Dear Dr. Virtanen:

We have addressed the referee comments below, and made appropriate changes to the manuscript. We sincerely look forward to a favorable decision on this revised submission.

Referee comments are in black font color, our responses are in **bold red**, and the revisions made to the manuscript and Supplemental Data are highlighted in *italicized red*.

Thank you.

Sincerely,

Rajan K. Chakrabarty, Apoorva Pandey, and co-authors.

Anonymous Referee #1

This manuscript presents emission factors for three types of solid fuels from India based on real-world tests and associated chemical speciation analysis. The results are novel, and provide relevant information on EFs related with indoor combustion in the Indian (and the broader South Asian) context. The manuscript is written well, and provides succinct information on the sampling methods and previous work on the topic. The observations are described well, but the results are not discussed in detail.

Comment: Please include a brief description of the sampling location in the methods section- dimensions of the room where sampling was conducted, ventilation type (or lack thereof) in the sampling area, and representativeness of the sampling location.

Response: A schematic of the room has been included in the Supplemental Data and a brief description was added to the main text:

Revision at Line114: *The test kitchen (Fig. S1) was on the first floor of house, separate from all other rooms. A permanently open door was the only entry from an open terrace. A partially covered window covered was a second ventilation source.*

Comment: Lines 115-118: Please include limits of detection for carbon analysis using IMPROVE_A protocol, and average particle mass based on gravimetric analysis.

Response: The following details were added to the main text:

Revision at Line 132: The mass of Teflon filter deposits ranged 50 – 300 µg.

Revision at Line 134: *The minimum detection limits of the TOR analysis are about 9 µg for OC and 1 µg for EC (Solomon et al., 2014).*

Comment: Each emission test included data on PM (size distribution, real-time PM data with Sidepak, sensors), and gases (CO, CO2) in addition to filter samples, but none of the data is discussed in the manuscript. Please consider including the profile of real-time emissions for a sample test for each fuel. Also, include average particle size, or representative particle size distributions for the different regimes (ignition, flaming and smoldering).

Response: A sample plot of real-time gas concentration and Sidepak particle mass concentrations is now shown in the Supplemental Data.

Comment: With respect to the differences between EFs for Bihar and UP dung, is it known if there are differences in composition of dung cakes?

Response: Bihar dung had a higher carbon content than U.P. dung (Table 1), any other differences in composition are not known.

Comment: Figure 5: It is interesting to note that the carbon profile for the present study is very different from the results in the CPCB (2011) report. It would be beneficial if the authors could comment on the reason for the differences (methods, sample type or something else?). Also, what could be causing OC3 fraction to contribute nearly 50% of the total OC mass? Please elaborate.

Response: The Central Pollution Control Board (CPCB) of India conducted source apportionment studies in six Indian cities (CPCB, 2011). As part of this study, a source profile was developed for $PM_{2.5}$ emissions from wood *chulhas*, through laboratory combustion tests. Further details of the burn cycle or sampling system used in this study are not known, but the source profile obtained has been uploaded to their website: http://www.cpcb.nic.in/Source_Apportionment_Studies.php. Although we cannot comment on the reason for the differences, we presented the findings because, to our knowledge, this is the only available profile of Indian cookstove emissions and has been used in the apportionment studies mentioned above.

Revision at Line 188: *We also compare our results with a source profile developed for $PM_{2.5}$ emissions from wood chulhas, as part of source apportionment studies conducted by the Central Pollution Control Board (CPCB) of India (CPCB (2011)). This profile was based on laboratory burns, but details of the test fuel and burn protocol are not known.*

Comment: Based on the observations from this analysis, what are the implications for the real world? Are we able to derive substantial conclusions that could be relevant for mitigation policies, or exposure reduction measures?

Response: The conclusions section has been expanded with more analysis of the effect of fuel type and combustion phase on emission rates. Further discussion of the implications of our findings has also been added. We cannot draw any conclusions as to exposure reduction or mitigation.

Revision at Line 206: *Earlier studies have shown that fuel burn rate (Habib et al., 2008) and fire-tending practices (Roden et al., 2009) affect cookstove emissions, therefore standardized burn protocols (typically a water boiling test) cannot replicate cookstove performance in the field. In a controlled test, variability in wood size could be expected to affect emissions from fuel-wood combustion, hence, laboratory studies typically used identical wood chunks. The four wood fuel types in this study were used as chunks in a range of sizes, and yielded similar EFs even though these samples were gathered from widely separated parts of India. Overall, the variability in emissions between multiple measurements for the same fuel was larger than or comparable to the differences in average emission rates for different fuels.*

Revision at Line 213: *A short period immediately following the ignition phase showed the highest and most variable $PM_{2.5}$ EFs for all fuels, consistent with previous observations of high emissions from lighting and refueling of stoves (Roden et al., 2009). The use of variable amounts of kerosene and other kindling materials, as required in any burn, may explain the large spread in emission measurements during ignition. The highest OC-to-EC ratios were observed during smoldering (no visible flame present), when pyrolysis is the dominant process.*

Revision at Line 217: *The OC3 thermal fraction contributed nearly 50% of the total carbon mass for emissions from all fuels, indicating that most of the PM emissions are non-evaporative. This could have*

implications for the climatic impact of cookstove emissions, especially since volatility of OC emissions is inversely related to light absorption efficiency in the near-UV and visible wavelengths (Andreae and Gelencsér, 2006). The thermal fractions found in this study differ from those in other biomass burning tests, including the source profile used an input to receptor models of particulate emissions in India (CPCB, 2011).

Revision at Line 225: *The EFs from this study could contribute toward improving evaluations of climate and health impacts of carbonaceous aerosols over India. Further investigation is needed on the relationship between the composition of OC emissions and their effects on climate and health.*

Comment: Lines 154-157: Does this mean that one household could use different fuel types depending on the type of cooking activity being undertaken?

Response: Yes, it is anecdotally known that a single household often uses a mix of fuels (Venkataraman et al., 2010). The household in this study also had an LPG stove but they continued to use freely available biomass (dung and wood) to reduce their cooking costs. However, available large-scale survey data (Census, 2011) only presents the distribution of primary cooking fuels across households.

Comment: Figure S1: Using a plot of PM concentrations from the optical sensors will be much more useful compared to the raw signal data currently included in the supplemental data. Was a secondary check performed to compare average concentration of Sharp sensor for each cooking event with gravimetric filter measurement or Sidepak measurements?

Response: A separate objective of this study was to validate the use of wireless sensors in the field (Patel et al., 2017). Reference measurements by the Sidepak were well-correlated with data from a collocated sensor. However, the Sidepak measurement being a measure of particle light scattering does not reflect the actual particle concentration, unless the instrument is independently calibrated for the given type of particles. Therefore, in this manuscript, we have not converted the sensor signals to equivalent Sidepak concentrations.

