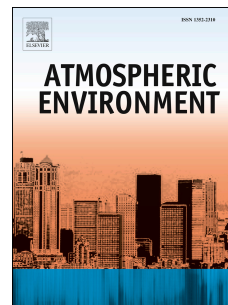


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Predicting Indoor Concentrations of Black Carbon in Residential Environments

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1 **Predicting Indoor Concentrations of Black Carbon in Residential**
2 **Environments**

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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.

Keywords: black carbon; exposure; modeling; estimation.

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ACCEPTED MANUSCRIPT

86 1.0 INTRODUCTION

87 Exposure to traffic-related air pollution has been associated with adverse health effects.^(1, 2)
88 Black carbon (BC) is an example of a traffic-related air pollutant and is used as a surrogate of
89 traffic-related particles.^(3, 4) During the cold season (September 1 – March 31), exposure to BC is
90 associated with cough among children.⁽⁵⁾ Black carbon is also linked to the prevalence of
91 bronchitis and asthma in children,⁽⁶⁾ and respiratory hospitalizations among the elderly.⁽⁷⁾

92
93 Black carbon is a descriptive term for light-absorbing particles that represent a continuum of
94 incomplete combustion residues ranging from larger charred materials that retain structural
95 information of parent materials to highly condensed refractory soot particles that are produced
96 from incomplete combustion.⁽⁸⁾ Soot particles include organic carbon and black carbon particles
97 derived from combustion.⁽⁹⁾ They are nanometer to submicrometer in aerodynamic diameter,⁽¹⁰⁾
98 and can be emitted from the exhausts of internal combustion engines.⁽¹¹⁾ Chars are large particles
99 that do not travel far. Consequently, in most filter-based measurements of airborne particulate
100 matter (PM), BC mainly consists of soot particles that usually contain other atoms and attached
101 organics such as polycyclic aromatic hydrocarbons.⁽⁸⁾ Soot is determined by optical methods and
102 chemical-thermal methods. Soot determined via optical methods is referred to as black carbon.
103 The term elemental carbon is used when soot is determined by chemical-thermal methods that
104 measure the amount of CO₂ evolved. There is a high correlation ($r = 0.95$) of soot results
105 obtained with optical methods and chemical-thermal methods.⁽¹²⁾ Due to this high correlation, the
106 terms black carbon and elemental carbon are often used interchangeably.⁽¹³⁾ Using light
107 absorption of colored particles at one or more wave lengths, optical absorption techniques have
108 been utilized to differentiate black carbon from other colored components such as particles from
109 cigarette smoke.^(8, 14) Majority of colored components of PM, such as cigarette smoke, are
110 colored organic carbon, and not black carbon, as they make sampling filters yellow-brown and
111 not black. It is estimated that <1% of PM emitted from burning cigarettes have light-absorbing
112 properties of black carbon.⁽¹⁵⁾

113
114 Black carbon (BC) can be emitted from any incomplete combustion source. For indoor
115 environments, examples of BC sources include lighting or extinguishing candles, using kerosene
116 lamps, charring food, and cooking or heating with solid fuels.^(11, 16, 17) Cleaning activities, such as
117 vacuuming carpets, can cause resuspension of indoor particles with aerodynamic diameter ≤ 10
118 μm , which results in increased indoor aerosol concentrations.⁽¹⁸⁾ In urban settings, exhaust
119 emissions from traffic and especially older diesel engines are one of the major contributors to
120 ambient BC.⁽¹¹⁾ Thus, the distance of a home to a road with high vehicular traffic may modify
121 indoor BC concentrations. Other factors are also associated with indoor BC levels. Quantifying
122 the factors which modify indoor BC should enhance any predictive model for indoor BC
123 concentrations. Modeling residential indoor BC concentrations is useful for estimating average
124 exposure, given that people typically spend 64 – 66% of their time indoors at their residences.⁽¹⁹⁻
125 ²¹⁾ In addition, subgroups such as infants, the elderly, stay-at-home parents and people who work
126 from home spend much higher fractions of their time at their residences. Modeling residential
127 indoor BC concentrations would facilitate risk assessment in public health when population
128 exposure to BC is estimated.

