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Black carbon cookstove emissions: A field assessment of 19 stove/fuel combinations

Charity Garland [a, *], Samantha Delapena [a], Rajendra Prasad [b], Christian L'Orange [c], Donee Alexander [d], Michael Johnson [a]

[a] Berkeley Air Monitoring Group, 1900 Addison St. Suite 350, Berkeley, CA, 94704, USA
[b] IIT Delhi, Hauz Khas, New Delhi, Delhi, 110016, India
[c] Colorado State University, 430 N College Ave, Fort Collins, CO, 80524, USA
[d] Global Alliance for Clean Cookstsoves, 1750 Pennsylvania Avenue Northwest, Washington, DC, 20006, USA

HIGHLIGHTS

- Results of optical black carbon (BC) measurements of 19 cookstoves is presented.
- An attenuation cross-section was determined for BC analysis using transmissometry.
- BC emission factors and BC/PM is presented for five stove classes.
- Potential relative climate impacts were estimated using CO_2-equivalents.

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ABSTRACT

Black carbon (BC) emissions from household cookstoves consuming solid fuel produce approximately 25 percent of total anthropogenic BC emissions. The short atmospheric lifetime of BC means that reducing BC emissions would result in a faster climate response than mitigating CO_2 and other long-lived greenhouse gases. This study presents the results of optical BC measurements of two new cookstove emissions field assessments and 17 archived cookstove datasets. BC was determined from attenuation of 880 nm light, which is strongly absorbed by BC, and linearly related between 1 and 125 attenuation units. A relationship was experimentally determined correlating BC mass deposition on quartz filters determined via thermal optical analysis (TOA) and on PTFE and quartz filters using transmissometry, yielding an attenuation cross-section (σ_{ATN}) for both filter media types. σ_{ATN} relates TOA measurements to optical measurements on PTFE and quartz ($\sigma_{ATN(PTFE)} = 13.7\ cm^{-2}\ \mu g$, $R^2 = 0.87$, $\sigma_{ATN(Quartz)} = 15.6\ cm^{-2}\ \mu g$, $R^2 = 0.87$). These filter-specific σ_{ATN}, optical measurements of archived filters were used to determine BC emission factors and the fraction of particulate matter (PM) in the form of black carbon (BC/PM). The 19 stoves measured fell into five stove classes; simple wood, rocket, advanced biomass, simple charcoal, and advanced charcoal. Advanced biomass stoves include forced- and natural-draft gasifiers which use wood or biomass pellets as fuel. Of these classes, the simple wood and rocket stoves demonstrated the highest median BC emission factors, ranging from 0.051 to 0.14 g MJ^{-1}. The lowest BC emission factors were seen in charcoal stoves, which corresponds to the generally low PM emission factors observed during charcoal combustion, ranging from 0.0084 to 0.014 g MJ^{-1}. The advanced biomass stoves generally showed an improvement in BC emissions factors compared to simple wood and rocket stoves, ranging from 0.0031 to 0.071 g MJ^{-1}. BC/PM ratios were highest for the advanced and rocket stoves. Potential relative climate impacts were estimated by converting aerosol emissions to CO_2-equivalent, and suggest that some advanced stove/fuel combinations could provide substantial climate benefits.

* Corresponding author. Tel.: +1 831 818 2038.
E-mail address: cgarland@berkeleyair.com (C. Garland).

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1. Introduction

Approximately 41% of the world's households, or about 2.8 billion people globally, depend on solid fuels for meeting daily cooking needs (Bonjour et al., 2013). Use of these fuels for cooking and heating results in the emissions of climate forcing pollutants such as methane and black carbon (BC) (Jetter and Kariher, 2009; MacCarty et al., 2008; Preble et al., 2014). Introduction of cleaner-burning stoves and fuels has been proposed and pursued as a means to reduce household pollutant emissions that influence global and regional climate. BC emissions from cookstoves are of particular interest, as BC is estimated to be second only to CO_2 in its warming impact (Ramanathan and Carmichael, 2008) and solid fuel burning for cooking of heating in homes produces approximately 25 percent of total anthropogenic BC emissions (Bond et al., 2013). Since the atmospheric lifetime of BC is only a few days, reducing BC emissions can produce near-term climate change mitigation, whereas benefits due to reductions in CO_2 and other long-lived greenhouse gases accrue over decades to centuries (Bond and Sun, 2005).

Quantifying emissions of short-term climate pollutants is important for both climate modeling as well as understanding the implications of promoting different stove/fuel interventions. Ideally, this information can be used to incentivize household energy programs and cooking technology developers to produce and promote solutions which maximize benefits. Frameworks which incentivize household energy programs to provide reductions in long-term climate benefits by quantifying and trading carbon offsets have been in place for several years, and now relatively new methodologies have been developed which provide a similar mechanism trading short-term climate benefits (The Gold Standard Foundation, 2015). Importantly, quantifying these short-term benefits requires a careful assessment of their aerosol emissions. Particulate matter (PM) emissions include both BC and organic carbon (OC). While BC has a strong warming impact, OC has a cooling effect, as it tends to scatter light rather than absorb it (Bond et al., 2013). Thus, characterization of the BC and OC aerosol emissions is fundamental to understanding and quantifying the climate benefits posed by household energy interventions.

To date, while there have been a handful of field studies which have reported on real-world black and organic carbon emissions from cookstoves (Johnson et al., 2008; Johnson et al., 2011a; Roden et al., 2006), there is still relatively little information on how various classes of stove/fuel intervention technologies may impact these emissions. Specifically, very little data is available on newer and more advanced types of technologies, such as forced-draft and pellet stoves, which may have the greatest potential for reducing emissions from solid fuels. Additional comparisons with data from controlled laboratory testing are also needed to help us better characterize the differences which have been observed between laboratory and field performance (Johnson et al., 2010; Roden et al., 2009). Finally, emissions sampling is relatively intensive and costly compared to other types of stove performance testing in homes, and thus the data sets are comprised of stove fuel combinations (e.g. 1–5 fuel/stove combinations) and/or small sample sizes (e.g. 5–20 homes or events). Complimenting and augmenting these studies with larger data sets is needed to provide a more definitive characterization of aerosol emissions from household energy technologies.