Minor comments:

Figure 2: Please change the resolution. The image gets blurred if the reader zooms in (same problem in case of Figure 4).

Revise the manuscript - words and/or punctuation marks are missing at some places (e.g., Lines 77, 79 and 138). In the supplement, the sentence "MCE is typically treated as an identifier of . . .. . . " seems to be incomplete.

Please edit. Line 89: The thermal fractions "found in" rather than "found it"

Response: These comments have been addressed.

Anonymous Referee #2

In the manuscript "Aerosol emission factors from traditional biomass cookstoves in India: Insights from field measurements" the real-world emission factors of PM components from different biomass fuels in food cooking are studied in an Indian household. The findings increase the understanding of real-world PM, OC, and EC emission factors for a wide range of combustion of solid fuels from different regions.

The results are compared with previous findings in standardized tests, and in general, the results of the manuscript show higher PM emissions and OC/EC ratios existing in the literature. In a big picture, the main findings are novel and significant enough for an ACP paper but the quality of presentation needs improvement (major revision). On the positive side, I like the title, abstract and conclusions parts of the manuscript.

Specific comments

Comment: Table 2 lists the experiments performed, indicating the number of reps and the foods that were cooked. The variation of the foods cooked was a bit random; apparently, the family was just living ordinary life. In the analysis, the food is not considered as a factor instead it is just causing deviation to different EFs for each fuel. Authors should analyze the role of the food cooked while discussing the results. Is the food itself emitting something, or is it only so that different foods require different amount of heat or time?

Response: See and Balasubramanian (2008) investigated emissions during various cooking operations on a gas stove stove. On average, they found particle mass concentrations of about 23 $\mu g/m^3$ from boiling and about 65 $\mu g/m^3$ from deep frying, showing that certain cooking processes could emit a significant number of particles. Particle emissions from gas stoves are low enough that the effect of the cooking process itself can be observed. Solid biomass cookstoves are known to emit over 5-10 times more particles than gas stoves under identical lab settings (Smith et al., 2000). In this study, we estimated particle mass concentrations of 500 $\mu g/m^3$-20 mg/m^3, from gravimetric measurements. Therefore, the contribution from food emissions is expected to be negligible.

Comment: Authors have used several instruments in the tests: PAMS, Sidepak analyzer, gas analyzer, minivol sampler and wireless optical particle sensors. First of all, locations of wireless optical sensors are not shown in Figure 1. Also it is not clear why minivol sampler could not use the "eight armed sampling probe" and adjacent tubing to be in parallel with other instruments. Figure 1 would be more informative if the whole room or house would be also sketched to have an idea how the exhaust flows in the space. I assume there is not a dedicated stack for exhaust gas ventilation.

Response: The Minivol samplers being ambient $PM_{2.5}$ sensors could not be connected to the tubing from the sampling probe. Given the constraints of the small kitchen, we tried to ensure that all instruments excepting the Minivol samplers be sampled from the same location (that is via the eight armed probe).

The position of the wireless sensors is now shown in Figure 1. A schematic of the room has been included in the Supplemental Data, and described briefly in the manuscript:

Revision at Line 114: *The test kitchen (Fig. S1) was on the first floor of house, separate from all other rooms. A permanently open door was the only entry from an open terrace. A partially covered window covered was a second ventilation source.*

Comment: The authors have corrected values from different sampling locations using wireless sensors and correction factor of about 1.04 (data shown in Figure S1). About half of the time both sensors were showing maximum concentrations, so that time they were not showing any meaningful results. Was the maximum concentrations also included in the regression analysis? I see that at many times the difference between sensors was much more than 4%. Also you should include all the experiments with wireless sensors to this regression analysis. I think getting this factor right is very crucial thing because gas analyzers measured from different location than the minivol sampler.

Response: We used the optical wireless sensors for a total of 6 runs. Following the reviewer's comments, the regression analysis was re-evaluated: we observed that data points close to the saturation limit of the sensors were strongly influencing the regression results. The following changes were made

Revision in Supplemental Data: *The saturation voltage for the sensors is close to 750 mV, discarding all values higher than 750 mV, regression analysis of the remaining points yields a slope of 0.96. However, if the saturation threshold was set at 745 mV, the slope changed to 0.89. This is probably because saturation behavior for these sensors is a soft-limit saturation, such that the input-response relationship becomes non-linear at some voltage lower than the final limiting value of 750 mV. If measurements from this non-linear region are included, the linear regression analysis would give erroneous results. Therefore, we systematically reduced the threshold values until we observed negligible change in the regression slope. Finally, we discarded the data points where either of the sensors had readings above the linearity threshold (720 mV). About 60% of all data points were used, and a slope of 0.63 (R^2=0.65) was obtained. The scatterplot of data from the two sensors is shown in the Supplemental Data.*

Revision at Line 145: *Wireless optical particle sensors (details available in Patel et al. (2017)) were attached to the Minivol sampler and the sampling probe during six experiments to check for any significant differences in the particle concentrations measured at the two locations. Measurements where either sensor was saturated were discarded, and a linear regression analysis performed on the valid data points. A correction factor of 1.6 was applied based on regression slope (Supplemental Data).*

Comment: It would be also interesting to see how gas concentrations change during combustion/non-combustion phases to see how much is the increase in CO and CO2 concentrations in the measurement locations. Authors could include a plot of this in the supplementary information.

Response: A sample plot of real time gas concentration and Sidepak particle mass concentrations is also now shown in the Supplemental Data.

Comment: The manuscript in current form presents only data from filter collections (Minivol sampler) while the authors also list other instruments that were used. For instance, authors should show time series of some individual test how the PM emission behaves during different phases of the combustion. Overall the role of combustion phase is highlighted in the results. Authors should also show and analyze particle size distributions measured with the PAMS instruments. This would help to e.g. identify whether the PM mass is in the particle size range of PAMS or above it.

Response: We were unable to draw any conclusions from the PAMS data because the instrument was typically saturated within the first ten minutes of a burn.

Comment: The results are focusing on the effects of combustion phase and fuel quality of the EFs. The discussion and conclusions of the results are so far in form where authors just focus on the measured values. The cause and effect analysis is much lacking in the current version of the manuscript. For instance, why different phases of combustion show different results? Why are the deviations in results such high? The authors should dedicate more effort in this.

Response: The conclusions section has been expanded with more analysis of the effects of fuel type and combustion phase on emission rates. Further discussion on the implications of our findings has also been added.

Revision at Line 206: *Earlier studies have shown that fuel burn rate (Habib et al., 2008) and fire-tending practices (Roden et al., 2009) affect cookstove emissions, therefore standardized burn protocols (typically a water boiling test) cannot replicate cookstove performance in the field. In a controlled test, variability*

in wood size could be expected to affect emissions from fuel-wood combustion, hence, laboratory studies typically used identical wood chunks. The four wood fuel types in this study were used as chunks in a range of sizes, and yielded similar EFs even though these samples were gathered from widely separated parts of India. Overall, the variability in emissions between multiple measurements for the same fuel was larger than or comparable to the differences in average emission rates for different fuels.

Revision at Line 213: *A short period immediately following the ignition phase showed the highest and most variable $PM_{2.5}$ EFs for all fuels, consistent with previous observations of high emissions from lighting and refueling of stoves (Roden et al., 2009). The use of variable amounts of kerosene and other kindling materials, as required in any burn, may explain the large spread in emission measurements during ignition. The highest OC-to-EC ratios were observed during smoldering (no visible flame present), when pyrolysis is the dominant process.*

Minor remarks

Figure quality. You should remove all borderlines from the figures. Font quality is bad in Figures 2 and 4.

Response: Higher quality figures are now embedded in the manuscript.

Row 79. Full stop apparently missing.

Response: Corrected.

Rows 131-133. You basically don't have to use value of 2.4% here since you have measured OC/EC yourself, or you can also compare the amount of non-CO or nonCO2 carbon based on your results to this value.

Response: We have measured OC/EC but we cannot estimate non-(CO and CO_2) carbon because we do not know the mass of fuel burned as a function of time. We cannot balance the fuel carbon with carbonaceous emissions.

Row 135. "Figure 2 compares EFs for the different fuels." Are these EF's from the "whole burning event" including all the phases from combustion.

Response: Yes, this figure includes all data points for each fuel.

Row 156. "is used". Should this be "was used"?

Figure 5. I imagine you could have all the bars in the same plot.

Row 185. "Fuel-wood and dung EFs. . ." You could rephrase this to be in more accurate, it is not emission of fuel-wood or dung after all.

Row 188. OC3 does not look to be more than 50% in Fig. 5

Response: The four comments above have been addressed.

References

Census: Houselisting and Housing Census Data New Delhi., 2011.

CPCB: Air quality monitoring, emission inventory and source apportionment study for Indian cities. National Summary Report, Central Pollution Control Board, 2011.

Patel, S., Li, J., Pandey, A., Pervez, S., Chakrabarty, R. K., and Biswas, P.: Spatio-temporal measurement of indoor particulate matter concentrations using a wireless network of low-cost sensors in households using solid fuels, Environ. Res., 152, 59-65, 2017.

See, S. W., and Balasubramanian, R.: Chemical characteristics of fine particles emitted from different gas cooking methods, Atmospheric Environment, 42, 8852-8862, 2008.

Smith, K. R., Uma, R., Kishore, V., Lata, K., Joshi, V., Zhang, J., Rasmussen, R., Khalil, M., and Thorneloe, S.: Greenhouse gases from small-scale combustion devices in developing countries, Phase IIa: Household Stoves in India, US Environmental Protection Agency, Research Triangle Park, NC, 98, 2000.

Venkataraman, C., Sagar, A., Habib, G., Lam, N., and Smith, K.: The Indian national initiative for advanced biomass cookstoves: the benefits of clean combustion, Energy for Sustainable Development, 14, 63-72, 2010.

Aerosol emission factors from traditional biomass cookstoves in India: Insights from field measurements

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Abstract. Residential solid biomass cookstoves are important sources of aerosol emissions in India. Cookstove emission rates are largely based on laboratory experiments conducted using the standard water-boiling test, but real-world emissions are often higher owing to different stove designs, fuels, and cooking methods. Constraining mass emission factors (EFs) for prevalent cookstoves is important because they serve as inputs to bottom-up emission inventories used to evaluate health and climate impacts. Real-world EFs were measured during winter, 2015, for a traditional cookstove ("*chulha*") burning fuel-wood (FW), agricultural residue (AG) and dung (DG) from different regions of India. Average (± 95% confidence interval) EFs for FW, AG, and DG were: 1) $PM_{2.5}$ mass: 10.5 (7.7 – 13.4) g kg^{-1}, 11.1 (7.7 – 15.5) g kg^{-1}, and 22.6 (14.9 – 32.9) g kg^{-1}, respectively; 2) elemental carbon (EC): 0.9 (0.6 – 1.4) g kg^{-1}, 1.6 (0.6 – 3.0) g kg^{-1}, and 1.0 (0.4 – 2.0) g kg^{-1}, respectively; and 3) Organic carbon (OC): 4.9 (3.2 – 7.1) g kg^{-1}, 7.0 (3.5 – 12.5) g kg^{-1}, and 12.9 (4.2 – 15.01) g kg^{-1}, respectively. The mean (± 95% confidence interval) OC-to-EC mass ratios were 6.5 (4.5 – 9.1), 7.6 (4.4 – 12.2), and 12.7 (6.5– 23.3), respectively, with OC and EC quantified by the IMPROVE_A thermal/optical reflectance protocol. These real-world EFs are higher than those from previous laboratory-based measurements. Combustion conditions have larger effects on EFs than the fuel-types. We also report the carbon mass fractions of our aerosol samples determined using the thermal-optical reflectance method. The mass fraction profiles are consistent between the three fuel categories, but markedly different from those reported in past literature − including the source profiles for wood stove $PM_{2.5}$ emissions developed as inputs to receptor modelling studies conducted by the Central Pollution Control Board of India. The OC3 thermal fraction contributed nearly 50% of the total carbon mass for emissions from all fuels.

1 Introduction

The Indian subcontinent is a regional hotspot for anthropogenic emissions (Ramanathan and Carmichael, 2008). Carbonaceous aerosol (black carbon – BC, and organic carbon – OC) in India is linked to surface dimming (Kambezidis et al., 2012), solar warming of the lower atmosphere (Ramanathan et al., 2001;2007), changing regional monsoon patterns (Chung and Seinfeld, 2005;Menon et al., 2002;Ramanathan et al., 2005), and accelerated melting of Himalayan glaciers (Ramanathan et al., 2007). Particulate matter (PM) emissions—particularly particles with aerodynamic diameters less than 2.5 μm ($PM_{2.5}$)—are also associated with numerous adverse consequences for human health (Pope et al., 2009;Pope and Dockery, 2006). The Global Burden of Disease study has identified indoor air pollution as the largest risk factor and outdoor air pollution as the seventh largest risk factor for disability-adjusted life years in India (Murray et al., 2013) .

The most recent emissions inventory for India indicated that residential biomass cookstoves are the largest contributors to total annual $PM_{2.5}$ emissions (Pandey et al., 2014;Sadavarte and Venkataraman, 2014). In 2010, 67% of Indian households, more than 160 million total, relied primarily on solid fuels for cooking (Census, 2011). Commonly used cooking appliances are mostly mud stoves, with some three-stone type brick stoves and metal stoves (Kar et al., 2012), that burn fuel-wood (FW), agricultural residues (AG), and dried cattle dung (DG). Traditional cookstoves have low combustion efficiencies, resulting in incomplete combustion and high PM emissions (Smith et al., 2000b).

Emissions performance of cookstoves is commonly expressed in terms of mass-basis emission factors (EFs) or mass of pollutant emitted per unit mass of fuel burned. PM emission rates depend on fuel properties, combustion device, operator behaviour and cooking patterns (Leavey et al., 2015;Sahu et al., 2011;Roden et al., 2009). Cookstove heating efficiencies and EFs and are often measured in a laboratory setting using a water-boiling test (WBT) with high- (boiling) and low- (simmering) power phases (Habib et al., 2008;Smith et al., 2000a;MacCarty et al., 2008). These standardized tests are useful for comparing different stove-fuel combinations, but they do not represent real-world stove behaviors found in the field (Roden et al., 2006;Roden et al., 2009;Smith, 2007). Habib et al. (2008) changed the amount of water boiled from 0.5 kg to 1.5 kg in the WBT test, thereby changing the fuel burn rate and burn cycle duration, and observed a factor of ~2.7 increase in the $PM_{2.5}$ EF and a factor of ~2 increase in the OC fraction. A real-world study of Honduran wood-burning cookstoves (Roden et al., 2006) found higher $PM_{2.5}$ EFs and OC content than those from previous laboratory studies. Roden et al. (2006;2009) found that real-world fire tending and cooking practices (and therefore burn conditions) were important factors determining PM EFs and compositions.

Real-world EFs for commonly used fuel types and cooking technologies in India are needed for accurate of bottom-up emission estimates (Bond et al., 2013;Bond et al., 2004;Pandey et al., 2014). Inventoried emission rates serve as inputs to regional and global atmospheric transport models that predict spatiotemporal profiles of pollutant burdens and associated impacts on climate and human health (Bond et al., 2013;Guttikunda and Calori, 2013;Sadavarte et al., 2016;Schulz et al., 2006). Alternatively, these inventories are used in conjunction with impact metrics such as intake fraction (Grieshop et al.,

2011) and global warming potential (Shindell et al., 2012) to evaluate mitigation policies (MHFW, 2015;Sagar et al., 2016). Such measurements identify key parameters to be monitored during laboratory testing and appliance certification.

With the above goals, we measured cookstove emission characteristics in a rural Indian household. Local meals were prepared with a traditional mud stove ("*chulha*") using biomass fuels collected from different regions of India. Real-time measurements of emitted gas concentrations were conducted and PM filter samples were collected at regular time intervals during each cooking cycle. $PM_{2.5}$, OC and EC EFs are reported here as a function of fuel-type and combustion phase. Thermal carbon fractions provided by the IMPROVE_A protocol are also examined.

2 Methods

Thirty separate cooking tests were conducted between December 19 and 30th of 2015 in a rural household on the outskirts of Raipur, a city located in the central Indian state of Chhattisgarh (abbreviated as Chh.). ~77% of Chhattisgarh households are located in rural areas and rely almost entirely on solid biomass fuels for cooking (Census, 2011). On a national level, fuel-wood, agricultural residue and dung are used as primary cooking fuels by 49%, 9% and 8% Indian households respectively (Census, 2011). Accounting for average combustion efficiencies and calorific values of these fuels, annual fuel usage estimates are 250 MT fuel-wood, 73 MT agricultural residue and 100 MT dung (Pandey et al., 2014). For this study, fuel-wood was obtained from Uttar Pradesh (U.P.), Rajasthan (Raj.), Andhra Pradesh (A.P.), Bihar, and Punjab which collectively account for 35% of the total fuel-wood user base in India. All wood fuels were in the form of chunks with typical dimensions of 5 – 15 cm. Cattle dung (in the form of dung cakes dried in the sun) was collected from U.P. and Bihar, which account for 60% of the dung use for cooking in India. Agricultural residues from of *tur* crops (a type of woody stalk) and rice straw were procured from a village near the study location. Test fuels were collected and stored in sealed bags, and later analysed for elemental (carbon, oxygen, hydrogen, nitrogen) composition and moisture content. Fuel compositions are compared in Table 1. Per real-world practice, fuel samples were naturally dried in the sun and stored indoors, bringing moisture contents to < 9%. These compositions are consistent with those reported in other tests (Habib et al., 2008;Smith et al., 2000a).

Table 1: Elemental composition and moisture content of the biomass fuels in this study.

Fuel	Elemental composition (%)				Moisture content (%)
	Carbon	Hydrogen	Oxygen	Nitrogen	
U.P. dung	33.1	4.0	30.0	1.6	7.5
Bihar dung	41.4	5.1	33.6	2.1	8.6
Chh. rice straw	40.7	5.5	39.0	0.8	5.3
Chh. *tur* stalk	48.4	6.5	42.7	0.6	4.8
Punjab wood	50.3	0.2	40.9	0.4	6.2
Raj. Wood	49.7	5.6	42.9	0.1	8.1
U.P. wood	49.9	0.1	41.8	0.2	5.6
A.P. wood	48.3	0.1	43.4	0.7	3.1

Table 2 describes the fuels used and the foods cooked; replicate tests were made for some of these combinations with at least three for each fuel. Dung (20-50 g) was doused with approximately 10 ml kerosene for initial ignition and the test fuel was added after a steady flame was achieved. Additional fuel of the same type was added as needed to complete the recipe. A ten-minute period following lighting of the fire or the addition of kindling materials is designated as *ignition* phase. The remainder of the cooking cycle was designated as the *flaming* phase when a visible flame was present. Combustion entered the *smoldering* phase when the flame died down. The U.P. dung and Chh. rice straw could not sustain the flaming phase for more than a few minutes. Dung is typically smoldered for low-power cooking applications, and it is used as kindling material for igniting fuel-wood in a typical rural household. The low carbon content of U.P. dung (Table 1) possibly hinders its ability to sustain a flame, more so than Bihar dung. Rice straw has a low material density and high surface-to-volume ratio, and therefore tends to burn out very quickly. It also produces large amounts of smoke, making its use as a standalone fuel impractical and harmful for the cook's health. To circumvent these limitations, a few experiments established a steady flame using U.P. dung/Chh. rice straw mixed with U.P. wood (approximately 2.5:1 ratio of test fuel mass to wood mass).

Table 2: List of cooking experiments conducted during the 10-day intensive study period. Abbreviations for Indian states: U.P. = Uttar Pradesh, Raj. = Rajasthan, A.P. = Andhra Pradesh, Chh. = Chhattisgarh.