129
130 As BC refers to light-absorbing particles, any indoor air quality (IAQ) intervention that aims at
131 reducing indoor particles, may also reduce indoor BC. Examples of IAQ interventions include

132 equipping the heating, ventilation and air conditioning (HVAC) systems with efficient air
133 filters,⁽²²⁾ operating kitchen exhaust hoods with recirculated air through a filter or outdoor
134 exhaust.^(23, 24) Indoor-outdoor air exchange modifies indoor pollutant levels.⁽²⁵⁾ The air exchange
135 rate is affected by infiltration and exfiltration via unintentional leaks in a building envelope, open
136 windows or doors and mechanical ventilation.⁽²⁶⁾ Pressurized fan tests are used to measure
137 building air tightness⁽²⁷⁾, an indicator of air infiltration via unintentional leaks in a building
138 envelope. Furthermore, air leakage attributable to different building components can be
139 estimated.⁽²⁸⁾

140
141 A model to accurately predict indoor BC can be developed based on the information about the
142 above-listed home characteristics. After such a predictive model is developed, its performance
143 should be assessed through validation methods, e.g., cross-validation to ensure the accuracy of
144 the model output.⁽²⁹⁾ There have been attempts to establish relationships between specific
145 environmental characteristics and the carbon particle levels in residential settings. Baxter et al.
146 used home characteristics, occupant activities and traffic indicators to predict indoor elemental
147 carbon determined by PM_{2.5} filter reflectance analysis.^(30, 31) Because the method of analysis used
148 by Baxter et al. is an optical method, the measured particles can be regarded as black carbon.

149
150 Baxter et al. developed two models, in which factors such as an increase in ambient BC and the
151 close proximity of a home with windows kept open to a road with high truck counts were both
152 significantly associated with an increase in indoor BC.^(30, 31) However, some home characteristics
153 and occupant activities that can potentially modify indoor PM, e.g., electrostatic/HEPA HVAC
154 filters were not incorporated into the models developed by Baxter et al. Therefore, the need
155 remains for an advanced predictive model incorporating real-life multiple housing characteristics
156 that can potentially modify indoor BC concentration. The goal of this study was to develop such
157 a predictive model for indoor BC. We utilized multiple housing characteristics as covariates and
158 assessed the predictive performance of the model with a leave-one-out cross-validation method.

159 160 **2.0 METHODS**

161 162 *2.1 Study Overview*

163 The study was conducted in 23 residential environments (single-family and apartment buildings)
164 in the Cincinnati-Kentucky-Indiana tristate region (Figure S1) from September 2015 through
165 August 2017. The BC levels were measured indoors and outdoors, and home characteristics
166 specific to each dwelling were documented. The homes in this study belonged to a cohort of
167 subjects from another ongoing study.⁽³²⁾ The ongoing study was focused on the efficiency of air
168 cleaners in removing indoor particles, but only baseline measurements (before the deployment of
169 air cleaners) were included in this study. All homes were located in neighborhoods with ≥ 0.33
170 $\mu\text{g}/\text{m}^3$ outdoor elemental carbon attributable to traffic, as determined in a previous study.⁽³³⁾ The
171 study received Institution Review Board (IRB) approval from the University of Cincinnati IRB.

172 173 *2.2 Environmental Monitoring*

174 Samples of airborne fine particulate matter (PM_{2.5}) were collected simultaneously from inside
175 and outside of each residence over 48 hours using single-stage Personal Modular Impactors
176 (SKC, Inc., Eighty Four, PA) equipped with 37-mm Teflon filters. Measurements were repeated
177 twice in each home. Indoor samples were collected in a bedroom and outdoor samples in the

178 immediate vicinity (backyard or in front) of the home. Sampling pumps were calibrated to a flow
179 rate of 3 L/min using a mass flow meter (TSI Inc., Shoreview, MN). Measurements were
180 repeated twice in each home with a 2-month gap between the two measurements. After
181 gravimetric determination of the PM_{2.5}, the filter samples were analyzed for BC by optical
182 absorption technique⁽⁸⁾, which has a published limit of detection (LOD) of 1.4 ng/mm² of the
183 filter (equivalent to an air concentration of 0.12 µg/m³ in this study). Media and field blanks were
184 collected in parallel at a rate equal to 10% of all filter samples. The mean concentration of BC in
185 the blank samples was 0.25 ng/mm² of the filter (equivalent to an air concentration of 0.07 µg/m³
186 in this study). This value was subtracted from the BC measured on the real samples.

188 2.3 Documenting Housing Characteristics

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

- 193 • Exhaust hood in the kitchen – yes or no.
- 194 • Presence of electrostatic filter or high-efficiency particulate air (HEPA) filter in the HVAC
195 system – yes or no.
- 196 • Lit candles during the sampling period – yes or no.
- 197 • Use of fireplace during the sampling period – yes or no.
- 198 • At least one open window during the sampling period – yes or no.
- 199 • Cleaning (vacuuming or sweeping or dusting) during the sampling period – yes or no.

