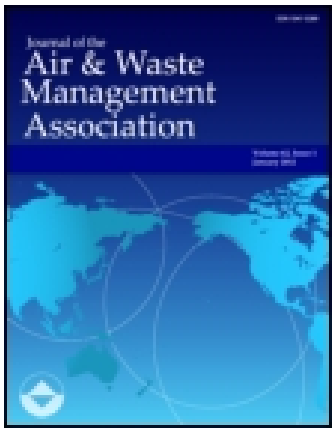

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Real-Time Indoor and Outdoor Measurements of Black Carbon in an Occupied House: An Examination of Sources

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ABSTRACT

Black carbon (BC) was measured every 5 min for two years (May 1998–May 2000) inside and immediately outside a northern Virginia house (suburban Washington, DC) occupied by two nonsmokers. Two aethalometers, which measure BC by optical transmission through a quartz fiber tape, were employed indoors and outdoors. Meteorological parameters were obtained on an hourly basis from nearby Dulles airport. Indoor activities were recorded to identify indoor sources such as combustion activities, which occurred 9% of the time during the first year and 4% of the time during the second year. At times without indoor sources, indoor/outdoor BC ratios averaged 0.53 in the first year and 0.35 in the second year.

The main outdoor source of BC was the general regional background, contributing 83–84% of the total during each of the two years. Morning rush hour traffic contributed 8–9% of the total BC. An evening peak in the fall and winter, thought to include contributions from wood burning, was responsible for ~8% of the annual average BC concentration. The main indoor sources of BC were cooking and candle burning, contributing 16 and 31%, respectively, of the annual average indoor concentrations in the two years. Relative humidity (RH) affected the outdoor aethalometer in both years. An artifact associated with the tape advance was noted for the aethalometer, but a correction factor was developed that reduced the associated error by a factor of 2.

IMPLICATIONS

Although unregulated at present, BC ("soot") has important health implications and, therefore, its main sources are of interest. Knowledge of the important sources and relative indoor/outdoor concentrations of BC is important for assessing human exposure and evaluating the effectiveness of regulating outdoor sources, such as diesel particulates, under the Clean Air Act. This two-year study at one suburban location near Washington, DC, identifies the regional background as supplying the bulk of the annual average outdoor concentration, with local traffic and wood burning responsible for ~8% each. Indoor sources, such as cooking and candle burning, were found to be important contributors to the annual average indoor concentration.

INTRODUCTION

Black carbon (BC) is one of the components of airborne particulate matter (PM) and is generated only by the combustion of fuels containing carbon.[1,2] Black carbon is operationally defined, as is elemental carbon (EC). Black carbon is determined by measuring visible light reflected or transmitted through a filter. Elemental carbon is measured using a thermal-optical method, which involves the sequential volatilization of organic carbon (OC) and EC.[3] Black carbon is presumed to be mainly EC.[4,5] The "black smoke" (BS) method employed at the time of the London fog episode is based on measuring BC and then multiplying by a factor to approximate the total PM mass.

The major component of what has been historically referred to as "soot" is BC. Exposure to soot became a health concern as early as 1775, when Percival Pott noted that chimney sweeps had an elevated incidence of scrotal cancer.[6] More recent studies have shown that chimney sweeps had an increased number of deaths from coronary heart disease, respiratory diseases (including lung cancer), esophageal disease, and liver cancer.[7]

Although BC itself is generally considered inert, the combustion process results in the BC being coated by organic matter such as nitroso compounds and particle-bound polycyclic aromatic hydrocarbons (PAH), some of which have been shown to have carcinogenic properties in animals.[8] The extractable organic matter from PM generated by traffic (diesel and gasoline) has been shown to be mutagenic.[9] Because most of the particles emitted by diesel engines are of submicron size, they can be inhaled

and deposited in the deep lung and cause cancer and other ailments.[10] Daily mortality in Amsterdam was associated with BS but not with PM_{10}.[11] Two studies of children in the Netherlands found increased respiratory symptoms[12] and reduced lung function[13] associated with BS.

The measurement of BC or EC is of public health interest because it can be used as a surrogate for diesel exhaust and other combustion processes. In a set of studies in southern California, EC made up ~75, 61, and 7% of the particle mass in the exhaust of heavy-duty and light-duty diesel engines and gasoline engines, respectively.[14-16] In a study in Colorado, EC emission rates from "medium"-emitting gasoline vehicles were 15 mg/mi compared with 241 and 1316 mg/mi for light-duty and heavy-duty diesel vehicles.[17,18] Also, BC has been found to be a tracer for wood burning, exposure to which has documented health effects.[19] Wood smoke consists of 3–15% EC and is emitted at a rate of 0.15–0.75 g/kg of wood burned.[20,21] Wood burning exposure has been linked with abnormal chest X-rays, pulmonary arterial hypertension, decreased pulmonary function,[22] increased hospital visits for asthma,[23] increased coughing and wheezing in young children,[24] increased respiratory symptoms, and chronic bronchitis.[25] In addition, some compounds found in wood smoke—benzo[a]pyrene and formaldehyde—may be carcinogenic to humans.[26]