To address these needs, here we present black and organic carbon emissions estimates from 19 stove/fuel combinations being used in Asia and Africa. All testing was conducted in homes during uncontrolled cooking events to provide real-world estimates of emissions performance.

2. Methods

2.1. Field campaigns

A combination of archived filters (N = 453) and newly collected filters (N = 44) were analyzed for this study, which is outlined in Table 1. Field campaigns occurred at seven locations in Asia and Africa. Brief descriptions of the stove and fuel types, sample sizes, and study locations for the field campaigns can be found in Table 1. Additional details and maps are provided in the supporting information.

2.2. Emissions sampling

All samples were collected following the same fundamental protocol. Emissions sampling was conducted during uncontrolled cooking events in participants' homes, for which the cook was instructed to prepare a meal as they normally would, without altering stove operation, cooking techniques, or fuel type. An example of the measurement scheme is shown in Fig. 1. The emissions species measured included carbon dioxide (CO_2), carbon monoxide (CO), particulate matter $\leq$ 2.5 μm in aerodynamic diameter ($PM_{2.5}$), and black carbon (BC). Additional measurements of elemental carbon (EC) and organic carbon (OC) were made in India and Cambodia.

Before and after each cooking event, all fuels were weighed separately. Ash and char were immediately removed from the stove, separated using an ash screen, and weighed. Scale type varied, but all scales were calibrated using NIST standard weights before use and were checked daily for drift throughout the field campaigns. Fuel moisture content was determined using either a two-pin resistance style moisture meter (Extech M0210) or a moisture analyzer balance (Precisa, Model - XM 60-HR). Fuel mass was converted to equivalent energy using energy densities determined via bomb calorimetry (C200, IKA Works Inc., USA) or default values provided by the Water Boling Test protocol version 4.2.4 (WBT Technical Committee, 2013).

Emissions were collected using the partial capture method and emissions factors were determined using the carbon balance approach. Details of partial capture and carbon balance methods have been described in previous publications (Johnson et al., 2011a; Roden et al., 2006; Smith et al., 2000), as well as the Water Boling Test protocol version 4.2.3 (WBT Technical Committee, 2013). Briefly, real-time concentrations of CO and CO_2 were measured using a TSI IAQ-CALC 7545 (TSI Inc., USA), and gravimetric measurements were taken to quantify $PM_{2.5}$ and BC. When Measuring EC and OC, two sample streams were drawn by constant-flow SKC sampling pumps (SKC Inc., USA), splitting after the sample exited a BGI Triplex cyclone (BGI, USA) at 0.75 L per minute (LPM) (1.5 LPM total through cyclone) from each line to remove particles larger than 2.5 μm in diameter. One sample line drew air through a PTFE filter to determine $PM_{2.5}$ mass deposition followed by a quartz filter (Advantec) to collect gas phase OC when measured. The other sample line passed air through only a quartz filter and collected both particle and gas phase OC and EC. Mass deposition was determined gravimetrically by weighing the Teflon filters before and after sampling in a constant humidity and temperature room on an electronic microbalance with 0.1 μg resolution (Mettler Toledo, USA).

Emissions factors were determined using the carbon balance approach, as has been done in previous studies of stove emissions and is described in the WBT 4.2.3 protocol (Johnson et al., 2011b; Roden et al., 2006; Smith et al., 2000; WBT Technical Committee, 2013). Flow rates and sample volumes were adjusted for temperature and pressure, which were recorded before and after each event.

Table 1
Archived filter data sets from Berkeley Air that were used for black carbon optical analysis.

Location	Stove Types/Descriptions	Fuel	Sample Size	Study Site
Cambodia	- Traditional metal bucket stove covered in baked clay	Wood	22	Peri-urban neighborhood of Phnom Penh, Cambodia.
	- Forced-draft, electrical grid or solar powered battery charging, with variable fan speed.	Wood	22	
Southern Vietnam (from archive)	- Traditional ceramic stove with upper and lower fuel shelves	Wood	19	Village of Châu Lăng, a rural community near the border of Cambodia in the An Giang district on the Mekong Delta.
	- "High efficiency" metal-clad stove with a ceramic liner	Wood, Charcoal	11, 4	
Northern Vietnam (from archive)	- Traditional metal frame support for pots, with cooking fires lit underneath	Wood	16	Phú Binh district in a rural, agricultural region of the Thai Nguyen province
	- Rice husk gasifier constructed of metal with air forced into the stove via a separate blower fan	Rice Husks	7	
Uganda (from archive)	- Three stone fire	Wood	16	Peri-urban community outside of Kampala on Wakiso road
	- Traditional charcoal varied in construction, primarily ceramic with fuel grate and 3 pot rests	Charcoal	5	
	- Forced-draft TEG[a] made with stainless steel combustion chamber	Wood	11	
Kenya (from archive)	- Kenyan ceramic jiko with metal-clad ceramic liner, three pot supports, and metal legs	Charcoal	22	Urban community of Kwangware in Nairobi
	- Stainless steel combustion chamber and an adjustable ash tray to regulate primary airflow	Charcoal	32	
India: Uttar Pradesh	- Traditional stationary u-shaped mud chulha	Wood	44	Uttar Pradesh: Khaga Block of District Fatehpur
	- Two-pot-mud stationary stove	Wood	40	
	- Metal rocket stove 1, primary air through fuel opening	Wood	44	
West Bengal (from archive)	- Metal forced-draft TEG with refractory combustion chamber	Wood	46	West Bengal: Bagnan Block of District Howrah
	- Metal rocket stove 2, primary air through intake holes and fuel opening	Wood	45	
	- Metal top-lit Updraft (TLUD)	Biomass	48	
	- Forced-draft pellet stove	Pellets	43	

[a] TEG: thermoelectric generator, which converts heat to electricity to power a fan or other devices.

More details of the fuel analysis, instrumentation, and sampling approach can be found in the supporting information.