200
201 Other housing characteristics assessed by the study team included the distance of the home to the
202 nearest state highway or federal interstate (major road) and the annual average rate of air
203 infiltration in the home. The distance of the homes to the nearest major road was calculated with
204 a geographical information system (ArcGIS 9.0, Environmental Systems Research Institute, Inc.,
205 Redlands, CA).⁽³³⁾ Data on the geographical location of the major roads were obtained from the
206 Ohio Department of Transportation (2004) and the Kentucky Transportation Cabinet (2006).⁽³³⁾
207 The annual average rate of air infiltration via unintentional leaks in the home was determined
208 from measurements of a blower door system in accordance with the ASTM standard for fan
209 pressurization tests.⁽²⁷⁾ The blower door system includes a fan, which is positioned at an exterior
210 door in a building. Before the start of the blower door measurement, all exterior doors and
211 windows in a home were shut, and the interior doors were opened. A baseline building pressure
212 was measured with the blower door system, and the blower door fan was utilized to induce
213 pressure differences between indoor and outdoor of 10 to 60 Pa with 5 Pa increments.⁽²⁷⁾
214 Building air tightness was derived from the blower door system by recording the airflow needed
215 to establish the above-indicated pressure differences (10 – 60 Pa), and a summary of the test was
216 reported through a proprietary software (TECTITE).⁽³⁴⁾ The software was programmed to
217 calculate an annual average rate of natural air infiltration based on the American Society of
218 Heating, Refrigerating and Air-Conditioning (ASHRAE) standard.⁽³⁵⁾ Results from the blower
219 door system give an estimation of the number of air changes per hour through unintentional leaks
220 such as cracks and holes in the building envelope.

222 2.4 Statistical Analysis

223 In the current study, each home was assigned an identification number. A linear mixed-effect
224 model was used for predicting indoor BC. Outdoor measured BC concentrations and documented
225 housing characteristics were assigned as fixed effects, and home identification numbers were
226 treated as random effects. Statistical analyses were done with R studio.⁽³⁶⁾ To assess the effect of
227 housing characteristics and outdoor BC concentration on indoor BC, the predictive model was
228 developed in three stages. First, an all subsets regression analysis was conducted using lowest
229 Bayesian information criterion (BIC) for model selection. Second, indoor sources of BC and
230 housing characteristics that represented the infiltration of BC, but were not included in the model
231 obtained from the all subset regression analysis were added one at a time. This was done because
232 all the documented home characteristics in this study were potential modifiers of indoor BC and
233 BIC is designed to penalize predictor variables as the sample size increases.⁽³⁷⁾ Third, each time a
234 new independent variable was added to the model obtained from the All Subset Regression
235 analysis, the predictive accuracy of the model was assessed with a leave-one-out cross-validation
236 method. The version of the model that yielded the highest out-of-sample R^2 and lowest root
237 mean squared error (RMSE) was selected as the final model for the prediction.

238
239 The method for utilizing leave-one-out cross-validation has been reviewed by Arlot et al.⁽²⁹⁾ In
240 summary, one observation from the dataset used to develop the predictive model was removed,
241 and the predictive model was rebuilt again. Regression estimates of this rebuilt model were used
242 to predict the indoor concentration of BC in the observation that was removed. The removed
243 observation was then returned to the dataset, and another observation was removed, after which
244 the predictive model was rebuilt again. Next, the indoor concentration of BC in the newly
245 removed data point was predicted with the regression estimates of the new predictive model.
246 This process was done 45 times (number of observations in the dataset).

247 248 *2.5 Handling Non-Detectable Measurements of BC*

249 Concentrations of indoor BC were skewed (geometric standard deviation = 3). Twenty-four
250 percent (24%) of indoor BC samples and 2% of outdoor BC samples were below the LOD of
251 $0.12 \mu\text{g}/\text{m}^3$. All samples below the LOD were replaced with the value of LOD/2 as
252 recommended by Hornung et al.⁽³⁸⁾

253 254 **3.0 RESULTS**

255 256 *3.1 Measurements and Housing Characteristics*

257 After measurements were repeated twice in the 23 homes, one observation was lost in one home
258 due to a pump failure. Consequently, there were 45 observations from the 23 homes. Table 1
259 presents data on sample collection in the current study stratified by seasons. Sample
260 measurements were obtained from the fall, winter, summer and spring seasons (22.2%, 24.4%,
261 28.9% and 24.4%, respectively). Table 2 presents categorical characteristics of the homes
262 utilized in this study. Of the 45 visits, at least one window was opened in 29 of them. Candles
263 were lit during 6 of the visits. Cleaning activities were performed in the homes during 28 visits.