Although it has been shown that BC is a useful marker for outdoor PM from diesel exhaust, indoor sources of BC have not been assessed. Characterization of indoor sources is of critical importance in assessing exposure and risk because we spend, on average, 89% of our time indoors,[27] and pollutants from indoor sources tend to occur at high concentrations. Because there are limited data on human exposure to indoor concentrations of BC, this study was designed to examine the concentration and source contribution of BC measured indoors and outdoors.

METHODS

The study home is a three-level, four-bedroom, 385-m^3 end townhouse in Reston, VA, a suburban area 25 mi northwest of Washington, DC. Reston is an unincorporated town of ~60,000 residents. A mixture of computer and software technology, government buildings, national associations, and service industries provides ~55,000 jobs. The main particle pollution sources would be expected to be commuter vehicles and construction. During the years of the study, very active construction of multistory office buildings occurred in the Reston Town Center, a region about 2–4 miles away from the townhouse. The predominantly western winds (~80% of the time) pass over mostly rural and suburban areas before crossing Reston, so that it would be unlikely to experience much of the pollution associated with the major urban area to the southeast.

The townhouse is located at the intersection of two suburban roads in Reston (Glade and Soapstone drives). It is ~80 m east of Soapstone and 100 m north of Glade. The Dulles toll road, (Route 267), a 6-lane highway enclosing the 4-lane Dulles airport access road, is located ~1.5 mi north of the house. The house is built on a hill, such that the basement is underground at the front (west) of the house but opens onto a patio at the back of the house. The townhouse is located in a cluster of 80 homes, which looks out to a 30-acre wooded area to the east.

Heating is central forced air with a gas furnace and standard furnace filter, gas hot water heater, and a vented gas dryer. All other homes in the cluster are also heated by natural gas and are, therefore, not expected to emit BC during home heating. Central air conditioning is also available, with an outdoor compressor near the patio. The basement is partially finished, with a carpeted floor in the east portion and a cement floor in the utility room, containing washer, dryer, furnace, and hot water heater. Two adult nonsmokers live in the house. The first floor contains a kitchen/dining area, a bathroom, and a living room with fireplace (unused). The second floor contains four rooms: a master bedroom with bath, a guest bedroom, and two rooms used as offices. Residents recorded source activities, including cooking and candle burning.

Natural ventilation was employed during the two-year experimental period, with windows typically open when temperatures were moderate. The fraction of time windows were open ranged from 10% in the winter to 60% in the summer. Air conditioning was used sparingly, only on the hottest days of the summer months. The central furnace fan was on ~90% of the time to promote air movement and good mixing. Typical air change rates under closed-window conditions ranged from 0.2 hr^{-1} during the summer to 0.6 hr^{-1} during the winter, and ranged up to 2 hr^{-1} when multiple windows were opened.

Black carbon was measured for two years (May 14, 1998–April 30, 1999, and July 15, 1999–May 30, 2000) using two aethalometers. This instrument operates by measuring the intensity of light transmitted through a quartz filter tape. A built-in flow meter controls the air sampling rate to 5 L/min. The tape automatically advances when the light transmittance is diminished to a preset level due to filter loading. The instrument records light transmission through the quartz tape and reports BC concentrations in ng/m^3, using an empirical coefficient based on studies of aerosols near combustion sources in urban areas and in remote regions.[1] The coefficient was similar for all aerosol types (~2000 m^2/g). Measurements were logged every 5 min. Recent studies comparing aethalometer results, in various locations, to the thermal-optical method found agreement to within ~20% between the BC and EC values.[4,5] The outdoor aethalometer was located on the

east-facing balcony of the first floor of the home, and the indoor monitor was located in the basement, also on the east side of the house. Both instruments were placed on the floor, with intakes ~1 m from the walls and 0.5 m high.

Weather data from the Dulles airport, which is located 7 miles northwest of the home, were used to represent meteorology conditions. These data were obtained from the NOAA National Climatic Data Center in Asheville, NC. Because these data were collected every hour, the hourly value was ascribed to each 5-min BC measurement. The variables used in this study were dry temperature, wet temperature, dew point, relative humidity (RH), wind speed, and wind direction. During the second year of the study, temperature and RH measurements were made indoors and outdoors at the house. Temperatures were measured using thermistors at one outdoor and nine indoor locations; RH was measured at one outdoor and five indoor locations. A portable meteorology instrument (Climatronics, Inc.) was placed on the rooftop and recorded horizontal wind velocity using ultrasound pulses (10 Hz). Wind speed and direction were recorded every 6 sec. The BC measurements, household activity data, and meteorological data were all combined into a single database.