Day	Primary fuel used	No. of replicate experiments	Food cooked
1	Bihar dung	1	lentil-rice
2	U.P wood	2	rice, vegetables
3	U.P wood	1	tea
	Raj. wood	4	lentils, rice, vegetables
4	U.P. wood	2	lentils
	A.P. wood	1	rice
5	A.P. wood	2	rice, vegetables
	Bihar dung	1	tea
6	Chh. *tur* stalks	3	rice, vegetables, tea
7	U.P. dung[a]	3	vegetables, rice
	Bihar dung	1	tea
8	Chh. rice straw[a]	3	rice, vegetables
	Chh. wood	1	tea
9	Raj. wood	4	water heated, rice and curry
10	Punjab wood	1	milk porridge

[a] Two experiments for each fuel conducted with fuel-wood mixed with the test fuel

The test kitchen (Fig. S1) was on the first floor of house, separate from all other rooms. A permanently open door was the only entry from an open terrace. A partially covered window covered was a second ventilation source. The emissions testing system is shown in Fig. 1. The combustion device was a traditional mud *chulha*, such stoves have poor heat insulation, poor combustion efficiency and do not allow proper mixing of fuel and air (Smith et al., 2000a;Venkataraman et al., 2010). An eight-armed stainless steel probe (based on Roden et al. (2006)) sampled naturally-diluted emissions at ~1.2 m above the top of the stove. Each arm of probe was 0.5 m in length, with 4 uniformly placed holes facing the plume. This probe was connected to three real-time instruments−a Kanomax Portable Mobility Particle Sizer (PAMS) (Kulkarni et al., 2016), a TSI Sidepak (Zhu et al., 2007), and a Testo-350 gas analyser (Wang et al., 2012). The PAMS recorded particle size distributions from 10 – 400 nm mobility diameter. The Sidepak provides a light-scattering (670 nm) surrogate for measured $PM_{2.5}$ mass that is calibrated with Arizona Road Dust (O'Shaughnessy and Slagley, 2002). PM concentrations exceeded the top range of these instruments for short periods during the plume monitoring. The Testo-350 gas analyser was factory-calibrated prior to the experiments for carbon monoxide (CO) and carbon dioxide (CO_2). Measured concentrations (acquired every second) were

at least five times the detection limits of 1 ppm CO and 0.01 % CO_2 by volume. $PM_{2.5}$ was collected on 47 mm Teflon-membrane and pre-baked quartz-fiber filters several times during a cooking cycle using Minivol (5 L min^{-1}) samplers (AirMetrics Model 4.2) with greased impactor inlets located in the plume ~0.9m above the stove. Filter sample durations ranged from 0.5 to 4 minutes, based on the continuous SidePak reports, to prevent filter overloading. Field blanks were collected (minimum sampling duration of 15 minutes) each day before testing. The Teflon filters were weighed before and after sampling to obtain the net mass deposit which was divided by the sample volume (flow rate times duration) to obtain the concentration. The mass of Teflon filter deposits ranged 50 – 300 µg. Quartz filters were analysed using the Interagency Monitoring of Protected Visual Environments – A (IMPROVE_A) thermal-optical reflectance (TOR) method (Chow et al., 2007b;2011) to determine elemental and organic carbon fractions in the sampled particulates. The minimum detection limits of the TOR analysis are about 9 µg for OC and 1 µg for EC (Solomon et al., 2014).

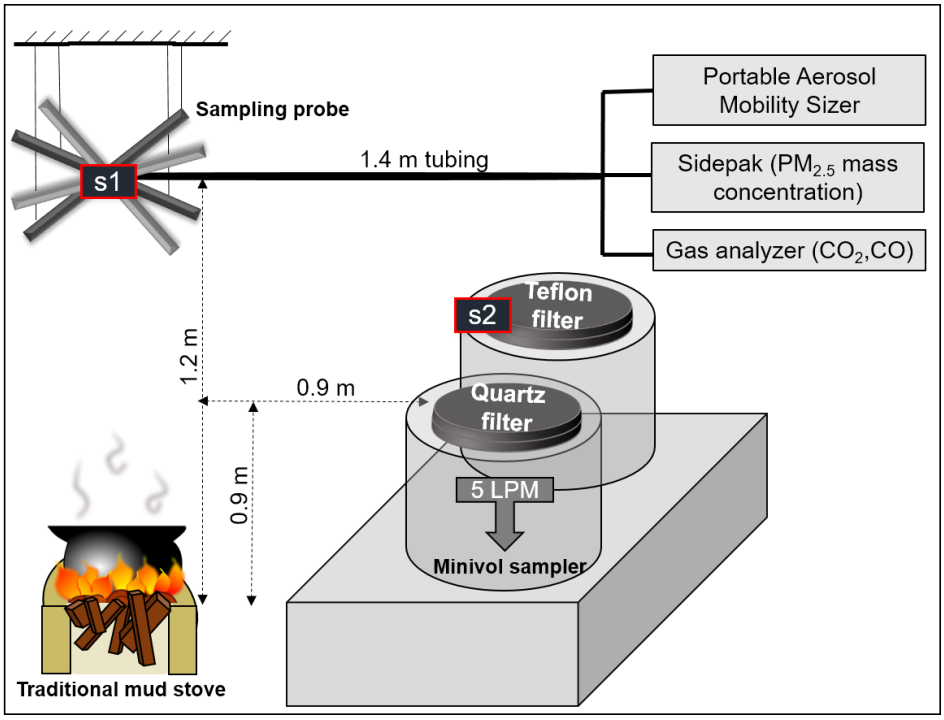

Figure 1: Schematic representation of the experimental setup. S1 and S2 denote the position of two wireless optical sensors.

Using the carbon mass balance technique, fuel-based EFs were calculated for each filter:

$$EF_i = CMF_{fuel} \frac{C_i}{\Delta C_{CO_2}\left(\frac{M_c}{M_{CO_2}}\right) + \Delta C_{CO}\left(\frac{M_c}{M_{CO}}\right)} \quad (1)$$

where EF_i is the EF of species *i* in grams emitted per gram of fuel consumed. CMF_{fuel} is the carbon mass fraction of the fuel, which ranged from 33% to 50% for the tested fuels. C_i is the concentration of emittant *i*, in this case $PM_{2.5}$, OC, or EC, in g m^{-3}, determined for each Teflon and quartz filter. ΔC_{CO2} and ΔC_{CO} are the concentrations above ambient levels of CO_2 and

CO in g m^{-3}, respectively. M_C, M_{CO2}, and M_{CO} are the atomic or molecular weights of C, CO_2, and CO in g $mole^{-}1$. Eq. (1) assumes that the carbon emitted in CH_4, NMHC, and PM is negligible compared to that in CO and CO_2.

Wireless optical particle sensors (details available in Patel et al. (2017)) were attached to the Minivol sampler and the sampling probe during six experiments to check for any significant differences in the particle concentrations measured at the two locations. Measurements where either sensor was saturated were discarded, and a linear regression analysis performed on the valid data points. A correction factor of 1.6 was applied based on regression slope (Supplemental Data). The equation above was also corrected to account for the small fraction of fuel carbon that gets converted to gaseous volatile organic carbon, rather than CO_2 or CO, assumed as 2.4% (Habib et al., 2008;Roden et al., 2006).

3 Results and discussion

Figure 2 compares EFs for the different fuels. There are no statistically significant (unpaired *t*-test, 95% confidence intervals) EF differences for wood-fuels from different regions of India. Bihar dung EFs exceeded those for U.P. dung, possibly owing to the addition of wood to sustain flaming. On average, $PM_{2.5}$ and OC emission factors for dung were higher than those for fuel-wood. EFs for dung, rice straw and *tur* stalk show a larger spread than corresponding EFs for fuel-wood.

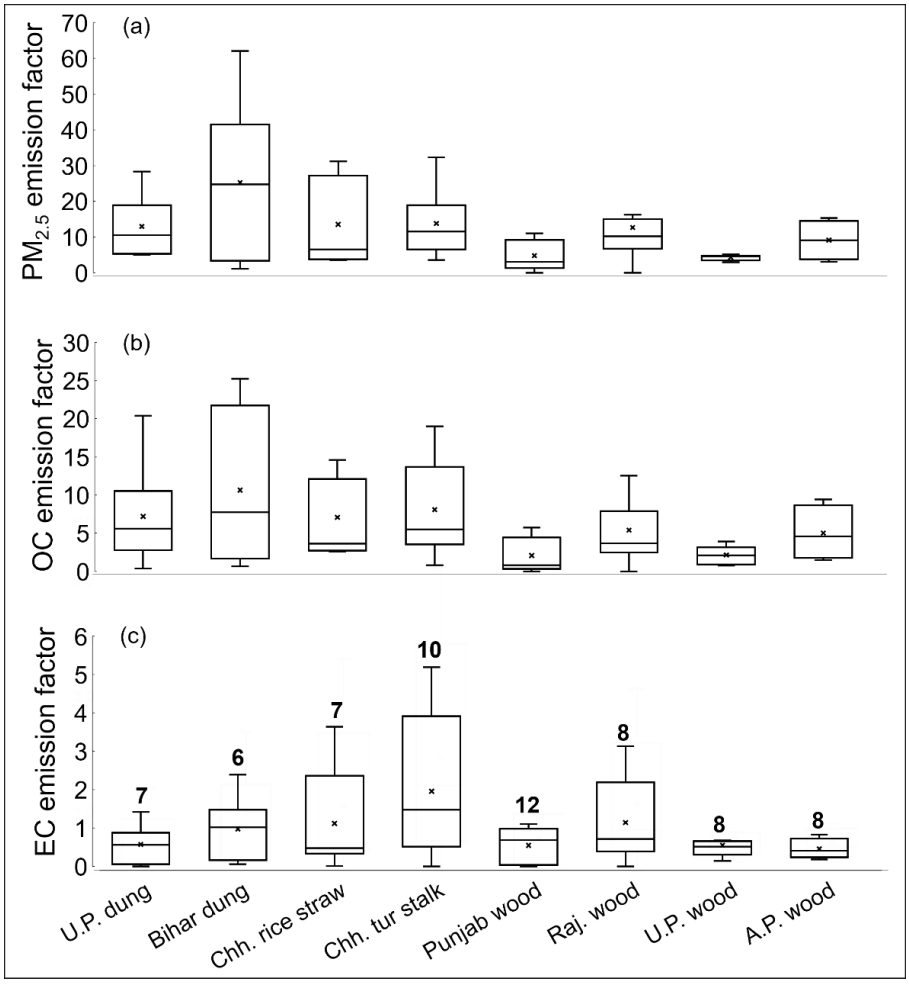

Figure 2: Box plots of (a) $PM_{2.5}$ emission factors, (b) OC emission factors, and (c) EC emission factors. All emission factorss are expressed in g-pollutant per kg of fuel burnt. Boxes denote lower and upper quartiles; whiskers are 1.5 times the interquartile ranges of the upper and lower quartiles. The numbers above the error bars in panel (c) indicate the number of samples for each fuel.

Figure 3 compares EFs for the different burning phases. $PM_{2.5}$ EFs are highest during the ignition phase for all fuels. The OC/EC ratio (Fig. 3b) increases from ignition and flaming, to smoldering for all fuels. CO EFs and modified combustion efficiencies (MCE, ratio of CO_2 concentration to $CO+CO_2$ concentration) show no correlation with $PM_{2.5}$ emission factors or OC-to-EC ratios (Supplemental Data). MCEs exceeded 0.9 – a value associated with flaming (Reid et al., 2005;Zhang et al., 2008) – for nearly 90% of the test duration, even when no flaming was observed.

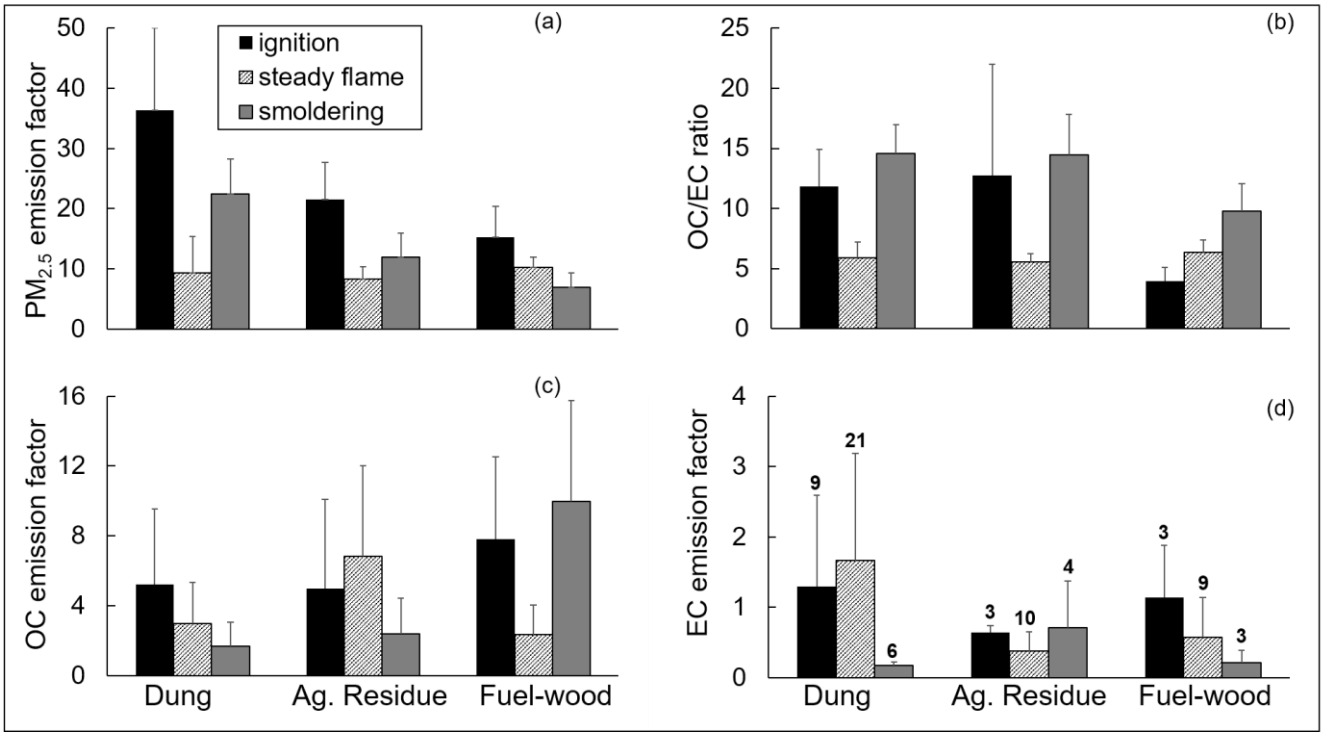

Figure 3: Fuel-wise average values of (a) $PM_{2.5}$ emission factors, (b) OC-to-EC ratios, (c) OC emission factors, and (d) EC emission factors, categorized by observed combustion phases. All emisssion factors are expressed in g-pollutant per kg of fuel burnt. One-sided error bars are shown to denote one standard deviation from the mean. The numbers above the error bars in panel (d) indicate the number of samples for each fuel and combustion phase.

Average EFs for the entire burn cycle were calculated as a time-weighted sum of EFs for each phase of combustion. Fuel-wood and agricultural residue are used predominantly in flaming conditions to carry out the bulk of cooking operations, resulting in weights of 17% ignition, 66% steady flame and 17% smoldering. For dung, the weights are 17% ignition, 17% flaming and 66% smoldering because dung was used for longer low-power operations such as heating water/milk and roasting vegetables. These EFs are compared with other reported EFs in Figure 4. Average fuel-wood $PM_{2.5}$ EFs and OC/EC ratios in this study are 20% larger than those reported by (Roden et al., 2006) for Honduran cookstoves, but they are 2-8 times as large as those reported for laboratory studies (Habib et al., 2008;MacCarty et al., 2008;Saud et al., 2012). For agricultural residue and dung, the average EFs and OC/EC ratios are 1.8 – 4.2 times and 1.3 – 2.2 times higher, respectively, compared to those reported by Saud et al. (2012) and Habib et al. (2008).

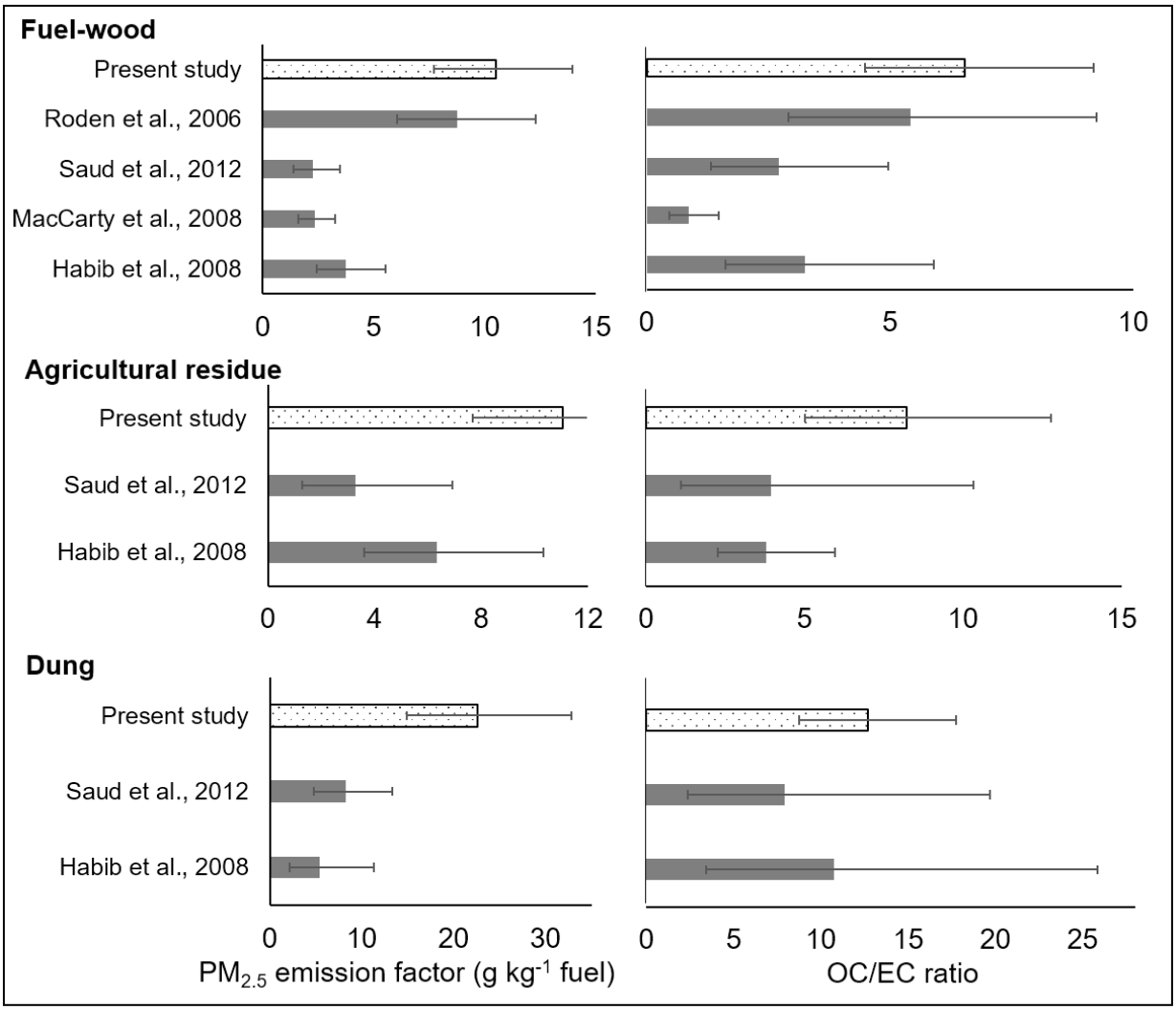

Figure 4: Average $PM_{2.5}$ emission factors and OC-to-EC ratios for the three fuel categories in this study, compared with relevant studies. Error bars for values estimated in this study denote 95% confidence intervals based on standard errors of the means. Error bars for other studies are the bounds reported within those studies.

Thermal fractions of total carbon constituted by the IMPROVE_A protocol are compared in Fig. 5. OC1, OC2, OC3 and OC4 refer to carbon that evolves at temperatures of 120 ºC, 250 ºC, 450 ºC, and 550 ºC respectively, in the inert helium atmosphere. OP denotes pyrolyzed carbon, OC charring in the inert helium carrier. EC1, EC2 and EC3 fractions evolve in a 2%O_2/98%He oxidizing atmosphere at 550 ºC, 700 ºC and 800 ºC, respectively. Fig.5 compares fractions from this study with those reported for controlled biomass (hardwood and softwood) burning reported in Chow et al. (2007a). We also compare our results with a source profile developed for $PM_{2.5}$ emissions from wood *chulhas*, as part of source apportionment studies conducted by the Central Pollution Control Board (CPCB) of India (CPCB (2011)). This profile was based on laboratory burns, but details of the test fuel and burn protocol are not known. OC3 was the most abundant fraction, ~50% of OC3 by mass, while

the profiles in literature ranged 10%-34% in the OC3 fraction. The OC1 fraction for all fuels in this study was uniformly less than 3%, a finding comparable only to the 5% OC1 reported for softwood, but not for the other two profiles.

Carbonaceous aerosol source profiles are useful for source apportionment, and they may also have implications for climate and health impact assessments. In an previous study (Pandey et al., 2016), we reported that light absorbing OC may play a larger role in light absorption by cookstove emissions than that from earlier work on biomass burning in the U.S.A. The difference in constituents of OC emissions from the two sources might contribute to the observed difference in their optical characteristics, since thermal stability is known to be inversely related to the light absorption efficiency of organic compounds (Andreae and Gelencsér, 2006;Saleh et al., 2013).

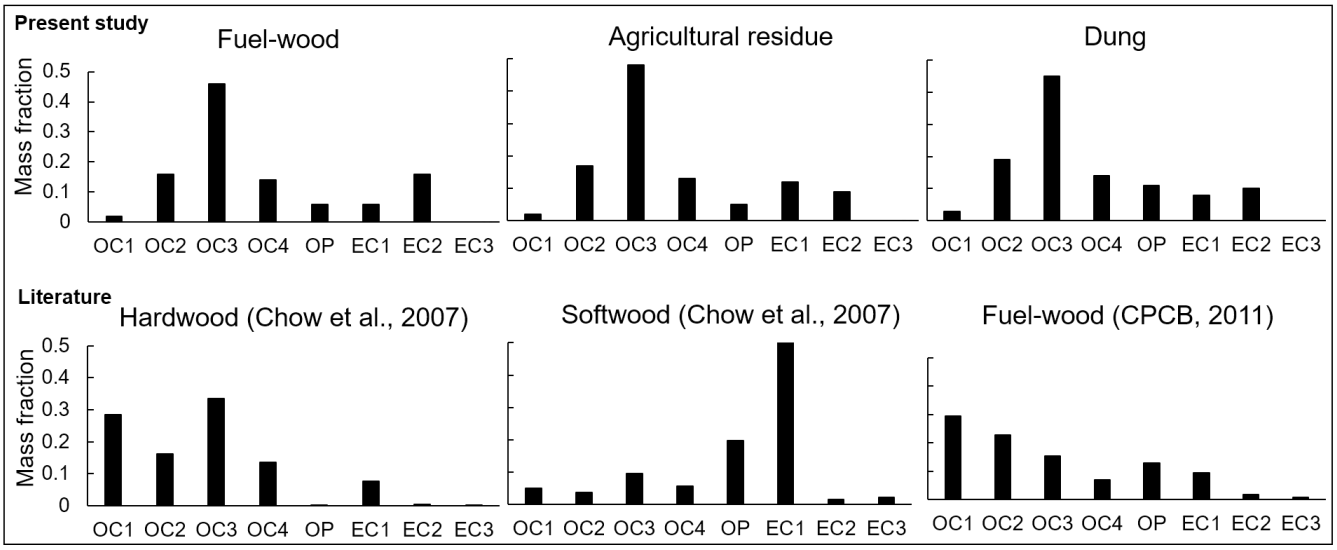

Figure 5: Fraction of total carbon contributed by the IMPROVE_A thermal carbon fractions.

4 Conclusions

We estimated $PM_{2.5}$, OC and EC mass emission factors from real-world combustion of commonly-used biomass fuels in India. EFs for fuel-wood and dung are more than twice those derived from laboratory tests for similar appliances and fuels. Earlier studies have shown that fuel burn rate (Habib et al., 2008) and fire-tending practices (Roden et al., 2009) affect cookstove emissions, therefore standardized burn protocols (typically a water boiling test) cannot replicate cookstove performance in the field. In a controlled test, variability in wood size may could be expected to affect emissions from fuel-wood combustion, hence, laboratory studies typically used identical wood chunks. The four wood fuel types in this study were used as chunks in a range of sizes, and yielded similar EFs even though these samples were gathered from widely separated parts of India. Overall, the variability in emissions between multiple measurements for the same fuel was larger than or comparable to the differences in average emission rates for different fuels.

A short period immediately following the ignition phase showed the highest and most variable $PM_{2.5}$ EFs for all fuels, consistent with previous observations of high emissions from lighting and refuelling of stoves (Roden et al., 2009). The use of variable amounts of kerosene and other kindling materials, as required in any burn, may explain the large spread in emission measurements during ignition. The highest OC-to-EC ratios were observed during smoldering (no visible flame present), when pyrolysis is the dominant process. The OC3 thermal fraction contributed nearly 50% of the total carbon mass for emissions from all fuels, indicating that most of the PM emissions are non-evaporative. This could have implications for the climatic impact effect of cookstove emissions, especially since volatility of OC emissions is inversely related to their light absorption efficiency in the near-UV and visible wavelengths (Andreae and Gelencsér, 2006). The thermal fractions found in this study differ from those found in other biomass burning tests, including the source profile used an input to receptor models of particulate emissions in India (CPCB, 2011). Our findings suggest that combustion conditions may have a larger influence on intrinsic properties of biomass combustion emissions than fuel variability. Connecting aerosol emissions from a given source to their effects on climate and human health requires knowledge of their mass emission rates and physiochemical properties. The EFs from this study could contribute toward improving evaluations of climate and health impacts of carbonaceous aerosols over India. Further investigation is needed on the relationship between the composition of OC emissions and their climate and health effects.

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