264
265
266

267 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%

268 Seasons = astronomical seasons (obtained from the National Centers for Environmental
 269 Information)⁽³⁹⁾

270

271

272 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

273

274

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

281

282

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

Home characteristics	Q1	Median	Mean	Q3	SD
Indoor BC ($\mu\text{g}/\text{m}^3$)	0.13	0.28	0.43	0.62	0.42
Outdoor BC ($\mu\text{g}/\text{m}^3$)	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

284 BC = black carbon, Q1 = 25th percentile, Q3 = 75th percentile, Indoor/outdoor ratio of BC is based on individual
 285 home ratios. LOD for BC samples = $0.12 \mu\text{g}/\text{m}^3$
 286

287 3.2 Inferential Information from the Predictive Model

288 Of the nine housing characteristics and occupant activities investigated, only five (outdoor BC,
 289 average annual air infiltration via unintentional leaks, HEPA/electrostatic HVAC filter,
 290 open/closed windows, candles) were selected as predictors of indoor black carbon in the chosen
 291 predictive model (equation 1).
 292

$$293 \text{Indoor BC concentration}_{it} = b_i + \beta_0 + \beta_1 \times \text{outdoor BC concentration}_{it} + \beta_2 \times \\
 294 \text{average annual air infiltration via unintentional leaks}_i + \beta_3 \times \text{HVAC filter}_i + \beta_4 \times \\
 295 \text{open windows}_{it} + \beta_5 \times \text{lit candles}_{it} + \epsilon_{it} \quad (1)$$

296
 297 Where, *indoor BC concentration*_{it} is the measured indoor BC in each home *i* on sampling visit *t*, *b_i* is a random
 298 intercept specific to each home *i*, *β₀* is the fixed intercept, *β₁* is the effect of the measured BC in the local outdoor
 299 environment of each home *i* on sampling visit *t*, *β₂* is the effect of average annual air infiltration via unintentional
 300 leaks in each home *i*, *β₃* is the effect of electrostatic/HEPA HVAC filter in each home *i*, *β₄* is the effect of open or
 301 closed windows in each home *i* during visit *t*, *β₅* is the effect of presence or absence of lit candles or absence of lit
 302 candles in each home *i* during visit *t*, *β₆* is the effect of the presence or absence of a kitchen exhaust hood in each
 303 home *i* and *ε_{it}* is the residual error in the model.
 304

305 Table 4 presents the results of the regression model obtained using the complete dataset. These
 306 results are based on 48-hour averages of the indoor BC levels in the 45 sampling events. The
 307 independent variables (covariates) that were significantly associated with indoor BC
 308 concentration were outdoor BC concentration, electrostatic/HEPA HVAC filters, and lit candles.
 309 An increase of $1 \mu\text{g}/\text{m}^3$ in outdoor BC was associated with $0.53 \mu\text{g}/\text{m}^3$ increase in indoor BC
 310 (Table 4). With other covariates being equal between the two groups, homes with efficient
 311 HVAC filters were associated with $0.26 \mu\text{g}/\text{m}^3$ decrease in indoor BC when compared to homes
 312 without HVAC filters. Homes where candles were lit, had $0.41 \mu\text{g}/\text{m}^3$ higher indoor BC when
 313 compared to homes where candles were not lit, with other covariates being equal between the

314 two groups. Indoor BC was positively associated with open windows and average air infiltration
 315 (albeit not significant) (Table 4).

316

317 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 *

318 ¹Results are applicable to 48-hour average of indoor BC, coefficient of multiple determination (R^2) = 0.71, root
 319 mean squared error = 0.71.

320 ²* indicates statistically significant variables ($P < 0.05$)

321 ³Reference group = No.