Instrument Precision and Bias

The two aethalometers were run side by side inside the house for 14 consecutive days (3980 measurements at 5-min intervals), from December 27, 1998, to January 10, 1999 (the middle of the year-long monitoring period), to determine their precision. During this evaluation, it was noted that an instrument measurement bias was associated with each tape advance, such that the instrument with the most recent advance always read higher than the other and maintained higher values until the other instrument's tape advance. The tape advance occurred once or twice a day, on average, with the outdoor instrument advancing more frequently due to the higher outdoor values. A typical increase following the tape advance was ~50%, with each instrument leapfrogging the other following the tape advance. Because no particular trend toward ever-increasing values was noted, we assumed that the initial increase resulted in a concentration above the correct value, followed by a downward trend, such that the final values preceding the next tape advance would be below the correct value. Other investigators have observed the same phenomenon; the cause is unknown.[28] Because the tape advance is triggered by measurement of optical depth, it was possible to correct for this short-term bias by fitting an exponential function of the optical depth to the observed concentration:

$$C_{obs} = AC_{true} \exp(-Bq) \quad (1)$$

where q is the standardized optical depth (observed optical depth/100), and A and B are constants, different for each instrument. Because both instruments are measuring the same aerosol, we can solve for C_{true} in the above equation for each instrument and set the two expressions equal. After some algebra, we arrive at the following equation:

$$\ln (C_1/C_2) = \ln (A_1/A_2) + B_2q_2 - B_1q_1 \quad (2)$$

where the C_i are the observed concentrations by the two instruments. The unknown parameters (A_1/A_2, B_1, and B_2) can then be determined by linear least squares. However, this approach gives only the ratio A_1/A_2 [$\ln A_1/A_2 = 0.076$ (0.007 SE)], not the individual values. Because the two instruments agreed with each other to within 7%, we can obtain the individual values of A_i by making the further assumption that neither instrument shows an overall bias. In that case, the sum of the corrected values (C_{true}) equals the sum of the observed values (C_{obs}) for each instrument. With these requirements, A and B were found to have the following values:

	A	B
aethalometer #15	1.3	0.584 (0.011 SE)
aethalometer #16	1.2	0.532 (0.011 SE)

where n = 3980, and the adjusted R^2 was 0.65. These values correspond with an overestimation of 20–30% immediately after the tape advance, and an underestimation of 16–19% just before the next tape advance. This leads to an apparent increase of 48–54% over the most recent value when the tape advances, as observed.

Correcting the observations using these values of A and B succeeded in reducing the average absolute error from 19.9 to 9.0% (Figure 1). These parameters were used to correct all readings from May of 1998 to April of 1999. A similar calculation was used to correct the values in the second year. Multivariate regressions were conducted to assess meteorological

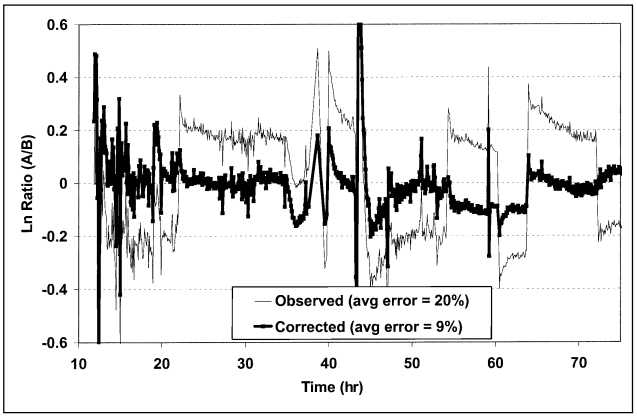

Figure 1. Effect of correcting collocated aethalometers (A = #15 and B = #16) for tape advance artifact.

influences on aethalometer measurements over the two-year monitoring period. Because the measurements were highly skewed, the analysis was conducted on the natural logarithm of the indoor and outdoor concentrations.

Collinearity between the variables used in the outdoor regression was investigated using Pearson correlations, scatterplots, variance inflation factors, and a forward stepwise multiple regression. The scatterplots were used to qualitatively assess associations between the variables. Variance inflation factors quantify the increase in variance due to multiple correlation among the covariates. The forward stepwise regression begins with an "empty" model and adds a variable per step in order of importance. If collinearity exists between variables, only the most significant of the collinear variables will be added to the model. Statistical analyses were conducted using Intercooled STATA 6.0 and Statistica.

Table 1. Summary statistics (May 14, 1998–April 30, 1999).

