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Indoor black carbon and brown carbon concentrations from cooking and outdoor penetration: Insights from the HOMEChem study

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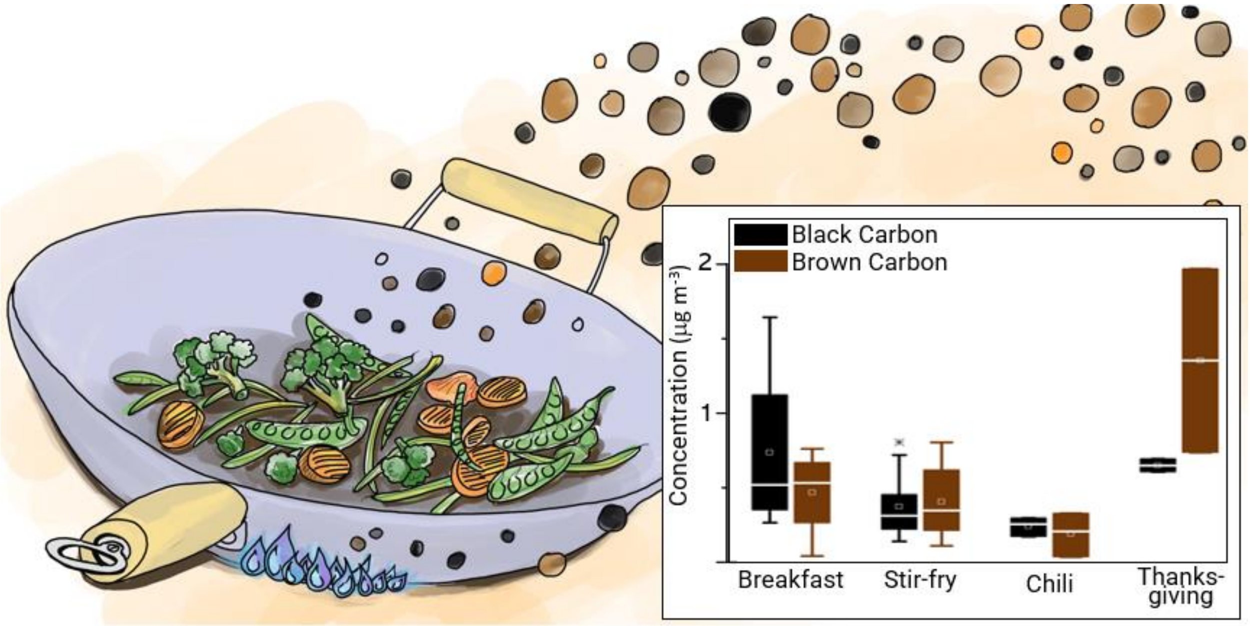
Black carbon and brown carbon emissions were investigated for different indoor activities and during periods of no activity in a test house as a part of the HOMEChem study.



Abstract

Particle emissions from cooking are a major contributor to residential indoor air pollution and could also contribute to ambient concentrations. An important constituent of these emissions is light-absorbing carbon, including black carbon (BC) and brown carbon (BrC). This work characterizes the contributions of indoor and outdoor sources of BC and BrC to the indoor environment by concurrently measuring real-time concentrations of these air pollutants indoors and outdoors during the month-long HOMEChem study. The median indoor-to-outdoor ratios of BC and BrC during the periods of no activity inside the test house were 0.6 and 0.7, respectively. The absorption Ångström exponent was used to characterize light-absorbing particle emissions during different activities and ranged from 1.1 to 2.7 throughout the campaign, with the highest value (indicative of BrC-dominated emissions) observed during the preparation of a simulated Thanksgiving Day holiday style meal. An indoor BC exposure assessment shows that exposure for an occupant present in the kitchen area was ~4 times higher during Thanksgiving Day experiments (primarily due to candle burning) when compared to the background conditions.

Environmental Significance Statement

Light-absorbing carbon, categorized as black carbon (BC) and brown carbon (BrC), can be emitted indoors from activities such as cooking and other combustion sources, and can also penetrate from outdoors. Real-time measurements of the absorption Ångström exponent in indoor environments performed in this study help characterize the indoor and outdoor sources of light-absorbing carbon attributable to indoor activities and outdoor penetration resulting from window opening. Studies characterizing the indoor air consequences of cooking can assist in future source-attribution efforts for local and regional BrC and BC. In addition, the BC exposure consequences from indoor cooking emissions are of potential concern for human health.

1. Introduction

Light-absorbing carbon (LAC) can be broadly classified as black carbon (BC) and brown carbon (BrC). The term BC refers to carbonaceous aerosols, which absorb light approximately uniformly across the visible portion of the light spectrum.[1] BC is emitted through combustion processes in the form of carbon spherules that are refractory and water-insoluble.[2] The health effects of BC are interrelated with those of overall particualte matter (PM) exposure, which include an increased risk of developing respiratory and cardiovascular ailments.[3] BC is a valuable additional air quality indicator to study the health risks associated with combustion-related activities. Studies have shown that chronic exposure to BC can lead to an inflammatory response and the development of benign and malignant carcinomas in rat lungs.[4] Magalhaes et al. reported that an average 1 $\mu g\ m^{-3}$ increase in short-term (< 7 day) BC exposure was associated with an increase in diastolic blood pressure.[5] Similar studies in indoor environments where cooking with biomass fuel is widely prevalent have also found BC exposure as a risk factor for high blood pressure in adults.[6,7] BC has also been shown to play an indirect role in toxicity by acting as a broad-spectrum carrier for semi-volatile organic compounds released from combustion sources.[8]

BrC consists of many types of organic compounds, including humic-like substances and tarry materials, which are generated during biomass burning and present a distinct light absorption spectrum from BC, with a sharp increase in light absorption in the near-ultraviolet (UV) portion of visible light.[2,9,10] In ambient environments, BrC compounds have been shown to act as a protective layer around heavy metals and carcinogens such as benzo[a]pyrene—formed during the incomplete combustion of carbonaceous material, thereby increasing the lung cancer risk associated with their personal exposure.[11–13] BrC emissions need to be studied from an indoor air quality perspective because there are not many studies published on health effects specifically related to BrC in comparison to other components of PM—even though environmental tobacco smoke has been established as a major contributor to indoor BrC.[14,15]

Indoor air quality has gained attention during recent decades owing to concerns over the potential health effects of a wide variety of indoor air pollutants[16–18] and the fact that people spend the majority of their time indoors, especially at home.[19–21] Indoor air pollution has been linked to harmful effects on respiratory and cardiovascular systems and has been associated with the risk of lung cancer.[22] To conserve energy, modern buildings have become more airtight with lower air exchange rates, which may lead to decreased exposure to air pollutants of outdoor origin.[17] However, these conditions also tend to increase the exposures and risks associated with pollutants of indoor origin, especially if the indoor emissions are not adequately vented outdoors.

Cooking is one of the biggest contributors to indoor air pollution due to the emission of PM and gaseous air pollutants such as nitrogen oxides and volatile organic compounds.[23–25] Health effects associated with cookstove emissions have been well documented in developing countries, especially when cooking is performed in poorly ventilated spaces using solid fuels.[26] According to the Global Burden of Disease study, 3.5 million premature deaths are linked to smoke exposure from solid fuel cooking.[27] Residential cooking could also be a material source of outdoor BC and BrC, germane for climate effects on a local (and potentially regional) scale. Recent studies have shown that volatile chemical products and indoor sources are becoming increasingly important for ambient air quality.[28,29] Moreover, indoor cooking activities have been shown as important contributors to organic aerosol concentrations in urban environments.[30–32] While indoor PM concentrations are much lower in developed countries due to the use of cleaner gas and electric stoves, cooking still constitutes an important indoor air pollution source that might adversely impact occupant health and may have potential effects on climate.

This work presents results obtained during the House Observations of Microbial and Environmental Chemistry (HOMEChem) study, an experimental campaign investigating how everyday indoor activities—such as cooking, cleaning, and human occupancy—affect the chemistry of indoor environments.[33] Specific objectives of this work were to characterize the impacts of cooking activities on indoor air quality in terms of BC and BrC concentrations, especially in comparison with outdoor contributions, and to determine the resulting absorption Ångström exponent (AAE) for different indoor conditions.

Real-time data collected in studies of this kind can improve our understanding of the generation of BC and BrC indoors and might be helpful for future studies on health effects due to personal exposure to these pollutants in indoor environments.

2. Methods

2.1 Measurement Site and Ventilation Conditions

The HOMEChem experiment was conducted in June 2018. Descriptions of the overall study goals, test house, experimental design, activities, and measurements are described in detail in Farmer et al.[33]. Briefly, the study was conducted in a 111-m^2 manufactured, three-bedroom test house located at the University of Texas at Austin research campus. The test house has been used previously in several studies on indoor environmental quality and building energy research.[34–36]

An internal fan and duct system recirculated and effectively mixed air throughout the house at a flow rate equivalent to 8 house volumes per hour. This recirculating system was coupled to a typical residential air conditioning system. The thermostat that controlled compressor operation maintained the house at a target temperature of ~25 °C for most of the time. An outdoor air supply system kept the test house at positive pressure and maintained an air exchange rate of ~0.5 h^{-1}. To assist with effective internal mixing, interior doors (except those to the bathrooms) were kept open. A ceiling fan was also used continuously in the living room area. No filters were present in the ventilation systems. The range hood above the stove was not used during this study.

2.2 HOMEChem Experimental Design

The HOMEChem campaign included different types of experiments in which prescribed activities were performed inside the test house. The present work focuses on three types of experimental days, briefly described below.

Each of three Sequential Stir-fry days entailed cooking four replicate vegetable stir-fry and rice meals (some using a propane-fueled stove and some on an electric hot plate) and included at least two "house open" periods, in which doors and windows of the test house were opened to the outdoors for 30 minutes in between cooking experiments. On the other hand, during cooking periods the external doors and windows remained closed. For each meal, the recipe and quantities were maintained constant to ensure that the cooking activity was controlled to some extent. However, different volunteers cooked meals leading to some variability in the cooking process and temperature.

Layered Day activities occurred on four days of the campaign. These were designed as "day in the life" simulations, investigating the potential interactions of cooking and cleaning performed by three house occupants. The occupants stayed inside the house from 8:25 am to ~6:00 pm (CDT), and all doors and windows remained closed during this period. The following scripted activities were undertaken: preparing breakfast (eggs, sausage, toast, and coffee), mopping the floor with a pine-scented cleaner, cooking lunch (the same stir-fry as in sequential stir-fry days), making coffee and toast, preparing dinner (lasagna on one day and beef chili on the remaining three days), mopping the floor with a bleach-based cleaner, and, before leaving the house, starting the automatic dishwasher.

Each of two Thanksgiving Day experiments simulated a holiday meal preparation by four volunteers from 8:40 am to 3:40 pm (CDT), including breakfast (the same breakfast as in layered day experiments). At ~4:00 pm, 12-14 additional volunteers entered the house as guests to partake in the meal. All occupants left the house at 5:00 pm after performing cleaning activities.

Additional data are shown for no-activity periods, which comprise all measurements collected when the house was closed and unoccupied - mostly during nighttime. Data collection for these periods started after particulate matter concentrations generated from the last activity of the day decayed to background levels and ended at 6:30 am every morning when the test house was reopened for instrument maintenance.

2.3 Instrumentation and Associated Calculations

Two portable aethalometers (microAeth MA200, Aethlabs, San Francisco, CA) concurrently measured the concentration of light-absorbing particles indoors and outdoors. Portable aethalometers are relatively inexpensive instruments that can be deployed easily to provide additional information to overall indoor PM measurements, including multiple optical properties. MicroAeth aethalometers have been used to monitor personal exposures in multiple previous studies owing to their compactness and ability to measure BC continuously for weeks.[37–39] The indoor unit was located on the kitchen countertop, with its inlet ~ 0.6 m from the stove. The outdoor unit was located in an air-conditioned trailer adjacent to the test house; it sampled outdoor air ~4 m above ground level and ~4 m north of the test house through ~2 m long × 6.4 mm inner diameter conductive tubing that traversed a trailer window and was mounted at the roof. The indoor unit was flow calibrated (microAeth MA series flow calibration kit, Aethlabs, San Francisco, CA) as part of a firmware update before the start of the campaign. The outdoor unit had been recently purchased and was deployed for the first time during the campaign, after factory calibration. For the June 25th Layered day and the June 27th Thanksgiving Day experiments, data correction for the indoor MA200 was not possible, so data from a different aethalometer (AE33, Magee Scientific, Berkeley, CA), also deployed throughout the HOMEChem campaign, were used. The AE33 aethalometer was located in the same air-conditioned trailer as the outdoor MA200 unit and was operated at 5 l min^{-1} and 1 second time resolution in "dual spot" mode; an algorithm that provides high quality data with real-time loading effect compensation.[40] The AE33 was connected to an inlet that continuously switched between indoor air (25 min) and outdoor air (5 min). Time series data from this instrument were then converted to 1 min averages for analysis.

We operated the MA200 aethalometers in "single spot" mode with a 100 ml min^{-1} sample flow rate and one-minute time resolution. These aethalometers measure light absorbing carbon concentrations based on the difference in light attenuation between a continuously loaded filter and a reference (blank) filter at five wavelengths: 375 nm, 470 nm, 528 nm, 625 nm, and 880 nm. As filtered particles accumulate on the sampling spot, the intensity of light transmittance (J) decreases compared to the reference spot (J_o), causing a change in light attenuation (ATN), where ATN = $-\ln(J/J_o)$. The concentration is then calculated for each channel using Equation 1:[41]

$$C_\lambda = \frac{\sigma_{abs}}{\alpha_{abs}} = \frac{1}{\alpha_{abs}}\left(\frac{A}{Q}\right)\left(\frac{\Delta_{ATN}}{\Delta t}\right) \qquad (1)$$

where C_λ is the concentration for wavelength λ, σ_{abs} is the particle absorption coefficient and α_{abs} is the mass absorption coefficient of the particle cross-section. The values of α_{abs} for each wavelength were provided by the manufacturer and are listed in Table S1. Other parameters are as follows: A is the cross-sectional area of the tape spot, Q is the sample air flow rate, Δ_{ATN} is the change in light attenuation for the time interval Δt.

The concentration measured at the 880 nm wavelength is referred to as the mass equivalent black carbon concentration and hereafter will be referred to as BC.[42] The concentration measured at the 375 nm wavelength is referred to as ultraviolet particulate matter (UVPM); it includes both BC and BrC contributions.[43] The brown carbon concentration was estimated by subtracting the predicted black carbon absorption (linearly extrapolated from σ_{abs} at 880 nm assuming AAE value of 1) from the total absorption at 375 nm, which was then converted to a concentration using α_{abs} at 375 nm (24 m^2 g^{-1}). This method of estimation for BrC is similar to those in previous studies apportioning BrC in both indoor and outdoor environments.[44,45]

It is important to note that this estimation method holds best for externally mixed aerosols. In the case of internal mixtures, the AAE for BC can be higher than 1 due to lensing effects.[46,47] This feature may lead to an underestimation of BrC for internally mixed aerosols. The method of BrC estimation used in this study has also been shown to introduce uncertainties in the range of +7% to -22%.[47] However for AAE values greater than 1.6, this method can be used with greater confidence.[46] We acknowledge that previously published values on the α_{abs} of LAC have been shown to exhibit a considerable amount of variability (5-39.5 m^2 g^{-1}) due to inherent measurement uncertainties and the mixing state of particles.[45,48] Therefore the BrC concentrations values reported in this study are meant as a semi-quantitative comparative analysis into the characterization of various indoor sources.

For our study, we calculated AAE using Equation 2:[49]

$$\mathrm{AAE} = -\frac{\log\left(\sigma_{abs,375nm}/\sigma_{abs,880nm}\right)}{\log\left(375\,nm/880\,nm\right)} \tag{2}$$

The AAE value can provide insight into the composition of emissions-associated particles, such as the relative preponderance of BC or BrC particles during an event.[50] The value of AAE has been used in previous studies of outdoor air for source apportionment to separate traffic from wood-burning emissions.[51–53] For these source-apportionment studies, the AAE of pure, uncoated BC is assumed to be 1 and the AAE value greater than 1 is attributed to non-BC emissions owing to increased light absorption in the ultraviolet range.[10,47,54] The corresponding AAE values calculated using linear fitting are usually defined as $\mathrm{AAE}_{\mathrm{TR}}$ for traffic emissions consisting mainly of BC and ranging between 0.8 and 1.1 and $\mathrm{AAE}_{\mathrm{WB}}$ for wood burning emissions to estimate BrC emissions, reported to be in the range of 0.9-3.5.[52]

Although the optical absorption literature is mature in the context of outdoor air pollution and atmospheric chemistry, that is not the case for indoor sources. Real-time AAE values can provide insight into the variability among different types of food preparation in terms of relative BC and BrC emissions and can help characterize the differences between indoor and outdoor source contributions to indoor PM.

2.4 Corrections Due to Loading Effect and Noise

We utilized an optimized noise-reduction averaging (ONA) algorithm in post-processing to reduce noise from the raw data with an ATN threshold setting of 0.01.[55] This algorithm uses increments of ATN value to determine periods of time averaging interval for BC data smoothing.

The post-processed data were corrected for loading effects using the procedure described in Virkkula et al.[41]

We calculated a correction factor, K_i, using Equation 3:[41]

$$K_i = \frac{1}{ATN(t_{i,\mathrm{last}})}\left(\frac{C(t_{i+1,\mathrm{first}})}{C(t_{i,\mathrm{last}})} - 1\right) \tag{3}$$

where $C(t_{i+1,\mathrm{first}})$ is the first measurement after the tape moves to a new spot, $C(t_{i,\mathrm{last}})$ is the last measurement result for filter spot i and $ATN(t_{i,\mathrm{last}})$ is the maximum preset ATN value for a given wavelength channel. Accordingly, the concentration for each wavelength channel in ng m^{-3} was calculated using Equation 4:[41]

$$C_{corrected} = (1 + K_i \times ATN) \times C \tag{4}$$

The corrected data were validated by plotting $\log(\lambda)$ versus $\log(\sigma_{\mathrm{abs}})$ to observe the power dependence of the absorption coefficient (σ_{abs}) in relation to the wavelength (λ) as per recommendations in Devi et al.[50] We excluded from the analysis measurements with $R^2 < 0.8$ for $\log(\lambda)$ versus $\log(\sigma_{\mathrm{abs}})$, which amounted to less than 1% of the entire dataset.

2.5 Quality Assurance and Quality Control

Diffusion dryers (0.45 m long × 0.07 m inner diameter) filled with self-indicating silica beads were attached to the inlet of each aethalometer to minimize the effects of relative humidity (RH), which is known to affect measured concentrations due to aerosol water uptake and subsequent changes in optical properties.[56] To determine the effectiveness of the diffusion dryers over the course of the campaign, we plotted the RH measurements reported by a sensor located near the kitchen of the test house and the aethalometer's internal RH measurements over the entire campaign (Fig. S1). Although the indoor aethalometer's RH values (12-18%) generally varied with the house RH (38-77%), overall conditions in the aethalometer were much drier and the amplitude of variation was smaller in the aethalometer than in the kitchen. A Nafion^{TM} membrane dryer was used at the inlet of the AE33 aethalometer.

Diffusion losses in the diffusion dryer were calculated for a particle size range of 10 nm to 1 µm.[57,58] For particle diameters greater than 100 nm, diffusion losses were less than 2% (Fig. S2). To further characterize particle losses for aethalometer measurements, an intercomparison was made between both aethalometers used in this study and, accordingly, a correction factor was obtained as shown in Fig. S3. Afterwards, a diffusion dryer was attached to the inlet of one of the aethalometers while both aethalometers sampled emissions from incense burning in a well-mixed chamber. A comparison plot between the two aethalometers shows good correlation ($R^2 = 1$) with a slope of ~1 for both BC and UVPM channels, suggesting minimal particle loss through the diffusion dryers, as shown in Fig. S4.

For this study, we were able to neglect noise effects due to changes in temperature and vibrations or sudden movement as instruments were stationary in air-conditioned buildings. No size-selective aerosol inlets were used for either instrument and the effective measurement size range is not provided by the manufacturer. The BC limit of detection (LOD) provided by the manufacturer is 30 ng m^{-3} but this value was determined for different flowrate and sampling conditions (5 min time base, 150 ml min^{-1} flow rate). For this study, we assumed an LOD value of 100 ng m^{-3} because this value matches the LOD of the microAeth AE51 series aethalometer model when operated under our study's sampling conditions.[56]

The effects of varying ambient RH on the BC background measurements were assessed by performing a laboratory experiment in which the ambient RH was cycled between 35% and 70%, while the aethalometer inlet was attached to a HEPA filter, as shown in Fig. S5. Although the raw data varied with RH, the data corrected for noise reduction (described in the previous section) didn’t vary with RH when the aethalometer was connected to the HEPA filter.[55] However, there appears to be a positive offset in both raw and corrected BC signal after HEPA filter was first attached to the inlet and it took ~30 minutes for the concentrations to reach the zero level.

An intercomparison assessment between the indoor MA200 and the AE33 for three different experimental days is presented in the supplemental file (Fig. S6 and Fig. S7). AE33 data from 370 nm and 950 nm wavelength channel was used for BC and BrC measurements and the corresponding AAE calculations assuming α_{abs} values of 18.47 $m^2 g^{-1}$ and 7.19 $m^2 g^{-1}$ for 370 nm and 950 nm wavelength channel respectively (values obtained from the instrument manual). On average, BC and BrC measurements from both instruments agreed to within ~10% and ~40% respectively. No consistent bias is apparent in the AE33 to MA200 mass concentration intercomparison. A similar intercomparison of the AAE time series for Thanksgiving Day and Sequential Stir-fry Day also shows similar trends for both aethalometers (average agreement within ~30%, Fig. S8), despite the instrument inlets being located on opposite sides of the kitchen and the AE33 unit also had a longer sampling inlet (~10 m).

3. Results and Discussion

3.1 BC and BrC Concentrations During HOMEChem Events

Fig. 1 depicts BC and BrC concentrations for various events during the campaign. The BC and BrC concentrations were calculated for each event by taking time-averaged concentrations integrated over an event's entire duration, including the associated decay phase period in case of a cooking event.

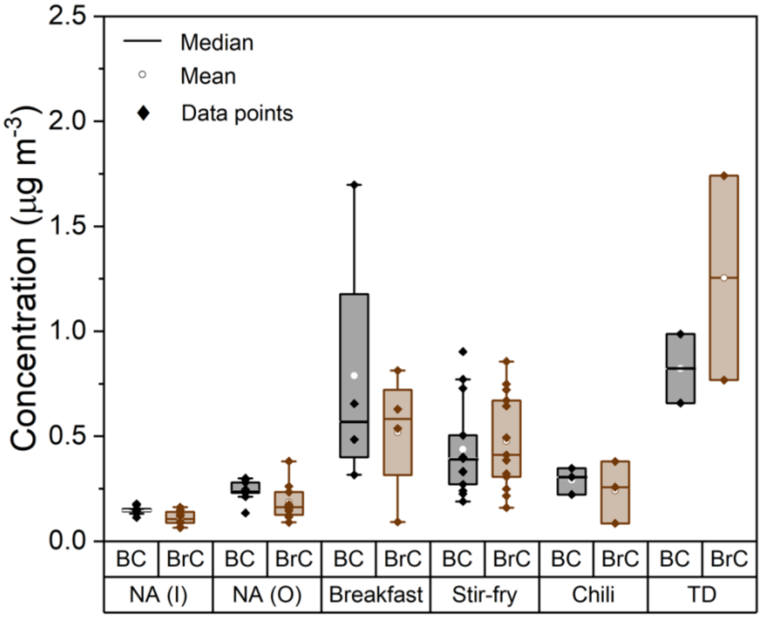


Fig. 1. Black (BC) and brown (BrC) carbon time-averaged concentrations in the test house kitchen during different activities. NA (I) and NA (O) represent indoor and outdoor concentrations during periods of no activity in the test house, respectively. TD represents Thanksgiving Day. On average, the duration of breakfast and chili was ~70 minutes, stir-fry events lasted for ~60 minutes and the average duration of Thanksgiving Day and no activity periods were close to 9 hours.

The mean BC concentration outdoors (0.24 µg m^{-3}) was about 60% higher than indoors (0.15 µg m^{-3}) during periods of no activity in the house. The mean BrC concentration outdoors (0.2 µg m^{-3}) was twice that of indoors (0.1 µg m^{-3}). We present a more detailed discussion of indoor versus outdoor concentrations in section 3.2.

Cooking any meal during this campaign led to significant increases in both BC and BrC compared to periods of no activity. During breakfast, BC (0.8 ± 0.6 µg m^{-3}) and BrC (0.5 ± 0.3 µg m^{-3}) concentrations (mean ± standard deviation) were higher than during lunch (stir-fry) and dinner (chili), also cooked on that experimental day. During breakfast, toast, sausages, eggs, and coffee were prepared near simultaneously and emissions associated with each activity could have contributed indoor concentrations. Both lunch (stir-fry) and dinner (chili) exhibited similar mean BC concentrations (0.4 ± 0.2 µg m^{-3} and 0.3 ± 0.1 µg m^{-3}, respectively), but stir-fry led to a BrC concentration approximately twice that of the chili preparation (0.5 ± 0.2 μg m^{-3} and 0.2 ± 0.1 μg m^{-3}, respectively). Differences in ingredients and cooking temperatures could have led to differences in BC and BrC concentrations between meals. These results highlight a need to further investigate the effect of different aspects of cooking processes (e.g., temperature of cooking, water content of food, type of oils used, and other ingredients) on BC and BrC emissions.

The mean concentrations of both BC and BrC were highest during the Thanksgiving Day experiment since this experimental day entailed a host of different meal preparation activities and combustion related activities, including 3 h of roasting activities inside the propane gas-fueled oven and candle burning. Moreover, this was the only event in which the mean BrC concentration (1.2 ± 0.7 μg m^{-3}) significantly exceeded that of BC (0.8 ± 0.2 μg m^{-3}). We discuss the activities performed on Thanksgiving Day and their associated emissions in more detail in later sections.

3.2 Indoor-to-Outdoor (I/O) Ratios of BC and BrC During Different Events

In this section, we present a direct comparison between BC and BrC levels during indoor activities and corresponding outdoor concentrations. Fig. 2 shows the distribution of I/O ratios of BC and BrC for different events during the HOMEChem campaign. To account for the time lag of aerosol infiltration into the indoor environment, the I/O ratios were calculated by integrating the indoor and outdoor concentrations over the entire duration of an event and then taking the ratios of those time integrals. More detail is presented in Table S2.

As an example of the typical temporal variability in indoor and outdoor measurements, we present a time series of BC and BrC concentrations for the June 8th Layered Day in Fig. S9. Transient indoor BC and BrC concentrations reached as high as ~7.5 μg m^{-3} and ~4.2 μg m^{-3}, respectively, with sharp increases during cooking periods. Outdoor BC and BrC concentrations peaked at ~1 µg m^{-3} and ~0.7 µg m^{-3} on that day, respectively, with smoother temporal behavior.

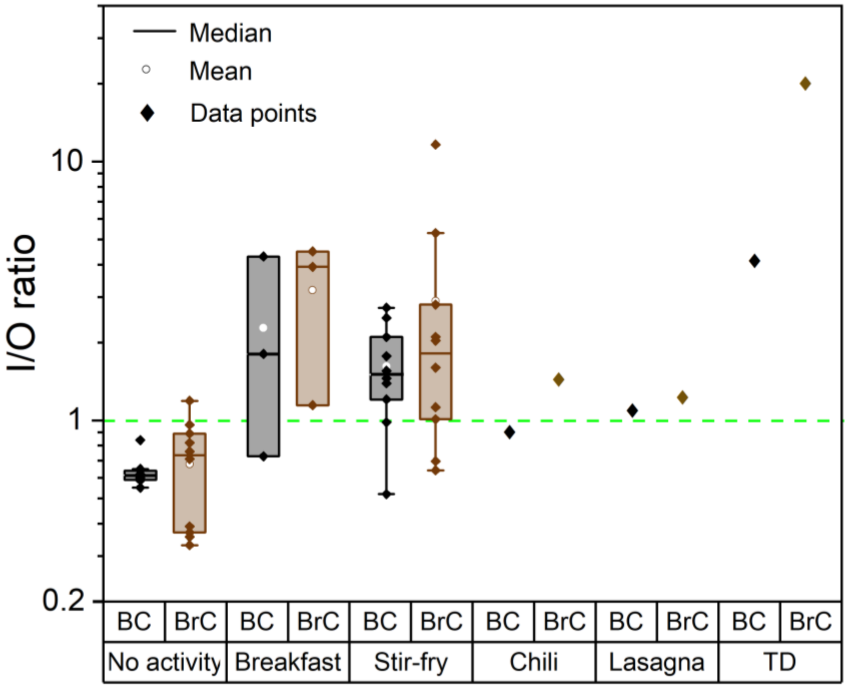


Fig. 2. Box plot showing the distribution of BC and BrC I/O ratios for different events throughout the HOMEChem campaign. Boxes represent the 25th to 75th percentiles, with means indicated by circles and medians in bars. Single data point events are represented by filled diamonds. TD represents the Thanksgiving Day of June 18th. For the Chili, Lasagna, and Thanksgiving Day experiments, only one dataset each was available for analysis. Each data point represents the I/O ratio for an entire experiment, which was calculated by averaging minutely indoor and outdoor concentrations over the entire duration of an event and then taking the ratios of those time integrals. The green line represents an I/O ratio value of 1.

The median I/O ratios for BC and BrC during the periods of no activity were 0.6 and 0.7, respectively. Since there were no known indoor sources of BC and BrC during these times, we attribute the concentrations measured indoors to the penetration and persistence of BC and BrC aerosols from outdoors. This I/O ratio for BC is comparable to the ratios reported by LaRosa et al.[59], in the range of 0.35-0.5, measured as a part of a two-year study focusing on BC exposure of household occupants. Similarly, Viana et al. showed that 70% of indoor BC originated outdoors in an urban building[60] and Reche et al. determined that indoor BC concentrations in urban and suburban schools were greatly dependent on distance to heavily trafficked roads.[61] Studies by Johnson et al.[62] and Avery et al.[63] on the indoor transport of ambient aerosols reported median BC I/O ratio of 0.61 for a mixed-use laboratory space and 0.4 and 0.55 for wintertime and summertime measurements in a university classroom.

All meal-cooking activities mostly led to I/O ratios >1.0. for both BC and BrC. Comparing different meals, breakfast presented the highest median I/O ratio for BC (1.8), followed by stir-fry (1.5), lasagna (1.1) and chili (0.9). A similar trend was also observed for BrC, with the highest median I/O ratio observed for breakfast (3.9), followed by stir-fry (1.8), chili (1.5) and lasagna (1.2). During the Thanksgiving Day experiment, the BrC concentrations indoors reached a level greater than 20 times that of outdoors. The higher temperature (>~200 °C) for oven-roasting activities in addition to a substantially larger meal quantity cooked over multiple stove-top burners may have led to an enhancement in BrC emissions compared to other cooking activities, which were limited to one or two stove-top burners.

3.3 Characterizing Emissions Using the Absorption Ångström Exponent (AAE)

In this section, we discuss the temporal variation of AAE regarding different activities performed during HOMEChem. First, we take as an example the Sequential Stir-fry Day on June 6 to characterize the emissions attributable to indoor cooking and outdoor penetration resulting from window opening, as shown in Fig. 3 (a similar plot for June 12 is shown in Fig. S10).

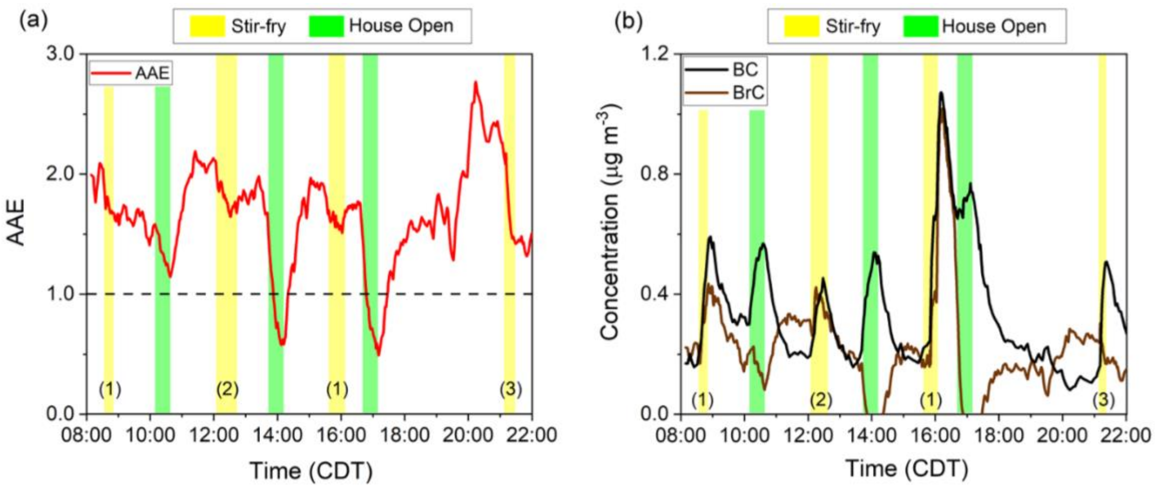


Fig. 3. Sequential Stir-fry Day (June 6): (a) AAE time series; (b) time series of BC and BrC concentrations throughout the day. Data were smoothed using a 10-minute moving average. (1) represents stir-fry cooked on gas stove in a steel wok; (2) represents stir-fry cooked on an electric hot plate (medium setting) in a steel wok; and (3) represents stir-fry meals cooked on gas stove in a cast-iron pan.

The indoor AAE value was >1.0 (indicative of an increased aerosol absorption at near UV wavelengths from BrC emissions) during most of the day, except for two instances when the house was opened to the outdoors. The mean AAE value for stir-fry meals cooked throughout the day was calculated to be 1.7 ± 0.2 whereas the mean value of AAE during the house open periods was 1.1 ± 0.3. With the exception of the house open event at ~10 am, the AAE values dropped below 1 during the remaining two house-open periods. These periods were also associated with a rise in BC concentrations, whereas the corresponding BrC concentrations apportioned during these periods declined below zero. Sudden changes in sample RH due to the opening of windows and doors can lead to evaporation or condensation from filter material which has been known to affect the optical paths of the reference and sensor channel.[37,56] This artifact can lead to sudden spikes in the absorption values for BC channel and therefore introducing uncertainties in BrC concentrations apportioned during the house open periods. RH measurements from the aethalometer's internal sensor (Fig. S11) show that house open periods were indeed associated with increases in RH values, trending towards matching ambient RH levels. Outdoor BC and BrC concentrations for that day can also be seen in Fig. S12. Similar instances of AAE values in the range of 0.5-1 have been observed in ambient environments in previous studies,[64,65] however the hypothesis of a humidity-driven artifact seems to be the most likely explanation for the observed AAE values in the present study.

The BC and BrC time series data also suggest that the cooking was a major source of indoor BrC emissions and both cooking as well as penetration from outdoors contributed substantially to BC levels indoors. It is also noteworthy that different stir-fry meals exhibited different BC and BrC emission patterns. Although most meals contributed both BC and BrC to the indoor air, the relative BC-to-BrC concentrations for each meal were different. This observation may be a result of different cooking temperatures achieved with each type of heating source and cooking surface and also due to differences in volunteer cook behavior while adding ingredients, even though the same ingredients (type and quantity) were used for each cooking episode.

Stir-fry temperature measurements taken during the campaign only provide a rough estimate of the temperature profile during each cooking event, as these values were recorded using an infrared temperature gun, pointing the laser onto the stir-fry ingredients or the cooking surface. The spatially and temporally varying temperature of the cooking surface and the food cannot be fully captured during such experiments. Consequently, we cannot definitively conclude what role cooking temperature or other factors may have had influencing the variability of BC/BrC emissions.

To demonstrate the effects of an intensive indoor cooking event in indoor BC and BrC concentrations, we present in Fig. 4 the results of a Thanksgiving Day experiment performed on June 18. The corresponding AAE values are also depicted.

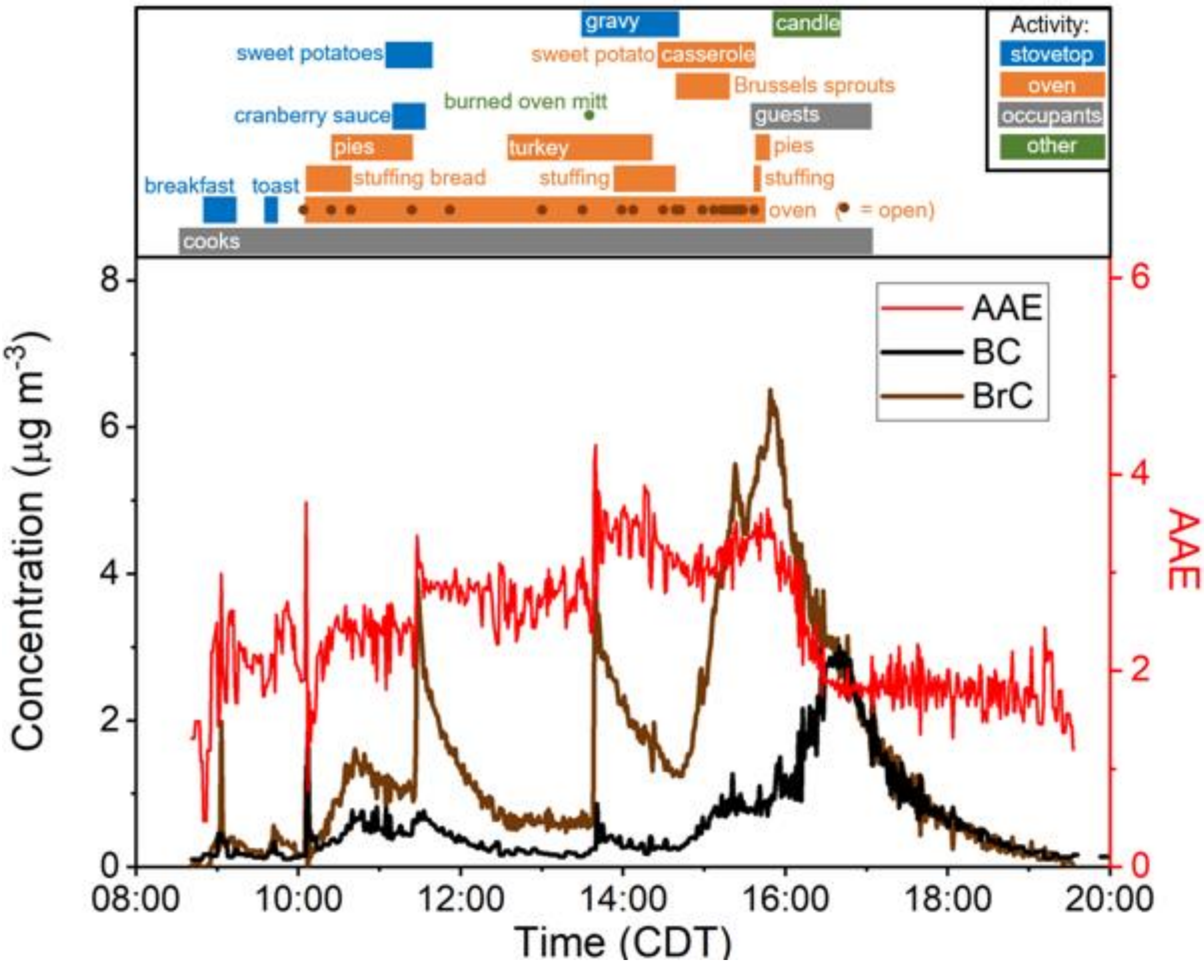


Fig. 4. BC and BrC concentrations during a simulated Thanksgiving Day experiment (June 18). The red trace (right-hand axis) shows the AAE values over time. The upper panel shows the timing and duration of the main activities performed throughout the day.

Fig. 4 shows that multiple activities during cooking and preparation led to peaks in BC and BrC concentrations throughout the day. A comparison of BC, BrC, and size-segregated PM (PM$_{0.5}$, PM$_1$, PM$_{2.5}$, PM$_{10}$) concentrations is presented in Fig. S13. PM mass data was obtained using particle sizing instruments also deployed in the kitchen area during HOMEChem and assuming unit density. Patel et al. presented a discussion of PM density during HOMEChem and demonstrated that the density of PM$_1$ varied between ~ 1.0 g cm^{-3} during cooking periods and ~ 1.5 g cm^{-3} during non-cooking periods.[66] BrC concentrations generally followed the same trends as PM mass throughout the day, but PM concentrations were about 2-3 orders of magnitude higher than BrC concentrations. BC concentrations did not follow the trend in PM as well as did the BrC concentrations. Detailed PM concentration results from HOMEChem can be observed in Patel et al.[66] and Tian et al..[67]

Dominant BC peaks were observed twice, at ~10:00 am, when the oven was first turned on to ~200 ºC and again starting at ~3:50 pm, when two scented candles were lit inside the house. During the remainder of the day, especially during high-intensity cooking activities, emissions were dominated by BrC.

At least six distinct BrC concentration peaks are observed in Fig. 4. The first, at ~9:00 am, coincided with the moment when cooks added tomatoes to a pan with hot, smoking oil (~230 ºC). The next peak coincided with toasting bread in an electric toaster (~9:45 am). Similarly, toasting bread in the oven for stuffing also caused an increase in BrC concentration (10:06 – 10:40 am). Multiple peaks in concentration were the result of specific actions or accidents during cooking. Baking two pies in the oven resulted in a sharp increase in BrC concentrations towards the end of baking, when pie filling briefly dripped into the oven. In addition to the above occurrences, accidentally burning an oven mitt, and roasting Brussels sprouts in the oven also contributed to distinct BrC concentration peaks during the Thanksgiving Day experiment.

The time series plot shows that, during cooking activities, AAE values were in the range of 2-4, whereas during the candle burning event, the AAE declined to less than 2. This observation suggests a difference between cooking emissions, dominated primarily by BrC, and candle emissions, substantially comprising soot (BC) particles. Overall, the AAE mean value throughout the day was 2.4 ± 0.7.

It is important to note that filter-based aerosol light absorption measurements have been shown to suffer a considerable positive bias—up to 100% for environments with organic aerosol (OA) concentrations > 12.5 µg m^{-3}.[47] During Sequential Stir-fry Day and Thanksgiving Day experiments, aerosol emissions from cooking may have resulted in a positive bias in aethalometer measurements. Therefore, the data reported for those periods might be an overestimate of true concentrations.

3.4 BC Indoor Exposure Assessment

In this study, time-averaged BC concentrations were used to estimate the exposure of an individual residing in the test house with the assumption that this person was present in the kitchen area over the duration of the entire cooking and decay periods. The resulting BC exposure for each event was calculated by multiplying the time-averaged concentrations with the duration of each event. In Fig. 5, we compare estimated BC exposures (in units of µg m^{-3} h) for different meals prepared during HOMEChem (breakfast, chili, and stir-fry) by accounting for periods of elevated BC concentrations associated with a particular meal. BC exposure during the breakfast meal was highest among all the discrete cooking events. The exposure values for individual meals are shown in Table S3.

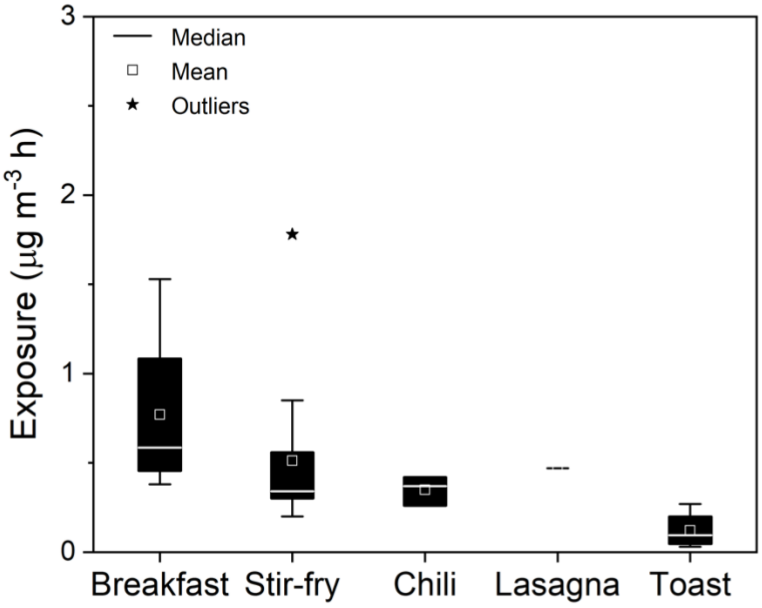


Fig. 5. BC exposure for different meals throughout the HOMEChem campaign.

The mean BC exposure during the breakfast (0.8 ± 0.5 μg m^{-3} h) was twice as high as that for chili preparation (0.4 ± 0.1 μg m^{-3} h). The mean BC exposure for stir-fry (0.5 ± 0.4 μg m^{-3} h) and cooking lasagna (0.5 μg m^{-3} h) were comparable to each other. The BC exposure during the toasting event was the lowest among all the meals (0.1 ± 0.1 μg m^{-3} h) because of its short duration (~ 5 min). It is important to mention that the lasagna was cooked inside the oven whereas all the other meals were cooked in the open space adjacent to the aethalometer inlet, so this value is representative of exposure at that specific, stationary location.

We can use the same approach to compare BC exposure values for an entire day, for periods starting from ~8:30 am until the time when the test house was closed at the end of the day, and compare those values with periods of no activity of similar durations (Fig. 6). The no-activity period represents a hypothetical scenario in which an occupant would be present in the closed house during a period of no indoor PM-emitting activities, thus representing a “best-case scenario” for BC exposure during the HOMEChem experiment. In reality, no occupants were present in the house when these measurements were taken. We also acknowledge that this “best-case scenario” could have been further improved if the test house ventilation system had been outfitted with a good-quality filter to remove PM from outdoor sources or with the use of a portable air filtration system in the house.

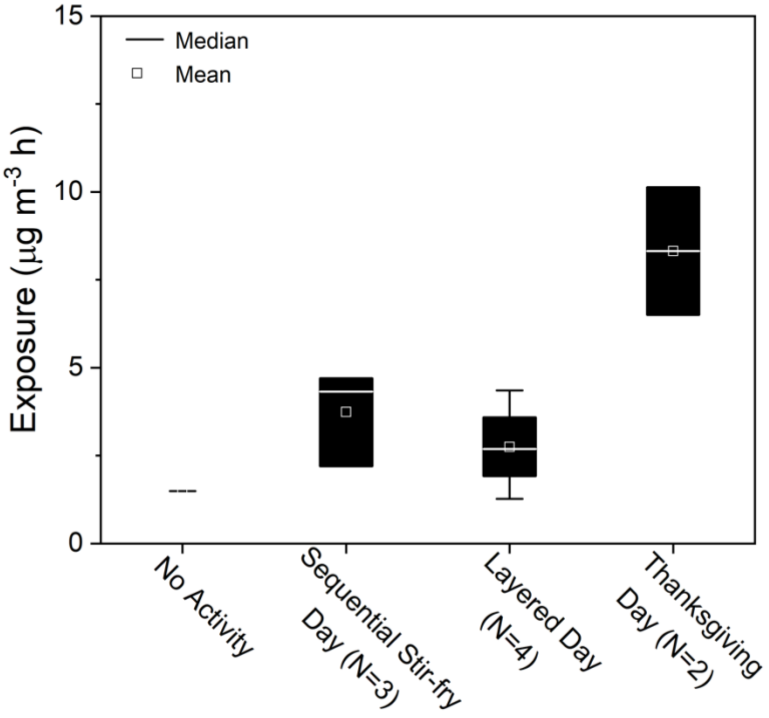


Fig. 6. All-day BC exposure for different experimental days during the HOMEChem campaign. In the Sequential Stir-Fry Days, the same stir-fry meal was prepared 3-4 times, and the house was opened for ~30 min 2-3 times throughout the day. In Layered Days, three meals were cooked throughout the day (breakfast, stir-fry lunch, and a dinner). In both Layered Days and Thanksgiving Days, the house was not opened for any significant period.

The mean BC exposure during Sequential Stir-fry Days (4 ± 1 µg m^{-3} h) and Layered Days (3 ± 1 µg m^{-3} h) were similar to each other and ~ 2× higher than the BC exposure during the periods of no activity (2 µg m^{-3} h). The mean BC exposure (8 ± 3 µg m^{-3} h) during Thanksgiving Day was highest among all the full-day experiments. That outcome is expected as Thanksgiving Day was designed to be a cooking intensive experiment and also included candle burning activities which were associated with the largest peaks in BC concentration observed throughout the day.

Note that during Stir-fry Day and Layered Day experiments, meals for ~3 people were cooked, whereas during Thanksgiving Day experiments, a meal was prepared for ~15 people. However, we cannot infer that it was the quantity of food alone that contributed to higher BC exposure because different meals were prepared during these experiments, each with distinct set of ingredients, temperature profiles, and heating source as well as other combustion activities (candle burning, accidentally burning an oven mitt, etc.). Each of these factors likely played a role influencing BC emissions and resulting exposures.

We also acknowledge that while BC exposures were calculated for daytime periods, indoor infiltration of BC from outdoor sources continues throughout the night which might lead to higher total BC exposure. Moreover, even though the indoor space of the test house was relatively well-mixed, there might be some noteworthy spatial variability in the kitchen area due to the short-term peaks associated with cooking emissions. According to Boedicker et al., particle number concentrations in the kitchen were up to ~70% higher than in other rooms during HOMEChem.[68] Therefore, the BC exposure values for occupants present in other rooms during cooking activities might be lower.

4. Conclusion

During the month-long HOMEChem experiment, cooking was a major contributor of BC and BrC indoors, leading to concentrations that were ~2-10× higher than in periods of no activity in the test house. The results also indicate that sporadic indoor sources of BC throughout the day lead to significantly higher exposure than outdoor infiltration. BrC concentrations generally followed the temporal trends of PM mass during Thanksgiving Day experiments. The median I/O ratios of BC (BrC) ranged from 0.6 (0.7) during periods of no activity to 4 (22) during the Thanksgiving Day. The investigation of AAE values for different experimental days showed an increase with the intensity of cooking activities due to the dominance of BrC particles in cooking emissions. The indoor BC exposure assessment performed in this study for a kitchen microenvironment showed that candle burning and cooking emissions are prominent indoor sources of BC and, in the case of our experimental setup, cooking 3-4 meals a day doubled the daytime BC exposure for an occupant residing in the kitchen area compared with exposure during background conditions. Although these types of personal exposure assessment studies represent idealizations of more complex realities, they can contribute to a growing body of literature on the impacts of cooking on pollutant exposures and associated health risks for people at home. Results from this study can help bring into perspective various indoor sources of BC and BrC, with the understanding that indoor PM can contribute to ambient air quality. The results from this study also invite more research into the type of compounds generally classified as BrC released primarily during cooking and their potential toxicological effects on human health.

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