322

323 3.3 Predictive Capability of the Model

324 Figure 1 shows a scatter plot of the measured indoor BC concentrations and the predicted indoor
 325 BC concentrations as obtained from the leave-one-out cross-validation algorithm. Measured
 326 indoor BC concentrations ranged from 0.06 to 2.18 $\mu\text{g}/\text{m}^3$ (mean = 0.43 $\mu\text{g}/\text{m}^3$, SD = 0.42);
 327 predicted indoor concentrations ranged from -0.09 to 1.70 $\mu\text{g}/\text{m}^3$ (mean = 0.43 $\mu\text{g}/\text{m}^3$, SD =
 328 0.38). Negative predicted values are assumed to be < LOD. The predicted indoor BC
 329 concentrations explained 78% of the variability in measured indoor BC concentrations (Out-of-
 330 sample $R^2 = 0.77$). The standard deviation of the unexplained variance in measured indoor BC
 331 concentration was 0.20 $\mu\text{g}/\text{m}^3$ (root-mean-squared error, RMSE).

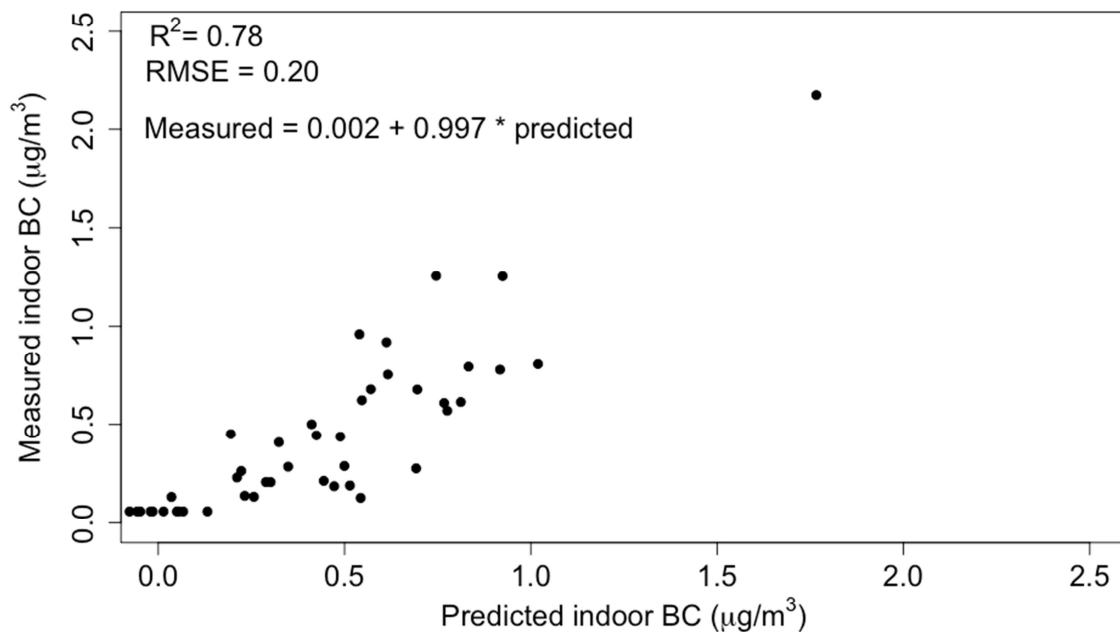
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333 3.4 Sensitivity Analysis

334 Using the complete dataset, the result of a univariate model that had only outdoor BC as a
 335 covariate yielded an R^2 of 49% (Table S1). This was a 22% loss in R^2 when compared to the
 336 final model that included indoor covariates in addition to outdoor BC (Table 4). Removing the
 337 insignificant covariates from the final model in Table 4 (average annual infiltration and open
 338 windows) and rerunning the model did not considerably change the regression estimates in Table
 339 4 (Table S2). Likewise, the out-of-sample R^2 obtained from the leave-one-out cross-validation
 340 method ($R^2 = 76\%$) (Figure S4) was similar to that obtained in the model that included the
 341 insignificant variables ($R^2 = 78\%$) (Figure 1). Sensitivity analysis, performed where season was
 342 added to the final model, showed that the effect of season on indoor BC was not significant
 343 (Table S3). Furthermore, results of the leave-one-out cross-validation method indicates that the
 344 model which includes season as a covariate had a slight increase in error (RMSE) and explained
 345 less variability in indoor BC (Figure S5) when compared to the final model (Figure 1).