Statistic	Corrected Outdoor BC (ng/m^3)	Corrected Indoor BC (ng/m^3)	Corrected Indoor BC (w/o Events) (ng/m^3)	Ratio of Indoor/Outdoor BC (w/o Events)
N	89,771	94,215	85,629	–
Arith. mean	741	462	393	0.53
Standard dev.	612	3197	278	–
Standard error	2	2	1	–
Geometric mean	573	319	308	0.54
GSD	2.1	2.2	2.1	–
0.1st percentile	–25	22	22	–
1st percentile	90	46	47	0.52
5th percentile	167	85	84	0.50
10th percentile	220	113	112	0.51
25th percentile	356	189	186	0.52
50th percentile	588	335	328	0.56
75th percentile	941	533	519	0.55
90th percentile	1412	792	754	0.53
95th percentile	1857	1025	932	0.50
99th percentile	2940	1774	1305	0.44
99.9th percentile	4802	12,772	1898	0.40

RESULTS

In both years, ~95% of the possible measurements were successfully collected. An example winter's day is shown in Figure 2. The figure shows a morning peak, attributed to rush hour traffic, and an evening peak, thought to be due mainly to wood burning. The artifactual increase in concentration occurring after a tape advance (which causes a 10-min break in measurement frequency) is also evident in both the indoor and outdoor records.

Summary statistics and selected percentiles from the BC distribution, recorded during the two years, are provided in Tables 1 and 2. To account for the effects of indoor sources such as cooking and candle burning, the times during which indoor sources were affecting indoor concentrations were identified either from activity diary entries or by observing sharp increases in indoor concentrations without corresponding outdoor increases. About 9% of the indoor BC measurements in the first year, and 4% in the second year, were identified as influenced by an indoor source. Both the outdoor and indoor BC concentration values are nearly log-normally distributed over most of the range of concentrations. The peak indoor concentration (99.9th percentile) of 12,772 ng/m^3 in the first year is attributable to the burning of a citronella candle for a few minutes. The mean indoor concentration of BC when no sources were evident was 393 ng/m^3 (N = 85,508). The mean value with indoor sources active was 1133 ng/m^3 (N = 8707). Thus, indoor sources produced 16% of the total BC indoor concentrations. In the second year, indoor sources produced 31% of the total indoor BC.

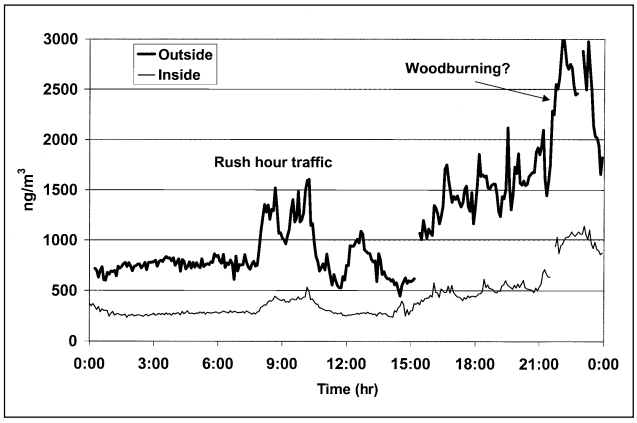

Figure 2. Example winter's day. Sources of peaks are tentatively identified as vehicles in the morning and wood burning at night. The artifactual increase in concentration associated with a tape advance can be seen following the "breaks" in both the outdoor and indoor traces.

Indoor BC levels in the first year were correlated with outdoor levels (Spearman's rho = 0.70, N = 88,411, p < 0.0001). This correlation increased to Spearman's rho = 0.74, N = 80,032, p < 0.0001 when the times with indoor BC sources were excluded from the distribution.

A multiple regression was performed on the logarithms of the corrected outdoor BC concentrations in each of the two years. Meteorological data were available from Dulles airport on an hourly basis. Because all airport meteorological measurements were taken at 51 min past the hour, the corresponding BC measurements (50 min past the hour) were selected for the regressions. The initial regressions

Table 2. Summary statistics (July 19, 1999–May 30, 2000).

Statistic	Corrected Outdoor BC (ng/m^3)	Corrected Indoor BC (ng/m^3)	Corrected Indoor BC (w/o Events) (ng/m^3)	Ratio of Indoor/Outdoor BC (w/o Events)
N	87,696	90,286	86,623	–
Arithmetic mean	680	348	241	0.35
Standard dev.	1214	2020	240	–
Standard error	4	7	1	–
Geometric mean	492	178	165	0.34
GSD	2.3	2.8	2.5	–
0.1st percentile	–354	–90	–95	–
1st percentile	–20	4	3	–
5th percentile	104	33	27	0.26
10th percentile	166	50	46	0.28
25th percentile	293	93	88	0.30
50th percentile	508	178	167	0.33
75th percentile	849	335	308	0.36
90th percentile	1328	584	514	0.39
95th percentile	1741	822	686	0.39
99th percentile	2929	2122	1155	0.39
99.9th percentile	5961	19,507	2017	0.34