2.3. Black and elemental carbon analysis

BC mass deposition on 497 filters used to collect $PM_{2.5}$ was determined via its black-body optical properties with an SootScan Model OT21 Optical Transmissometer (Magee Scientific, USA). The transmissometer measures light attenuation by PM deposited on filter media at two wavelengths: 880 nm attenuation provides a quantitative measurement of BC, and 370 nm attenuation gives a qualitative indication of certain organic compounds. For the purposes of this study, only the 880 nm channel was used. Attenuation (ATN) is defined as:

$$ATN = -100 \ln(I/I_o)$$

Where I and I_o represent the light transmitted through the sample filter and a blank, reference filter, respectively. The attenuated 880 nm light is directly proportional to the BC deposited on the filter, in units of BC mass per filter area ($\mu g/cm^2$). Linearity is limited, however, due to light saturation at high BC loading. The linear range suggested by the manufacturer is from 0 to 125 ATN units. Using a light attenuation technique for BC estimates allows archived samples to be analyzed since BC is generally non-volatile and does not degrade, and thus quantification can occur years after samples are collected (Chow et al., 2010; Dutkiewicz et al., 2014; Husain et al., 2008).

Conversion from attenuation to BC mass was done using a BC attenuation cross-section (σ_{ATN}). The σ_{ATN} was determined by comparing the attenuation with elemental carbon (EC) from two sites where filters compatible with EC analysis were used. In India, 310 glass fiber filters (Whatman, USA) were used to collect $PM_{2.5}$ mass, a subset (N = 29) of which was analyzed to determine EC. In Cambodia, PTFE filters (Pall Scientific, USA) were used for $PM_{2.5}$ mass determination and pre-fired quartz (SKC Inc, USA) for EC were collected simultaneously for 44 samples. The glass fiber and quartz filters were analyzed at Colorado State University (Fort Collins, USA) for EC and OC via thermal-optical analysis (TOA). The TOA analysis method is based upon NIOSH 5040 (Birch and Cary, 1996). NIOSH Method 5040 calls for the use of a quartz filter as opposed to glass fiber; however, glass fiber filter media is typically more mechanically robust then quartz filters. The robustness of glass fibers has certain advantages when collecting samples that requires

Fig. 1. Emissions sampling setup in Phnom Penh, Cambodia.

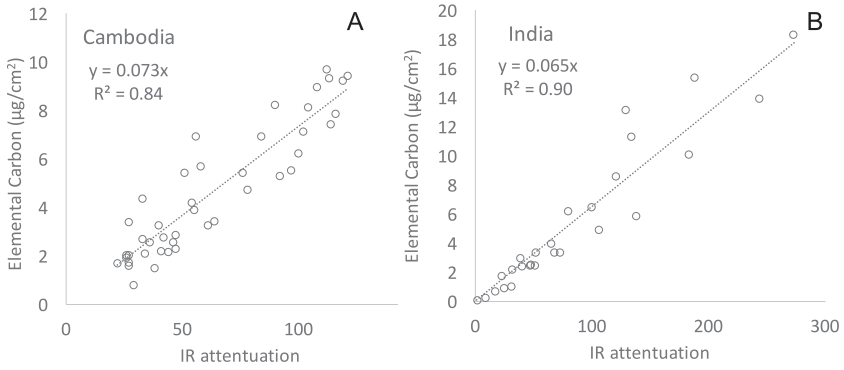

Fig. 2. Correlation of EC from TOA and absorbance of 880 nm IR light during transmissometry of samples collected during wood-burning emissions samples in Cambodia (N = 43) (A), and in India (N = 29) (B).

extensive handling and shipping (such as remote field samples). Quartz filters have the advantage of typically resulting in lower sampling artifacts. However, these artifacts can be addressed through proper filter blank collection. Glass fiber filter media has successfully been applied to quantifying carbon aerosols using thermal analysis techniques in previous studies (Cachier et al., 1989; Huntzicker et al., 1982; Lin and Friedlander, 1988). Thermal-optical measurements were conducted using a Sunset Laboratory OC-EC Aerosol Analyzer. The OC-EC Aerosol Analyzer thermally desorbs material off filter media which is then subsequently analyzed using a flame ionization detector. By first desorbing in an inert helium atmosphere and then an oxidizing atmosphere, the split between OC and EC can be determined.

Following a common approach applied in several studies (Chow et al., 2010; Dutkiewicz et al., 2014; Gundel et al., 1984; Japar et al., 1986; Liousse et al., 1993; Petzold and Niessner, 1995), a simple linear relationship was used to relate light attenuation to TOA derived EC in terms of µg BC per cm^2 of filter area (Fig. 2). EC mass was assumed to be approximately the same as BC mass, which is a common assumption for source characterization studies (T. C. Bond et al., 2004). All Cambodian samples used to determine σ_{ATN} demonstrated absorbance values below 125 ATN units, which falls within the manufacturer's recommended linear range. Filters from India contained some samples outside of this range, however, those with both EC and BC measurements show linearity up to 275 ATN units (Fig. 2). Samples greater than 275 ATN units occurred only in the Indian data set, for which only 1 percent (N = 3) of the attenuation measurements were between 275 and 300 ATN units. This small subset was not excluded, as exclusion could bias Indian stove BC estimates lower, although it should be noted that the mean estimates may be slightly underestimated due to nearing instrument response saturation.

Cambodian and Indian data sets both had good linear agreement between EC mass deposition and attenuation (see Fig. 2). The σ_{ATN} was determined by taking the inverse of the slope of the line relating EC and attenuation (15.4 cm^{-2} µg for the Indian dataset and 13.7 cm^{-2} µg for the Cambodian dataset). One outlying data point from the Cambodian data set was excluded (not shown in Fig. 2A), which would have shifted the value for σ_{ATN} by 1.0 cm^{-2} µg (or 6.8%). Sampling form notes and information did not indicate any problems or irregularities for the sample filters for this data point; however, it is possible it was either compromised or a transcription error occurred. Further outlier analysis can be found in the supporting information.