346

347 Table S4 presents the final model that was developed from the complete dataset but with the
 348 exclusion of one influential observation. Removing the influential observation resulted to a 10%
 349 loss in R^2 (Table S4). In this model, $1 \mu\text{g}/\text{m}^3$ increase in outdoor BC was associated with 0.43
 350 $\mu\text{g}/\text{m}^3$ increase in indoor BC (compared to $0.53 \mu\text{g}/\text{m}^3$ in the final model containing the
 351 influential observation). Other regression estimates in both models (models with and without the
 352 influential observation) were similar (Table S4 and Table 4). The influential observation was the
 353 observation with the maximum measured indoor and outdoor BC ($2.2 \mu\text{g}/\text{m}^3$ and $3.6 \mu\text{g}/\text{m}^3$,
 354 respectively) (Figure 1). Using the dataset that did not contain the influential observation, the
 355 result of a univariate model that only had outdoor BC as a covariate yielded an R^2 of 21% (Table
 356 S5). This is a 40% decrease in R^2 when compared to the model R^2 obtained from using both
 357 indoor factors and outdoor BC as predictors of indoor BC (Table S4). Cross-validation of the
 358 model without the influential observation showed that the model explained less variation in
 359 indoor BC (Figure S6) when compared to the validation of the final model (Figure 1).



360 Figure 1: Scatter plot of measured indoor BC and predicted indoor BC levels obtained from leave-one-out cross-
 361 validation. RMSE = root-mean-squared error of the predictive model.
 362

363
 364 Using measurements of average local outdoor BC concentration and average air infiltration rate
 365 of a building, combined with the home conditions in the presented final model, estimates of
 366 average indoor BC can be obtained in real-life scenarios (equation 2).
 367

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

371
 372
 373

4.0 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.,^(30, 31) 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 = $0.28 \mu\text{g}/\text{m}^3$, outdoor = $0.68 \mu\text{g}/\text{m}^3$) 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 $0.49 \mu\text{g}/\text{m}^3$ and $0.55 \mu\text{g}/\text{m}^3$ indoors and outdoors, respectively (I/O = 0.89).⁽³¹⁾ Coombs et al. reported median BC in Cincinnati to be $0.99 \mu\text{g}/\text{m}^3$ and $0.94 \mu\text{g}/\text{m}^3$ in indoor and outdoor environments, respectively (I/O = 1.05).⁽⁴⁰⁾ Furthermore, 48-hour mean I/O of BC in New York City was 0.93 and 0.84 during the summer and winter seasons, respectively.⁽⁴¹⁾ 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 $\text{PM}_{2.5}$ 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 $1 \mu\text{g}/\text{m}^3$ in outdoor BC was associated with an increase of $0.53 \mu\text{g}/\text{m}^3$ 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.,⁽³⁰⁾ 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,⁽¹¹⁾ 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 $0.41 \mu\text{g}/\text{m}^3$ increase in indoor BC when compared to homes where candles were not lit. Paraffin wax is a common type of candle wax that contains

420 heavy hydrocarbon chains with carbon chain lengths that can be greater than 50 (C_{50}).⁽⁴²⁾ This
421 may explain why the effect of burning candles in only 6 of the 45 sampling periods in this study
422 (13%) was sufficient enough to have a significant increase in indoor BC. Interestingly, despite
423 the relatively large proportion of study homes with lit candles (26%), there was no statistically
424 significant increase in indoor BC attributable to lit candles in the study by Baxter et al.⁽³¹⁾ A
425 reason for this discrepancy could be that some types of candles emit negligible amounts of BC.
426 Further research into BC emissions from different types of candle wax will aid the understanding
427 of the observed differences. Already, it is known that scented candles emit ultrafine particles
428 (size of BC particles)⁽⁴³⁾ about twice less in concentration when compared to pure wax
429 candles.⁽⁴⁴⁾ Moreover, the concentration of BC particles emitted from unsteady burning candles
430 (light and extinguish) is greater than that of steady burning candles.⁽⁴⁵⁾

431
432

433 *4.2 Housing Characteristics/Occupant Activities Associated with a Decrease in Indoor BC*

434 In the study homes, electrostatic or HEPA HVAC filter was the most significant variable that
435 reduced indoor BC. Homes with electrostatic or HEPA HVAC filters had $0.26 \mu\text{g}/\text{m}^3$ decrease in
436 indoor BC when compared to homes without such filters. This finding is expected, given that the
437 efficiency of HEPA filters is $> 99.97\%$.⁽⁴⁶⁾ Electrostatic filters reduce PM by charging and
438 trapping PM on oppositely charged plates.⁽⁴⁷⁾ It would be interesting to distinguish between the
439 effects of HEPA and electrostatic HVAC filters on indoor BC. However, studying the effect of
440 the specific type of filter in the HVAC systems was outside the scope of the study.

