INDOOR BLACK AND BROWN CARBON FROM COOKING ACTIVITIES AND OUTDOOR PENETRATION: INSIGHTS FROM THE HOMECHEM STUDY

by

SUMIT SANKHYAN

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written by Sumit Sankhyan

has been approved for Department of Mechanical Engineering

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Dr Marina Vance

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Dr Shelly Miller

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Dr Michael Hannigan

Date_____

The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.

Abstract

Sankhyan, Sumit (MS, Mechanical Engineering)

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

Thesis directed by Assistant Professor Marina Vance

Particulate matter emissions from cooking activities are a major contributor to indoor air pollution in households. A major part of these emissions consists of light absorbing aerosols known as black carbon (BC) and brown carbon (BrC). The goal of this work was to characterize the contributions of indoor and outdoor sources of BC and BrC to the indoor environment by measuring real-time concentrations of these components indoors and outdoors concurrently during the month-long HOMEChem field study in June 2018. We quantified the penetration of BC and BrC into the house from outdoor sources and characterized the impacts of cooking activities on indoor air quality in terms of BC and BrC concentrations, including exposure and dose calculations. The BC exposure was at least 4 times higher during the preparation of any cooked meal than during a comparable period of no activity. The exposure and dose during a simulated *Thanksgiving Day* were highest with BrC concentrations peaking at 6390 ng m^{-3}. The Power law fitting approach was used to calculate angstrom exponent (α) for characterizing aerosol emissions during different activities. The value of α ranged from 1.1 to 3.67 during the entire campaign, with the lowest value (indicative of BC-dominated aerosols) observed in periods of no activity and the highest value (indicative of BrC-dominated aerosols) observed during the *Thanksgiving Day* experiments. Real-time data collected in this study improves our understanding of the generation of BC and BrC indoors and the effects of outdoor air pollution on indoor air quality.

Acknowledgements

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1. Introduction

The field of indoor air quality has seen notable research impetus in the last two decades due to the severe health effects associated with a wide variety of indoor air pollutants.[1–3] To save energy, buildings are being made more airtight and with lower air exchange rates, which may lead to decreased exposure to air pollutants of outdoor origin, but might result in increased exposure to indoor air pollutants. In developed countries people spend 90 percent of their time indoors, which leads to increased exposure from indoor air pollutants and results in increased risk of developing health issues.[4,5] Cooking indoors is one of the most significant contributors to indoor air pollution due to the release of ultrafine and fine particulate matter (PM) as well as gaseous air pollutants such as nitrogen oxides (NOx) and polycyclic aromatic hydrocarbons (PAH) when combustion is used as heating source[6]. Pope et al.[7] found a relationship between a decrease in fine particulate pollution and an increase in life expectancy, which emphasizes the need to study the factors that might contribute to a reduction of exposure to indoor air pollutants. Health effects associated with cooking emissions have been well documented in developing countries, in which cooking usually takes place in unventilated spaces using solid fuels. According to the global burden of disease study, 3.5 million premature deaths have been linked to smoke exposure from solid fuel cooking.[8]

Cooking aerosols contain carbonaceous particles that have light absorbing properties. Optically absorbing carbonaceous aerosols are broadly described as black carbon (BC) and brown carbon (BrC). BC particles are known to absorb light of 880 nm wavelength efficiently, while BrC consists of inorganic and organic compounds that absorb light most efficiently in the ultraviolet (UV) range of light (~375 nm wavelength).

BC is a major component of soot released during combustion processes. As such, BC can be used as a tracer for PM sources like diesel exhaust, wood smoke, and other combustion-related activities. LaRosa et al.[9] reported rush-hour traffic and wood burning activities as the major

outdoor BC sources and cooking and burning candles as the primary indoor sources. BC is a valuable indicator of air quality in environments dominated by combustion particles. Therefore, real-time monitoring of BC concentrations may be instrumental in characterizing indoor cooking sources.

The health effects of BC are closely associated with PM exposure, which includes an increase in the risk of developing respiratory and cardiovascular ailments.[10,11] Multiple studies showed that chronic exposure to BC can lead to inflammatory response and the development of benign and malignant carcinomas in rat lungs.[12,13] Additionally, BC has been shown to play an indirect role by acting as a universal carrier for semi-volatile organic compounds released from combustion sources. Nikula et al. showed that the pulmonary carcinogenicity of "bare" BC and diesel exhaust were fairly similar, indicating that BC was the main driver of observed carcinogenic effects.[13] The documented adverse health effects of BC inhalation make it essential to quantify BC exposure and dose during combustion-related activities.

The primary sources of outdoor BrC emissions are biomass combustion such as wildfires and wet trash burning in developing countries.[14,15] Not many studies have been published on health effects specifically related to BrC exposure. However, BrC sources—such as biomass combustion—have been associated with various long-term, chronic health effects, such as cardiovascular ailments and chronic obstructive pulmonary disease (COPD).[16]

This work investigates results obtained during the House Observations of Microbial and Environmental Chemistry (HOMEChem) study. HOMEChem was an indoor air chemistry field campaign conducted in June 2018 in Austin, TX to investigate how everyday indoor activities—like cooking, cleaning, and human occupancy—affect the chemistry of indoor environments.

The goal of this work was to characterize the contributions of indoor and outdoor sources of black and brown carbon to the indoor environment by measuring real-time concentrations of BC and BrC indoors and outdoors concurrently. Specific objectives were to quantify the penetration of BC and BrC into the house from outdoor sources and to characterize the impacts of cooking activities on indoor air quality in terms of BC and BrC concentrations. Real-time data collected in this study was used to improve our understanding of the generation of BC and BrC indoors and the effects of outdoor air pollution on indoor air quality.

2. Materials and Methods

2.1 Measurement Site

The study was conducted in a 111-m^2 manufactured three-bedroom, two-bathroom test house located at the University of Texas at Austin research campus. The test house has been used for several studies on indoor environmental quality and building energy research projects.[17–19]

To promote even and constant mixing in the air flow throughout the test house, two separate ventilation systems were operated during HOMEChem. An internal ventilation system recirculated air inside the house at a flow rate equivalent to 8 air changes per hour (ACH). This internal ventilation system was coupled with a dehumidifier and a typical air conditioning system connected to a thermostat set to maintain the house at 24.4 °C. An additional outdoor air supply system was operated to keep the test house at positive pressure and to maintain the external air exchange rate between 0.5 - 0.8 ACH. No filters were used in either ventilation system for this study to avoid variability due to filter type and loading effects. For similar reasons, the exhaust hood over the stove was also not used during this study. House parameters, such as relative humidity (RH), temperature, and energy consumption were continuously recorded.

Indoor sampling was performed on a kitchen countertop, with the instrument inlet located approximately 0.6 m from the stove. Outdoor sampling was performed adjacent to the test house—with the instrument housed inside a trailer—and the inlet was located at approximately 4 meters height from the ground and approximately 4 meters north of the test house.

2.2 Instrumentation

Two Microaeth aethalometers (MA200, Aethlabs, San Francisco, CA) were used to measure the concentration of light-absorbing aerosols indoors and outdoors concurrently. It calculates the concentration of light-absorbing aerosol particles based on the difference in light attenuation measured between a continuously-loaded filter and a reference (blank) filter across five wavelength channels: 375 nm, 470 nm, 528 nm, 625 nm, and 880 nm. This instrument has been used to monitor personal exposures in multiple previous studies due to its compactness and its ability to measure BC continuously for weeks.[20–22]

The aethalometers were operated in "single spot" mode with a 100 ml min^{-1} sample flow rate and a one-minute measurement time resolution. As particles get deposited on the sampling spot, the intensity of light transmittance (I) keeps on decreasing as compared to the reference spot (I_0), thereby causing a change in light attenuation (ATN), where $ATN = -\ln(I\ I_0^{-1})$. The concentration is the calculated for each channel using Eq 1.[23]

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

where C_λ is the concentration for wavelength channel λ, σ_{abs} is particle absorption coefficient and α_{abs} is the mass absorption coefficient of the cross-section. A is the cross-sectional area of the tape spot, Q is the sample air flowrate, $\Delta ATN\ \Delta t^{-1}$ is the change in light attenuation for the time interval Δt. The concentration measured at the 880 nm wavelength is referred to as the BC

concentration whereas that measured at the 375 nm wavelength is referred to as the BrC concentration.

Diffusion dryers (0.45 m length and 0.07 m annular diameter) filled with self-indicating silica beads were attached to the inlet of each aethalometer to avoid sudden changes in RH to affect measured concentrations due to aerosol water uptake and subsequent changes in optical properties. Since both instruments were kept indoors—one in the temperature-controlled house and instrument trailer—throughout the study, potential noise effects due to changes in temperature and vibrations or sudden movement can be neglected.

2.3 HOMEChem Experimental Design

The month-long campaign was divided into different types of experiments based on the specific activities that were performed inside the test house. Only the experiments from which data was used in this work are described below:

A *Response Day* experiment was conducted to monitor the time required for the house to reach "steady state" conditions after it has been flushed with outdoor air. This experiment was performed during one day by repeatedly opening the doors and windows of the house for 30 min. This was followed by closing the house and keeping it closed and undisturbed for at least 90 min. The air conditioning system was kept off for most of this day, but box fans and the air handling unit were kept on during some replicates to enhance air movement within the house.

Three *Sequential Stir-fry* days were performed and consisted of four stir fry events in each day, conducted using different heating methods and utensils. This experiment was performed during three days. An electric heating plate and a propane-fuelled stove were used to obtain variability in heating methods. For utensils, a cast iron and a stainless steel wok were used. The experimental meal consisted of stir-fried vegetables topped with hot sauce and teriyaki sauce

with a side of white rice. For each meal, the recipe and quantities were maintained constant to ensure that the cooking experiment was controlled. External doors and windows were kept closed during all sequential experiments. After each sequential experiment replicate, all windows and doors were opened and left open for 30 min and, after closing them, the house was left unoccupied for 80 min until volunteers re-entered for the next replicate.

Layered Day activities were performed during four days of the campaign and focused on studying the potential interactions of cooking and cleaning perturbations performed by three occupants who stayed inside the house from 9:00 AM to 5:30 PM with no one leaving the or entering the house within that time period. The occupants prepared breakfast (eggs, sausage toast and coffee), mopped floors with a pine-scented cleaner, then prepared lunch (the same vegetable stir fry as in *Sequential Stir-fry* days), then occupants prepared coffee and toast, then prepared dinner (lasagne for one day and beef chili for three days), mopped floors with a bleach-based cleaner, ran the dishwasher, and left the house.

There were also two *Thanksgiving Day* experiments that simulated the process of a holiday meal preparation by 4 volunteers from 8:30 AM to 3:40 PM, including breakfast (the same breakfast that was prepared during *Layered Day* experiments). At approximately 4:00 PM, 12 - 14 occupants entered the house as guests to partake in the meal. All occupants left the house at 5:30 PM after performing cleaning activities and starting the dishwasher.

2.4 Data Analysis

The aerosol light absorption at the visible range follows a power law dependence given by $\sigma_{abs} = K \lambda^{-\alpha}$, where K is a constant, λ is the wavelength of light, and α is called the angstrom exponent. A power-law fitting approach was used to estimate α for different events to characterize the aerosol optical behaviour, which may be tied to chemical properties. The value

of α for pure, uncoated BC has been fixed at 1. For emissions from non-BC sources, the α value is greater than 1 due to light absorption in the ultraviolet range.[24]

The raw data was corrected for loading effects using the procedure given in Virrakula et al.[23]

The correction factor K_i was calculated using the Eq 2:

$$Ki = \frac{1}{ATN(ti,last)}\left(\frac{BC(ti+1,first)}{BC(ti,last)} - 1\right) \quad (2)$$

Where BC(ti +1,first) is the first measurement after the tape moves to a new spot, BC (ti, last) is the last measurement data for filter spot I and ATN (ti, last) is the maximum pre-set ATN value. Accordingly the corrected BC measurement ($BC_{corrected}$) can be calculated using Eq 3:

$$BCcorrected = (1 + K.ATN)\mathrm{BC} \quad (3)$$

The corrected data was validated by plotting log (λ) versus log (σ_{abs}) to observe the wavelength dependence of the absorption coefficient (σ_{abs}) as per recommendations presented in Devi et al.[25] Datasets with $R^2 < 0.8$ were removed from the analysis. Additionally, the EPA's optimized noise-reduction algorithm (ONA) was used to account for noise reduction based on recommendations put forward by Hagler et al.[26] The ONA algorithm along with an ATN threshold setting of 0.01 was used to filter out noise from real-time data.

Penetration factor for BC was calculated using the I/O ratios as per the methodology presented in Thatcher et al.[32] An overall deposition rate was determined for BC concertation using Eq 4:

$$\lambda d = \left(\frac{1}{t}\right) * \ln\left(\frac{Ci}{C}\right) - \lambda v \quad (4)$$

Where λ_d is the overall deposition rate (hr^{-1}), λ_v is the infilitration rate (hr^{-1}), and $\left(\frac{1}{t}\right) * \ln\left(\frac{Ci}{C}\right)$ represents the slope of first order decay curve for BC concetration calculated during window open events. The penetration factor P can be calculated using the Eq 5:

$$P = \left(\frac{Cin}{Co}\right) * \frac{\lambda d + \lambda v}{\lambda v} \quad (5)$$

$\left(\frac{Cin}{Co}\right)$ is the indoor to outdoor ratio of BC calculated for periods of no activity.

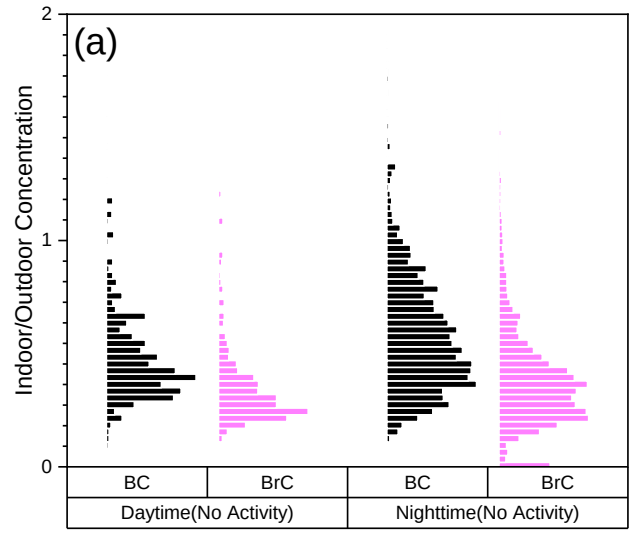
The aerosol absorption of light at the visible spectrum follows a power law dependence given by $B_{abs} = K\lambda^{-\alpha}$ where K is a constant, λ is wavelength and α is called Angstrom Exponent[27]. A fitting approach was used to calculate α for different events during the HOMEChem experiment, using average light absorption values for the duration of each activity. The value of α for pure, uncoated BC has been fixed at 1. For emissions from non-BC sources, the α value is usually greater than 2 due to the absorption in ultraviolet range.

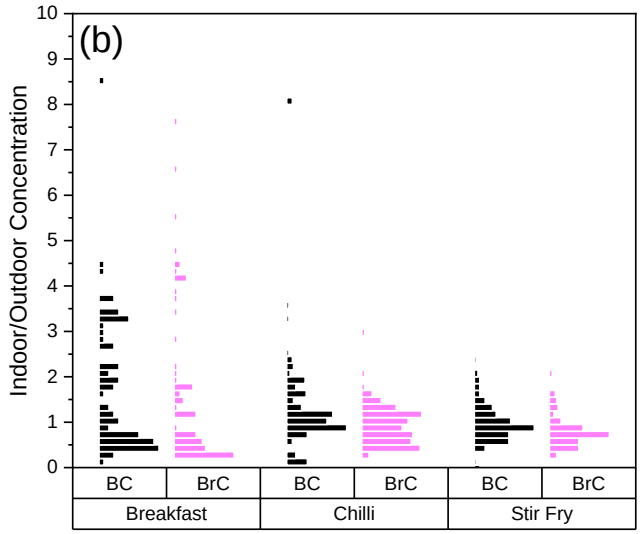
In this study, an exposure assessment for different meals cooked during HOMEChem was performed using the kitchen as a microenvironment. The dose of BC was calculated by multiplying the average exposure measured for each activity with the inhalation rate and the duration of each activity. The exposure was calculated over the duration for which the cooking heat source, either the stove or electric hot plate, was in use. An inhalation rate of 11 l min^{-1} was assumed to represent an adult engaged in home activities according to information published by Dons et al.[28] and Allan et al.[29]

3. Results and Discussion

3.1 Indoor-to-outdoor ratios of BC and BrC during different events

Figure 1 shows the ratios between indoor and outdoor concentrations of BC and BrC for different events during the HOMEChem campaign. Cooking-intensive events—such as *Thanksgiving Day* and *Layered Day* meals—showed higher median ratios for both BC and BrC as compared to periods of no activity inside the house. Mean and median ratio values for all activities can be seen in Table S1.


(a)
Indoor/Outdoor Concentration
0
1
2
BC
BrC
BC
BrC
Daytime(No Activity)
Nighttime(No Activity)


(b)
Indoor/Outdoor Concentration
0
1
2
3
4
5
6
7
8
9
10
BC
BrC
BC
BrC
BC
BrC
Breakfast
Chilli
Stir Fry

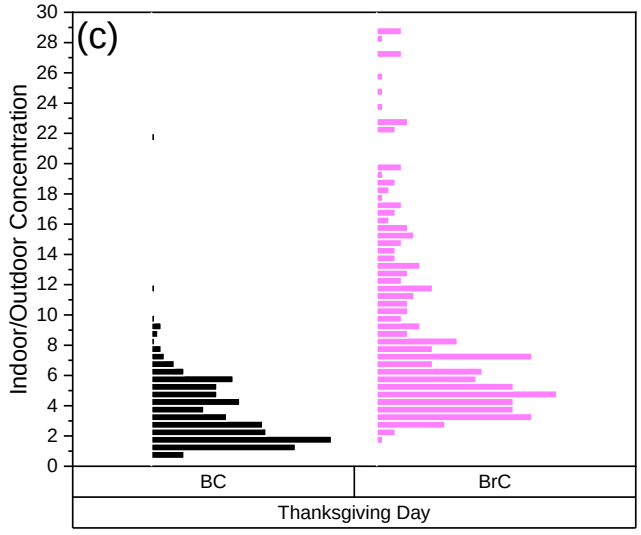

(c)
Indoor/Outdoor Concentration
0
2
4
6
8
10
12
14
16
18
20
22
24
26
28
30
BC
BrC
Thanksgiving Day

Figure 1. Indoor-to-outdoor BC (black) and BrC (magenta) ratios for (a) periods of no activity inside the house, (b) different meals cooked during HOMEChem, including *Layered Days* and *Sequential Stir-fry Days*, and (c) *Thanksgiving Day*. Note the significant differences in the y-axis scale among panels.

Figure 1(a) shows the distribution of indoor-outdoor ratios (I/O) for BC and BrC during day-time and night-time during periods of no activity with the test house closed and unoccupied. Since there were no indoor sources of BC and BrC, the concentrations measured indoors can be attributed to the penetration of organic and elemental carbon from outdoors. Indoor concentrations of BC and BrC were 0.42 ± 0.01 and 0.29 ± 0.01 compared to outdoors, respectively. These results indicate that BC and BrC particles likely exist in different size ranges, thus resulting in differences in their I/O ratios. Thornburg et al.[30] found that I/O ratios of PM were highest in aerodynamic diameters between 0.2 and 0.5 μm, with an I/O > 0.4 for accumulation mode (0.1 – 1 μm) and with curves sloping down outside this range. This indicates that BC might be present in the accumulation mode while BrC might fall outside it. Penetration factor for BC particles during periods of no activity was calculated to be 0.97. Since the penetration factor is slightly less than 1 this means the buiding envelope wasn't effective in filtering out outdoor BC from penetrating indoors.

The BC I/O values measured for the test house were comparable to the ratios reported by LaRosa et al.[9], in the range of 0.35 - 0.5, measured as a part of a two-year study focusing on BC exposure of household occupants. A study performed by Meng et al.[31] investigated the contribution of outdoor $PM_{2.5}$ to indoor concentration for three homes as a part of the RIOPA project and found out that the median contribution of ambient $PM_{2.5}$ to indoor concentration is 56%. Since organic carbon is co-emitted as a part of combustion-generated PM emissions, the infiltration behaviour of BC and BrC can be expected to approximate that of $PM_{2.5}$.

Figure 1(b) represents the I/O ratios observed during the preparation of different meals combined for all four *Layered Day*—which included breakfast, stir-fry lunch, and chili dinner—and *Sequential Stir-fry* experiments. For BC, breakfast and chili exhibited I/O ratios slightly larger than 1.0 (i.e., indoor concentrations slightly higher than outdoors), indicating a larger contribution from indoor sources than from outdoors. Breakfast presented the highest median I/O ratio (1.12 ± 0.2) for BC compared to chili (1.08 ± 0.07) and stir-fry (0.85 ± 0.03). According to the Wilcoxon rank-sum test, the BC I/O ratio distribution for the breakfast meal was significantly different from chili and stir-fry (p=0.8 and 0.5, respectively), while chili and stir-fry were statistically similar to each other (p = 0.0003).

In the case of breakfast, different meal elements such as sausages, tomatoes, toast, scrambled eggs, and coffee were prepared within a span of 30 min. Because each of these meal elements emitted different amounts of BC and BrC, the distribution corresponding to breakfast has a visibly higher variance relative to chili and stir-fry.

During the *Thanksgiving Day* experiment, the BrC I/O ratios (Fig. 1,c) ranged between < 1 and 28.63 indicating that the concentration indoors reached approximately 29× that of ambient levels. Concentrations indoors exceeded 10× that of outdoors during 30.4% of the time. We hypothesize that high temperature (>~200 °C) oven-roasting activities have led to an enhancement in BrC emissions compared to other activities during the HOMEChem experiment.

A combined analysis of all BC and BrC data shown here indicates that these aerosol fractions behaved differently from each other. A Wilcoxon rank-sum test resulted in p-values < 0.05 for every event. Table S2 presents all calculated p values.

3.2 BC and BrC concentrations during different cooking activities

Figure 2 depicts average concentrations of BC and BrC for different events during the campaign, including "no activity" periods, in which the test house was left closed and unoccupied, *Response Day* experiments, in which all doors and windows were repeatedly open for 30 min throughout the day—leading to indoor concentrations that can be assumed to approximate outdoor levels, and during four cooking-related activities: the preparation of breakfast, stir-fry, beef chili, and a simulated *Thanksgiving Day* event.

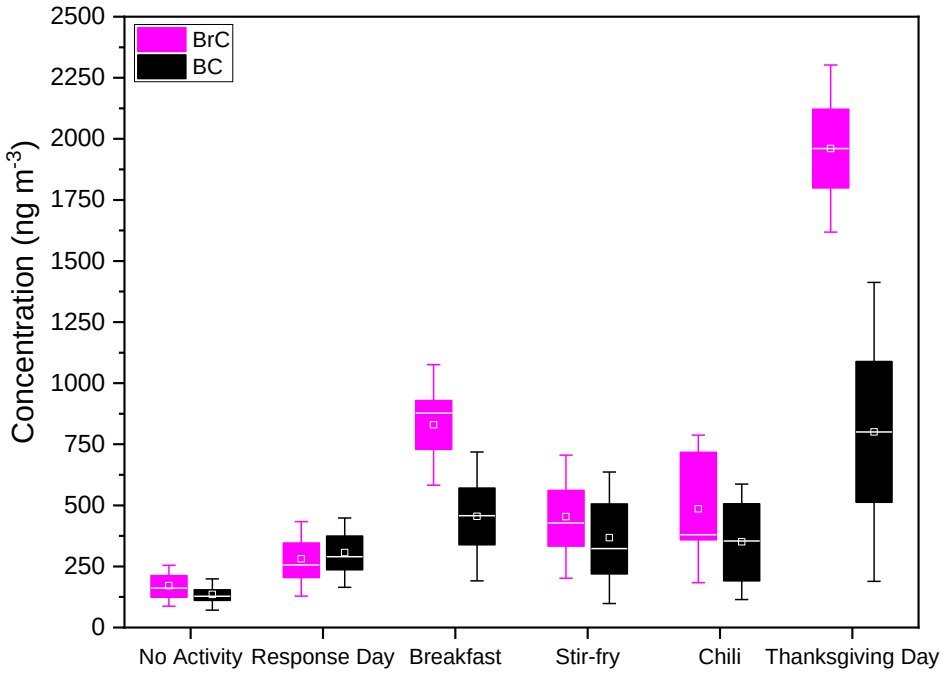


Figure 2. Distributions of black and brown carbon concentrations in the test house kitchen during different HOMEChem activities. Each box represents the 25th to 75th percentiles with whisker limits bounded by 1.5 times the standard deviation. The median value is represented by horizontal line and the □ represents the mean.

The average BC concentration in the test house during *Response Day* (306.4 ± 3.4 ng m^{-3}) was approximately twice that of no periods with activity in the closed house (135.26 ± 41.3 ng m^{-}

^{3}). Moreover the mean BC concentration was higher than that of BrC which is indicative of BC emissions from outdoors due to traffic related activity.

In the case of breakfast, the median concentration of BrC (829.2 ± 142.6 ng m^{-3}) was almost twice higher than that of BC (454.7 ± 152.1 ng m^{-3}), likely due to cooking of sausages and tomatoes. If a comparison is drawn in terms of BrC emissions during stir-fry and chili, the average concentration for chili (485.3 ± 350.7 ng m^{-3}) is slightly higher than that of stir-fry (453.7 ± 162.6 ng m^{-3}). This slight increase in mean can be attributed due to browning of beef during chili preparation (timeseries data not shown).

The median concentration for both BC and BrC was highest for *Thanksgiving Day* as compared to other events. *Thanksgiving Day* was an intensive cooking experiment, with BC and BrC concentrations reaching maximum values of 1189 ng m^{-3} and 7015 ng m^{-3}, respectively. The median BrC concentration (1960.5 ± 161.2 ng m^{-3}) was almost 10 times higher than the median BrC concentration during periods of no activity (171.1 ± 53.9 ng m^{-3}), indicating the strong potential of cooking-intensive events for affecting indoor air quality.

3.3 Aerosol light absorption properties during a *Thanksgiving Day*

Figure 3 shows a time series of light-absorbing aerosol particle concentrations throughout a *Thanksgiving Day* experiment. Similar data, collected for the other *Thanksgiving Day* experiment (June 27) can be seen in Figure S1 Total concentrations peaked in the range of 7000 ng m^{-3} which were higher than any other experimental day during HOMEChem.

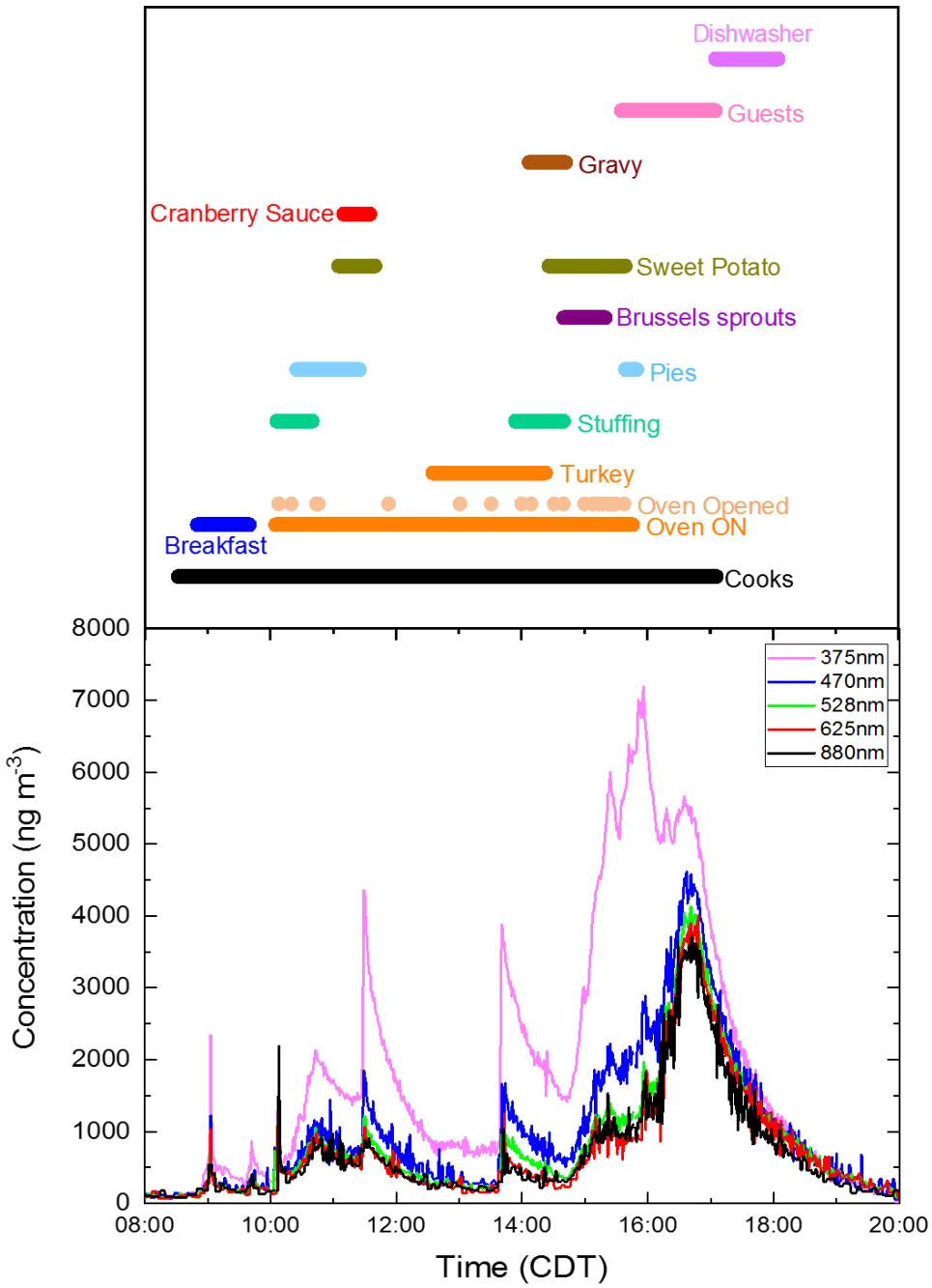


Figure 3. BC and BrC concentrations for *Thanksgiving Day* (June 18). The upper panel (a) shows the duration of each activity during the day. The lower panel (b) shows the real-time concentrations of aerosols determined through aethalometer measurements for five different light wavelengths: 375 nm ('UV channel'), 470 nm, 528 nm, 625 nm, and 880 nm ('BC channel').

It can be inferred from Figure 3 that multiple cooking activities lead to a clear peak separation in the UV channel (375 nm), indicative of BrC emissions. Only one discrete peak in BC (2197 ng m^{-3}) was observed when the oven was first turned on to ~200 ˚C. During the remainder of the day, emissions were dominated by BrC. Contributions from individual activities can be seen earlier in the day, but after ~Specifically, BrC emission peaks were observed while cooking tomatoes in a hot (~100 ºC) pan (2346 ng m^{-3}), toasting bread in an electric toaster (867 ng m^{-3}), toasting bread for stuffing in the oven (2135 ng m^{-3}), removing two pies from the oven (4362 ng m^{-3} earlier and 7016 ng m^{-3} later in the day), briefly and accidentally burning an oven mitten (3884 ng m^{-3}), roasting brussels sprouts (6005 ng m^{-3}), and roasting stuffing (6390 ng m^{-3}). These results indicate that oven-cooking emissions contained BrC particles which led to stronger absorption in UV range leading to distinct peaks as compared to higher wavelength channels.

3.4 Characterisation of emissions during different days using the angstrom exponent (α)

The power-law fitting between the wavelength of light (λ) and the particle absorption coefficient (σ_{abs}) for different events is shown in Figure 4. The α values ranged from 1.1 to 3.67 during the entire campaign, with the lowest value (indicative of BC) observed in periods of no activity and the highest value (indicative of BrC) observed during the *Thanksgiving Day* experiments. In general, cooking activities led to an increase in value of α, which can be attributed to BrC emissions.

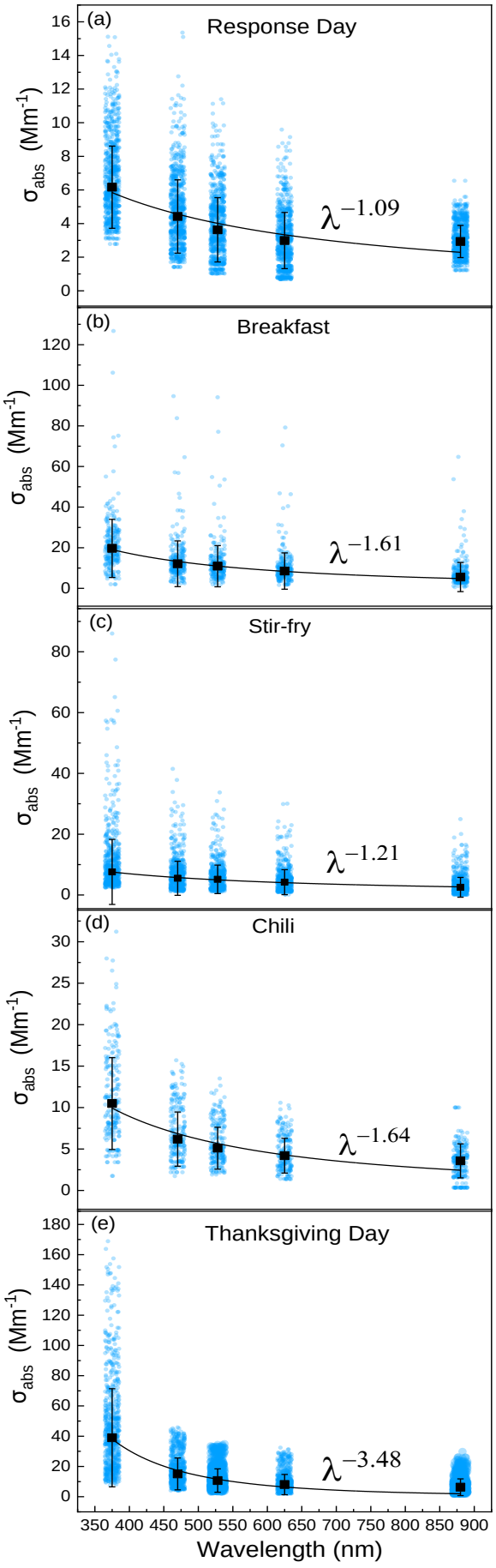

(a)
Response Day
$\lambda^{-1.09}$
(b)
Breakfast
$\lambda^{-1.61}$
(c)
Stir-fry
$\lambda^{-1.21}$
(d)
Chili
$\lambda^{-1.64}$
(e)
Thanksgiving Day
$\lambda^{-3.48}$
σ_{abs} (Mm^{-1})
Wavelength (nm)

Figure 4. Average values of σ_{abs} at five wavelengths (λ) fitted using the power law relationship for (a) *Response Day* (b) All *Breakfast* meals combined (b) All Stir-fry meals combined (c) All Chili meals combined (d) The *Thanksgiving Day* of June 27, showing the coefficient of determination for each dataset (R^2) and the angstrom exponent (α) associated with λ.

During the *Response Day* experiment (Fig. 4a) there were no activities taking place inside the house. The value of α (1.09) can be attributed to BC sources outdoors. Our data is in agreement with results reported by Sandradewi et al. [32] in which median α values were between 1 and 1.1 for summer months in an Alpine valley in Switzerland. During winter months, their reported α values were higher than 2, which was attributed to increased BrC emissions from wood burning activities for residential heating.

Another study performed by Ran et al.[34] studied temporal variability of α for two years in China. For non-heating seasons the value of α was lower than winters due to increased coal burning emissions. In the summer seasons the lower value of α was attributed to the freshly emitted BC from fossil fuel combustion.

During stir-fry activities (Fig. 4c), the value fitted for α was 1.21. Cooking emissions contain both BC and BrC particles which would lead to $\alpha > 1$.When α values for stir-fry are compared with breakfast (1.61) and chili (1.63), higher values associated with breakfast and chili emissions might be attributed to BrC emissions from meat cooking.

Similarly, in the case of *Thanksgiving Day* (Fig 4e), the α value (3.48) is much higher than 2, which indicates that BrC absorption was dominant as discussed in the previous section. BrC peaks were observed in association with multiple oven-cooking activities, making oven-related emissions a potential source for indoor BrC.

3.5 Black carbon exposure and dose during various activities

In exposure assessment studies that use fixed-station monitoring, there are many challenges in modelling the exposure of occupants. Multiple factors need to be taken into account, including modelling pollutant drift into surroundings, temporal and spatial variability, etc.[35]. In the case of the HOMEChem study, because the kitchen microenvironment was being monitored in close proximity to the stove and oven (~0.6 m), an assumption can be made that measurements were Figure 5 represents the average exposure and dose for different meals during the HOMEChem campaign: breakfast, lunch (stir-fry), and dinner (beef chili) compared with a *no activity* period of 30 min. This *no activity* period represents a hypothetical scenario in which an occupant would be present in the closed house during a period of no activities, thus representing a "best-case scenario" for BC and BrC inhalation exposure during the HOMEChem experiment. In reality, no occupants were present in the house when these measurements were taken. It must be mentioned here that this "best-case scenario" could have been further improved if the test house ventilation system had been outfitted with a filter to remove PM from outdoor sources or with the use of portable air filtration in the kitchen. BC doses were calculated by multiplying the real time concentrations with an inhalation rate of 11 l min^{-1} and taking the sum over entire duration for which each activity took place.

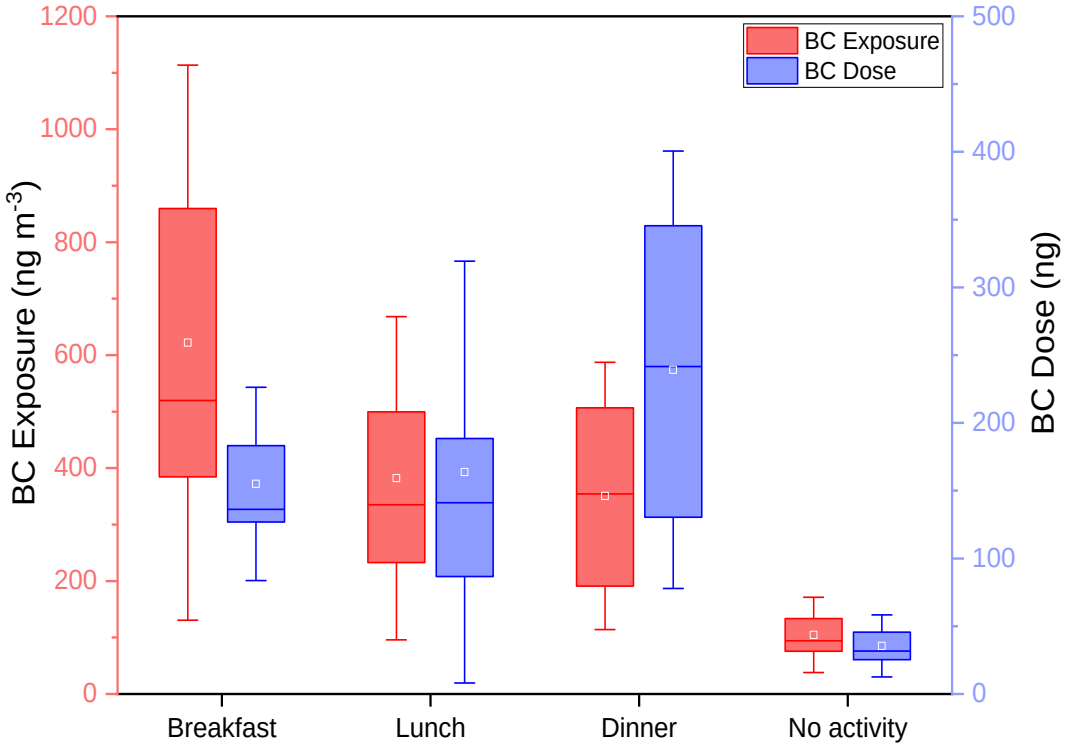


Figure 5. Exposure and Dose for different meals during HOMEChem campaign (breakfast, stir-fry lunch, beef chili dinner, and no activity period). Each box represents the 25th to 75th percentiles with whisker limits bounded by 1.5 times the standard deviation. The median value is represented by the horizontal line and the □ represents the mean.

According to Figure 5, the BC exposure was approximately 4.3 times higher during the preparation of any meal than during a comparable period of no activity in the test house; BC dose was approximately 5.2 times higher. BC exposure levels were the highest during the breakfast event (616 ± 42 ng m^{-3}). Although the exposure level is the highest during breakfast, the dose (153.1 ± 23.8 ng) was comparatively lower than the other meals because the average duration of breakfast (21 min) was shorter than stir-fry (30 min) and chili (65 min) and also because BC emissions occurred in short-term, but intense peaks of emissions (e.g., during toast preparation). Both lunch (stir-fry, 382.1 ± 46.1 ng m^{-3}) and dinner (chili, 350.7 ± 89 ng m^{-3})

behaved similarly. However, the dose associated with chili was higher because this cooking event lasted about twice longer.

The cumulative dose of a full day spent inside the test house (8:25 am – 6 pm) can be assessed by observing real-time BC concentrations during a *Layered Day* experiment (which included breakfast, lunch, and dinner preparation). House occupancy during that day amounted to 21% of BC dose while cooking activities amounted to 79% of BC dose. The average exposure during meal times (421.5 ng m^{-3}) was 2.5 times higher than that of house occupancy period (158.4 ng m^{-3}).

Dons et al.[22] studied the time activity patterns for 62 individuals and calculated BC exposure and dose accordingly. Sleep and home-based activities contributed 51.7% of daily exposure levels whereas the contribution of these two activities to the daily BC dose was 35.7%. Outdoor transport contributed to 29.8% of daily BC dose. Figure 6 depicts a comparison of BC dose and exposure for a Thanksgiving Day compared with values that would be obtained if the house were occupied in a day of no activities during HOMEChem.

Figure 6 depicts a comparison of BC exposure and dose for a Thanksgiving Day compared with values that would be obtained if the house were closed and unoccupied in a day with no activities during HOMEChem (a *best-possible-case* scenario for the test house in these experimental conditions).

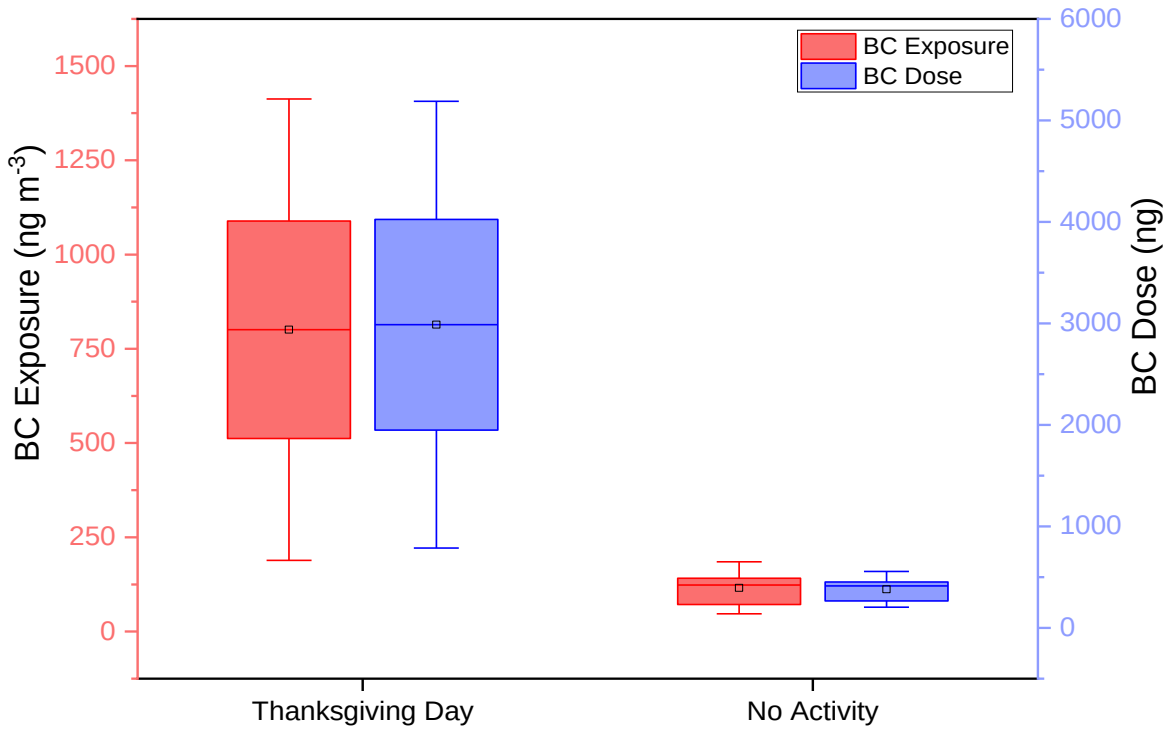


Figure 6. A comparison between BC exposure and dose for Thanksgiving Day combined as compared to a period of no activity for the same time interval. Each box represents the 25th to 75th percentiles with whisker limits bounded by 1.5 times the standard deviation. The median value is represented by the horizontal line and the □ represents the mean.

The dose of BC was calculated for the period in which the oven was on during a *Thanksgiving Day* experiment, which amounted to 342 min. Figure 5 shows that the average dose received during Thanksgiving Day (2988 ± 734 ng) would be approximately 25 times higher than a time period in the house with no cooking activities taking place (115.9 ± 18.3 ng).

4 Conclusion

Real-time data collected in this study improves our understanding of the generation of BC and BrC indoors and the effects of outdoor air pollution on indoor air quality. An analysis of continuous BC and BrC relative concentrations showed that there were significant differences between the outdoor penetration of these aerosol fractions, indicating that they may exist in different size ranges. The average concentrations of BC and BrC during some cooking events were significantly higher than outdoor concentrations demonstrating the contribution of cooking to the deterioration of indoor air quality. The power law fitting approach performed in this study to calculate α can aid in future studies to link aerosol optical properties to chemical composition. The values calculated for exposure and dose of BC during the preparation of different meals, while representing a rough estimate — because we lack size-segregated BC and BrC particle data— contributes to a growing body of literature in exposure studies in indoor microenvironments and provides numbers that can be used in in vitro and in vivo inhalation studies for BC toxicity assessment.

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Appendix

Supplementary Figures and Tables

Table S1 Table showing mean and median values for BC and BrC during different events

	Mean		Median ± SE	
Event	*BC*	*BrC*	*BC*	*BrC*
Day-time (No Activity)	0.50	0.35	0.42 ± 0.01	0.29±0.01
Night-time (No Activity)	0.59	0.39	0.53±0.01	0.35±0.01
Breakfast	1.80	1.48	1.12±0.20	0.70±0.18
Chilli	1.30	0.94	1.08±0.07	0.93±0.03
Stir Fry	0.95	0.80	0.85±0.03	0.73±0.03
Thanksgiving Day	3.42	8.72	2.95±0.12	6.83±0.32

Table S*2* Table showing p-values between BC and BrC distribution during different events

Event	*p-value*
Day-time (No Activity)	1.7758e-72
Night-time (No Activity)	0
Breakfast	0.0390
Chilli	9.6974e-07
Stir Fry	4.5237e-05
Thanksgiving Day	2.7327e-54

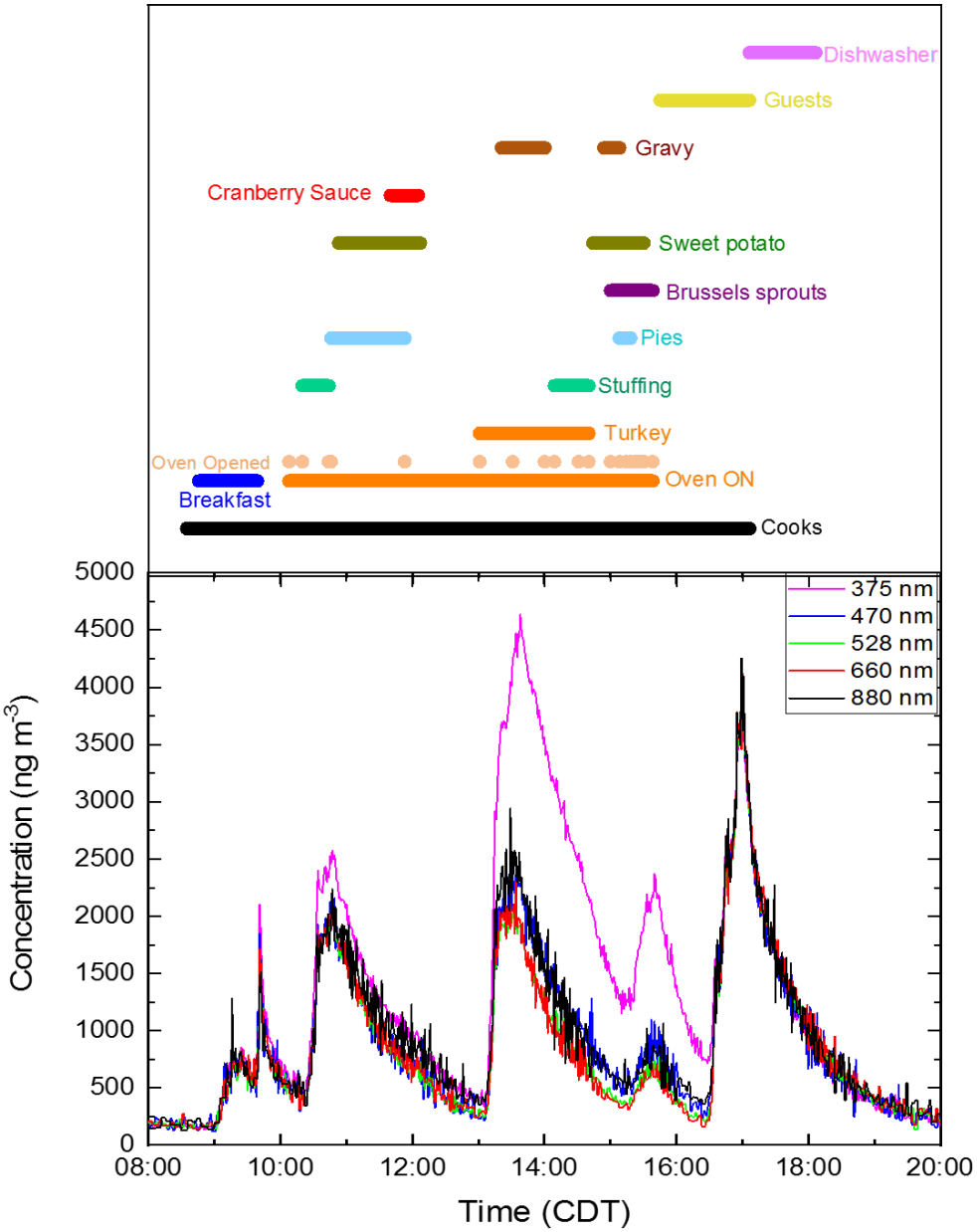

Figure S1. BC and BrC concentrations for *Thanksgiving Day* (June 27). The upper panel (a) shows the duration of each activity during the day. The lower panel (b) shows the real-time concentrations of aerosols determined through aethalometer measurements for five different light wavelengths: 375 nm (UV channel), 470 nm, 528 nm, 625 nm, and 880 nm.