The σ_{ATN} were applied to transmittance measurements of filter samples using the following equation to determine the BC mass loading (S_{BC}) on a filter in units of BC/cm^2:

$$S_{BC} = ATN/\sigma_{ATN}$$

The σ_{ATN} from the India data set was applied to all data from the India study, where glass fiber filters were used. The σ_{ATN} from the Cambodia dataset was applied to all other sites where the PTFE filters were used.[1]

Filter transmittance, along with gravimetric $PM_{2.5}$, event sample times, fuel mass consumed, sample flow, and fuel energy content, allow determination of BC emission factors (g/kg, g/MJ), mass percentage BC of PM (BC/PM), and emission rates (g/min). Additional details for estimating organic carbon emissions are also presented in the supporting information.

2.4. Organic carbon estimation

OC was only directly measured for this study on quartz filters from Cambodia, using the NIOSH thermal optical method (Birch and Cary, 1996). The gas phase OC filter artifact was corrected by subtracting the OC on a quartz filter placed inline after the Teflon filter. For the remaining samples for which only Teflon filters were available, OC was estimated by looking at the relationship between OC and the non-BC fraction of particulate matter. Fig. S8 (see supporting information) shows this relationship for a variety of biomass stoves from previous studies which measured elemental and organic carbon using the thermal optical technique with the resulting linear relationship (OC = 0.567*[PM-BC]-0.119) ($r^2 = 0.885$). This relationship was then used to estimate the OC on the Teflon filters based on the difference between its PM and BC mass estimates.

2.5. Conversion to CO_2-equivalent

The net warming or cooling impact of a given stove/fuel combination's aerosol was estimated using 20-year global warming potentials (GWPs), which normalize the impact of a warming or cooling species to the equivalent impact of CO_2 over 20 years (IPCC, 2013). The specific calculation is as follows:

[1] The σ_{ATN} used for wood samples was also applied to charcoal samples. While the intention was to calculate a charcoal-specific σ_{ATN} from previous emissions studies where both PTFE and quartz filters were collected during charcoal emissions tests, PM and BC mass depositions were too low from sampling charcoal stoves to determine the relationship between ATN and BC.

$$CO_2e = EF_{BC}{}^{*}FC{}^{*}GWP_{BC} + EF_{OC}{}^{*}FC{}^{*}GWP_{OC},$$

where CO_2e is the CO_2-equivalent emitted per person per meal, EF_{BC} and EF_{OC} are the black and organic carbon emission factors in g/kg, FC is the fuel consumption per person-meal, and GWP_{BC} and GWP_{OC} are the global warming potentials for BC (2421) and OC (−244) published by the International Panel on Climate Change (IPCC, 2013). Similar approaches using GWPs to estimate net climate impact of stove emissions have been applied in previous studies (Edwards et al., 2003; Johnson et al., 2009; MacCarty et al., 2008) and are used in methodologies for quantifying and trading climate benefits (The Gold Standard Foundation, 2015; UNFCCC, 2012).

3. Results

BC emission factors for all 19 stove types and average BC emission factors based on stove class are shown in Fig. 3-a and Fig. 3-b, respectively. The boxes designate the stove class and correspond to the y-axis labels in Fig. 3-b. The simple wood stoves and the rocket stoves had the highest BC emissions factors. Within the simple wood stoves, there is substantial variability. Indian traditional chulhas, two-pot-mud stoves, and Cambodian traditional stoves had the highest median BC emission factors at 0.091, 0.12, and 0.13 g/MJ, respectively. Rocket stoves show similar BC emission factors as simple wood stoves, both with median values around 0.09 g/MJ. Advanced stoves, which include one natural-draft TLUD stove with the remainder being forced-draft stoves, on average, had lower median BC emission factors when compared to average rocket and simple wood stove performance, ranging from 0.0055 to 0.074 g/MJ, with the lowest and highest advanced stove BC emission factors belonging to the rice husk gasifier and the forced-draft wood stoves, respectively.

The lowest BC emission factors were from charcoal stoves, with medians ranging from 0.008 to 0.015 g/MJ. Low PM production from charcoal stoves is partially due to the combustion process associated with carbonizing the fuel, which drives off large amounts of PM-forming material. Also, charcoal typically combusts through surface oxidation (a process in which PM formation is much less likely) as opposed to diffusion flames where PM forms in fuel rich zones. Of importance when considering overall climate impacts from solid fuel use, is the contribution of PM to total emissions made during the carbonization process of wood and agricultural residue to produce charcoal. Although to our