441

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

446

447 *4.3 Housing Characteristics/Occupant Activities that Explained Less Variability in Indoor BC*

448 Of the five housing characteristics in the predictive model, average annual air infiltration via
449 unintentional leaks and open windows did not significantly modify indoor BC. Including these
450 variables added some predictive power to the model (higher R^2). One reason for the non-
451 significant findings can be a result of low statistical power ($n = 45$). Furthermore, the effect of
452 open windows is complex, as it facilitates outdoor-indoor transport of particles from outdoor
453 sources, but can also facilitate exfiltration of particles produced from indoor sources. This may
454 explain why the effect of windows kept open was not significant. PM infiltrates from local
455 outdoor environments,⁽⁴⁸⁾ and indoor pollutants can accumulate in homes with very tightly sealed
456 building envelopes.⁽⁴⁰⁾ Our results show that one unit increase in air exchange rate via
457 unintentional leaks in a building envelope was associated with $0.09 \mu\text{g}/\text{m}^3$ increase in indoor BC.
458 This increase was not significant, likely due to the losses through Brownian diffusion of $\text{PM} \leq$
459 $0.1 \mu\text{m}$ ⁽⁴⁹⁾ (size of BC particles)⁽⁴³⁾ which decreases the infiltration factor.⁽⁵⁰⁾

460

461 We initially assessed nine housing characteristics that potentially modified the concentration of
462 indoor BC, and five housing characteristics were selected through the model development phase
463 of the current study. Overall, we suggest that these five characteristics serve as better proxies or
464 predictors of indoor BC than other housing characteristics and conditions documented in the
465 study (presence/absence of kitchen exhaust hoods, use of fireplace, cleaning activities, and

466 distance to the nearest major road). An explanation for the low variation of indoor BC explained
467 by the presence/absence of kitchen exhaust hood, could be the unknown frequency of the use of
468 kitchen exhaust hoods during the sampling periods. In the questionnaires administered, the
469 subjects were only asked how often they used an exhaust hood in the kitchen. However, they
470 were not explicitly asked if they operated their exhaust hoods during the sampling periods.

471
472 It was unexpected that the use of fireplace was not a proxy for indoor BC. The main reason could
473 be the low number of samples collected while a fireplace was being used: in only 2 of the 45
474 visits. Furthermore, different types of woods used in fireplaces and differing intensities of the
475 fire emit varying levels of BC.⁽⁵¹⁾ Results from measurements of PM made during the burning of
476 six types of wood in fireplaces indicate that at least 80% of PM emitted from burning woods in
477 fireplaces are organic carbon and not black carbon.⁽⁵¹⁾ In addition, most PM emitted from a
478 wood-burning fireplace is directed to the chimney.⁽⁵²⁾ Therefore, the particle transport from the
479 fireplace to other parts of the indoor environment is limited. In contrast, combustion particles
480 produced by burning candles may remain airborne for extended periods, which increases indoor
481 BC.

482
483 The relative adhesive force of PM on surfaces increases as particle aerodynamic diameter
484 decreases.⁽⁵³⁾ Consequently, the settled BC particles (which are ultrafine) may not be easily
485 removed by air turbulence and human activities, which are naturally associated with cleaning.⁽⁵³⁾
486 This may explain why vacuuming, sweeping, and dusting were not found to be good predictors
487 of indoor BC level. It is acknowledged that the dominating source of outdoor BC is from outdoor
488 sources such as diesel vehicular emissions.⁽¹¹⁾ Thus, distance to a major road is likely a proxy of
489 outdoor BC concentrations. This gives a possible reason why outdoor BC concentration and not
490 distance to the nearest major road explained more variation in indoor BC.

491 492 *4.4 Application of the predictive model*

493 The model can be used as a predictive model to support risk assessment in public health. The
494 model provides the first step at anticipating cumulative exposure levels to BC, because
495 cumulative exposure level is a function of indoor concentration (which the models provide),
496 outdoor concentration and time spent indoors and outdoors (duration of exposure).^(54, 55) In some
497 regions, outdoor levels of BC can be obtained from stationary monitoring stations and output of
498 predictive models.⁽⁵⁶⁾ This suggests that the estimation of average exposure level to BC is
499 achievable when time spent indoors and outdoors is known.

