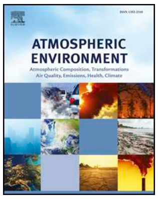
Atmospheric Environment 201 (2019) 223–230

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv



Predicting indoor concentrations of black carbon in residential environments

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ARTICLE INFO

Keywords:
Black carbon
Exposure
Modeling
Estimation

ABSTRACT

Black carbon (BC) is a descriptive term that refers to light-absorbing particulate matter (PM) produced by incomplete combustion and is often used as a surrogate for traffic-related air pollution. Exposure to BC has been linked to adverse health effects. Penetration of ambient BC is typically the primary source of indoor BC in the developed world. Other sources of indoor BC include biomass and kerosene stoves, lit candles, and charring food during cooking. Home characteristics can influence the levels of indoor BC. As people spend most of their time indoors, human exposure to BC can be associated to a large extent with indoor environments. At the same time, due to the cost of environmental monitoring, it is often not feasible to directly measure BC inside multiple individual homes in large-scale population-based studies. Thus, a predictive model for indoor BC is needed to support risk assessment in public health. In this study, home characteristics and occupant activities that potentially modify indoor levels of BC were documented in 23 homes, and indoor and outdoor BC concentrations were measured twice. The homes were located in the Cincinnati-Kentucky-Indiana tristate region and measurements occurred from September 2015 through August 2017. A linear mixed-effect model was developed to predict BC concentration in residential environments. The measured outdoor BC concentrations and the documented home characteristics were utilized as predictors of indoor BC concentrations. After the model was developed, a leave-one-out cross-validation algorithm was deployed to assess the predictive accuracy of the output. The following home characteristics and occupant activities significantly modified the concentration of indoor BC: outdoor BC, lit candles and electrostatic or high efficiency particulate air (HEPA) filters in heating, ventilation and air conditioning (HVAC) systems. Predicted indoor BC concentrations explained 78% of the variability in the measured indoor BC concentrations. The data show that outdoor BC combined with home characteristics can be used to predict indoor BC levels with reasonable accuracy.

1. Introduction

Exposure to traffic-related air pollution has been associated with adverse health effects (Katsoulis et al., 2014; Bowatte et al., 2015). Black carbon (BC) is an example of a traffic-related air pollutant and is used as a surrogate of traffic-related particles (Janssen et al., 1997; Power et al., 2011). During the cold season (September 1 – March 31), exposure to BC is associated with cough among children (Patel et al., 2009). Black carbon is also linked to the prevalence of bronchitis and asthma in children (Kim et al., 2004), and respiratory hospitalizations among the elderly (Bell et al., 2009).

Black carbon is a descriptive term for light-absorbing particles that represent a continuum of incomplete combustion residues ranging from larger charred materials that retain structural information of parent materials to highly condensed refractory soot particles that are produced from incomplete combustion (Yan et al., 2011). Soot particles include organic carbon and black carbon particles derived from combustion (Petzold et al., 2013). They are nanometer to submicrometer in aerodynamic diameter (D'Anna, 2009), and can be emitted from the exhausts of internal combustion engines (World Health Organization (WHO), 2012). Chars are large particles that do not travel far. Consequently, in most filter-based measurements of airborne particulate matter (PM), BC mainly consists of soot particles that usually contain other atoms and attached organics such as polycyclic aromatic

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https://doi.org/10.1016/j.atmosenv.2018.12.053
Received 15 August 2018; Received in revised form 16 December 2018; Accepted 19 December 2018

hydrocarbons (Yan et al., 2011). Soot is determined by optical methods and chemical-thermal methods. Soot determined via optical methods is referred to as black carbon. The term elemental carbon is used when soot is determined by chemical-thermal methods that measure the amount of CO_2 evolved. There is a high correlation (r = 0.95) of soot results obtained with optical methods and chemical-thermal methods (Kinney et al., 2000). Due to this high correlation, the terms black carbon and elemental carbon are often used interchangeably (Han et al., 2010). Using light absorption of colored particles at one or more wave lengths, optical absorption techniques have been utilized to differentiate black carbon from other colored components such as particles from cigarette smoke (Yan et al., 2011; Lawless et al., 2004). Majority of colored components of PM, such as cigarette smoke, are colored organic carbon, and not black carbon, as they make sampling filters yellow-brown and not black. It is estimated that $< 1\%$ of PM emitted from burning cigarettes have light-absorbing properties of black carbon (National Institute for Occupational Safety and Health (NIOSH), 1995).

Black carbon (BC) can be emitted from any incomplete combustion source. For indoor environments, examples of BC sources include lighting or extinguishing candles, using kerosene lamps, charring food, and cooking or heating with solid fuels (World Health Organization (WHO), 2012; LaRosa et al., 2002; Habre et al., 2014). Cleaning activities, such as vacuuming carpets, can cause resuspension of indoor particles with aerodynamic diameter $\leq 10\,\mu m$, which results in increased indoor aerosol concentrations (Corsi et al., 2008). In urban settings, exhaust emissions from traffic and especially older diesel engines are one of the major contributors to ambient BC (World Health Organization (WHO), 2012). Thus, the distance of a home to a road with high vehicular traffic may modify indoor BC concentrations. Other factors are also associated with indoor BC levels. Quantifying the factors which modify indoor BC should enhance any predictive model for indoor BC concentrations. Modeling residential indoor BC concentrations is useful for estimating average exposure, given that people typically spend 64–66% of their time indoors at their residences (Buonanno et al., 2013; Brasche and Bischof, 2005; Leech et al., 2002). In addition, subgroups such as infants, the elderly, stay-at-home parents and people who work from home spend much higher fractions of their time at their residences. Modeling residential indoor BC concentrations would facilitate risk assessment in public health when population exposure to BC is estimated.

As BC refers to light-absorbing particles, any indoor air quality (IAQ) intervention that aims at reducing indoor particles, may also reduce indoor BC. Examples of IAQ interventions include equipping the heating, ventilation and air conditioning (HVAC) systems with efficient air filters (Sadiktsis et al., 2016), operating kitchen exhaust hoods with recirculated air through a filter or outdoor exhaust (Lunden et al., 2015; Rim et al., 2012). Indoor-outdoor air exchange modifies indoor pollutant levels (Sexton et al., 1983). The air exchange rate is affected by infiltration and exfiltration via unintentional leaks in a building envelope, open windows or doors and mechanical ventilation (Ng et al., 2015). Pressurized fan tests are used to measure building air tightness (ASTM, 2010), an indicator of air infiltration via unintentional leaks in a building envelope. Furthermore, air leakage attributable to different building components can be estimated (American Society of Heating, 2017).

A model to accurately predict indoor BC can be developed based on the information about the above-listed home characteristics. After such a predictive model is developed, its performance should be assessed through validation methods, e.g., cross-validation to ensure the accuracy of the model output (Arlot and Celisse, 2010). There have been attempts to establish relationships between specific environmental characteristics and the carbon particle levels in residential settings. Baxter et al. used home characteristics, occupant activities and traffic indicators to predict indoor elemental carbon determined by $PM_{2.5}$ filter reflectance analysis (Baxter et al., 2007a, 2007b). Because the method of analysis used by Baxter et al. is an optical method, the measured particles can be regarded as black carbon.

Baxter et al. developed two models, in which factors such as an increase in ambient BC and the close proximity of a home with windows kept open to a road with high truck counts were both significantly associated with an increase in indoor BC. (Baxter et al., 2007a, 2007b) However, some home characteristics and occupant activities that can potentially modify indoor PM, e.g., electrostatic/HEPA HVAC filters were not incorporated into the models developed by Baxter et al. Therefore, the need remains for an advanced predictive model incorporating real-life multiple housing characteristics that can potentially modify indoor BC concentration. The goal of this study was to develop such a predictive model for indoor BC. We utilized multiple housing characteristics as covariates and assessed the predictive performance of the model with a leave-one-out cross-validation method.

2. Methods

2.1. Study overview

The study was conducted in 23 residential environments (single-family and apartment buildings) in the Cincinnati-Kentucky-Indiana tristate region (Fig. S1) from September 2015 through August 2017. The BC levels were measured indoors and outdoors, and home characteristics specific to each dwelling were documented. The homes in this study belonged to a cohort of subjects from another ongoing study (Cox et al., 2018). The ongoing study was focused on the efficiency of air cleaners in removing indoor particles, but only baseline measurements (before the deployment of air cleaners) were included in this study. All homes were located in neighborhoods with $\geq 0.33\,\mu g/m^3$ outdoor elemental carbon attributable to traffic, as determined in a previous study (Ryan et al., 2008). The study received Institution Review Board (IRB) approval from the University of Cincinnati IRB.

2.2. Environmental monitoring

Samples of airborne fine particulate matter ($PM_{2.5}$) were collected simultaneously from inside and outside of each residence over 48 h using single-stage Personal Modular Impactors (SKC, Inc., Eighty Four, PA) equipped with 37-mm Teflon filters. Measurements were repeated twice in each home. Indoor samples were collected in a bedroom and outdoor samples in the immediate vicinity (backyard or in front) of the home. Sampling pumps were calibrated to a flow rate of 3 L/min using a mass flow meter (TSI Inc., Shoreview, MN). Measurements were repeated twice in each home with a 2-month gap between the two measurements. After gravimetric determination of the $PM_{2.5}$, the filter samples were analyzed for BC by optical absorption technique (Yan et al., 2011), which has a published limit of detection (LOD) of $1.4\,ng/mm^2$ of the filter (equivalent to an air concentration of $0.12\,\mu g/m^3$ in this study). Media and field blanks were collected in parallel at a rate equal to 10% of all filter samples. The mean concentration of BC in the blank samples was $0.25\,ng/mm^2$ of the filter (equivalent to an air concentration of $0.07\,\mu g/m^3$ in this study). This value was subtracted from the BC measured on the real samples.

2.3. Documenting housing characteristics

Questionnaires on the housing conditions and appliances were administered to the participants of the study. In addition, the homes were inspected during each visit, and the questionnaire data were verified and documented. Information from the questionnaires contained the following characteristics:

- Exhaust hood in the kitchen – yes or no.
- Presence of electrostatic filter or high-efficiency particulate air (HEPA) filter in the HVAC system – yes or no.
- Lit candles during the sampling period – yes or no.

- Use of fireplace during the sampling period – yes or no.
- At least one open window during the sampling period – yes or no.
- Cleaning (vacuuming or sweeping or dusting) during the sampling period – yes or no.

Other housing characteristics assessed by the study team included the distance of the home to the nearest state highway or federal interstate (major road) and the annual average rate of air infiltration in the home. The distance of the homes to the nearest major road was calculated with a geographical information system (ArcGIS 9.0, Environmental Systems Research Institute, Inc., Redlands, CA) (Ryan et al., 2008). Data on the geographical location of the major roads were obtained from the Ohio Department of Transportation (2004) and the Kentucky Transportation Cabinet (2006) (Ryan et al., 2008). The annual average rate of air infiltration via unintentional leaks in the home was determined from measurements of a blower door system in accordance with the ASTM standard for fan pressurization tests (ASTM, 2010). The blower door system includes a fan, which is positioned at an exterior door in a building. Before the start of the blower door measurement, all exterior doors and windows in a home were shut, and the interior doors were opened. A baseline building pressure was measured with the blower door system, and the blower door fan was utilized to induce pressure differences between indoor and outdoor of 10–60 Pa with 5 Pa increments (ASTM, 2010). Building air tightness was derived from the blower door system by recording the airflow needed to establish the above-indicated pressure differences (10–60 Pa), and a summary of the test was reported through a proprietary software (TECTITE) (The Energy Conservatory, 2014). The software was programmed to calculate an annual average rate of natural air infiltration based on the American Society of Heating, Refrigerating and Air-Conditioning (ASHRAE) standard (American Society of Heating, 1993). Results from the blower door system give an estimation of the number of air changes per hour through unintentional leaks such as cracks and holes in the building envelope.

2.4. Statistical analysis

In the current study, each home was assigned an identification number. A linear mixed-effect model was used for predicting indoor BC. Outdoor measured BC concentrations and documented housing characteristics were assigned as fixed effects, and home identification numbers were treated as random effects. Statistical analyses were done with R studio (RStudio, 2016). To assess the effect of housing characteristics and outdoor BC concentration on indoor BC, the predictive model was developed in three stages. First, an all subsets regression analysis was conducted using lowest Bayesian information criterion (BIC) for model selection. Second, indoor sources of BC and housing characteristics that represented the infiltration of BC, but were not included in the model obtained from the all subset regression analysis were added one at a time. This was done because all the documented home characteristics in this study were potential modifiers of indoor BC and BIC is designed to penalize predictor variables as the sample size increases (Vrieze, 2012). Third, each time a new independent variable was added to the model obtained from the All Subset Regression analysis, the predictive accuracy of the model was assessed with a leave-one-out cross-validation method. The version of the model that yielded the highest out-of-sample R^2 and lowest root mean squared error (RMSE) was selected as the final model for the prediction.

The method for utilizing leave-one-out cross-validation has been reviewed by Arlot et al. (Arlot and Celisse, 2010). In summary, one observation from the dataset used to develop the predictive model was removed, and the predictive model was rebuilt again. Regression estimates of this rebuilt model were used to predict the indoor concentration of BC in the observation that was removed. The removed observation was then returned to the dataset, and another observation was removed, after which the predictive model was rebuilt again. Next, the indoor concentration of BC in the newly removed data point was predicted with the regression estimates of the new predictive model. This process was done 45 times (number of observations in the dataset).

Table 1
Samples collected by seasons.

Season	Duration	Percentage of samples
Fall	September 22 – December 21	22.2%
Winter	December 22 – March 20	24.4%
Summer	June 21 – September 22	28.9%
Spring	March 21 – June 20	24.4%

Seasons = astronomical seasons (obtained from the National Centers for Environmental Information) (National Centers for Environmental Information, 2017).

2.5. Handling non-detectable measurements of BC

Concentrations of indoor BC were skewed (geometric standard deviation = 3). Twenty-four percent (24%) of indoor BC samples and 2% of outdoor BC samples were below the LOD of 0.12 $\mu g/m^3$. All samples below the LOD were replaced with the value of LOD/2 as recommended by Hornung et al. (Hornung and Reed, 1990)

3. Results

3.1. Measurements and housing characteristics

After measurements were repeated twice in the 23 homes, one observation was lost in one home due to a pump failure. Consequently, there were 45 observations from the 23 homes. Table 1 presents data on sample collection in the current study stratified by seasons. Sample measurements were obtained from the fall, winter, summer and spring seasons (22.2%, 24.4%, 28.9% and 24.4%, respectively). Table 2 presents categorical characteristics of the homes utilized in this study. Of the 45 visits, at least one window was opened in 29 of them. Candles were lit during 6 of the visits. Cleaning activities were performed in the homes during 28 visits.

Table 3 presents the summary statistics for numerical home characteristics. The average annual air infiltration rate in the study homes ranged from 0.02 to 5.07 air changes per hour. The nearest distance of a home to a major road was 32 m, and the home farthest from a major road was 3.90 km away. Homes with and without electrostatic/HEPA HVAC filters had median indoor/outdoor BC ratios of 0.18 and 0.62, respectively. The median fraction of BC in the indoor and outdoor $PM_{2.5}$ samples was 0.04 and 0.09, respectively (Figs. S2 and S3).

3.2. Inferential information from the predictive model

Of the nine housing characteristics and occupant activities investigated, only five (outdoor BC, average annual air infiltration via unintentional leaks, HEPA/electrostatic HVAC filter, open/closed windows, candles) were selected as predictors of indoor black carbon in the chosen predictive model (equation (1)).

Table 2
Descriptive statistics of the categorical characteristics examined in the 45 Visits.

Categorical characteristics	Yes	No
HVAC filter (electrostatic/HEPA)	20	25
Exhaust hood in kitchen	34	11
At least one window opened	29	16
Lit candles	6	39
Use of fireplace	2	43
Cleaning activities (vacuuming, sweeping, or dusting)	28	17

Table 3
Descriptive statistics of measurements in the study (n = 45).

Home characteristics	Q1	Median	Mean	Q3	SD
Indoor BC (µg/m^3)	0.13	0.28	0.43	0.62	0.42
Outdoor BC (µg/m^3)	0.53	0.68	0.85	1.16	0.58
Total indoor/outdoor ratio of BC	0.17	0.47	0.50	0.69	0.35
Indoor/outdoor ratio of BC in homes with HVAC filter (electrostatic/HEPA)	0.13	0.18	0.33	0.40	0.27
Indoor/outdoor ratio of BC in homes without HVAC filter (electrostatic/HEPA)	0.47	0.62	0.64	0.84	0.35
Annual air infiltration via unintentional leaks (h^{-1})	0.29	0.42	0.61	0.66	0.77
Distance to major road (m)	288	393	651	744	822

BC = black carbon, Q1 = 25th percentile, Q3 = 75th percentile, Indoor/outdoor ratio of BC is based on individual home ratios. LOD for BC samples = 0.12 µg/m^3

$$\begin{aligned} \textit{Indoor BC concentration}_{it} = {} & b_i + \beta_0 + \beta_1 \times \text{outdoor BC concentration}_{it} \\ & + \beta_2 \times \text{average annual air infiltration via unintentional leaks}_i \\ & + \beta_3 \times \textit{HVAC filter}_i + \beta_4 \times \textit{open windows}_{it} + \beta_5 \times \textit{lit candles}_{it} + \varepsilon_{it} \end{aligned} \quad (1)$$

Where, *indoor BC concentration*$_{it}$ is the measured indoor BC in each home i on sampling visit t, b_i is a random intercept specific to each home i, β_0 is the fixed intercept, β_1 is the effect of the measured BC in the local outdoor environment of each home i on sampling visit t, β_2 is the effect of average annual air infiltration via unintentional leaks in each home i, β_3 is the effect of electrostatic/HEPA HVAC filter in each home i, β_4 is the effect of open or closed windows in each home i during visit t, β_5 is the effect of presence or absence of lit candles or absence of lit candles in each home i during visit t, β_6 is the effect of the presence or absence of a kitchen exhaust hood in each home i and ε_{it} is the residual error in the model.

Table 4 presents the results of the regression model obtained using the complete dataset. These results are based on 48-h averages of the indoor BC levels in the 45 sampling events. The independent variables (covariates) that were significantly associated with indoor BC concentration were outdoor BC concentration, electrostatic/HEPA HVAC filters, and lit candles. An increase of 1 µg/m^3 in outdoor BC was associated with 0.53 µg/m^3 increase in indoor BC (Table 4). With other covariates being equal between the two groups, homes with efficient HVAC filters were associated with 0.26 µg/m^3 decrease in indoor BC when compared to homes without HVAC filters. Homes where candles were lit, had 0.41 µg/m^3 higher indoor BC when compared to homes where candles were not lit, with other covariates being equal between the two groups. Indoor BC was positively associated with open windows and average air infiltration (albeit not significant) (Table 4).

3.3. Predictive capability of the model

Fig. 1 shows a scatter plot of the measured indoor BC concentrations and the predicted indoor BC concentrations as obtained from the leave-one-out cross-validation algorithm. Measured indoor BC concentrations ranged from 0.06 to 2.18 µg/m^3 (mean = 0.43 µg/m^3, SD = 0.42); predicted indoor concentrations ranged from −0.09–1.70 µg/m^3 (mean = 0.43 µg/m^3, SD = 0.38). Negative predicted values are assumed to be < LOD. The predicted indoor BC concentrations explained 78% of the variability in measured indoor BC concentrations (Out-of-sample R^2 = 0.77). The standard deviation of the unexplained variance in measured indoor BC concentration was 0.20 µg/m^3 (root-mean-squared error, RMSE).

Table 4
Results from the final model containing the complete dataset (n = 45)[1].

Effects	Regression estimate (β)	Standard error	P – Value[2]
Intercept	−0.06	0.09	0.52
Outdoor BC concentration	0.53	0.05	< 0.001 *
Average annual air infiltration via unintentional leaks	0.09	0.05	0.12
Electrostatic/HEPA HVAC filter (yes vs. no) [3]	−0.26	0.10	0.02 *
Open windows (yes vs. no) [3]	0.12	0.08	0.12
Lighting candles (yes vs. no) [3]	0.41	0.08	< 0.001 *

[1] Results are applicable to 48-h average of indoor BC, coefficient of multiple determination (R^2) = 0.71, root mean squared error = 0.71.
[2] * indicates statistically significant variables ($P < 0.05$).
[3] Reference group = No.

3.4. Sensitivity analysis

Using the complete dataset, the result of a univariate model that had only outdoor BC as a covariate yielded an R^2 of 49% (Table S1). This was a 22% loss in R^2 when compared to the final model that included indoor covariates in addition to outdoor BC (Table 4). Removing the insignificant covariates from the final model in Table 4 (average annual infiltration and open windows) and rerunning the model did not considerably change the regression estimates in Table 4 (Table S2). Likewise, the out-of-sample R^2 obtained from the leave-one-out cross-validation method (R^2 = 76%) (Fig. S4) was similar to that obtained in the model that included the insignificant variables (R^2 = 78%) (Fig. 1). Sensitivity analysis, performed where season was added to the final model, showed that the effect of season on indoor BC was not significant (Table S3). Furthermore, results of the leave-one-out cross-validation method indicates that the model which includes season as a covariate had a slight increase in error (RMSE) and explained less variability in indoor BC (Fig. S5) when compared to the final model (Fig. 1).

Table S4 presents the final model that was developed from the complete dataset but with the exclusion of one influential observation. Removing the influential observation resulted to a 10% loss in R^2 (Table S4). In this model, 1 µg/m^3 increase in outdoor BC was associated with 0.43 µg/m^3 increase in indoor BC (compared to 0.53 µg/m^3 in the final model containing the influential observation). Other regression estimates in both models (models with and without the influential observation) were similar (Table S4 and Table 4). The influential observation was the observation with the maximum measured indoor and outdoor BC (2.2 µg/m^3 and 3.6 µg/m^3, respectively) (Fig. 1). Using the dataset that did not contain the influential observation, the result of a univariate model that only had outdoor BC as a covariate yielded an R^2 of 21% (Table S5). This is a 40% decrease in R^2 when compared to the model R^2 obtained from using both indoor factors and outdoor BC as predictors of indoor BC (Table S4). Cross-validation of the model without the influential observation showed that the model explained less variation in indoor BC (Fig. S6) when compared to the validation of the final model (Fig. 1).

Using measurements of average local outdoor BC concentration and average air infiltration rate of a building, combined with the home conditions in the presented final model, estimates of average indoor BC can be obtained in real-life scenarios (equation (2)).

$$\begin{aligned} \textit{Predicted indoor BC} = {} & -0.06 + 0.53 \times \textit{outdoor BC concentration} \\ & + 0.09 \times \textit{air infiltration via leaks} \\ & - 0.26(\textit{if HVAC filter present}) \\ & + 0.12(\textit{if at least one window is opened}) \\ & + 0.41(\textit{if at least one candle is lit}) \end{aligned} \quad (2)$$

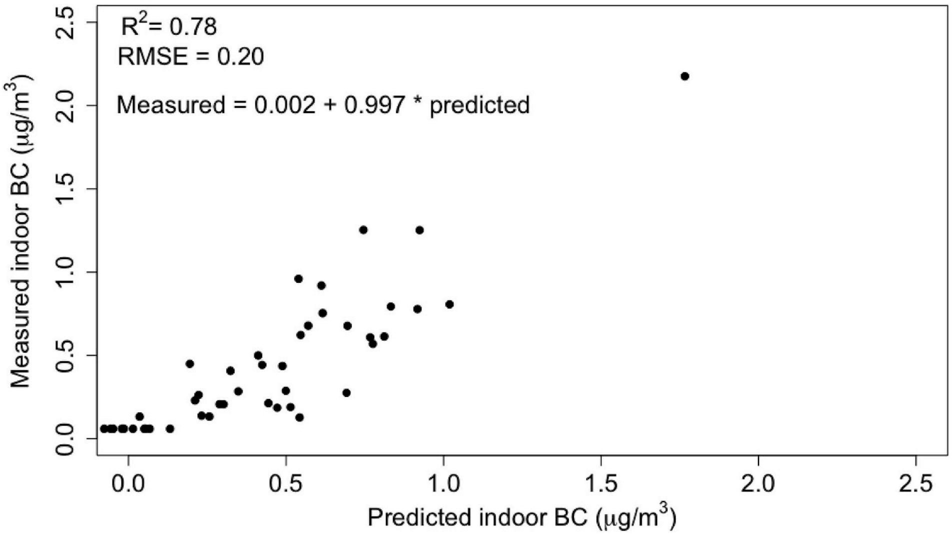

Fig. 1. Scatter plot of measured indoor BC and predicted indoor BC levels obtained from leave-one-out cross-validation. RMSE = root-mean-squared error of the predictive model.

4. Discussion

A linear mixed-effect model was developed to predict indoor BC concentrations by using the measured outdoor BC concentrations and home characteristics as predictors. Predicted indoor BC concentrations explained 78% of the variability in the measured indoor BC. As compared to the models by Baxter et al., 2007a, 2007b, the present model allowed incorporating electrostatic/HEPA HVAC filters, which potentially decrease indoor BC levels.

The median levels of BC in the current study (indoor = 0.28 µg/m^3, outdoor = 0.68 µg/m^3) and the indoor/outdoor ratio (I/O) of BC (0.47) were lower than the levels and ratios reported in other studies. In the current study, median I/O in homes with electrostatic/HEPA HVAC filters was 0.18 and 0.62 in homes without HVAC filters. This finding confirms that electrostatic/HEPA HVAC filters reduced indoor BC, as estimated in the regression model. Baxter et al. reported median BC in Boston homes as 0.49 µg/m^3 and 0.55 µg/m^3 indoors and outdoors, respectively (I/O = 0.89) (Baxter et al., 2007b). Coombs et al. reported median BC in Cincinnati to be 0.99 µg/m^3 and 0.94 µg/m^3 in indoor and outdoor environments, respectively (I/O = 1.05) (Coombs et al., 2016). Furthermore, 48-h mean I/O of BC in New York City was 0.93 and 0.84 during the summer and winter seasons, respectively (Kinney et al., 2002). The low I/O ratio in the current study indicates that there are other unstudied housing characteristics that reduce indoor BC in the study homes. It may be possible that the variation in the actual number of windows opened during the study sampling period can act as unstudied black carbon sinks in homes that had indoor sources of black carbon. This is because windows were documented as a categorical variable in the study (i.e., at least one window opened during the sampling period or all closed). Our sampling results show that the median fraction of BC in the sampled $PM_{2.5}$ mass was 0.09 outdoors, but much lower indoors (0.04). The data suggest that BC was not a major indoor pollutant in the study homes, except when emitted from a few indoor sources as observed in the current study.

4.1. Housing characteristics/occupant activities associated with an increase in indoor BC

Based on the amount of variation contributed to indoor BC by the covariates, outdoor BC was the most significant contributor to indoor BC. An increase of 1 µg/m^3 in outdoor BC was associated with an increase of 0.53 µg/m^3 in indoor BC. The data suggest that on average roughly half of the level of increase in outdoor BC infiltrated indoor environments. Similarly, in a univariate model presented by Baxter et al. (2007a), there was a significant positive relationship (R^2 = 0.49) between outdoor and indoor BC levels. This relationship is identical to the positive relationship found from the univariate model of indoor and outdoor BC in the current study (R^2 = 0.49). The observed result is expected, given that vehicular exhaust emissions are major sources of ambient BC (World Health Organization (WHO), 2012), and all the homes used in this study were in proximity to a highway or interstate (median distance = 393 m). It was observed that including the data pair of maximum indoor and outdoor BC (influential observation) strengthened the relationship between indoor and outdoor BC, and added accuracy to the model output. This finding suggests that having measurements of pollutants that range at least one order of magnitude provides better representation of data for optimum model development.

Lit candles were the second most significant contributors to indoor BC after outdoor BC. Homes, where candles were lit, were associated with 0.41 µg/m^3 increase in indoor BC when compared to homes where candles were not lit. Paraffin wax is a common type of candle wax that contains heavy hydrocarbon chains with carbon chain lengths that can be greater than 50 (C_{50}) (Kuszlik et al., 2010). This may explain why the effect of burning candles in only 6 of the 45 sampling periods in this study (13%) was sufficient enough to have a significant increase in indoor BC. Interestingly, despite the relatively large proportion of study homes with lit candles (26%), there was no statistically significant increase in indoor BC attributable to lit candles in the study by Baxter et al. (2007b) A reason for this discrepancy could be that some types of candles emit negligible amounts of BC. Further research into BC emissions from different types of candle wax will aid the understanding of the observed differences. Already, it is known that scented candles emit ultrafine particles (size of BC particles) (Anthonisen et al., 1994) about twice less in concentration when compared to pure wax candles (Afshari et al., 2005). Moreover, the concentration of BC particles emitted from unsteady burning candles (light and extinguish) is greater than that of steady burning candles (Zai et al., 2006).

4.2. Housing characteristics/occupant activities associated with a decrease in indoor BC

In the study homes, electrostatic or HEPA HVAC filter was the most significant variable that reduced indoor BC. Homes with electrostatic or HEPA HVAC filters had 0.26 µg/m^3 decrease in indoor BC when compared to homes without such filters. This finding is expected, given that the efficiency of HEPA filters is > 99.97% (American National

Standards Institute/Air-Conditioning et al., 2013). Electrostatic filters reduce PM by charging and trapping PM on oppositely charged plates (Agrawal et al., 2010). It would be interesting to distinguish between the effects of HEPA and electrostatic HVAC filters on indoor BC. However, studying the effect of the specific type of filter in the HVAC systems was outside the scope of the study.

In the current study, the reducing effect (0.26 $\mu g/m^3$) of electrostatic or HEPA HVAC filters on indoor BC does not account for the potential difference in efficiency of these filters that may be observed in new versus older filters. It also does not account for the potential differences that can be observed in buildings of different volumes.

4.3. Housing characteristics/occupant activities that explained less variability in indoor BC

Of the five housing characteristics in the predictive model, average annual air infiltration via unintentional leaks and open windows did not significantly modify indoor BC. Including these variables added some predictive power to the model (higher R^2). One reason for the non-significant findings can be a result of low statistical power ($n = 45$). Furthermore, the effect of open windows is complex, as it facilitates outdoor-indoor transport of particles from outdoor sources, but can also facilitate exfiltration of particles produced from indoor sources. This may explain why the effect of windows kept open was not significant. PM infiltrates from local outdoor environments (Matson, 2005), and indoor pollutants can accumulate in homes with very tightly sealed building envelopes (Coombs et al., 2016). Our results show that one unit increase in air exchange rate via unintentional leaks in a building envelope was associated with 0.09 $\mu g/m^3$ increase in indoor BC. This increase was not significant, likely due to the losses through Brownian diffusion of PM ≤ 0.1 μm (Liu and Nazaroff, 2001) (size of BC particles) (Anthonisen et al., 1994) which decreases the infiltration factor (Rim et al., 2010).

We initially assessed nine housing characteristics that potentially modified the concentration of indoor BC, and five housing characteristics were selected through the model development phase of the current study. Overall, we suggest that these five characteristics serve as better proxies or predictors of indoor BC than other housing characteristics and conditions documented in the study (presence/absence of kitchen exhaust hoods, use of fireplace, cleaning activities, and distance to the nearest major road). An explanation for the low variation of indoor BC explained by the presence/absence of kitchen exhaust hood, could be the unknown frequency of the use of kitchen exhaust hoods during the sampling periods. In the questionnaires administered, the subjects were only asked how often they used an exhaust hood in the kitchen. However, they were not explicitly asked if they operated their exhaust hoods during the sampling periods.

It was unexpected that the use of fireplace was not a proxy for indoor BC. The main reason could be the low number of samples collected while a fireplace was being used: in only 2 of the 45 visits. Furthermore, different types of woods used in fireplaces and differing intensities of the fire emit varying levels of BC (Fine et al., 2001). Results from measurements of PM made during the burning of six types of wood in fireplaces indicate that at least 80% of PM emitted from burning woods in fireplaces are organic carbon and not black carbon (Fine et al., 2001). In addition, most PM emitted from a wood-burning fireplace is directed to the chimney (Stone, 1969). Therefore, the particle transport from the fireplace to other parts of the indoor environment is limited. In contrast, combustion particles produced by burning candles may remain airborne for extended periods, which increases indoor BC.

The relative adhesive force of PM on surfaces increases as particle aerodynamic diameter decreases (Hinds, 1982). Consequently, the settled BC particles (which are ultrafine) may not be easily removed by air turbulence and human activities, which are naturally associated with cleaning (Hinds, 1982). This may explain why vacuuming, sweeping, and dusting were not found to be good predictors of indoor BC level. It is acknowledged that the dominating source of outdoor BC is from outdoor sources such as diesel vehicular emissions (World Health Organization (WHO), 2012). Thus, distance to a major road is likely a proxy of outdoor BC concentrations. This gives a possible reason why outdoor BC concentration and not distance to the nearest major road explained more variation in indoor BC.

4.4. Application of the predictive model

The model can be used as a predictive model to support risk assessment in public health. The model provides the first step at anticipating cumulative exposure levels to BC, because cumulative exposure level is a function of indoor concentration (which the models provide), outdoor concentration and time spent indoors and outdoors (duration of exposure) (Dimitroulopoulou et al., 2001; Zeger et al., 2000). In some regions, outdoor levels of BC can be obtained from stationary monitoring stations and output of predictive models (Gryparis et al., 2007). This suggests that the estimation of average exposure level to BC is achievable when time spent indoors and outdoors is known.

One variable that still requires actual measurement for the utilization of the model presented in the current study is air infiltration. However, estimates of air infiltration can be made based on existing models, which are discussed. Infiltration is a function of air leakage area, stack coefficient, difference between indoor and outdoor temperature, wind coefficient and average windspeed (American Society of Heating, 2017). Building age, building size, and other household features have been used to predict air leakage area and the models have been presented in peer-reviewed studies (Chan et al., 2005; Chan, 2013). Furthermore, the ventilation and infiltration chapter of the ASHRAE Fundamentals contains empirical values of stack and wind coefficients based on wind speed, direction, and building shape and geometry (American Society of Heating, 2017). Therefore, one can conveniently obtain air infiltration upon readily available weather data and building information (e.g., age, size, shape), and incorporate the value obtained into the model in the current study to provide an anticipated level of indoor BC. Due to the cost of environmental monitoring, it is often not feasible to directly measure BC inside multiple individual homes in large-scale population-based studies. The presented model for indoor BC can be used when regional estimates of indoor BC are needed to support risk assessment in public health practice.

5. Limitations

The subjects in this study were not specifically asked if they used their kitchen exhaust hoods during each 48-h sampling period. However, we expected that subjects would make use of this appliance for the preparation of at least one out of the six meals in a 48-h period. Consequently, a cumulative effect of exhaust hoods on indoor BC was assessed. Our sample size was not large enough to detect seasonal differences that may be associated with BC. Indoor BC was only performed in the bedroom and may not be close to an indoor source of BC. In addition, information on lit candles, and window opened during the sampling period were based on questionnaire data which can have recall bias. However, this method was employed in order to reduce the severity of subject recall bias which may occur when subjects are asked to quantify the frequency of window opening and the number of candles lit during the sampling periods. Due to the categorical structure of the variable on candle use, the model does not account for extreme situations where home occupants light numerous candles that are not typical for the average home occupant. Furthermore, the concentration of BC migrating through a window may vary with the window area and weather conditions. There is a potential for selection bias which can reduce the generalizability of the study findings, as samples of BC were collected only from houses with ≥0.33 $\mu g/m^3$ outdoor elemental carbon attributable to traffic. However, the study provides information that can be used to conduct a similar study in remote locations where elemental carbon attributable to traffic is likely to be < 0.33 μg/m (Janssen et al., 1997).

6. Conclusions

The data show that home characteristics and outdoor BC concentrations can be used to predict indoor BC levels with reasonable accuracy. In the current study, the most significant sources of indoor BC were outdoor BC and lit candles, whereas the HVAC system with HEPA/electrostatic filters was the most significant home appliance that reduced indoor BC. It is recommended that occupants, who burn candles and/or have homes situated in locations with high outdoor BC levels, consider installing HEPA filters in their HVAC systems. Housing conditions that include the presence of electrostatic or HEPA filter in the HVAC system and no lit candles facilitate low indoor BC concentrations.

Conflicts of interest

None.

Acknowledgment

This study was supported by the United States Department of Housing and Urban Development (Grant OHHHU0027-14). K.I. was funded by the University of Cincinnati Graduate Assistantship and Graduate Scholarship. Additional support was provided by the National Institutes of Health (Grant P30ES009089).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2018.12.053.

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