included eight independent variables: RH, temperature, wind speed, wind direction (two binary variables corresponding to components along the north-south and east-west axes), a binary variable indicating day (0) or night (1), another binary variable for the spring-summer (0) versus fall-winter (1) seasons, and a binary variable for weekend (0) versus weekday (1). However, during each year, the correlation between the temperature and season variables was strong (absolute value >0.6), so two new regressions were run, dropping each variable in turn. Of these two regressions, the one including temperature had the highest R^2 value, so the season variable was dropped. The seven remaining variables had no correlation greater than 0.4. The westerly component of wind direction had only a small effect in each year, so the final regressions had six variables (Table 3).

In each year, the strongest independent variable was wind speed. An increase in wind speed of one standard deviation (SD) produced a decrease in the logarithm of the BC concentration of 0.32 and 0.35 SD, respectively, in the two years. The second strongest variable in the first year was RH; an increase in RH of one SD produced an increase in the logarithm of the BC concentration of 0.27 SD. Higher temperatures resulted in higher BC values; they were higher during the day than during the night, and higher during workdays compared with weekend days. Finally, BC concentrations increased when the wind had a southerly component but were nearly unaffected by wind with a westerly component. All variables gave results that were similar in sign and strength in both years.

The second year included measurements of indoor RH. Therefore, a multiple regression was performed on the logarithms of the indoor BC concentrations, with the independent variables being the logarithms of the outdoor BC concentrations and the average indoor RH. Times when indoor sources were present were excluded. Indoor BC concentrations were strongly dependent on outdoor BC concentrations [$\beta = 0.64$, R^2 (adj.) = 0.40, N = 79759], but indoor RH had little effect [$\beta = -0.03$, R^2 (adj.) < 0.001]. We then dropped the indoor RH variable and performed a simple regression on the logarithm of indoor

Table 3. Multiple regression results.

(a) Year 1 (1998–1999) *N* = 5711 R^2 (adj.) = 0.278 F(6,5704) = 367

Variable	Beta	SE of Beta	*B*	SE of *B*	*t*	*p*
Intercept			5.799	0.0520	111.4	<0.000001
Wind speed (knots)	-0.32	0.013	-0.065	0.0026	-25.4	<0.000001
RH (%)	0.27	0.013	0.009	0.0004	20.8	<0.000001
Temperature (^{0}F)	0.10	0.012	0.004	0.0005	8.2	<0.000001
North	-0.08	0.012	-0.055	0.0088	-6.3	<0.000001
Night	-0.17	0.012	-0.251	0.0180	-13.9	<0.000001
Workday	0.18	0.011	0.296	0.0182	16.3	<0.000001

(b) Year 2 (1999–2000) N = 5571 R^2 (adj.) = 0.232 F(6,5564) = 281

Variable	Beta	SE of Beta	*B*	SE of *B*	*t*	*p*
Intercept			6.061	0.0606	100.0	<0.000001
Wind speed (knots)	-0.35	0.013	-0.078	0.0029	-26.9	<0.000001
RH (%)	0.12	0.013	0.005	0.0005	9.2	<0.000001
Temperature (^{0}F)	0.12	0.012	0.006	0.0006	9.6	<0.000001
North	-0.14	0.013	-0.120	0.0108	-11.1	<0.000001
Night	-0.14	0.013	-0.240	0.0230	-10.4	<0.000001
Workday	0.08	0.012	0.156	0.0234	6.7	<0.000001

Note: Beta is the coefficient of normalized variable with mean 0 and SD = 1; Dependent variable is the natural logarithm of corrected outdoor BC (ng/m^3); North: wind from the north = 1, from the south = -1; Night: day (6:00 a.m.-6:00 p.m.) = 0, night = 1; Workday: weekend = 0, weekday = 1.

BC versus the logarithm of outdoor BC. Indoor BC was again strongly linked to outdoor BC [$\beta = 0.75$, R^2 (adj.) = 0.56, $N = 79807$]. Even though in this multiple regression indoor RH seemed to have little effect, a bivariate analysis of variance (ANOVA) suggested a strongly nonlinear effect, with a "hockey-stick" shape (BC concentrations flat until indoor RH = 60%, followed by a sharp increase for higher RH levels).

To characterize BC concentration trends with time, ANOVAs were conducted. Figure 3 depicts how indoor and outdoor BC concentrations change hourly for the spring–summer and fall–winter months. Indoor and outdoor BC concentrations show a year-round peak between 5:00 a.m. and 11:00 a.m. In the fall and winter months only, another peak is seen in the evening between 4:00 p.m. and 2:00 a.m. Because it is unclear what value of RH corresponds with an accurate measurement of BC by the aethalometer, it is not possible to correct the data for the RH effect. Therefore, comparisons of values at different RHs must be viewed cautiously.

DISCUSSION

Our mean outdoor BC concentration of 741 ng/m^3 is close to the values reported in several studies for rural and suburban areas, and somewhat below values reported for urban areas. For example, studies in urban areas report mean EC values of 400–3500 ng/m^3 in five eastern cities;[5] 500–1700 ng/m^3 in two eastern cities;[29] 300–1100 ng/m^3 in two cities;[30] 1400 ng/m^3 in Seattle;[31] and 1300–2700 ng/m^3 in five California cities.[32] The median value in these 15 urban areas was 1400 ng/m^3. Studies in rural areas report values of 500 ng/m^3 in each of two eastern rural areas;[29] 1900 and 3300 ng/m^3 in two Colorado towns;[33] 100–400 ng/m^3 in the Smoky Mountains of Tennessee;[34] 700 ng/m^3 in a Washington town;[35] 200–700 ng/m^3 in six rural areas;[30] 400 ng/m^3 in a rural location 150 km inland from the mid-Atlantic coast;[36] 400–3300 ng/m^3 in 10 California locations;[37] and 100 ng/m^3 in the Grand Canyon.[38] The median value in these 24 rural areas was 700 ng/m^3. The only study[29] targeting a suburban location (in Reading, MA) found a mean value of 700 ng/m^3.

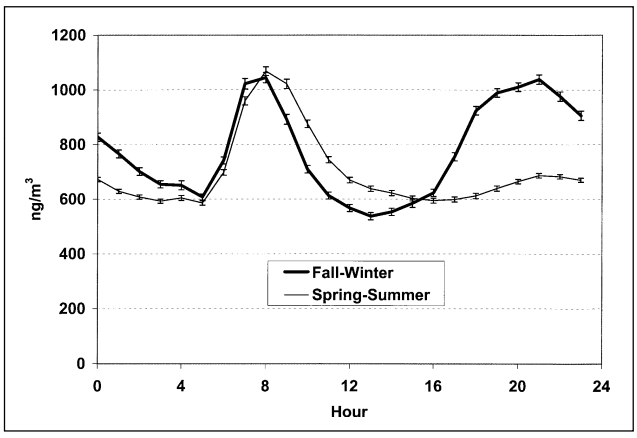

Figure 3. Diurnal variation of outdoor BC during the first year (1998–1999), averaged over 89,770 measurements. An evening peak thought to reflect wood burning occurs in fall and winter, but not in spring and summer.

During the first year, indoor BC concentrations were about half the outdoor values. In the second year, the indoor/outdoor ratio was lower, by about a third. The difference may be due to the fact that in the second year, the central fan was on continuously to promote better mixing during a series of air change measurements. About six house volumes of air per hour were forced through the ducts, with corresponding increased particle deposition. Also, in April 2000, an electrostatic precipitator (ESP) filter was installed in the return air duct and was found to be highly effective (>90% efficient, as measured by particle counters upstream and downstream) in removing particles of all sizes between 0.3 and >10 μm. The indoor-outdoor ratio in May 2000, when the ESP was on nearly 100% of the time, dropped to 0.28 from the average value of 0.34 during the rest of the year. The variable with the strongest effect on outdoor BC concentrations was wind speed, with higher wind speeds associated with lower BC concentrations. This would be expected if periods of stagnation allowed BC concentrations to build up.

The second strongest variable was outdoor RH. However, indoor RH appeared to have less or perhaps no effect on indoor BC concentrations. Because the outdoor aethalometer was exposed to outdoor conditions, a possible explanation for the apparent effect of RH would be extremely hot or cold conditions. Therefore, a multiple regression was performed on the outdoor BC in a restricted range of temperature corresponding with the observed indoor range from ~21 to 27 ºC. The results showed an even stronger effect of RH in these restricted temperature ranges, with one standard deviation change in RH associated with a 0.62 SD change in the logarithm of the BC concentration.

Another possibility is that most of the outdoor RH effect is concentrated at the upper RH levels. To test this, from 1999 to 2000, a series of multiple regressions was performed on outdoor BC, restricting the data to RH concentrations less than 90, 80, 70, 60, and 50%, successively. In all cases, the RH variable continued to be either first or second strongest of the seven independent variables. If the outdoor aethalometer is affected by RH and the indoor aethalometer is not, we would expect to see the outdoor/indoor ratio increase with increasing outdoor RH. We tested this idea by performing multiple regressions for both years on the natural logarithm of the outdoor/indoor ratio, with outdoor RH, temperature, wind velocity, season, day-night, and wind direction as independent variables.

In the second year, the measured air exchange rate was available as an additional independent variable. In both years, the outdoor/indoor ratio increased with increasing

outdoor RH, by 0.22 and 0.14 SD, respectively, for one SD change in RH [N = 87,887, R^2 (adj.) = 0.081 in the first year; N = 2140, R^2 (adj.) = 0.147 in the second year]. Outdoor RH was either the first or second strongest of the variables in affecting the outdoor/indoor ratio, although, of course, the air exchange rate was the strongest variable in the second year. (A high air exchange rate allows more outdoor air inside and tends to equalize the indoor and outdoor levels.) We conclude that the effect of outdoor RH on outdoor BC as measured by an aethalometer exposed to outdoor conditions is strong and reproducible from year to year. Therefore, for individuals or agencies employing outdoor aethalometers, it may be important to dehumidify the sampled air. However, whether indoor RH affects the aethalometer was difficult to determine from our data. We are unable to say from these data whether an indoor aethalometer sampling outdoor air would be sensitive to outdoor RH. Further research should be done to determine if dehumidification is necessary under these conditions.

The third variable with a strong effect was wind with a southerly component. Further analysis indicated that wind from the southwest had a strong positive effect, with wind from the southeast also increasing BC concentrations, but to a lesser extent. Wind from the northwest was associated with strongly decreased BC concentrations, and wind from the northeast produced slightly decreased BC concentrations. No obvious source southwest of Reston is known. Two possible sources of BC could be diesel trucks and buses in the city of Washington, DC, ~20 miles southeast of Reston, or diesel trucks in the main trucking corridor (Highway 95 and the 495 Beltway), ~10 miles east of Reston. Because winds with an easterly component did not appreciably increase BC concentrations, it does not appear that Highway 95 could have been a major source. Also, since Washington, DC, is twice as far as Highway 95 from Reston, it is unclear whether it could be the source of the increased BC with southeast winds. Long-range transport may be a possible explanation. Southwest winds form a large portion of the wind rose for Washington, DC, and air parcels from the industrial Midwest have been shown to swing south before turning north again.

Two remaining variables with relatively strong effects on BC concentrations were the diurnal and weekday-weekend variables. Daytime and workday concentrations of BC were higher, most likely due to the influence of rush hour traffic.

A variety of indoor and outdoor BC sources, varying with season, were hypothesized from the concentration profile data and time activity information. A major outdoor source of BC appeared to be morning traffic, because a peak occurred during the 6:00 a.m.–9:00 a.m. rush hour throughout the year. On workdays, the mean 6:00 a.m.–9:00 a.m. value was 1088 ng/m^3 compared with the weekend mean of 607 ng/m^3. Measurements of indoor and outdoor particle concentrations were made concurrently with the BC measurements at the Reston townhouse and did not show the pronounced morning peak that we have related to rush hour traffic.[39] This is consistent with other studies that have shown that EC, BC, and BS are better tracers for mobile source air pollution than is particle mass or number. For example, BS (as measured by a reflectance technique that measures mainly BC) increased over background by 300% at Amsterdam roadways, compared with only a 30 and 37% increase for PM_1 and PM_{10}, respectively.[40] The same increases (300% for elemental carbon and 30% for $PM_{2.5}$) were found in a study in Harlem, NY.[41] A similar 30% increase for PM_{10} and $PM_{2.5}$ was found at a street site in Arnhem, the Netherlands, compared with a 160% increase in black smoke.[42]

A similar outdoor peak occurred in the evening in the fall and winter, but not in summer or spring. The source for this peak is less clear. Three possibilities are considered: the evening rush hour, nighttime inversions, and wood burning.

If the main source was the evening rush hour, we should see, as we did for the morning rush hour, an effect throughout the year; however, the evening peak occurs only in the fall and winter. A rush hour source should also have a strong day-of-the-week effect. However, an ANOVA calculation on the 4:00 p.m.–7:00 p.m. period during the fall and winter (November through February) showed no significant difference ($p < 0.05$) between weekday and weekend concentrations. Furthermore, the evening peak occurs at 10:00 p.m., well after the evening rush hour traffic should have dissipated. Based on this evidence, it appears unlikely that the evening rush hour is the source of the evening BC peak.

Overnight inversions and reduced mixing heights would produce higher BC concentrations. This could account for the observed increased evening concentrations in winter, which has longer nights than in summer. However, the fall and spring have identical periods of light and darkness, and yet the fall shows the high BC peaks and the spring does not. Also, if the mixing height were playing an important role, for periods of comparable source activity, we should see high concentrations at night (low mixing height) relative to daytime hours, when the mixing height is increased. To test this idea, we calculated the geometric mean and standard deviation of the outdoor concentrations at 4:00 p.m. on Sunday (little traffic and wood burning) with those at 4:00 a.m. on Monday (also little traffic and wood burning) throughout the year. The logarithm of the geometric mean 4:00 a.m. value was 5.99 (SD 0.71, n = 528) compared with 5.87 (SD 0.72, n = 494) at 4:00 p.m.—little evidence for a strong effect of the inversion by itself. Therefore, we do not believe that reduced mixing heights account for the observed increase.

Wood burning is the remaining possibility. There are 80 homes in the Soapstone cluster containing the

experimental townhouse, many of which are equipped with fireplaces. An inspection of the neighborhood revealed 15 woodpiles, and wood smoke can often be smelled in the fall and winter. Another line of evidence comes from the relationship between BC levels and temperature. If wood burning were the source, we would expect to see a negative association with temperature during evening hours. A regression of the logarithm of the BC concentrations versus outdoor temperatures, between 7:00 p.m. and midnight between November and February, resulted in a strong negative relationship: one standard deviation decrease in temperature produced 0.28 (0.01 SE) standard deviations increase in the BC concentration ($N = 4804$, $p < 0.0000001$).

Finally, if wood burning were the cause, we would expect to see higher concentrations on traditional holidays. Christmas 1998 had the highest evening BC concentration (4891 ng/m^3) of all days in December. New Year's Eve 1998 was unfortunately not recorded, because the instruments had been brought indoors for a 14-day test of precision. New Year's Eve 1999 had the highest concentration (6891 ng/m^3) and Christmas 1999 the second highest concentration (3917 ng/m^3) in that month (apart from some days when experiments took place outdoors and indoors, involving citronella candle burning). Based on the different lines of evidence presented previously, we conclude that the evening increases in BC concentrations in the fall and winter may be mainly due to wood burning, although some influence of the evening rush hour and mixing height cannot be ruled out.

At times with little traffic and no wood burning, BC levels did not fall to zero, indicating a general or regional background source of BC. By averaging concentrations over these times, we calculated that the regional background source of BC was at the level of 613 ng/m^3 in the first year and 573 ng/m^3 in the second year, or ~83 and 84% of the annual average in the two years, respectively. Using the 5:00 a.m.–12:00 p.m. period to correspond with rush hour on weekday mornings, we calculated a mean value of 934 ng/m^3 ($N = 19157$) for the first year and 821 ng/m^3 ($N = 18349$) for the second year, corresponding with a contribution from rush hour traffic of 9.2 and 7.6% of the annual total outdoor BC concentration, respectively. Using the times from 4:00 p.m. to 3:00 a.m. in the fall and winter months to correspond with the evening wood burning peak, we calculated average BC concentrations of 866 and 783 ng/m^3 in the two years, corresponding with a contribution from wood burning of 8.0% of the annual average BC concentrations in both years. We experimented by choosing different times for our rush hour and wood burning periods and found that, although the peak concentrations changed, the calculated contribution to the total was quite stable.

During the first year, the mean indoor level of BC during times affected by indoor sources was 1124 ng/m^3 (SD = 10441 ng/m^3, $N = 8732$) compared with 393 ng/m^3 (SD = 278 ng/m^3, $N = 85,629$) during times with no indoor source. Therefore, indoor sources accounted for 16% of the total BC measured indoors. During the second year, the mean indoor level of BC during times affected by indoor sources was 2869 ng/m^3 (SD = 9716 ng/m^3, $N = 3581$) compared with 241 ng/m^3 (SD = 240 ng/m^3, $N = 86,623$) during times with no indoor source. In this second year, indoor sources accounted for 31% of the total indoor BC concentration. Further analyses were conducted to determine which indoor source contributed the most to overall exposure. The two major sources in this experimental home were cooking (broiling, toasting, sautéing) and the burning of citronella candles. Cooking generated low levels of BC frequently, whereas candle burning generated high levels infrequently.

CONCLUSIONS

The major contributor to outdoor BC concentrations in this suburban location is the regional background, providing 83–84% of the total in each of the two years. Traffic contributed 8–9% of the total, and wood burning contributed, at most, 8% each year. The single strongest indoor source of BC was the burning of citronella candles. Another common source of indoor BC was cooking. The indoor sources accounted for 16–31% of the indoor total during the two years. The indoor-outdoor BC ratio ranged from 0.53 the first year to 0.35 the second. Increased use of the central furnace fan and a filter installed in the return air duct during the second year may account for the decreased ratio.

The two aethalometers used in this study showed an artifactual increase in estimated BC concentrations associated with the tape advance. It is recommended that studies using the aethalometer test for whether this phenomenon is occurring. It is possible to partially correct for this artifact using an empirical exponential function. Based on this study, outdoor aethalometers are affected by RH. Because it has not been possible to correct for this effect, it may be necessary to use a drying system to remove the moisture from the air before it enters the aethalometer.

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