 study area. Black carbon, a significant component of woodsmoke, was used as an indicator compound for woodsmoke. Although indoor concentrations are reduced relative to adjacent outdoor concentrations, woodsmoke particles generated in the near-field are relatively persistent in the indoor environment when compared to many other ambient pollutants. In addition, for homes with active fireplace usage, indoor concentrations were higher than outdoor concentrations by more than a factor of 2, showing that generation had a more significant impact on indoor concentrations than infiltration of outdoor particles. With penetration factors approaching unity and deposition rates nearing zero, the overall exposure to newly generated woodsmoke particles of an individual inside is close to the exposure of an indi-

vidual outside. A simple indoor/outdoor model adequately represented indoor conditions when using measured values of infiltration, penetration, and deposition rates. The model results indicated that the homes studied had air exchange rates which were representative of typical residential homes, meaning that the results should translate to other homes of a similar age. For the predominant source of black carbon from newly generated woodsmoke, an average indoor/outdoor ratio of 0.78 shows the minimal shielding a home provides. This is an important conclusion for sensitive individuals who try to avoid inhalation by seeking protection inside a home.

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