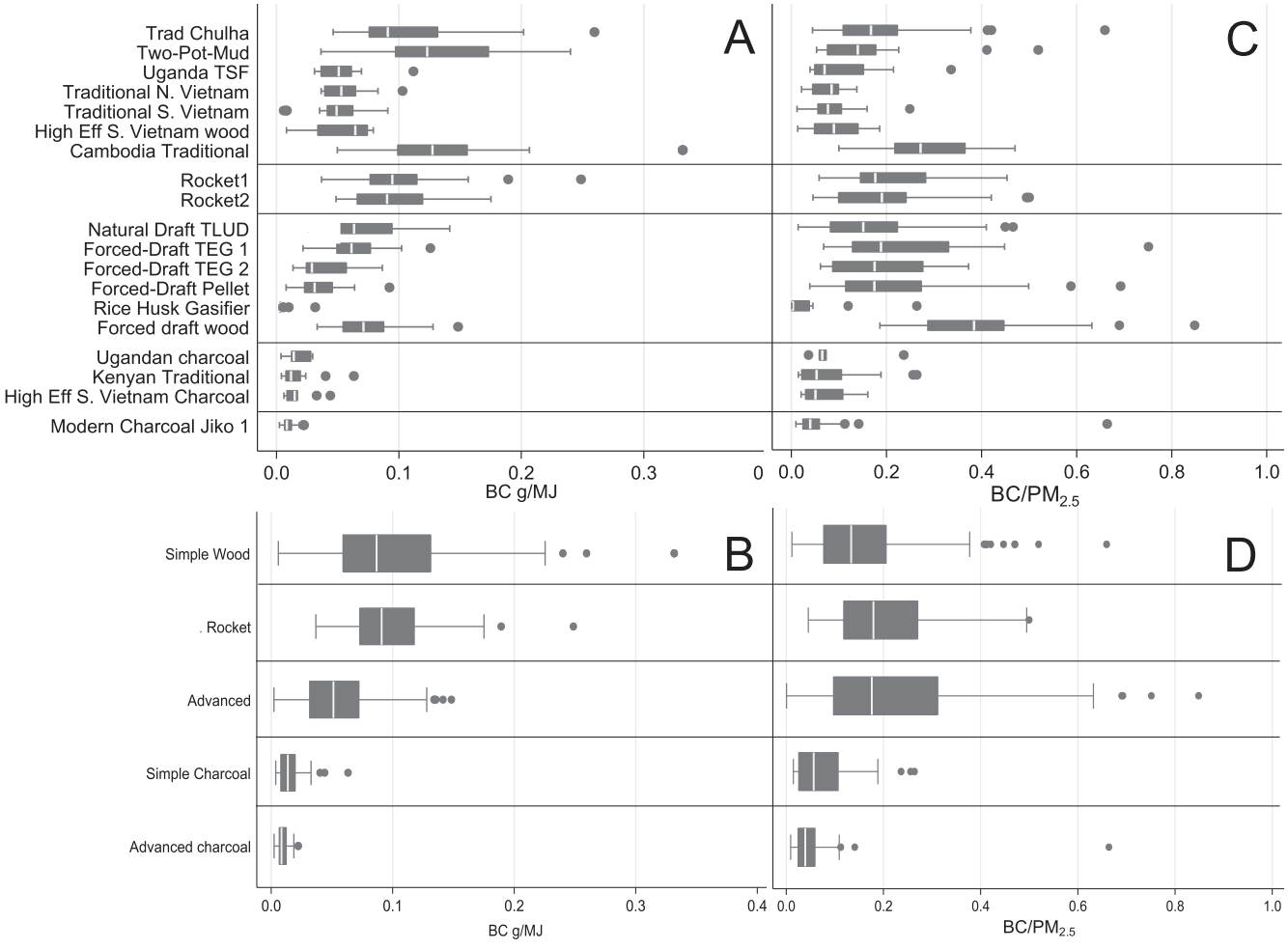

Fig. 3. (a) BC emission factors for all 19 stoves measured by optical transmission in terms of mass of BC emitted per unit of fuel energy consumed (g/MJ). Boxes represent groupings of stove types. (b) Stove class-specific averages of BC emission factors in terms of g BC/MJ fuel. Corresponding boxes from 3-a represent the stove types for individual stoves. (c) Fraction of PM emitted as BC for all 19 stoves measured by optical transmission in terms of BC/PM. Boxes represent groupings based on stove class. (d) Stove class-specific fraction of PM emitted as BC. Corresponding boxes from 3-a represent the stove types for individual stoves. Gray boxes denote the interquartile range (IQR), with medians shown in the white center line, and whisker ends encompass the most extreme values within Q3+1.5(Q3-Q1) and Q1-1.5*(Q3-Q1). All points beyond the whiskers are shown as gray dots.

knowledge there are no reported emission factors for BC and OC during charcoal production, it has been shown that PM emission factors are approximately 2.6 g/kg wood (Bond et al., 2004), which would be equivalent to approximately 0.14 g/MJ wood, if using the standard wood energy density of 19 MJ/kg (Bailis, 2007). Although literature is sparse, Cachier et al. report the fraction of PM assumed to be black carbon during charcoal production as 8% (Cachier et al., 1996). Using this coarse approximation, BC emission factors for charcoal production would be around 0.21 g/kg wood, or 0.011 g/MJ. The combination of approximate upstream and end-user BC emissions yields a BC emission factor of about 0.019–0.026 g/MJ for charcoal use, which is still quite low when compared to simple wood stoves.

The fraction of $PM_{2.5}$ emitted as BC is shown in Fig. 3-c (each stove) and Fig. 3-d (stove/fuel class). The BC/PM ratio is a good indicator of an aerosol's climate forcing, as BC absorbs solar radiation and the majority of the remaining PM fraction is made up of organic matter, which tends to reflect solar radiation (an analysis of approximate organic matter and organic carbon emissions is provided in the supporting information). The reflection of radiation back to space by organic matter has a cooling effect, which offsets the BC warming impact. Assuming the non-BC portion of PM is organic, for the same amount of total PM emitted, a smaller BC/PM ratio would result in less warming and vice-versa (Bond et al., 2013).

Of the wood burning stoves, the simple wood stoves generally had lower median BC/PM ratios, with the lowest being the Ugandan three stone fire (TSF) at 0.065 and the highest being the traditional Cambodian stove at 0.27. Both rocket stoves had BC/PM ratios around 0.19. The advanced stoves had the greatest variability with both the lowest and the highest median BC/PM stoves of the entire stove set belonging to this group. The rice husk gasifier had the lowest BC/PM of all 19 stoves at 0.003 and the forced-draft wood had the highest at 0.38. Charcoal stove BC/PM ratios were all low, with medians between 0.067 and 0.077, likely due to the smoldering combustion during start up, which generally emits lighter colored, organic PM. The higher PM/BC ratio seen in advanced stoves may be due to higher combustion temperatures and burning off more of the organic material in vapor form than a simple wood stove (Rau, 1989).

Summary statistics of BC emissions performance are provided in the supporting information, as well as additional estimates of OC emissions.

4. Discussion

4.1. Implications for climate finance

This study provided a relatively broad set of BC and OC emissions performance estimates for traditional and new stove technologies. The emission factors (see Table 2 in supporting information) can be used for climate models as well as to help inform approaches aiming to quantify and the short-term climate benefits associated with stove or fuel interventions. A common approach for comparing relative climate impacts is to convert the emission species to CO_2e, as shown in Fig. 4. While calculating CO_2e is a useful tool for guiding climate-finance toward projects more likely to have a positive impact, it is also important to note that more comprehensive climate modeling would be needed to better characterize the full warming or cooling implications of a given stove/fuel combination.

Fig. 4 illustrates how CO_2e can be used to compare the net short-term climate implications from aerosol emissions for the stove/fuel combinations measured at the India field sites. The only stove that

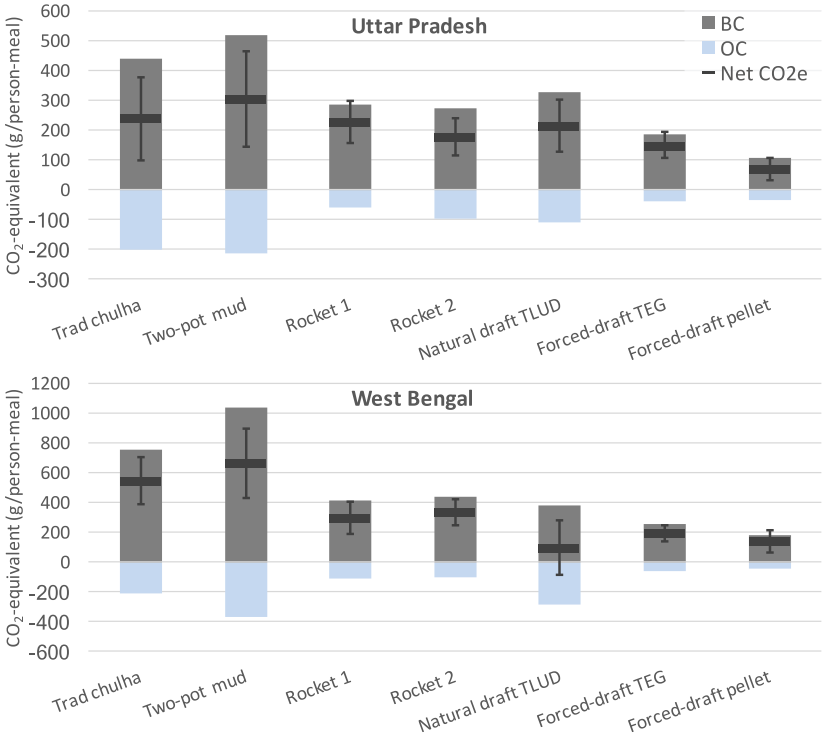

Fig. 4. CO_2-equivalent emissions from BC and OC associated with the stoves measured in India, presented on a per person-meal basis. The CO_2e estimates here do not include other co-emitted species, such as carbon monoxide and sulfates. Error bars represent ±95% confidence intervals of the net CO_2e.

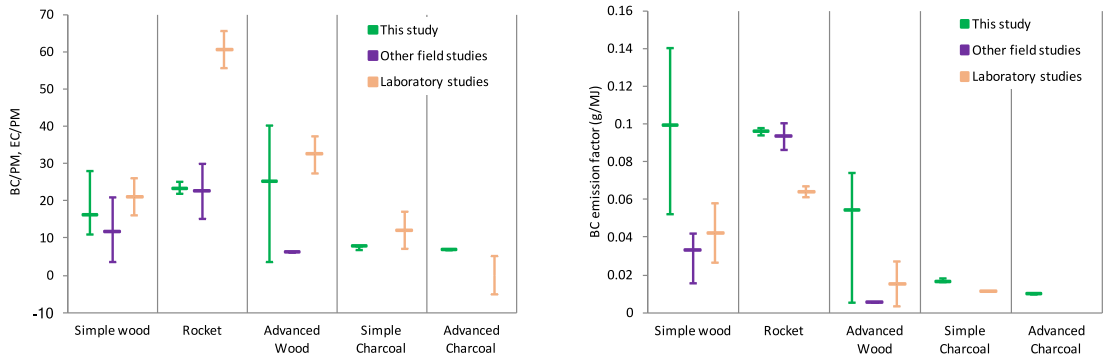

Fig. 5. Comparison of laboratory- and field-based BC emission factors (left panel) and BC/PM (right panel), including measurements taken for this study. Stove types measured for the respective studies were classified under the five main stove classes. The middle line is the mean of the individual measurements (this study) or the mean of the mean results from lab or field studies of the stoves tested within the respective classes. The error bars represent the range of the individual measurements (this study) or the range of mean results observed in the lab and field studies used for comparison.

appeared to have higher CO_2e aerosol emissions than the traditional chulha was the two-pot mud stove, although the rocket stove and natural-draft TLUD stoves in Uttar Pradesh had similar CO_2e emissions. The forced-draft-stoves generally had the lowest CO_2e in both study locations, although the natural-draft TLUD in West Bengal also had comparably low CO_2e. The difference in CO_2e emissions for the TLUD highlights the need to assess a technology for a given location, as there are clear performance differences for this stove between study sights. Finally, the TLUD aerosol emissions also illustrate how BC and OC contribute to CO_2e. While the TLUD had relatively low CO_2e in West Bengal, this was primarily due to higher OC emissions offsetting the impact of the BC. Although positive from a climate perspective, this implies that it's total PM emissions were still high, which has critical health implications. Ideally, both CO_2e and $PM_{2.5}$ are mitigated by a given intervention, such as the case for the forced-draft pellet stove, but it should not be assumed that reductions in one imply reductions in the other.

4.2. Comparison with previous laboratory and field studies

Measuring cookstove performance in the field is generally more resource intensive, technically complicated, and logistically demanding compared to similar measurements in the laboratory, and therefore is not as common in the literature as lab-based studies (Berkeley Air, 2012). A small set of field-based measurements of BC do exist, however, with comparisons shown in Fig. 5 and Table S1 (see supporting information). Most of these studies report EC rather than BC, which we will consider equivalent for the purpose of this comparison. Elemental and black carbon are typically differentiated based upon the analysis technique used for quantification; thermal volatilization vs light absorption, respectively (Andreae and Gelencsér, 2006; Bond and Bergstrom, 2006). Although the differences between BC and EC have implications in certain scientific analyses (such as atmospheric chemistry), BC and EC are highly correlated. For the purposes of this study grouping EC and BC introduces minimal error. A study of emissions from Honduran traditional stoves by Roden et al. (2006) reports EC emission factors and EC/PM ratios determined from 9 tests of a simple mud wood stove and 14 tests of a rocket stove. EC emission factors were reported in g/kg, which are slightly different than what is reported here, but assuming an energy density of 19 MJ/kg for wood (WBT Technical Committee, 2013), the median emission factors and EC/PM ratios for both the simple wood stove and rocket stoves were similar to the measurements made in this study, with differences in median values ranging from about 13% (rocket stove emission factors) to 65% (rocket stove median BC/PM).

Uncontrolled cooking test emission factors for traditional and intervention stoves (specifically a rocket stove, a wood-burning chimney stove, and a fan gasifier with pellets, measured in Uganda, Nepal, and India, respectively) were measured by Johnson et al. (2011a). Mean EC emission factors for traditional stoves and the Nepal wood-burning chimney stove, all simple wood stoves, were between 0.032 g/MJ and 0.042 g/MJ, assuming an energy density of 19 MJ/kg for wood. These are relatively low BC emission factors compared to the simple wood stoves reported in this study, with a median value of 0.087 g/MJ. The rocket stove measured by Johnson et al. (2011a) in Uganda had a mean BC emission factor of 0.10 g/MJ, which is very similar to the rocket stove performance reported here. The advanced stove measured in that work, a forced-draft pellet stove, had a mean BC emission factor of 0.0053 g/MJ, which is on the low end of the range measured for advanced stoves in this study. Measures of mean BC/PM reported by Johnson et al. (2011a) for simple wood, rocket, and advanced stoves were 0.11, 0.15, and 0.06, respectively. Results from this study show higher mean BC/PM for all stove classes, with simple wood, rocket, and advanced stove mean BC/PM of 0.16, 0.23, and 0.25, respectively. This is not unexpected, however, as Johnson et al. 2011a's measurements are of a single stove type with BC/PM values falling within the observed range of the respective stove classes measured during this study, all which include measurements of multiple stove types.

Johnson et al. (2008) presented lab- and field-based emission factors for traditional open fires in Mexico, although the sample sizes were small (N = 6 and N = 8, respectively) and may not be fully representative of traditional stove performance in Mexico. Lab and field EC emission factors were reported to be 0.057 and 0.016 g/MJ, respectively, and the EC/PM ratios were 0.22 and 0.034, respectively. Both of the lab-derived metrics more closely match the median measurements of simple wood stoves reported in this study, with the field-based measurements of EC emission factors and EC/PM both falling within the lower 3rd percentile of the simple wood stoves measured in this study. Although on the lower end of the performance range, the Mexican traditional open fires are still within the observed simple wood stove performance range for this study.

Fig. 5 provides comparisons of BC emission factors and EC/PM ratios of the different stove classes with the corresponding laboratory-based measurements, including three stone fires, rocket stoves, a natural-draft gasifier, forced-draft gasifiers, and a charcoal stove (Habib et al., 2008; Johnson et al., 2008; Just et al., 2013; MacCarty et al., 2008). Lab-based measurements of emission factors by MacCarty et al. (2008) and Just et al. (2013) are similar;

however, both sets of lab-based EC emission factors are lower than those measured in this study, when grouping by stove class. The ratio of EC/PM measured by MacCarty et al. and Just et al. (2013) is higher for all stove classes than those measured in this study, other than Just et al.'s (2013) EC/PM value for the simple wood stove, which is about 50% lower than the average measured for the same class reported here. Habib et al. (2008) reports an average EC emission factor and ratio from lab measurements of an Indian simple wood burning cookstove of 0.026 g/MJ and 0.16, respectively. While the emission factor is lower than what was measured here, the emission ratio is very similar to what was observed in this study.

Overall, the lower emission factors and higher BC/PM ratios found in the laboratory should not be surprising given the differences in stove operation between controlled laboratory testing and normal daily use in homes. Perhaps most relevant to understanding the reasons for these differences is that smoldering, which is characterized by more organic carbon emissions (Rau, 1989), is minimized during controlled testing by the careful and diligent manner in which fuel is fed and tended. Chen et al. (2012) and Roden et al. (2009), both showed that climate forcing emissions from cookstoves are more variable in the field, and that non-steady-state transition events, which again, are minimized in the laboratory, contribute substantially to aerosol emissions (Chen et al., 2012; Roden et al., 2009). This trend is shown in the inverse relationship between the $PM_{2.5}$ emission factors and the BC fraction of particulate matter (see Fig. 6). The higher $PM_{2.5}$ emission factors are characterized by relatively low BC content, suggesting substantial amounts of smoldering occurred and the aerosols were largely made of organic matter. The set of laboratory based data shows that the PM emission factors are generally lower, and correspondingly, the BC/PM ratios are higher than what is observed in the field.

5. Limitations

The use of optical measurements of light absorbing carbon have been shown repeatedly to accurately approximate BC mass (Ahmed et al., 2009; Chow et al., 2010; Dutkiewicz et al., 2014; Gundel et al., 1984; Husain et al., 2008; Wang et al., 2011). Limitations are still present, however, and should be considered when interpreting these results.

Emissions samples are taken from directly over the stove, however, ambient air is sampled along with the direct stove emissions. Background ambient CO, CO_2, and $PM_{2.5}$ are subtracted from the emissions samples based on pre- and post-sample concentrations measured for 10 min each in real time. Since background black and organic carbon samples were not taken, an ambient subtraction was not possible. The contribution of black and organic carbon from ambient air is generally negligible compared to the concentrations present in the emissions plume, however, and unlikely to influence the measurement. Previous measurements have shown that background PM concentrations are much lower than those measured for emissions samples, with typical ambient background PM concentrations in the study regions approximately 75 ± 50 μg/m^3, which is common for other village level ambient PM concentrations (Zuk, 2007). Plume PM concentrations are on the order of 5000–10,000 μg/m^3 for the emissions measurements taken for this study, meaning ambient PM contributes <1% to the PM measurement. In certain sampling environments, such as enclosed kitchens with low ventilation, emissions may be re-sampled and this assumption could overestimate emission factors.

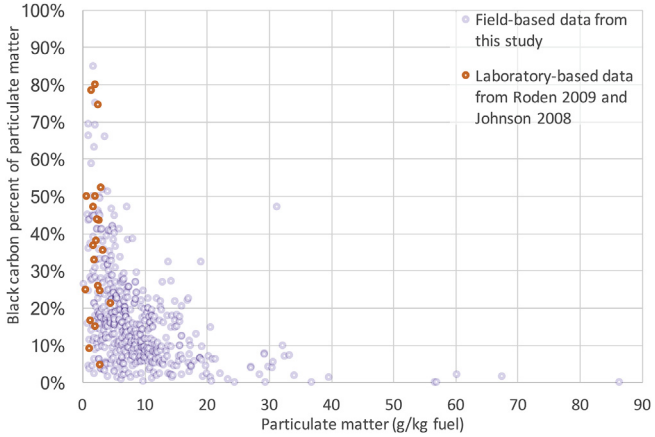

Fig. 6. Scatterplot showing the relationship between the magnitude of the particulate emission factors and their black carbon composition.

Optical properties of PM are variable and dependent on the source, age, and mix of the aerosol (Liousse et al., 1993). BC itself is difficult to define and there is not a universally accepted scientific definition. BC is the strongly light absorbing component of particulate matter formed during combustion, which is usually an impure form of EC (Andreae and Gelencsér, 2006; Buseck et al., 2012), and may contain some light-absorbing organic matter called "brown carbon". Although "brown carbon" absorbs more strongly at wavelengths smaller than the 880 nm light used by transmissometry, its presence may influence the optical measurement of BC and move the EC:BC measurement relationship away from 1:1 (Rehman et al., 2011). Although the scientific community is yet to come to agreement regarding how to define EC, BC, and brown carbon (Andreae and Gelencsér, 2006; Long et al., 2013); BC is often believed to be comprised of EC and light absorbing organic matter (i.e. "brown carbon"). Regardless, EC can be, and frequently is, used as a proxy for BC (Ahmed et al., 2009; Chow et al., 2010; Dutkiewicz et al., 2014; Gundel et al., 1984; Husain et al., 2008; Wang et al., 2011). As brown carbon fractions increase, the EC:BC ratio will decrease. When using optical methods, high brown carbon concentrations would result in an over estimation of EC.

The differences in optical properties of PM, especially the presence of brown carbon, emitted from cookstoves may be influenced by fuel type, combustion temperature, the presence or absence of oxygen, and flaming versus smoldering combustion. The attenuation coefficient used for determining BC for all stoves in this study is based on the BC light attenuation relationship from Indian and Cambodian stove emission samples. In reality, this relationship may vary slightly depending on the source of the smoke and how much brown carbon is present in the PM. To date, few studies have done extensive characterization of the brown carbon fractions found in the emissions of biomass cookstoves. Further research is needed to adequately characterize the potential contributions of brown carbon to total biomass cookstove emitted pollution.

With the exception of our data from Cambodia measured with the thermal-optical technique, OC was estimated indirectly from the difference between PM and BC. While the relationship we observed showed good correlation ($r^2 = 0.885$) and potentially provides a fairly simple and inexpensive means to estimate OC, this was a new approach and should be further investigated to see how stable this relationship is with other emission sources. Ideally, more comprehensive compositional analysis of PM could provide an indication of ratios of OM to OC as well as other non-organic contributions.

All emissions data reported here are based on measurement during events which occurred in a single season (generally the country's dry season) for logistical reasons. There may be significant seasonal effects on BC emissions and BC/PM from cookstoves.

To our knowledge, there has not yet been published data describing the seasonal influence on cookstove emissions, however, it has been shown that indoor air pollution in Bangladesh is strongly influenced by season, with winter concentrations significantly higher than summer (Gurley et al., 2013). This trend was also seen in indoor air pollution and personal exposure $PM_{2.5}$ in India (Andresen et al., 2005). Although the authors do not provide a clear explanation for these seasonal influences, it highlights the importance of characterizing stoves in all seasons to fully understand their impact on climate and health. Certainly, seasonal changes in fuel types or conditions would be expected to have an effect on climate forcing emissions. Repeated BC measurements over different seasons would allow stove performance to be more accurately described.

Although this study represents a large set of data from Asia and Africa, it is certainly not comprehensive with respect to technology nor geography. We observed differences in emissions characteristics for the same stove between states in India, indicating that even relatively small changes in location can be meaningful, and thus care should be taken in applying emission factors from this study to stoves used in other locations. At a high level, there is still little field emissions data available from Africa especially outside of East Africa. South America, to our knowledge, also has little or no published data on field emissions. These geographies, along with any project-specific sites where cooking and heating technologies are being planned for scale, should be targeted for further field emissions studies.

In order to fully understand climate impacts, additional information is required. Co-emitted species such as methane and total non-methane hydrocarbons, which also influence climate, would need to be included for an overall picture of a cooking technology's climate influence. In addition, the impact of cookstove emissions on climate is geographically specific and depends on factors such as weather patterns; transport of particulates to sensitive areas, such as glaciers; geographic location; and potential for emissions to contribute to secondary organic aerosols (Hansen and Nazarenko, 2004; Menon, 2002; Ramanathan and Carmichael, 2008).

6. Conclusion

Field-based black carbon emissions estimates from cookstoves have been limited (Johnson et al., 2011a, 2008; Roden et al., 2006). This study helps address this gap by providing real-world BC emissions and can be used to inform models, stove design, and build emissions inventories. The limited studies generally suggest that more advanced stove designs can provide meaningful climate benefits. More research, however, is needed to determine how fuel type, seasonal differences, and geography affect black carbon emissions. Integrating black carbon emissions estimates within programmatic activities would also aid in understanding the full range of benefits that stoves provide users and the environment. Recognizing that emissions measurements are generally technically difficult and relatively expensive, creative and cost-effective ways for programs to fund or undertake these studies should be pursued. Collaborations with research institutes, pooling resources to make measurements across various programs, or building regional capacity to make emissions measurements are potential means for making field emissions studies more feasible.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2017.08.040.

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