500
501 One variable that still requires actual measurement for the utilization of the model presented in
502 the current study is air infiltration. However, estimates of air infiltration can be made based on
503 existing models, which are discussed. Infiltration is a function of air leakage area, stack
504 coefficient, difference between indoor and outdoor temperature, wind coefficient and average
505 windspeed.⁽²⁸⁾ Building age, building size, and other household features have been used to
506 predict air leakage area and the models have been presented in peer-reviewed studies.^(57, 58)
507 Furthermore, the ventilation and infiltration chapter of the ASHRAE Fundamentals contains
508 empirical values of stack and wind coefficients based on wind speed, direction, and building
509 shape and geometry.⁽²⁸⁾ Therefore, one can conveniently obtain air infiltration upon readily
510 available weather data and building information (e.g., age, size, shape), and incorporate the value
511 obtained into the model in the current study to provide an anticipated level of indoor BC. Due to

512 the cost of environmental monitoring, it is often not feasible to directly measure BC inside
513 multiple individual homes in large-scale population-based studies. The presented model for
514 indoor BC can be used when regional estimates of indoor BC are needed to support risk
515 assessment in public health practice.

516

517 **5.0 LIMITATIONS**

518

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

537

538 **6.0 CONCLUSIONS**

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

547

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553

554 **Competing Interests**

555 Declarations of interest: none.

556

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731

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)

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 ($\mu\text{g}/\text{m}^3$)	0.13	0.28	0.43	0.62	0.42
Outdoor BC ($\mu\text{g}/\text{m}^3$)	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 = $0.12 \mu\text{g}/\text{m}^3$

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 *

¹Results are applicable to 48-hour average of indoor BC, coefficient of multiple determination (R^2) = 0.71, root mean squared error = 0.71.

²* indicates statistically significant variables ($P < 0.05$)

³Reference group = No.

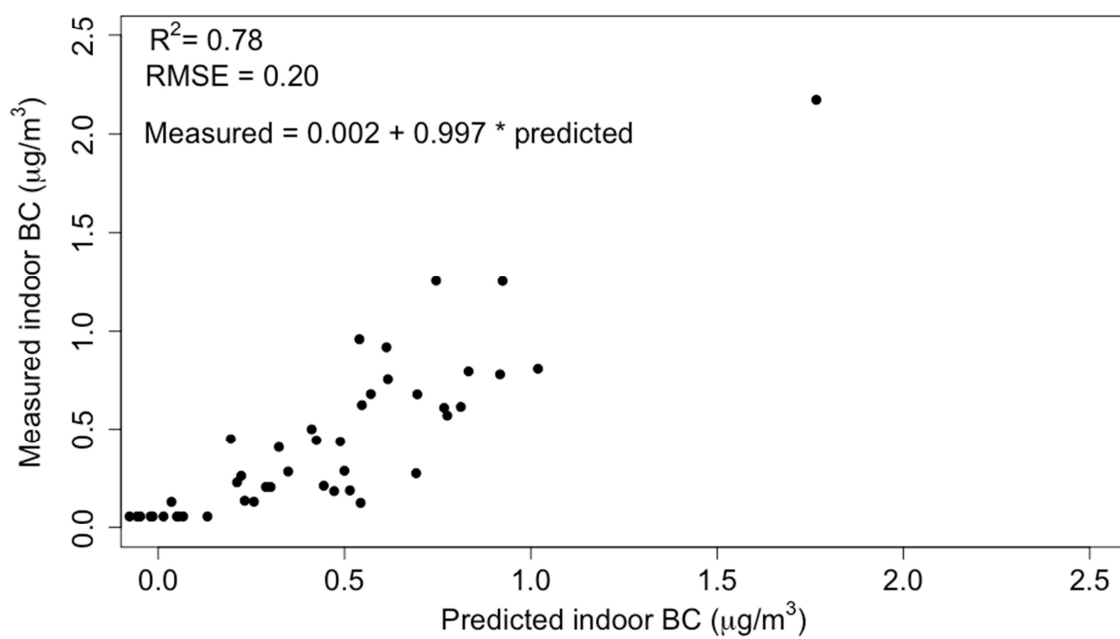


Figure 1: Scatter plot of measured indoor BC and predicted indoor BC levels obtained from leave-one-out cross-validation. RMSE = root-mean-squared error of the predictive model.

- Indoor black carbon (BC) can be predicted from outdoor BC and home characteristics.
- Increased outdoor BC and burning candles are associated with increased indoor BC.
- Outdoor BC explained the most variability in indoor BC.
- Electrostatic or high efficiency particulate air filters reduce indoor BC.
- The median fraction of BC/PM_{2.5} mass is very low (0.09 outdoors and 0.04 indoors).

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: