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The Contribution Of The Residential Environment To Indoor Air Pollutants

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Abstract:

Rationale: Little is known about the home environment and associated indoor exposures to brown carbon and black carbon, components that make up fine particulate air pollution.

Objective: Identify how features of the residential environment contribute to indoor measurements of brown and black carbon.

Methods: Between November 2012 and December 2014, 125 veterans who were part of a COPD cohort were recruited for this study. At roughly 3 month intervals, participants received a particle sampler to measure air pollutants in their home for a 1-week period. The filters within the samplers were analyzed for levels of black and brown carbon using the OT21 Transmissometer. Home environmental questionnaires were completed at baseline and for each measurement period. Outdoor black carbon averages were measured at a central site. Multivariate linear mixed effect modeling with a backward elimination strategy was utilized to generate specific parsimonious models for the dependent variables of indoor black and brown carbon levels.

Main Results: 131 different home addresses were included in the final sample. Indoor candle or incense use, home type, season, air conditioning use and outdoor levels of black carbon significantly predicted indoor black carbon levels in multivariate analysis. Heat type and season significantly predicted indoor brown carbon levels. Additionally, the mean indoor measurements of black carbon (0.688 ± 0.282 µg/m³) were approximately 20% higher than the mean external central site measurements (0.568 ± 0.232 µg/m³).

Conclusions: Home characteristics and the residential environment are associated with indoor air pollutants. Decreased exposure to black carbon and brown carbon, through altering variables in the residential setting, could improve indoor air pollution levels.

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Introduction:

Air pollution is the most significant environmental cause of early morbidity and mortality worldwide, specifically fine particulate matter or particulate matter ≤ 2.5µg ($PM_{2.5}$) (Schraufnagel et al., 2019; Landrigan et al., 2018). $PM_{2.5}$ has been associated with increased risk of many chronic diseases such as cardiovascular disease, chronic obstructive pulmonary disease (COPD), asthma and cancer (Landrigan et al., 2018; Schraufnagel et al., 2019; Kurt, Zhang & Pinkerton, 2016). In recent years, public health studies and initiatives have focused on decreasing exposure to $PM_{2.5}$, as this environmental pollutant has been cited as the fifth leading cause of death worldwide, resulting in over 3.2 million deaths per year (Schraufnagel et al., 2019; Kurt et al., 2016). Previous studies have primarily measured outdoor exposure to air pollution, though indoor exposure is likely more reflective of individual risk. Indoor exposure to $PM_{2.5}$ reflects both indoor and outdoor sources and is often higher than levels measured outdoors (Apelberg et al., 2013; US EPA, 1987). As Americans typically spend most of their time indoors (approximately 90%), it is important to assess how the residential environment contributes to indoor air pollutants (US EPA, 1989).

$PM_{2.5}$ is made up of multiple components, such as organic carbons, ammonium containing compounds and dust (Hao et al., 2005). Carbonaceous aerosols, such as black carbon and brown carbon, are specific components of $PM_{2.5}$ that have been linked to the increased morbidity and mortality associated with $PM_{2.5}$ exposure. In a recent study of patients with COPD, indoor black carbon exposure was associated with increased inflammatory markers (Garshick et al., 2018). Furthermore, black carbon has been associated with increased cardiovascular and lung cancer mortality (Grahame, Klemm & Schlesinger, 2014; Petzold et al., 2013). In population-based mortality studies, it is estimated that reducing a unit of black carbon rather than reducing a unit of $PM_{2.5}$, would improve life expectancy 4-9 times more (Grahame et al., 2014). Black carbon and brown carbon have also been associated with climate change and other negative health effects (Petzold et al., 2013).

Black carbon is an organic carbonaceous material that is produced from the incomplete combustion of biomass and fuels (Petzold et al., 2013; Presler-Jur, Doraiswamy, Hammond & Rice, 2017). It appears black and absorbs light at 880 nm (Brown, Lee, Roberts & Collett, 2016). In the United States, transportation is the major source of black carbon. In developing countries, industrial coal burning and agricultural burns are more prevalent sources of black carbon (Grahame et al., 2014). Although indoor coal burning, smoking and candle use have been associated with higher levels of black carbon, it is thought that the majority of indoor black carbon is from outdoor infiltration (Pagels et al., 2009; Monn, 2000). Therefore, many existing studies use environmental sampling from a centralized monitoring station to estimate residential black carbon exposure rather than directly measuring indoor exposure and little is known about how residential factors impact indoor exposures.

Brown carbon is an organic carbon that is produced from a smoldering, inefficient combustion process, such as wood burning. In contrast to black carbon, it appears brown and absorbs light towards the ultraviolet portion of the spectrum (Grahame et al., 2014). Brown carbon is estimated by the difference between the measured light absorption at 370 nm and the measured black carbon (Petzold et al., 2013; Brown et al., 2016). As brown carbon is a more recently recognized air pollutant, it has not been widely studied (Petzold et al., 2013).

The OT21 Transmissometer is a newer, cost-effective method that has been used to detect black carbon and other sources of air pollution on filters (Presler-Jur et al., 2017) potentially enabling easier area or individual air pollution sampling. In studies, the OT21 Transmissometer has evaluated biomass burning and the production of outdoor air pollutants, where the absorption measurements at different wavelengths can be used to determine the concentration of black carbon and estimate brown carbon (Brown et al., 2016). The OT21 Transmissometer has been shown to effectively measure carbonaceous aerosols and can be used to directly measure indoor air pollutants (Forder, 2014). Per our knowledge, the OT21 Transmissometer has not been previously used to evaluate indoor residential levels of brown and black carbon.

The primary aim of this study is to identify how features of the residential environment contribute to indoor measurements of brown and black carbon, as measured by the OT21 Transmissometer. The authors hypothesize that residential characteristics will contribute to indoor sources of black carbon and brown carbon. Such information could be used to reduce indoor air pollutant levels in the home environment.

Methods:

This study is an observational environmental health study measuring levels of indoor air pollutants within homes. The methods in this paragraph have been previously described in Garshick et al., 2018 and Grady et al., 2018. Between November 2012 and December 2014, 125 veterans at the VA Boston Healthcare System who were part of a COPD cohort examining associations between indoor air quality and health were recruited for this study. All patients were at least 40 years old and had an FEV1/FVC < 0.70. At roughly 3 month intervals, each participant received a Micro-environmental Automated Particle Sampler to measure air pollutants in their home for a 1-week period. Participants completed a baseline home environmental questionnaire at their initial study visit and a questionnaire on residential exposures throughout each particle sampler measurement period. Outdoor black carbon averages for each sampling period were measured at a central site (Francis A. Countway Library, Boston, MA) using an aethalometer (MageeScientific Company, model AE-16, Berkeley, CA), as previously described in Garshick et al., 2018 and Kang et al., 2010.

The Micro-environmental Automated Particle Samplers were collected after each weeklong sampling period. The integrated Teflon filters within the samplers were then analyzed for levels of black carbon and brown carbon using the SootScanTMModel OT21 Transmissometer, a new method that simultaneously measures UV and IR light attenuation at 370 nm and 880 nm to quantify levels of indoor air pollutants. In total, 380 filter measurements were collected on 131 different home addresses.

Initially, bivariate linear mixed (fixed and random) effects models were conducted separately to determine the association of black carbon with brown carbon as well as each of the other study variables. Next, multivariate linear mixed effect modeling was utilized, along with a backward elimination strategy incorporating a significance level to stay of p=0.05, to generate specific parsimonious models for the dependent variables of indoor black carbon level and indoor brown carbon level. To control for multiple comparisons, the Tukey-Kramer method was utilized to obtain an artificial p-value. Statistical significance was defined as p=0.05. All modeling was conducted using SAS version 9.4 (SAS 9.4; SAS Institute, Inc., Cary, NC).

Results:

One hundred thirty one different home addresses were included in the final sample. House characteristics are summarized in Table 1. The most common type of house in this study was a single family home (48.1%). The majority of houses had radiant heat (68.7%) and window unit air conditioning (56.5%). Gas was the most common fuel source (50.4%).

Table 1: House Characteristics of Study Sample (N=131)

House Type	
Single family home	63 (48.1)
Multi-family home	25 (19.1)
Apartment building	40 (30.5)
Trailer or mobile home	3 (2.3)
House Age (years)	65.33 ± 35.00 (8-205)
Distance to nearest major roadway (meters)	240.1 ± 17.8 (0.2 – 2162.9)
Heat Source	
Radiator	90 (68.7)
Forced air	40 (30.5)
Electric space heater	31 (23.7)
Open stove/ Fireplace/ Wood stove	9 (6.9)
Heat Fuel	
Gas	66 (50.4)
Electric	24 (18.3)
Oil	36 (27.5)
Air Conditioning (AC)	
No AC	22 (16.8)
Window units only	74 (56.5)
Central AC only	34 (26.0)
Window units and central AC	1 (0.8%)

* Table values are mean ± SD (range) for continuous variables and N (%) for categorical variables. Numbers may not sum to total (N=131) due to missing data, and percentages may not sum to 100% due to rounding.

Eighteen categorical variables were included in the final linear mixed effects regression analysis. The demographic characteristics of all studied categorical variables and the mean unadjusted indoor levels of brown and black carbon are listed in Table 2. Overall, the filters were collected throughout all seasons. As this study was designed to minimize indoor sources of black carbon, smoking, fireplace use and candle/incense burning were very infrequent during sampling periods. The variables with a p-value < 0.10 on unadjusted analysis are bolded in Table 2.

Table 2: Descriptive Characteristics of Categorical Variables with Mean Indoor Levels of Air Pollutants (Unadjusted)

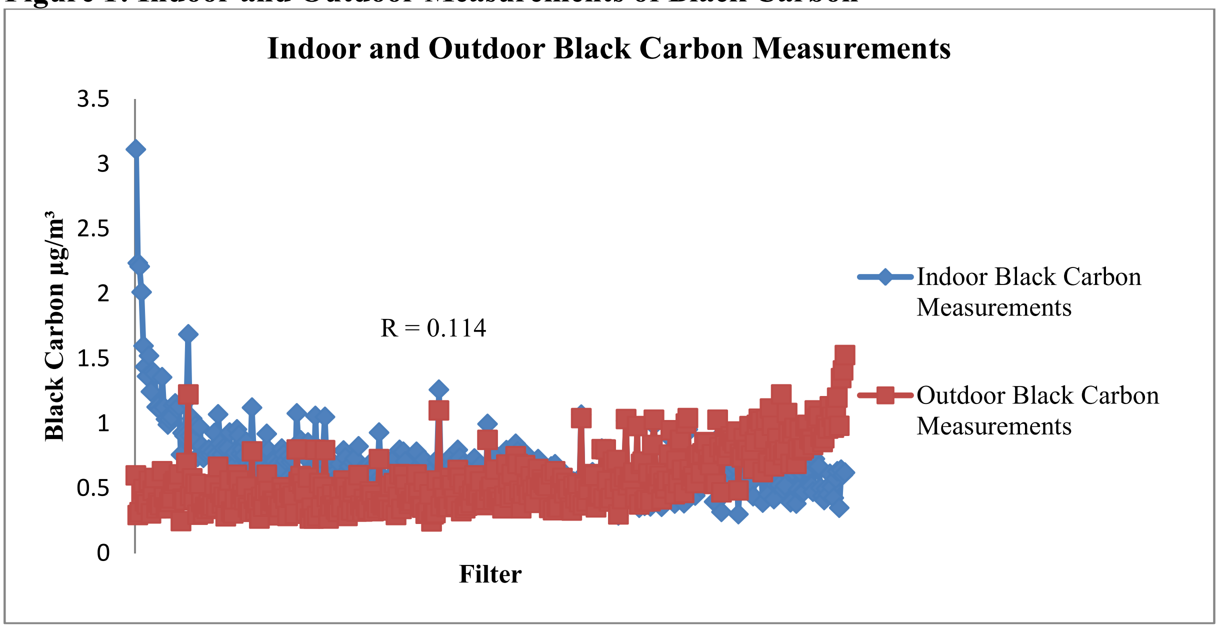
Variable Name	Description	N (%)*	Black Carbon		Brown Carbon	
			Estimate (μg/m³)	p-value	Estimate (μg/m³)	p-value
Season	1 = Winter (Dec, Jan, Feb)	82 (21.6)	0.686	**<0.001**	0.313	**<0.001**
	2 = Spring (Mar, Apr, May)	96 (25.3)	0.573		0.269	
	3 = Summer (Jun, Jul, Aug)	99 (26.1)	0.634		0.153	
	4 = Fall (Sep, Oct, Nov)	103 (27.1)	0.695		0.125	
Home Type	1 = Single family home	183 (48.2)	0.616	**0.059**	0.167	**0.062**
	2 = Multi-family home	68 (17.9)	0.716		0.181	
	3 = Apartment building	120 (31.6)	0.653		0.264	
	4 = Trailer or mobile home	9 (2.4)	0.653		0.236	
Home Age	1 = Up to 40 years old	90 (24.1)	0.675	0.407	0.209	0.767
	2 = 41-90 years old	199 (53.2)	0.631		0.198	
	3 = 91-205 years old	85 (22.7)	0.649		0.178	
Nearest Street Type	1 = Other	217 (57.1)	0.637	0.524	0.176	0.242
	2 = Cross street	33 (8.7)	0.622		0.252	
	3 = Main street	130 (34.2)	0.666		0.219	
Traffic	1 = Bus and car volume low	191 (50.3)	0.639	0.301	0.193	0.976
	2 = Bus or car volume medium	111 (29.2)	0.629		0.195	
	3 = Bus or car volume high	78 (20.5)	0.686		0.202	
Dust	1 = Located near idle, dust or lot	183 (48.2)	0.676	**0.028**	0.200	0.780
	0 = Not located near idle, dust or lot	197 (51.8)	0.620		0.192	
Heat Type	1 = Electric	76 (20.0)	0.647	0.430	0.290	**<0.001**
	2 = Oil or gas	238 (62.6)	0.654		0.156	
	3 = Forced heat with oil or gas	58 (15.3)	0.612		0.292	
Electric Space Heater Used	1 = Yes	46 (12.1)	0.648	0.940	0.296	**0.031**
	0 = No	334 (87.9)	0.645		0.185	
Fireplace/ Open Stove Used	1 = Yes	4 (1.1)	0.712	0.592	0.477	0.234
	0 = No	376 (99.0)	0.645		0.195	
Windows Open	1 = Yes	177 (46.6)	0.643	0.795	0.175	0.122
	0 = No	203 (53.4)	0.648		0.217	
AC Units On	1 = Yes	104 (27.4)	0.625	0.212	0.128	**<0.001**
	0 = No	276 (72.6)	0.654		0.228	
Smoking in Home	1 = Yes	11 (2.9)	0.676	0.664	0.289	0.347
	2 = No	369 (97.1)	0.645		0.194	
Pilot Light for Stove/Oven	1 = Yes	65 (17.1)	0.655	0.725	0.152	0.114
	0 = No	315 (82.9)	0.644		0.207	
Vented Fan in Kitchen	1 = No	205 (54.0)	0.656	0.385	0.201	0.735
	0 = Yes	175 (46.1)	0.635		0.191	
Humidifier Used at Home	1 = Yes	24 (6.3)	0.669	0.593	0.335	**0.046**
	0 = No	356 (93.7)	0.644		0.189	
Air Purifier Used at Home	1 = Yes	7 (1.8)	0.509	**0.066**	0.328	0.312
	0 = No	373 (98.2)	0.649		0.194	
Candle or Incense Burned	1 = Yes	24 (6.3)	0.935	**<0.001**	0.142	0.251
	0 = No	356 (93.7)	0.631		0.200	
Indoor Hours at Home Per Day	1 = Less than 17 hours	158 (41.6)	0.644	0.892	0.171	**0.098**
	2 = Greater than or equal to 17 hours	222 (58.4)	0.647		0.216	

* Numbers may not sum to total (N=380) due to missing data, and percentages may not sum to 100% due to rounding.

The mean outdoor levels of black carbon during each sampling period, as measured at a central site, were also included in the analysis for indoor levels of black carbon (Figure 1). In this study, indoor measurements of black carbon were not highly correlated with external central site measurements (R=0.114), though the measuring techniques differed. The mean indoor measurements of black carbon (0.688 ± 0.282 μg/m³), as measured via the OT21

Transmissometer, were approximately 20% higher than the mean external central site measurements (0.568 ± 0.232 μg/m³). Given this finding, the external central site measurements associated with each indoor filter measurement were included in the linear effects regression analysis for black carbon. Of note, outdoor levels of brown carbon were not available for analysis.

Figure 1: Indoor and Outdoor Measurements of Black Carbon



Indoor candle or incense use, home type, season, air conditioning use and outdoor levels of black carbon significantly predicted indoor black carbon levels in multivariate analysis (Table 3). Specifically, single family vs. multi-family homes, winter vs. spring and spring vs. fall were significantly different (Table 4).

Heat type and season were the only variables that significantly predicted indoor brown carbon levels in the parsimonious model (Table 3). Significant differences between seasons and heat types are listed in Table 4. As winter and spring were significantly different from summer or fall, this finding could reflect heat use during these periods. However, the season interaction with each variable included in the final models for both brown carbon and black carbon was not statistically significant.

Table 3: Final Multivariate Model for Indoor Brown and Black Carbon Levels

Variable	Level	Estimate	Std. Error	DF	t Value	F Value	Pr > F
Black Carbon (µg/m³)							
Home Type	Single family home Multi-family home Apartment building Trailer or mobile home	0.727 0.842 0.773 0.748	1.044 1.057 1.047 1.137	234 234 234 234	<0.001 0.002 <0.001 0.024	2.66	0.049
AC Units On	No	1.105	1.046	234	2.22	4.93	0.027
Candle	No	0.654	1.070	234	-6.28	39.38	<0.001
Season	Winter (Dec, Jan, Feb) Spring (Mar, Apr, May) Summer (Jun, Jul, Aug) Fall (Sep, Oct, Nov)	0.815 0.699 0.772 0.804	1.060 1.059 1.054 1.053	234 234 234 234	-3.51 -6.26 -4.91 -4.18	5.67	<0.001
Outdoor Black Carbon	Weekly Average Level	1.408	1.075	234	4.75	22.54	<0.001
Brown Carbon (µg/m³)							
Season	Winter (Dec, Jan, Feb) Spring (Mar, Apr, May) Summer (Jun, Jul, Aug) Fall (Sep, Oct, Nov)	0.356 0.311 0.194 0.145	1.153 1.145 1.158 1.141	236 236 236 236	-7.29 -8.63 -11.13 -14.66	11.31	<0.001
Heat Type	Electric Oil or gas Forced heat with oil or gas	0.305 0.163 0.265	1.176 1.099 1.198	236 236 236	-7.33 -19.32 -7.35	7.19	<0.001

Table 4: Indoor Air Pollutant Mean Differences and Adjusted p-values for Multiple Categorical Variables*

Variable	Black Carbon Estimate (ug/m^3)	Adjusted p-value (Tukey-Kramer)	Brown Carbon Estimate (ug/m^3)	Adjusted p-value (Tukey-Kramer)
Single vs. Multi-Family Home	0.864	**0.030**		
Electric vs. Oil or Gas			1.874	**0.002**
Oil or Gas vs. Forced Heat			0.615	**0.041**
Winter vs. Spring	1.166	**0.001**		
Winter vs. Summer			1.831	**0.006**
Winter vs. Fall			2.450	**<0.001**
Spring vs. Summer			1.600	**0.038**
Spring vs. Fall	0.869	**0.005**	2.140	**<0.001**

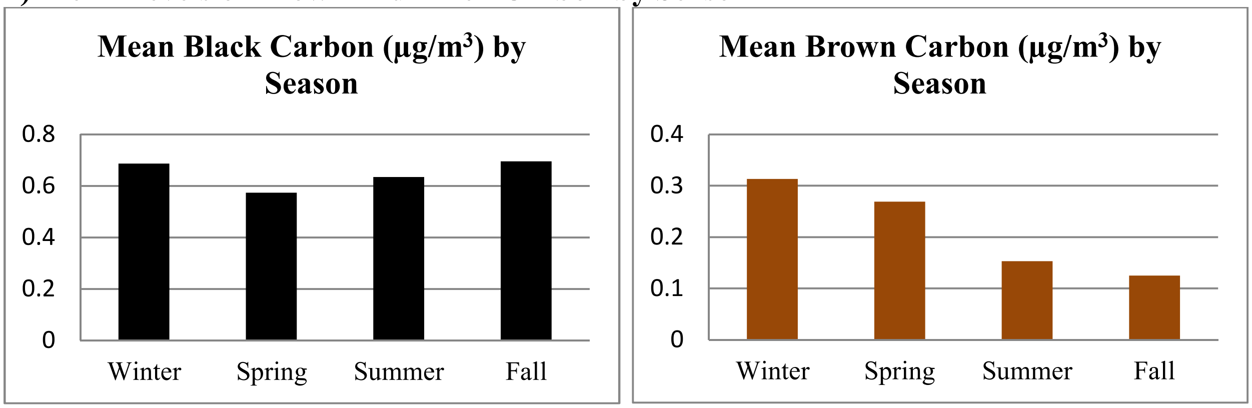
*Mean differences and adjusted p-values only reported for p-values <0.05

The seasonal variation of mean indoor black and brown carbon levels is shown in Figure 2. Mean indoor levels of black carbon were highest in fall and winter and lowest in spring. In contrast, mean indoor levels of brown carbon were highest in winter and lowest in fall (Figure

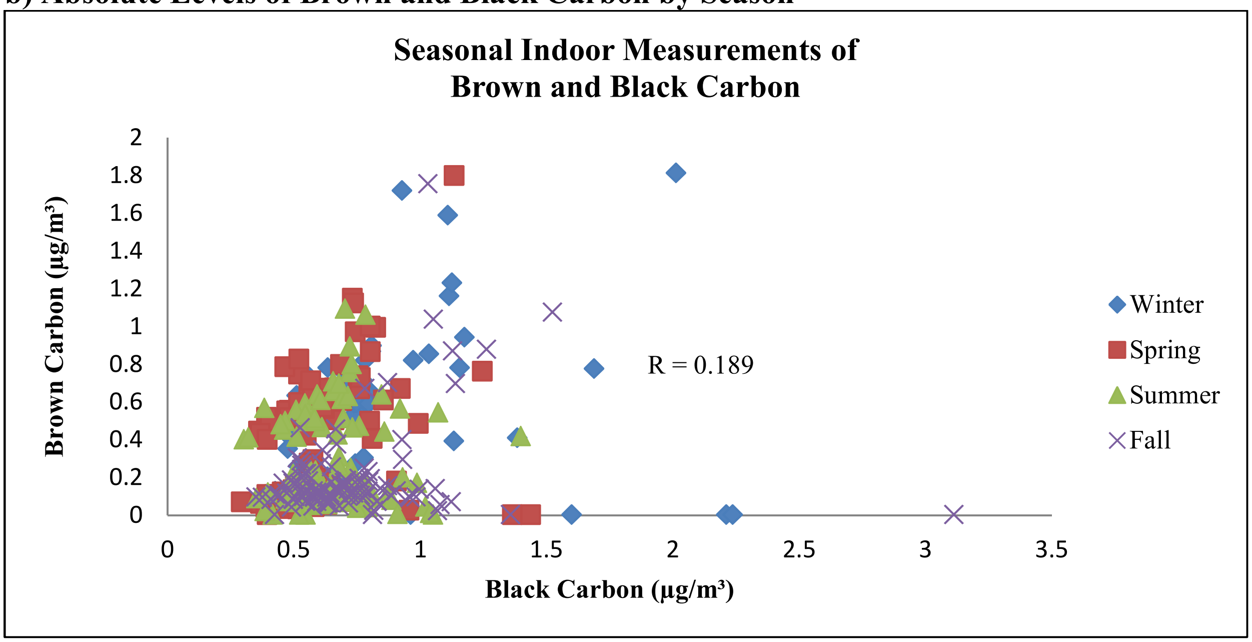
2a). Indoor measurements of brown and black carbon, as shown in Figure 2b, are not highly correlated (R = 0.189).

Figure 2: Seasonal Variation of Indoor Black and Brown Carbon

a) Mean Levels of Brown and Black Carbon by Season

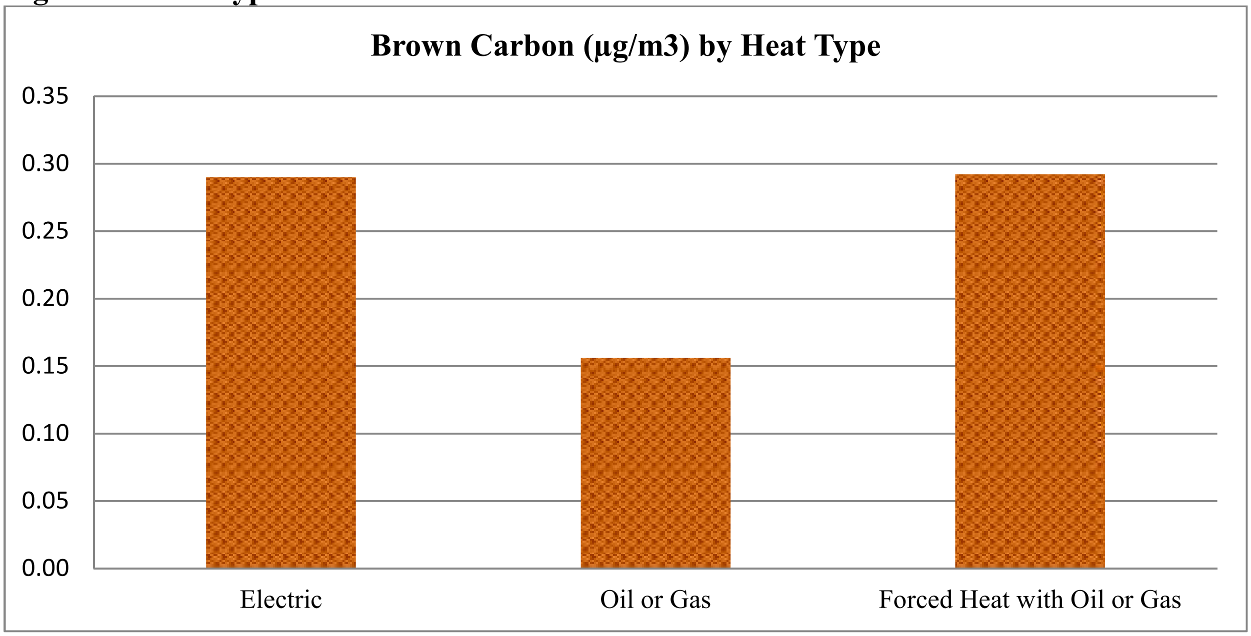


b) Absolute Levels of Brown and Black Carbon by Season



As previously discussed, heat type significantly predicted indoor brown carbon measurements. Highest levels of brown carbon were found in homes that had forced heat with oil or gas while lowest levels were found in homes that had oil or gas without forced heat (Figure 3).

Figure 3: Heat Type and Mean Indoor Brown Carbon Measurements



Discussion:

This study demonstrates that characteristics of the residential environment can predict indoor levels of air pollutants. Heat type, specifically, predicts indoor brown carbon levels. Home type, indoor candle/incense use, air conditioning use and outdoor levels of black carbon predict indoor black carbon levels. Higher levels of black carbon found in multi-family homes as compared to single-family homes suggest that black carbon levels in one unit affect other units in the home. Seasonal variation in indoor levels of brown carbon and black carbon also exists, with levels of both being highest in the winter. This seasonal variation of air pollutants is consistent with other studies (Huang et al., 2018).

These findings add to the existing literature on indoor particulate air pollution. Indoor sources of brown carbon, especially in developed countries, have not been widely studied. Our study is the first to suggest that forced heat increases levels of brown carbon in the home. Likewise, home characteristics, such as multiple units and not using air conditioning can increase levels of indoor air pollutants. This implies that people can modify their exposure to indoor brown carbon and black carbon by choosing their type of home and heating/cooling source.

For black carbon specifically, this study showed that central site measurements of outdoor black carbon levels are not highly correlated with indoor levels. For brown carbon, outdoor measurements were not available for comparison. As previously discussed, the measured indoor levels of black carbon were approximately 20% higher than the mean outdoor levels of black carbon even though this study was designed to minimize indoor sources. This suggests that interior variables in the residential environment contribute to indoor levels of black carbon. Directly measuring indoor levels of black carbon, rather than estimating exposure via central site monitoring, is more reflective of individual exposure. As people spend a majority of their time in the home, it is important to directly measure indoor levels of air pollutants to more accurately quantify total exposure from indoor and outdoor sources. Future air pollution studies should more accurately evaluate levels of indoor air pollutants through the use of direct filter measurements rather than estimating via a central site monitor. Modifying the home environment to reduce indoor air pollutant exposure in susceptible individuals may further improve morbidity and mortality in this population and should be a target of future research.

In our study, the mean central site measurements of outdoor black carbon levels were the only exterior variable that predicted indoor black carbon levels. Other outdoor variables affecting black carbon, such as traffic, did not significantly predict indoor air pollution. This finding differs from existing $PM_{2.5}$ literature and other air pollution studies (Jhun et al., 2019; Pateraki et al., 2019), possibly due to the inherent limitations of questionnaire data.

Overall, the indoor levels of brown carbon and black carbon measured in this study are low. By specifically limiting known indoor sources of black carbon, this study enabled the identification of lesser known sources of indoor black carbon. This study also included multiple measurements of each home, thus decreasing the effect of the variability of indoor home characteristics during sample collection. Weaknesses of this study include the use of a survey to measure home characteristics and the overall homogeneity of the sample population. Recall bias and the Hawthorne effect are likely present in the collected responses.

Conclusions:

Home characteristics and the residential environment are associated with indoor air pollutants. Decreased exposure to black carbon and brown carbon, through altering variables in the residential setting, could improve indoor air pollution levels.

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