

Comments by reviewer #1 are reproduced in the sans-serif font below. Our responses follow each comment in a blue, italicized, serif font. Text additions to the manuscript appear in the manuscript in red color. Deletions from the manuscript are described in the responses below.

In addition to the changes triggered by reviewer comments, other changes were made to improve readability. The significant additional additions or changes also show up in red color in the manuscript.

Pg 2, line 21: Please give a brief description of each stove type so that the reasons behind the differences in emissions are clearer.

The text was amended to clarify what the stoves are generally used for and the primary combustion type. Additions to the text are highlighted in red. The authors agree that these additions will assist the readers in making sense of the different emissions from the stoves that are described later.

“This time, we measured emissions in field conditions from two traditional, locally-made cookstoves, the *chulha* and the *angithi*. The former is a primarily flaming stove with generally higher modified combustion efficiencies or concentration ratios of carbon dioxide to the sum of carbon dioxide and carbon monoxide (average dung-*chulha* 0.865), used to cook village meals. The *angithi* is largely smoldering with lower modified combustion efficiencies (average dung-*angithi* 0.819) and is primarily used for cooking animal fodder and simmering milk.”

Additional text was added on page 3.

Pg 3, line 1: Was the dung:brushwood ratio known for each mixed fire to accurately estimate fuel C content?

Yes it was. The following sentence was added to clarify this point.

“When mixtures of dung and brushwood were used, both were individually weighed to accurately determine the carbon mass burned.”

Pg 4, line 12: “The background filter mass was adjusted to match the flow rate of the sample filter by assuming the flow rate is proportional to the filter mass.” This sentence is confusing. How would mass “match” a flow rate? Are the authors saying the sampled volume of the background filter was not always the same as that of the PM sample and therefore the background mass was scaled up or down according to the ratio of the sample volumes? Also, in Figure 1, the flow rate of the background filter is nominally the same as the PM filter, so were these marginal adjustments? Please rephrase this sentence.

We agree this was confusing as written. We clarified that Teflon A (sample) and Teflon C (background) of Figure 1 were reserved for gravimetric analysis, whereas Teflon B was not used in this paper. With this it becomes clear that the background mass needs to be adjusted for flow rate, which is now described as matching sampling volumes in the text. We also explicitly state our assumption that the amount of PM collected on the filter is proportional to the sampling flow rate through the filter.

Pg 7, line 12: In the comparison of EFs between this work and Stockwell et al., some consideration should be given to MCE. MCE could account for some of the EF differences for brushwood given the higher MCE of this study compared to Stockwell, although it is unlikely to explain the differences for

dung. Is there sufficient information in Stockwell et al. (2016) to calculate EFs in g/kg C that could be overlaid on Figure 3 (or use g/kg in figure 3)?

Thank you for making this suggestion. We plotted their raw EF data in the supplemental information (Figure S2.2). There is more agreement in our datasets than it first appeared. This spurred a conversation with the authors of Stockwell et al., 2016 during which we learned that much of the discrepancy could come from adjusting their laboratory EFs to better match field observations. We also added text to explain this in the paper (page 7-8).

Section 3.2: The overall discussion section would likely flow better if the MCE section is moved earlier, as most of the preceding discussion requires consideration of MCE.

We believe that the emission factors are the most impactful contribution of this manuscript. Therefore, we decided to show first the emission factors in the chemical composition section. The statement that emissions generally have an inverse relationship with MCE is common knowledge, and does not warrant extensive discussion at the beginning of the paper. Rather, we focus on compounds/compound classes that have interesting relationships with MCE.

Pg 7, line 28: In Figure 3b, the EFs from the chulha stove appear to follow a clear linear trend with MCE regardless of fuel type. In contrast, emissions from the angithi stove show no apparent trend with MCE. The benzene case is also similar. Could these “complicated” MCE relationships be due to differences between the two stoves? The authors should discuss the different MCE trends observed between the different stove types.

We agree with this interpretation, and convey that the fuel-stove combination is important for determining emissions (not just MCE) in the second paragraph of the MCE section 3.2. A sentence was added to emphasize this point.

“Knowledge of the cook fire MCE alone is not sufficient to determine the EF of ethane. **Combustion specific to the fuel-stove combination is a significant factor in cook fire emissions.**”

Pg 7, line 30: Alkyne emissions from the chulha stove clearly show increasing EF with decreasing MCE (negative slope). This contradicts the discussion on pg 6, line 34 that alkynes are predominantly emitted from flaming combustion, which would show a positive EF vs. MCE slope. Please reconcile those two points.

At lower MCEs there is a positive slope, this is largely encompassed by the dung-burning stoves. As there is more flaming combustion, we do in fact measure higher emissions of alkynes. However, it is well known that overall emissions are lower at higher MCEs. If one compares emission factors from ethane, alkenes, and alkynes in Figure 3, alkynes are highest at MCEs > 0.95.

Pg 8, line 36: “aromatics make up on average roughly 95% of SOA precursors for all cook fires.” This is not at all surprising considering that nearly all of the other compounds measured are either too light to contribute significantly to SOA production or don’t have a reported SOAP value (Table S3). A simple disclaimer is warranted stating as such and that the contribution of aromatic hydrocarbons to SOAP here is likely an upper limit depending on the composition of the unmeasured fraction of VOCs in cookstove emissions. The authors may additionally want to compare to Stockwell et al. 2015 (Atmos. Chem. Phys., 15, 845-865), who measured laboratory cooking fires using PTR-TOFMS and observed many other compounds that could act as SOA precursors.

While Stockwell et al. 2015 did measure EFs from laboratory cook fires using PTR-TOF-MS, we chose to focus on Stockwell et al. 2016 because they use similar fuels and some of these measurements are in the

field. We do acknowledge that we did not quantify every SOA precursor which could add up to a significant amount of SOA. We clarified that the 95% comes from only species measured in this study on page 9.

Pg 9, lines 7 and 12: Do these predicted ozone mixing ratios represent the excess ozone produced from only cooking fires? Clarify the text.

This was clarified by adding phrases “due solely to cook stove use”, “from cooking.”, and “excess” in front of the predicted ozone levels.

Section 4: The structure would make more sense if the paragraphs were swapped.

We chose to keep the order of the paragraphs in the “Atmospheric implications and conclusions” section. To us, the first paragraph gives us an implication for the work, which is the quantification of ozone production in a village from cooking. Then the last paragraph holds take home conclusions for the readers, the order given in the subtitle.

Technical corrections:

Pg 2, line 14: typo in ‘Alternatvely’

Thank you for catching this. It is now spelled correctly.

Pg 2, line 17: typo in “a simulated village houses”.

The grammar was corrected.

Equation 1: It’s redundant to use the summation sign and ‘+’ signs.

Equation 1 was fixed.

Comments by reviewer #2 are reproduced in the sans-serif font below. Our responses follow each comment in a blue, italicized, serif font. Text additions to the manuscript appear in the manuscript in red color. Deletions from the manuscript are described in the responses below.

1. There is a wide diversity of stove and fuel types globally and the stove and fuel types explored here are but a small fraction of those used in real world. So while the gas-phase speciation offers a detailed view of the emissions from these stove-fuel sources, how are the stove-fuel sources in this work representative of the stove-fuel combinations in India and globally? More specifically, what fraction of the gas-phase emissions from cookstoves come from the stove-fuels described in this work? And depending on the answers to the previous question, how can this speciation, if at all, be used to inform the gas-phase emissions speciation in large-scale atmospheric models?

The objectives of the current paper are not to produce representative values for global emissions. Stockwell et al., 2016 and this work are the most comprehensive measurements to date in producing speciated gas phase emission factors. The stoves and fuels measured in the current study are predominant in the Indo-Gangetic plains. As a part of this collaborative project, the Seinfeld group at Caltech is modeling secondary organic aerosol formation using CMAQ to describe rural North India.

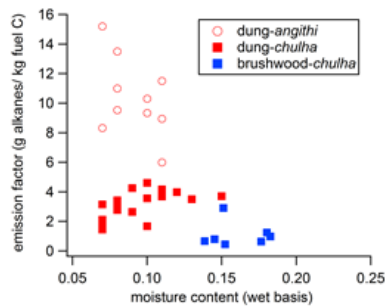
2. Since multiple tests were done with each stove-fuel combination, were other important variables recorded and/or controlled during the test? For example, fuel moisture content, environmental conditions (e.g., temperature, relative humidity), fuel size and burn rate, cooking pots, meals cooked. According to the authors, are any of these variables important in explaining the variability? Citing relevant literature on the factors affecting cookstove emissions variability would be helpful.

Moisture content of the fuels did not correlate with emission factors; in all cases the $r^2 < 0.3$. In one case, we did see a negative relationship between emission and moisture content (Benzene, dung-angithi, $r^2 = 0.718$). We did plot the emission factors as a function of fuel moisture content for a fraction of compounds as examples shown below. We found that meal cooked is not a significant variable ($p > 0.05$). We added the recorded data for each cook fire to the supplemental information, including variable information and emission factors so the dataset is publically accessible. The following statements were added to make this clear to the reader.

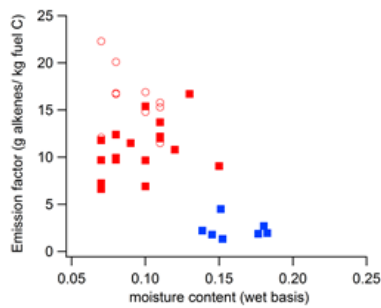
“Fuel moisture content, fuel mass burned, and meals cooked were noted for each cook fire, and can be found in the supporting information.” (Page 3)

“There are many factors that may lead to variability in biomass burning emissions including pyrolysis temperature (Chen and Bond, 2010), fuel moisture content (Tihay-Felicelli et al., 2017), and the wind speed/direction (Surawski et al., 2015), among others. Relationships between emissions and fuel moisture content (Figure S1) or meal cooked were not found to be significant for any compounds ($p < 0.05$). This paper focuses on the relationships between emissions and fuel-stove combination.” (Page 6)

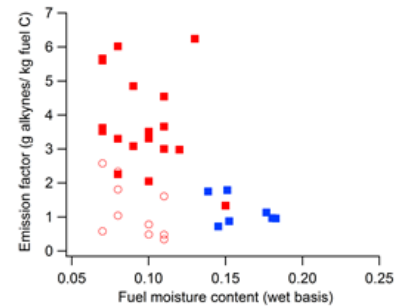
a) Total alkanes



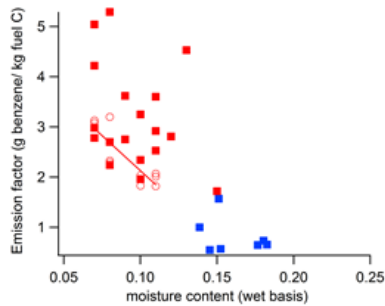
b) Total alkenes



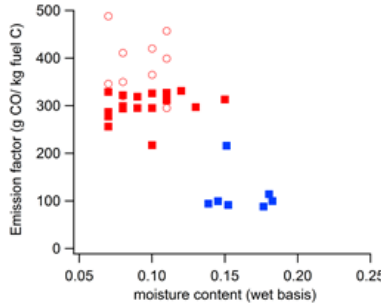
c) Total alkynes



d) Benzene



e) Carbon monoxide



3. Some more detail on the fuels and stoves for the less informed reader would be helpful. What animals was the dung from? Presumably cow? What is an Angithi stove? What is a chulha? How are these different? What is brushwood? Is the brushwood from a particular plant? The use of pictures could help.

The following was added to the introduction, to explain the hypothesis that these stove will lead to different emissions.

“The former is a primarily flaming stove with generally higher modified combustion efficiencies or concentration ratios of carbon dioxide to the sum of carbon dioxide and carbon monoxide (average dung-chulha 0.865), used to cook village meals. The *angithi* is largely smoldering with lower modified combustion efficiencies (average dung-angithi 0.819) and is primarily used for cooking animal fodder and simmering milk.”

The pictures of stoves and the kitchen are already published in Fleming et al., 2018, but the reader is referred to this publication for more extensive information about the materials used in the cook fires. The following sentences were added to the experimental methods section.

“Animal fodder simmers upon smoldering dung in a clay bowl, referred to as an *angithi*. *Chulha* stoves are made from bricks and a covering of clay, and the availability of oxygen from the packing of biomass fuels results in primarily flaming combustion. The *chulha* is used to cook most meals for the family in this village. Buffalo and cow dung patties and brushwood, in the form of branches and twigs, were used in *chulha* stoves, and for the 13 mixed fuel cooking events dung and brushwood were combined in a ratio determined by the cook’s preference.”

4. In equations 1-3, how are m_T and $m_{T,c}$ estimated/calculated?

The language in red was added to clearly show how m_T and $m_{T,C}$ are obtained.

“Fuels were weighed before they were burned, and the dry mass was calculated based on moisture content measurements. The ash was weighed after the cooking event and subtracted from the dry mass of the fuel giving the net dry fuel burned for the cooking event, m_T . When mixtures of dung and brushwood were used, both were individually weighed to more accurately determine the carbon mass burned. The fraction of carbon in the fuel used to yield $m_{T,C}$ was taken to be 0.33 for buffalo dung and 0.45 for brushwood fuels based on Smith et al. 2000.”

5. Page 4, line 11: What is the limit of detection and limit of quantification for the filter measurement? The 0.75 μg blank seems quite low.

We determined the method detection limit to be 9.3 micrograms based off the standard deviation of the field blanks multiplied by 3. All masses for the sample and background filters are above this limit of detection.

6. Page 5, line 1: I am not sure what the point is of normalizing the SOA production from a species to that of toluene? Why not report the SOA production in absolute values of g/kg-fuel when the presentation of results in Figure 2(c) is done in relative format anyways?

The SOAP value for a particular VOC, as described by Derwent et al., 2010, is the change in SOA mass concentration when this VOC is introduced into the photochemical transport model, divided by the change in SOA mass concentration for when toluene is introduced. We use these SOAP values published in Derwent et al., 2010 to determine SOA forming potential for each quantified VOC. We are simply inheriting these data from the literature to estimate the relative magnitude of the effects, with the hope that more detailed modeling work would follow. The dataset is publicly available, so if one wished to generate absolute g SOA mass/ kg fuel burned by estimating the SOA yield for toluene in the plume, this is possible.

7. Why was the maximum incremental reactivity approach used to determine the ozone potential? If an alternative method was used (e.g., MOIR, EBIR), do the findings change?

Maximum incremental reactivity was chosen because a VOC limited regime gives us the highest sensitivity in ozone generation to cooking emissions. Carter 1994 concludes that the MIR scenario, since it is based on integrated ozone production rather than maximizing peak ozone levels (MOR scenario,) is generally less dependent on exact NO_x inputs. This high- NO_x scenario might be more realistic as well since we are focusing on smoke plumes. A sentence was added to the experimental section 2.7 summarizing this explanation for the use of the MIR scenario.

8. Can more details about the SOA formation be added? Were the SOA yields for low or high NO_x conditions? What OA mass concentration as the absorbing mass was used to determine the SOA yield? Were they corrected for vapor wall losses? NO_x and vapor wall loss corrections are species dependent (see Zhang et al., 2014) and may change the apportionment shown in Figure 2(c).

The photochemical trajectory model is outfitted with the Master Chemical Mechanism (MCM 3.1), which dictates gas-phase reactions and SOA formation. NO was initialized at 2 ppb, and NO₂ to 6 ppb. This is now described in section 2.5.

9. Page 7, lines 3-21: Are the brushwood results in this study comparable to the hardwood results from Stockwell et al. (2016)? If yes, why? It is unclear what the point of the comparison to the Stockwell study is since the manuscript only has a few sentences on this comparison. Is it just to show that the emissions in this study were lower than those in Stockwell et al. (2016). The explanations offered for the lower emissions was not satisfactory. Did the authors try to compare the emissions on a normalized basis with each other? Do they correlate?

Stockwell et al. 2016 and this paper are the most comprehensive gas-phase inventories to date in terms of cook fire emissions. It made sense to the authors to cross-compare the results. “Hardwood” and “brushwood” emissions appear to be more similar to each other than emissions from dung-based fuels. There are some real differences in emissions from cook fires between the studies, and we do our best to explain in the text these differences for the reader to compare. In S2.1 we plot the EFs in Stockwell et al. 2016 against those reported in our paper. In the review process, we’ve learned more about how the laboratory EFs were adjusted to field observations in Stockwell et al., 2016, and we believe these adjustments may have contributed to the discrepancy in the reported EFs. We’ve added Figure S2.2 where we plot the unadjusted emission factors in both studies, and there is an encouraging degree of agreement in the actually measured (unadjusted) values.

10. Section 3.3 for SOA: Was there an estimate for emissions of total non-methane organic gases (NMOG)? What fraction did the speciated compounds account for of the total NMOG? Were any intermediate volatility and semi-volatile organic compounds targeted? What fraction of the NMOG was unspciated? What implications does this unspciated fraction that may include lower-volatility vapors have for SOA formation?

We agree that there are semi and intermediate volatility compounds that were not measured in this study. Many of these will contribute to SOA and ozone formation. We did not attempt to quantify NMOG. This was not possible using our carbon balance method and an unknown dilution factor. We have added a “disclaimer” to the manuscript in the “SOA formation potential” section, printed below.

“We would like to emphasize that the SOAP-weighted emissions are reflective of only the measured VOCs, and there are likely semi-volatile and intermediate volatility compounds that are not measured but also contribute to SOA formation.”

11. Section 4: Could atmospheric implications for SOA formation also be determined for this village similar to those for O3? How would the SOA formation compare to the primary PM2.5 emissions?

Unfortunately the SOA forming potentials generated using Derwent et al. 2010 only give relative amounts of SOA formation. From this analysis we concluded that a dung-chulha cook fire would produce almost 3 times more SOA than a brushwood-chulha cook fire. However, with this methodology we cannot compare the absolute magnitudes of primary and secondary emissions. Further work on addressing this exact question is in progress with the Seinfeld group at Caltech to model SOA formation in a simulated village.

Emissions from village cookstoves in Haryana, India and their potential impacts on air quality

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Abstract. Air quality in rural India is impacted by residential cooking and heating with biomass fuels. In this study, emissions of CO, CO₂, and 76 volatile organic compounds (VOCs) and fine particulate matter (PM_{2.5}) were quantified to better understand the relationship between cook fire emissions and ambient ozone and secondary organic aerosol (SOA) formation. Cooking was carried out by a local cook and traditional dishes were prepared on locally built *chulha* or *angithi* cookstoves using brushwood or dung fuels. Cook fire emissions were collected throughout the cooking event in a Kynar bag (VOCs) and on PTFE filters (PM_{2.5}). Gas samples were transferred from a Kynar bag to previously evacuated stainless steel canisters and analyzed using gas chromatography coupled to flame ionization, electron capture, and mass spectrometry detectors. VOC emission factors were calculated from the measured mixing ratios using the carbon-balance method, which assumes that all carbon in the fuel is converted to CO₂, CO, VOCs, and PM_{2.5} when the fuel is burned. Filter samples were weighed to calculate PM_{2.5} emission factors. Dung fuels and *angithi* cookstoves resulted in significantly higher emissions of most VOCs (p < 0.05). Utilizing dung-*angithi* cook fires resulted in twice as much of the measured VOCs compared to dung-*chulha*, and four times as much as brushwood-*chulha* with 84.0, 43.2, and 17.2 g measured VOC/ kg fuel carbon, respectively. This matches expectations, as the use of dung fuels and *angithi* cookstoves results in lower modified combustion efficiencies compared to brushwood fuels and *chulha* cookstoves. Alkynes and benzene were exceptions and had significantly higher emissions when cooking using a *chulha* as opposed to an *angithi* with dung fuel (for example, benzene emission factors were 3.18 g/ kg fuel carbon for dung-*chulha* and 2.38 g/ kg fuel carbon for dung-*angithi*). This study estimated that three times as much SOA and ozone in the Maximum Incremental Reactivity (MIR) regime may be produced from dung-*chulha* as opposed to brushwood-*chulha* cook fires. Aromatic compounds dominated as SOA precursors from all types of cook fires, but benzene was responsible for the majority of SOA formation potential from all *chulha* cook fire VOCs, while substituted aromatics were more important for dung-*angithi*. Future studies should investigate benzene exposures from different stove and fuel combinations and model SOA formation from cook fire VOCs to verify public health and air quality impacts from cook fires.

1 Introduction

35 Parts of rural India are comprised of densely populated villages with ambient ozone and PM_{2.5} levels that affect air quality for inhabitants (Bisht et al., 2015; Ojha et al., 2012; Reddy, 2012). For example, in the rural area of Anantapur in Southern India, monthly mean ozone levels varied between 29 ppbv (parts per billion by volume) in August during the monsoon season and 56 ppbv in April (Reddy, 2012). In Pantnagar, a semi-urban city, the maximum observed ozone concentration was 105 ppbv for one

day in May, with the lowest average monthly maximum of 50 ppbv being in January (Ojha et al., 2012). In terms of PM_{2.5} levels, Bisht et al. (2015) observed an average of 50 µg m⁻³ of PM_{2.5} over July-November 2011 in rural Mahabubnagar. While measurements of O₃ and PM_{2.5} in rural India are relatively scarce, it has become clear that household combustion is a major contributor to ambient levels of these pollutants. For example Balakrishnan et al. (2013) measured PM_{2.5} concentrations in households and observed 24-hour concentrations of 163 to 609 µg m⁻³. Over the last half-decade, several researchers have, through independent studies, come to the conclusion that a significant fraction (22-52%) of ambient PM_{2.5} is directly emitted from residential cooking and heating (Butt et al., 2016; Chafe et al., 2014; Conibear et al., 2018; GBD MAPS Working Group, 2018; Guttikunda et al., 2016; Klimont et al., 2017; Lelieveld et al., 2015; Silva et al., 2016).

Residences in India were estimated to consume 220, 86.5, and 93.0 Tg per year of dry matter of wood fuel, agricultural residues, and dung, respectively in the year 1985 (Yevich and Logan, 2003). While the fraction of Indians using biomass cook fuels is decreasing, the total population is increasing such that biomass fuels are still being utilized at approximately the same overall level (Pandey et al., 2014). Emissions of primary PM_{2.5} from residential cooking in India were estimated to be 2.6 Tg per year based on a compiled emissions inventory (Pandey et al., 2014). Additionally, Pandey et al. (2014) estimated that 4.9 Tg of non-methane volatile organic compounds (NMVOCs) are produced annually in India from residential cooking. This suggests that additional PM_{2.5} mass may be formed via secondary pathways from the oxidation of NMVOCs and either nucleation of new particles or condensation onto existing PM_{2.5}. Alternatively, these non-methane VOCs could contribute to photochemical ozone production in the presence of NO_x (Finlayson-Pitts and Pitts, 2000).

In this study, we quantified emissions of CO, CO₂, and 76 different VOCs from 55 cook fires carried out by a local cook in a village home cooking typical meals. It is thus a substantially updated version of the work done in simulated village houses in India and China in the 1990s, where 58 fuel-stove combinations were measured in semi-controlled conditions using water boiling tests including a number of non-biomass stoves although a similar set of pollutants were measured (Smith et al., 2000b, 2000a; Tsai et al., 2003; Zhang et al., 2000). This time, we measured emissions in field conditions from two traditional, locally-made cookstoves, the *chulha* and the *angithi*. The former is a primarily flaming stove with generally higher modified combustion efficiencies or concentration ratios of carbon dioxide to the sum of carbon dioxide and carbon monoxide (average dung-*chulha* 0.865), used to cook village meals. The *angithi* is largely smoldering with lower modified combustion efficiencies (average dung-*angithi* 0.819) and is primarily used for cooking animal fodder and simmering milk. We measured emissions from cookstoves with two kinds of biomass; the most popular biomass type brushwood (Census of India, 2011) and dung cakes. Our first objective is to characterize emissions of select VOCs and PM_{2.5} from these fuel-stove combinations. Subsequently, with the aid of secondary organic aerosol (SOA) potential values from Derwent et al. (2010), incremental reactivity first described in Carter, (1994), and second-order rate coefficients with OH combined with our emission factors, we estimate SOA forming potentials, ozone formed in a VOC-limited regime, and OH reactivity, respectively. Given their widespread use in India, emissions from these biomass-burning stoves are estimated to impact regional air quality due to both primary and secondary organic aerosol and ozone formation.

2 Experimental Methods

2.1 Field site and sample collection

The field office was located at the SOMAARTH Demographic, Development, and Environmental Surveillance Site in Palwal District, Haryana, India run by the International Clinical Epidemiological Network (INCLIN). The site consists of 51 villages in the area with roughly 200,000 inhabitants (Balakrishnan et al., 2015; Mukhopadhyay et al., 2012; Pillarisetti et al., 2014).

5 Samples were collected from cookstove emissions between August 5 and September 3, 2015. Cooking events occurred at a village kitchen in Khatela, Palwal District. A local cook was hired to prepare meals for human consumption consisting of either *chapatti* or rice with vegetables using a *chulha* stove, as well as animal food using an *angithi* stove.

10 Animal fodder simmers in a pot set upon smoldering dung in a clay bowl, referred to as an *angithi*. *Chulha* stoves are made from bricks and a covering of clay, and the availability of oxygen from the packing of biomass fuels results in primarily flaming combustion. The *chulha* is used to cook most meals for families in this village. Buffalo and cow dung patties and brushwood, in the form of branches and twigs, were used in *chulha* stoves, and for the 13 mixed fuel cooking events dung and brushwood were combined in a ratio determined by the cook's preference. Stoves and food ingredients were produced and fuels procured by the household or village. Fuel moisture content, fuel mass burned, and meals cooked were noted for each cook fire, and can be found in the supporting information. Additional information regarding the cooking events and set-up can be found in Fleming et al., 2018.

15 The sampling scheme is illustrated in Figure 1. Air sampling pumps (PCXR-8, SKC Inc.) created a flow of emissions through the sampling apparatus. Emissions were captured with three-pronged probes that were fixed 60 cm above the cookstove. PM_{2.5} emissions and gases were sampled through cyclones (2.5 μm, URG Corporation). The resulting flow of particles was captured on either quartz or PTFE filters, while gases were collected in an 80 L Kynar bag throughout the entire cooking event. Flows were measured both before and after sampling to ensure they did not change by more than 10% using a mass flowmeter (TSI 4140).
20 After the cooking event, pumps were turned off and a whole air sampler, consisting of a stainless steel canister (2L) welded to a Bellows-sealed valve (Swagelok), was filled to ambient pressure from the Kynar bag. Whole air samplers were thoroughly flushed and evacuated in the Rowland-Blake laboratory before being shipped to India. At the end of the measurement campaign, whole air samplers were shipped back to the laboratory and analyzed within two months of the end of the field measurements. A "grab" whole air sample was collected before cooking commenced each day. This served as a background for all cooking events
25 sampled on that day.

2.2 Gas chromatography analysis

Colman et al., 2001 described the VOC analysis protocol in detail. Briefly, a known amount of the whole air sample (WAS) flowed over glass beads inside a U-shaped trap cooled by liquid nitrogen. The flow was regulated by a mass flow controller and resulted in a 600 Torr drop in the pressure in the whole air sampler. High volatility gases such as O₂ and N₂ passed over the
30 beads, while lower volatility gases adsorbed onto the beads. Compounds were re-volatilized by immersing the trap in hot water and were injected into a He carrier gas stream where the flow was split equally to 5 columns housed in 3 gas chromatographs (HP-6890). The compounds were separated by gas chromatography and subsequently detected by two electron capture, two flame ionization, and one quadrupole mass spectrometer detectors. Peaks corresponding to compounds of interest were integrated manually. CO/CO₂ and CH₄ were analyzed using separate GC systems equipped with thermal conductivity and flame
35 ionization detectors as described in Simpson et al., 2014. The CO/CO₂ GC-FID system is equipped with a Ni catalyst that converts CO into detectable CH₄.

2.3 Gas and PM_{2.5} Emission factor calculations

Emission factors (EFs) were calculated using the carbon-balance method, which assumes that all carbon in the fuel is converted to CO₂, CO, VOCs, and PM when the fuel is burned. The total gas-phase carbon emissions were approximated with the concentrations of CO₂, CO, as well as 76 detected VOCs measured using WAS. This is a good approximation since most emitted carbon resides in CO₂ and CO (> 95 %), so the error associated with VOCs that are not detected is relatively small (Roden and Bond, 2006; Smith et al., 1993; Wathore et al., 2017; Zhang et al., 2000). The mass of carbon in species *i* (m_{i,C}) was calculated using equation (1).

$$m_{i,C} \text{ (g)} = \frac{C_{i,C} \text{ (g/m}^3\text{)}}{C_{\text{CO}_2,C} + C_{\text{CO},C} + C_{\text{CH}_4,C} + \dots + C_{\text{C}_6\text{H}_6,C} \text{ (g/m}^3\text{)}} \cdot m_{T,C} \text{ (kg)} \cdot \frac{1000 \text{ g}}{1 \text{ kg}} \quad (1)$$

Where C_{i,C} represents the mass concentration of carbon for species *i*, and m_{T,C} refers to the carbon mass of the fuel, adjusted for ash and char carbon. Fuels were weighed before they were burned, and the dry mass was calculated based on moisture content measurements. The ash was weighed after the cooking event and subtracted from the dry mass of the fuel giving the net dry fuel burned for the cooking event, m_T. When mixtures of dung and brushwood were used, both were individually weighed to more accurately determine the carbon mass burned. The fraction of carbon in the fuel used to yield m_{T,C} was taken to be 0.33 for buffalo and cow dung and 0.45 for brushwood fuels based on Smith et al. 2000. Carbon in ash was estimated as 2.9% and 80.9% of the measured char mass for dry dung and dry brushwood, respectively (Smith et al., 2000b). For each species with n_{c,i} carbon atoms in the formula and molecular weight MW_i, the emissions factor (EF_i) was calculated using equation (2).

$$EF_i \left(\frac{\text{g VOC}_i}{\text{kg fuel}} \right) = \frac{m_{i,C} \text{ (g)} \cdot \frac{MW_i \text{ (g/mol)}}{n_{c,i} \times 12.00 \text{ (g/mol)}}}{m_T \text{ (kg)}} \quad (2)$$

In addition to the emission factor normalized by the total fuel mass, emission factors were normalized to the total carbon mass in the fuel, calculated via equation (3).

$$EF_i \left(\frac{\text{g VOC}_i}{\text{kg fuel C}} \right) = EF_i \left(\frac{\text{g VOC}_i}{\text{kg fuel}} \right) \cdot \frac{m_T \text{ (kg)}}{m_{T,C} \text{ (kg)}} \quad (3)$$

PM_{2.5} mass was determined gravimetrically using Teflon filters (PTFE, SKC Inc., 47 mm) weighed on a Cahn-28 electrobalance with a repeatability of ±1.0 µg after equilibrating for a minimum of 24 hours in a humidity and temperature-controlled environment both before and after sample collection (average temperature 19.5± 0.5 °C; average relative humidity 49 ±5%). Another gravimetric filter was collected in the background during the cooking event and was equilibrated and weighed in the same way (Figure 1, Teflon C). Four field blanks filters were prepared by opening filters and then immediately closing and sealing the filters the same way as all samples; these filters had negligible mass loading (average: -0.75 µg) relative to samples (average: 1.57 mg). The background filter mass (Figure 1, Teflon C) was adjusted by matching the sampling volume to that of the sample gravimetric Teflon filter (Figure 1, Teflon A). This assumes that the mass of PM_{2.5} collected on the filter is directly proportional to the flow rate through the filter. The background mass was then subtracted from the sample mass to obtain the mass of PM (m_{PM}) in equation (4) below.

$$\frac{EF_{PM}}{EF_{CO}} = \frac{m_{PM}/V_{air}}{m_{CO}/V_{air}} \quad (4)$$

2.4 Modified Combustion Efficiency (MCE)

Modified combustion efficiency (MCE) is defined as follows:

$$\text{MCE} = \frac{\Delta\text{CO}_2}{\Delta\text{CO} + \Delta\text{CO}_2} \quad (5)$$

5 where ΔCO and ΔCO_2 are background-subtracted mixing ratios of CO and CO₂ for the time-integrated whole air sample. The total carbon mixing ratio is approximated by the sum of carbon monoxide and carbon dioxide in this definition.

2.5 SOA Forming potential

10 Relative SOA forming potential from measured VOCs was estimated using secondary organic aerosol potential (SOAP) values from Derwent et al., 2010, who used a photochemical transport model to simulate the SOA mass increase from the instantaneous emission of a particular VOC in a single parcel of air travelling across Europe. The model was outfitted with the Master Chemical Mechanism (MCM v.3.1) and the UK National Atmospheric Emission Inventory. The model was initialized with 2 ppbv NO and 6 ppbv NO₂ (Derwent et al., 1998). SOA mass was estimated assuming equilibrium partitioning of oxidation products. Partitioning coefficients were calculated using absorptive partitioning theory of Pankow, 1994. In the SOAP approach, all SOA mass increases from a particular VOC (*i*) are normalized to that of toluene as shown in equation (6).

$$\text{SOAP}_i = \frac{\text{Increment in SOA mass concentration with species } i}{\text{Increment in SOA mass concentration with toluene}} \times 100 \quad (6)$$

15 SOA forming potential was calculated from the published SOAP values using equation (7).

$$\text{SOA potential} = \sum_{i=0}^n \text{SOAP}_i \times \text{EF}_i \left(\frac{\text{g VOC}_i}{\text{kg fuel C}} \right) \quad (7)$$

Table S1 lists the SOAP values used to calculate SOA forming potential in this study. We emphasize that the SOA forming potential presented here is a relative value, and does not represent an absolute SOA yield.

2.6 OH reactivity

20 Total OH reactivity normalized by the mixing ratio of CO was calculated using equation (8).

$$\text{OH reactivity} \left(\frac{1}{\text{s} \cdot \text{ppbv CO}} \right) = \sum_{i=0}^n k_{\text{OH},i} \left(\frac{\text{cm}^3}{\text{molec} \cdot \text{s}} \right) \times \text{ER}_i \left(\frac{\text{pptv VOC}_i}{\text{ppbv CO}} \right) \times 2.46 \times 10^7 \left(\frac{\text{molec}}{\text{cm}^3 \cdot \text{pptv}} \right) \quad (8)$$

25 Second-order rate constants (k_{OH}) at 25°C were taken from the NIST chemical kinetics database (Manion et al., 2015). Table S1 reproduces the k_{OH} constants used in the study. ER_i is the emission ratio for compound *i* in pptv of VOC per ppbv of CO. The last term serves as a conversion factor from VOC mixing ratio to concentration at standard ambient temperature and pressure (25°C, 1 atm). By using the emission ratio to CO, we can track OH reactivity from VOCs depending on the extent of dilution from the plume. From here forward, the OH reactivity (s⁻¹) reported is the average at the location of the sampling probes, or roughly 60 cm above the cookstove.

2.7 Ozone-forming potential (OFP)

30 The ozone-forming potential was estimated from the incremental reactivity of VOCs tabulated in Carter, 2010. Incremental reactivities were calculated by comparing ozone formation before and after a VOC was introduced in a box model simulation.

The Maximum Incremental Reactivity (MIR) scenario sets high NO_x concentrations, optimized to yield the largest incremental ozone production. In other words, ozone production was VOC limited. Because of this, the OFPs given here represent a scenario where VOC emissions from cooking have the largest impact on ozone production. This high-NO_x scenario was chosen for high sensitivity in ozone production from cooking emission VOCs. In addition, it is expected to be more realistic for a smoke plume in which NO_x is co-emitted with VOCs. OFPs were calculated using equation (9).

$$\text{OFP} \left(\frac{\text{g O}_3}{\text{kg fuel C}} \right) = \sum_{i=0}^n \text{MIR} \left(\frac{\text{g O}_3}{\text{g VOC}_i} \right) \times \text{EF}_i \left(\frac{\text{g VOC}_i}{\text{kg fuel C}} \right) \quad (9)$$

OFPs used in this study are listed next to the corresponding compound in Table S1.

2.8 Statistical analysis

One-way analysis of variance (ANOVA) with Tukey Post-Hoc testing was utilized to determine if there were significant differences ($p < 0.05$) in emissions of specific VOCs among categorical variables, i.e., stove and fuel types. All analyses were performed in R version 3.4.0 and RStudio version 1.0.143 (RStudio Inc, 2016).

3 Results and discussion

3.1 Chemical composition

Average VOC and PM_{2.5} EFs (g/ kg dry fuel) as well as MCE are given in Table 1. The compounds are grouped by fuel-stove combination, with major species (CO₂, CO, CH₄, and PM_{2.5}) listed first, followed by sulfur-containing compounds, halogen-containing compounds, organonitrates, alkanes, alkenes, alkynes, aromatics, terpenes, and oxygenated compounds. The sample size (n) used for calculating the average values and standard deviations was n=18 for dung-*chulha*, n=14 for brushwood-*chulha*, n=13 for mixed-*chulha*, and n=10 for dung-*angithi*. For the majority of the compounds, the standard deviations are smaller than or comparable to the average values, indicating fair reproducibility. There are many factors that may lead to variability in biomass burning emissions including pyrolysis temperature (Chen and Bond, 2010), fuel moisture content (Tihay-Felicelli et al., 2017), and the wind speed/direction (Surawski et al., 2015), among others. Relationships between emissions and fuel moisture content (Figure S1) or meal cooked were not found to be significant for any compounds (all $p < 0.05$). This paper therefore focuses on the relationships between emissions and fuel-stove combination.

Figure 2a visually shows the mass fraction attributed to each compound class for the measured gas-phase emissions. The total EFs given below the pie charts are normalized by fuel carbon in Figure 2a in order to compare between cook fires generated with dung, wood, and wood-dung mixtures which have different carbon contents. The total measured VOC emissions from dung-*angithi* were roughly twice that of dung-*chulha* in terms of gram per kilogram fuel carbon. Further, dung-*chulha* emitted more than twice that of brushwood-*chulha*. The most prominent difference is non-furan oxygenates, making up almost half of all brushwood-*chulha* emissions and a smaller fraction for other fuel-stove combinations. While oxygenates make up a higher fraction of brushwood-*chulha* emissions, the absolute EFs for oxygenates from dung-burning and *angithi* cook fires are higher as discussed later in more detail.

Table 2 shows EFs (g/ kg fuel C) for select VOCs. The differences in mean EFs for each fuel-stove combination are also included in Table 2. Mean differences in EFs reported for *chulha* and *angithi* stoves were calculated for cook fires utilizing only dung fuels. Likewise, mean EFs for wood and dung cook fires only represent cooking events using the *chulha*. This was done to isolate a single variable—either fuel or stove type. For all alkanes and most alkenes, we measured higher emissions for dung-

angithi cook fires (Table 2). Also, from the mean differences in EFs, we found that stove specific combustion conditions impacts emissions more than the selection of fuel type. The difference is so dramatic for alkanes and most alkenes that the mean difference in EFs for cookstoves burning dung is always larger than the mean EF of that compound. For comparison, the mean difference in EFs for *chulha* cookstoves is always lower than the overall mean EF. Ethene was an exception; there was no relationship between ethene emissions and stove type. On the other hand, the mean EF of ethene by dung cook fires was very large compared to mean EFs from brushwood cook fires with a mean difference in EFs of 4.05 g/ kg fuel C. Some alkenes with two double bonds were also exceptions. For 1,3-butadiene ($p= 0.06$) and 1,2-butadiene ($p= 0.089$), stove and EF may or may not have a significant relationship. 1,2-Propadiene emissions from *chulha* cookstoves are higher ($p< 0.01$). All three compounds still show a significant relationship to fuel type with EFs being higher for dung cook fires.

Similar to alkanes and alkenes, aromatics, oxygenates, halogen-, and sulfur-containing compounds all had higher emissions per kilogram of fuel carbon when dung fuels and *angithi* stoves were utilized compared to brushwood fuels and *chulha* stoves, respectively. We focus on the behavior of the most interesting groups of compounds in the discussion below.

The chlorine-containing organic compounds are generally expected to come from cook fires in large quantities. However, we observed an interesting practice in which the cook often used plastic bags to start the fire, which could be a source of chlorine-containing compounds if composed of polyvinyl chloride. Carbonyl sulfide (OCS) is largely responsible for the yellow sulfur-containing fraction in Figure 2a and biomass burning is a well-known source of OCS in the atmosphere (Crutzen et al., 1979). Similar to other VOCs, OCS was significantly emitted in higher quantities when *angithi* stoves and dung fuels were utilized.

Benzene had higher emissions from *chulha* stoves, which had higher MCEs when cooking with dung fuels compared to *angithi* stoves (dung-*chulha* 3.18 g/ kg fuel C and dung-*angithi* 2.38 g/ kg fuel C). As the simplest aromatic compound, benzene also had the largest average difference in fuel type EFs compared to other aromatics (2.18 g/ kg fuel C, dung-wood). This information is relevant for exposure assessment, as benzene is a known human carcinogen. Higher benzene emissions from *chulha* cook fires could lead to higher benzene exposures which is a potential public health concern. However, it should be noted that the cook usually cannot control the stove used, as the *angithi* and *chulha* are used to prepare different types of meals, and exposure to benzene is not straightforward from its emission factors.

Higher emissions of alkynes were observed from dung fuels and *chulha* cookstoves. The latter observation is consistent with the literature showing flaming combustion generates more alkynes (Barrefors and Petersson, 1995; Lee et al., 2005). *Chulha* cook fires always had higher MCE than *angithi* cook fires (Table 1) which rely on smoldering combustion. Approximately the same difference in alkyne emissions results from comparing the *chulha* to the *angithi* using dung, in relation to using wood versus dung in combination with the *chulha*. There were two exceptions in stove type for 1-butane ($p= 0.055$) and 2-butane ($p>> 0.05$). The former may or may not have a relationship with stove type, while the latter does not. Emissions of some compounds did not show a relationship with either fuel or stove type, and are listed in Table S2.

VOC emissions from Stockwell et al. (2016) are also provided in Table 1 for comparison of VOC EFs. Samples in Stockwell et al. (2016) were collected in April 2015 in and around Kathmandu and the Tarai plains, which border India. While both are EFs from cookstoves using similar fuels, there are differences in the studies that should be noted. Stockwell et al. (2016) collected measurements of simulated cooking in a laboratory and from cooking fires in households; it was not noted in the latter case what meals were cooked. EFs were calculated from similar WAS measurements, but as grab samples in an area of the kitchen away from the fire (as opposed to the time-integrated approach used here). Emissions were assumed to be well-mixed in the kitchen

prior to sampling. Stockwell et al. (2016) also used a range of stoves, including the traditional single-pot mud stove, open three-stone fire, *bhuse chulo*, rocket, chimney, and forced draft stoves. “Dung” cook fires sometimes used a combination of fuels, such as wood. Finally, our study also has a larger sample size than Stockwell et al. (2016) with $n=49$ versus $n\approx 10$.

5 The emission factors for most compounds determined in this study were lower compared to those reported in Table 4 of the Stockwell et al. (2016) paper. Figure S2.1 visually shows that the EFs were generally lower in the present study. In some cases, EFs in this study were an order of magnitude lower, most notably n-pentane and n-hexane. We also found that our emission factors were always higher for dung-*chulha* compared to brushwood-*chulha*, which was not always the case in Stockwell et al. (2016). The EFs in Stockwell et al. (2016) could be biased high due to calculations rather than real differences in emissions. For example, ignoring ash and char carbon and using the same carbon content inflates the EFs reported in our paper by 7% for dung and 24% for brushwood emissions. However, this is a small percentage compared to the observed differences between EFs between the two measurements. By examining the EFs reported in the supporting information section of the Stockwell et al. (2016), we found that the disagreement resulted from the way the final recommended EFs were obtained from the measurements. Because Stockwell et al. (2016) measured both laboratory and field cook fires, they elected to adjust the laboratory EFs to account for the lower MCEs observed in the field. It has been shown by Roden et al. (2009) and Johnson et al. (2008) that cooking activities can strongly influence emissions, for example due to the cook tending to the cook fire differently, thus affecting combustion conditions. However, such adjustments should be done with caution because EF and MCE do not always follow a linear trend, as explained in the next section. Figure S2.2 plots the unadjusted laboratory and field emission factors as a function of MCE for all the measurements in Stockwell et al. (2016), as well as this study. Plotting the unadjusted EFs resolves most of the differences in observed EFs between the two studies. The data show an encouraging level agreement despite the differences in the experimental design between the two studies.

3.2 Modified combustion efficiency

The use of dung and *angithi*, rather than brushwood and *chulha*, respectively, results in lower modified combustion efficiencies as shown in Figure 3. In general, at lower MCEs we measured higher emissions of gas-phase compounds as discussed in Section 3.1. For example, emissions of ethane (Figure 3a) and other alkanes increase with decreasing MCE. However the dependence of ethane EF on MCE is not linear as observed in previous studies (Liu et al., 2017; Selimovic et al., 2018). For other VOCs, the dependence of the EF on MCE deviates from the linear trend even more, with the maximum EF observed at intermediate MCE values. For example, the ethene EF (Figure 3b) increases with decreasing MCE at MCEs > 0.85 , but it has the opposite trend at MCEs < 0.85 . Previously, we discussed that there is no relationship between ethene EF and stove type and we see this more clearly in Figure 3b. Alkynes have the same relationship to MCE as ethene, but it is even more pronounced (Figure 3c). Benzene (Figure 3e) stands apart from other aromatics with a relationship with MCE similar to ethene, while other aromatics have an EF versus MCE curve similar to alkanes and most other VOCs. In Figure 3e, we see again that emissions from brushwood-*chulha* and dung-*angithi* cook fires result in lower emissions of benzene compared to dung-*chulha*. Alkenes with two double bonds generally have a negative correlation between emissions and MCE, such as 1,3-butadiene in Figure 3f. The 1,3-butadiene EF versus MCE plot is not necessarily representative of all analogous plots for alkenes with two double bonds, as they have different shapes. 1,3-butadiene was chosen as its emission is high compared to other compounds in its subcategory and it also has health implications. It also happens to have a more linear relationship with MCE, albeit noisy.

It is of interest to compare EFs obtained from different fuel-stove combinations but with the same MCE. In the case of ethane, different cook fire types yield vastly different EFs at the same MCE. For example, at $MCE\approx 0.87$, mixed-*chulha* has an EF of

roughly 1.5 g/ kg fuel C, dung-*chulha* is 2.5 g/ kg fuel C, and dung-*angithi* is 5.5 g/ kg fuel C. Knowledge of the cook fire MCE alone is not sufficient to determine the EF of ethane. **Combustion conditions specific to the fuel-stove combination is a significant factor in determining cook fire emissions.** A similar conclusion can be reached for most of the measured gases, including non-ethene alkenes in Figure 3d.

5 3.3 Secondary pollutant formation and reactivity

OH reactivity and ozone forming potential

Total OH reactivity (s^{-1}) based on the measured VOCs in Figure 2b is given per ppbv of CO. Predicted OH reactivities in the village due to a single cooking event are 10.2, 6.73, 4.93, and 4.83 s^{-1} for emissions from dung-*angithi*, dung-*chulha*, mixed-*chulha*, and brushwood-*chulha* cook fires, respectively. This assumes a CO mixing ratio of 338 ppbv, which we measured as the
10 average background mixing ratio over the whole campaign. The relative total OH reactivity is over twice as high for dung-*angithi* cook fires as it is for brushwood-*chulha* cook fires.

The classes of compounds that act as the most important OH radical sinks in descending order are alkenes, oxygenates, furans, terpenes, and aromatics. Alkenes make up more than 50% of OH reactivity for all cook fire types. Ethene (by fuel type) and propene (by fuel and stove combination) are mostly responsible for the differences in fuel-stove combination results for alkenes.
15 For oxygenates, methanol ($p < 0.01$) and acrolein ($p < 0.05$) have significantly higher OH reactivity with wood fuel, while acetaldehyde has significantly higher OH reactivity with the *angithi* stove ($p < 0.001$). Differences in OH reactivity due to furans were observed for stove type but not fuel type. All three of the measured furans significantly contribute ($p < 0.001$) to a 6% increase in the fraction of OH reactivity due to furans for dung-*angithi* (12%) as opposed to dung-*chulha* (6%). The percentage of OH reactivity due to aromatics is constant at ~4% for the fuel-stove combinations. However, different aromatic compounds
20 are responsible for this ~4% contribution depending on the cook fire type. Benzene dominates OH reactivity due to aromatics for *chulha* cook fires. For *angithi* cook fires aromatics other than benzene, in particular toluene, dictate the OH reactivity for aromatics. Isoprene is solely responsible for the differences in OH reactivity due to terpenes.

Figure 2d shows the total ozone forming potential (g/ O₃ kg fuel) in the MIR scenario, as well as contributions to OFP by compound class. A critical step in photochemical ozone production is VOC reacting with OH. Therefore, the ozone forming
25 potential contributions by compound class are similar to those for OH reactivity (Figure 2c). Total OFP is nearly a factor of 3 higher for dung-*chulha* compared to brushwood-*chulha*, while it's twice as high for dung-*angithi* as compared to dung-*chulha*.

SOA formation potential

SOAP-weighted emissions relate SOA production from the different cook fire types in a qualitative manner. The contribution of each compound class to the total SOA formation potential is shown in Figure 2c. **We emphasize that the SOAP-weighted
30 emissions are reflective of only the measured VOCs, and there are likely semi-volatile and intermediate volatility compounds that are not measured but also contribute to SOA formation.** The sum of the contributions by each compound class, or the total SOA forming potential, is also shown below the pie charts. Dung fuels and *angithi* stoves yield larger amounts of SOA. However, fuel type is more important than stove type in terms of SOA formation. SOAP-weighted emissions are a factor of three higher for dung-*chulha* compared to brushwood-*chulha*. We discussed previously that benzene emissions are significantly higher
35 from *chulha* cook fires compared to *angithi* cook fires. These higher benzene emissions directly impact public health and also dictate SOA formation for *chulha* emissions. Benzene emissions are responsible for at least half of the SOA formation from the

chulha cook fire VOCs we measured. Beyond benzene, aromatics make up on average roughly 95% of SOA precursors measured in this study for all cook fires. While benzene is prominent for *chulha* cook fires, C₈-C₉ aromatics, toluene, and benzene contribute in approximately equal proportions to SOA formation in dung-*angithi* smoke plumes.

4 Atmospheric implications and conclusions

5 The extent of ozone formation hinges on the villages' overall NO_x levels as well as VOC emissions. However, in a VOC-limited regime, with each household in this village cooking three meals a day using the *chulha* and mixed fuels (brushwood + dung), 3.3 x 10⁵ g ozone per day is expected to be produced due solely to cookstove use. This was estimated based off the Census of India 2011 data for the village of Khatela and assuming the same fuel consumption as that used in this study. Over the lunch hour, when solar radiation is most intense, 30 ppbv of excess ozone is predicted. For this calculation we assumed a calm wind speed of
10 0.5 m/s, and confined our analysis to the village of Khatela with a boundary layer height of 1 km and village length of 1 km. In a similar way we calculated the amount of ozone that could be generated from cooking animal fodder. We assumed that every household prepares animal fodder every three days in addition to the assumptions already discussed, resulting in an additional 7.9 x 10⁴ grams of O₃ produced per day from cooking. If we assume every household in the village prepares animal fodder in the same hour, excess ozone levels of 7 ppbv are predicted, using the same assumptions described earlier for the lunch hour. We
15 should note that these estimations are approximate and a regional air quality model with detailed household level inputs should be used to more precisely predict the impact of cook fire emissions on ozone levels.

Using dung patties as opposed to brushwood has a large impact on local PM_{2.5} and ozone levels. Measured PM_{2.5} concentrations were more than a factor of two higher for dung-*chulha* compared to brushwood-*chulha* in grams emitted per kilogram of fuel carbon burned. In addition to this, the total SOA forming potential is three times higher for dung-*chulha* than that of brushwood-
20 *chulha*. We also estimated that dung-*chulha* cook fires produce roughly 3 times more ozone in the MIR regime than brushwood-*chulha* cook fires (163 g O₃ kg fuel C versus 56.9 g O₃ kg fuel C). However, compounds such as benzene are emitted in higher quantities from the *chulha* (1.03 g kg⁻¹ dry fuel) versus *angithi* (0.373 g kg⁻¹ dry fuel), and this public health concern should be investigated in more detail.

Acknowledgements

25 We thank the village of Khatela and our cook for welcoming us and for participating in the study. We also want to acknowledge Sneha Gautam's role in supporting the field work. This research was supported by EPA STAR grant R835425 Impacts of household sources on outdoor pollution at village and regional scales in India. The contents are solely the responsibility of the authors and do not necessarily represent the official views of the US EPA. The US EPA does not endorse the purchase of any commercial products or services mentioned in the publication.

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Figure 1. Sampling train for collecting cookstove emissions. PCXR8 (blue) are sampling pumps, WAS or whole air samples (green) are the air samplers, and orange boxes are Teflon or quartz filters used to collect PM_{2.5}.

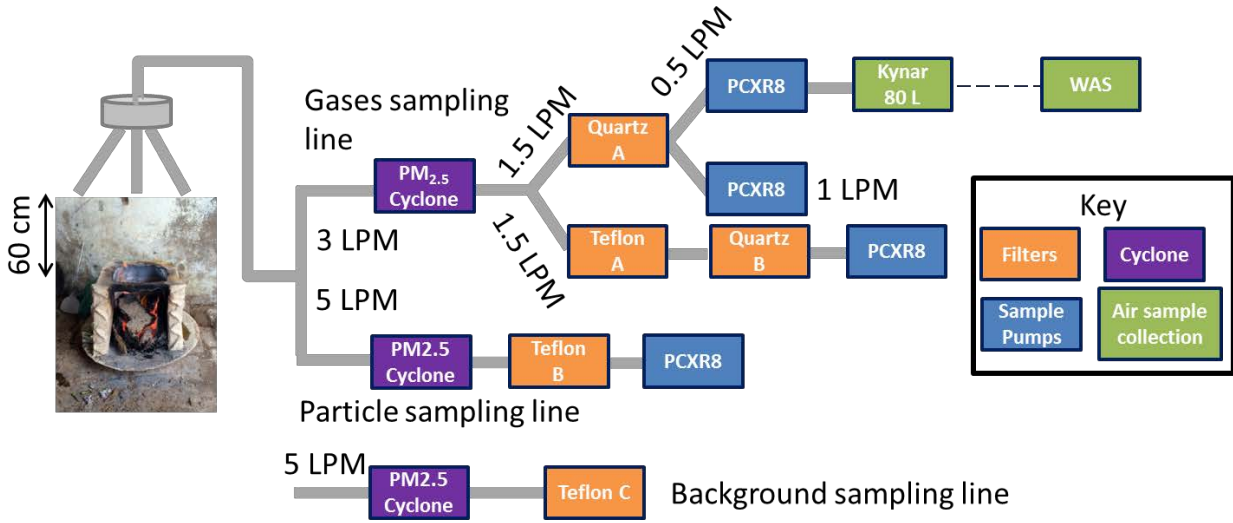
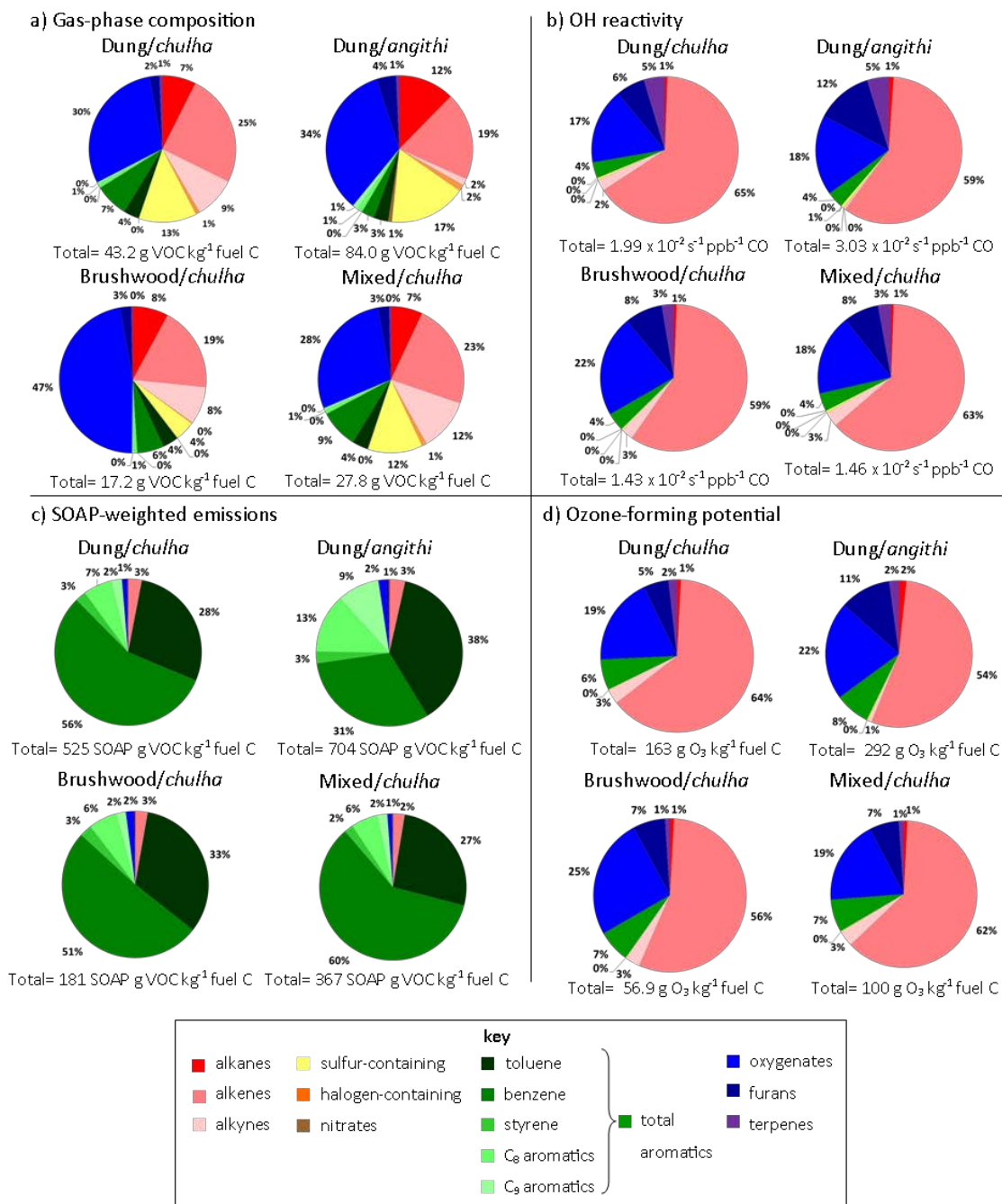
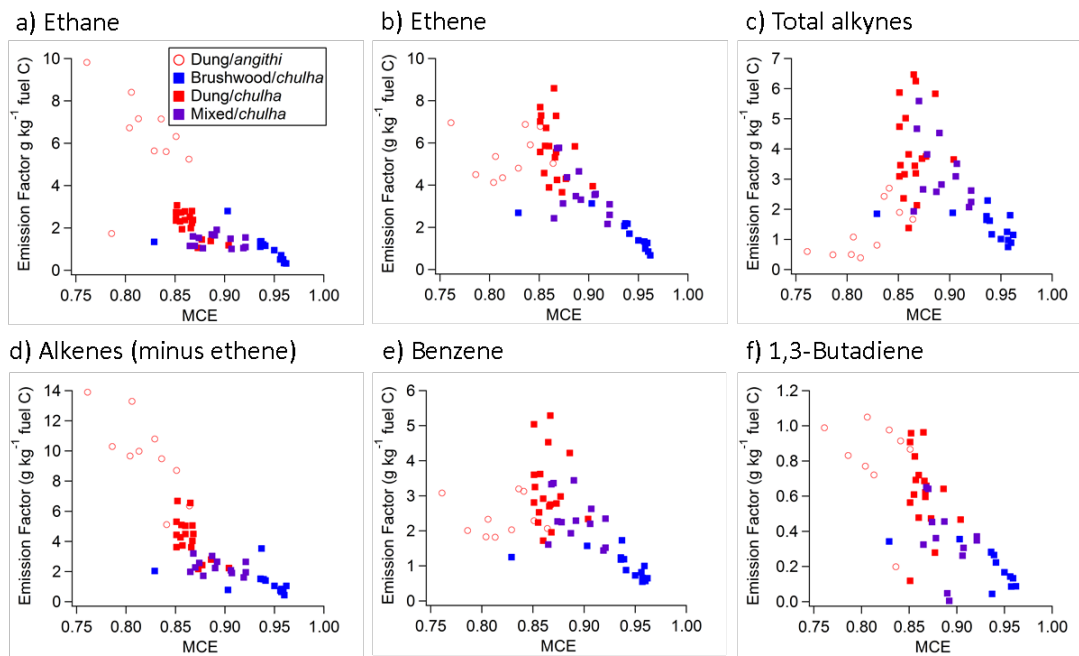


Figure 2. Pie charts showing the contribution of each species class to gas-phase composition (a), OH reactivity (b), SOAP-weighted emissions (c), and ozone-forming potential (d). For (b) and (d), total aromatics are shown rather than the breakdown of aromatics shown in (a) and (c). Sums of all components are shown below the pie chart.



^a1-Buten-3-yne is grouped in with alkynes

Figure 3. Emission factors as a function of modified combustion efficiency (MCE) for select species. Open circles indicate cooking events conducted with *angithi* stoves, whereas filled squares indicate *chulha* stoves. Color indicates fuel, either brushwood (blue), dung (red), or mixed (purple).



^a1-Buten-3-yne is grouped in with alkynes

Table 1. Averaged emission factors and standard deviation of PM_{2.5} and gas-phase species (g kg⁻¹ dry fuel) for dung-*chulha*, brushwood-*chulha*, mixed-*chulha*, and dung-*angithi* cook fires. Previously published emission factors (g kg⁻¹ dry fuel) from dung and hardwood cook fires are shown for comparison (Stockwell et al. 2016). Sample sizes for the current study (n) were n=18 for dung-*chulha*, n=14 for brushwood-*chulha*, n=13 for mixed-*chulha*, and n=10 for dung-*angithi*.

Compound (formula)	Dung- <i>chulha</i> Average (SD)	Brushwood- <i>chulha</i> Average (SD)	Mixed- <i>chulha</i> Average (SD)	Dung- <i>angithi</i> Average (SD)	Stockwell et al. (2016) Dung Average (SD)	Stockwell et al. (2016) Hardwood Average (SD)
Modified Combustion Efficiency	0.865 (0.014)	0.937 (0.035)	0.892 (0.021)	0.819 (0.031)	0.898	0.923
PM _{2.5}	19.2 (7.1)	7.42 (5.67)	11.0 (2.0)	33.2 (7.6)	14.73 (0.33)*	7.97 (3.80)*
Carbon dioxide (CO ₂)	984 (23)	1242 (61)	969 (31)	888 (48)	1129 (80)	1462 (16)
Carbon monoxide (CO)	97.7 (9.5)	53.0 (30.1)	74.8 (16.0)	125 (20)	80.9 (13.8)	77.2 (13.5)
Methane (CH ₄)	6.92 (1.23)	4.80 (2.09)	4.84 (0.89)	15.1 (2.6)	6.65 (0.46)	5.16 (1.39)
Sulfur-containing						
Carbonyl sulfide (OCS)	0.124 (0.040)	1.44 (0.54) x 10 ⁻²	8.50 (2.42) x 10 ⁻²	0.352 (0.217)	0.148 (0.123)	1.87 (1.15) x 10 ⁻²
DMS (C ₂ H ₆ S)	9.69 (4.54) x 10 ⁻³	1.39 (1.34) x 10 ⁻³	4.81 (2.26) x 10 ⁻³	4.34 (3.11) x 10 ⁻²	2.37 (0.08) x 10 ⁻²	0.255 (0.359)
Halogen-containing						
Dichloromethane (CH ₂ Cl ₂)	4.46 (3.94) x 10 ⁻⁴	2.18 (3.13) x 10 ⁻⁴	4.04 (6.44) x 10 ⁻⁴	4.56 (2.73) x 10 ⁻⁴	nm	nm
Chloromethane (CH ₃ Cl)	1.78 (0.70)	0.280 (0.157)	1.02 (0.42)	4.58 (1.89)	1.60 (1.53)	2.36 (1.62) x 10 ⁻²
Bromomethane (CH ₃ Br)	6.57 (2.78) x 10 ⁻³	7.92 (2.13) x 10 ⁻⁴	4.35 (1.81) x 10 ⁻³	1.43 (0.57) x 10 ⁻²	5.34 (3.02) x 10 ⁻³	5.61 (3.01) x 10 ⁻⁴
Iodomethane (CH ₃ I)	6.10 (4.78) x 10 ⁻⁴	9.62 (2.31) x 10 ⁻⁵	2.41 (0.66) x 10 ⁻⁴	8.83 (1.62) x 10 ⁻⁴	4.39 (1.78) x 10 ⁻⁴	1.23 (1.11) x 10 ⁻⁴
Ethyl chloride (C ₂ H ₅ Cl)	2.54 (1.17) x 10 ⁻³	4.22 (3.72) x 10 ⁻⁴	1.59 (0.67) x 10 ⁻³	9.11 (3.50) x 10 ⁻³	nm	nm
Dichloroethane (C ₂ H ₄ Cl ₂)	8.80 (2.98) x 10 ⁻⁴	2.55 (2.17) x 10 ⁻⁴	1.21 (2.32) x 10 ⁻³	1.47 (0.91) x 10 ⁻³	4.97 x 10 ⁻³ (-)	1.24 (0.30) x 10 ⁻⁴
Nitrates						
Methyl nitrate (CH ₃ ONO ₂)	1.83 (5.18) x 10 ⁻³	5.34 (14.4) x 10 ⁻³	6.60 (11.7) x 10 ⁻³	0.170 (0.339)	1.46 (1.94) x 10 ⁻²	6.96 (5.73) x 10 ⁻³
Ethyl nitrate (CH ₃ ONO ₂)	2.37 (3.86) x 10 ⁻⁴	5.54 (10.2) x 10 ⁻⁴	2.27 (6.40) x 10 ⁻³	4.53 (11.6) x 10 ⁻²	nm	nm
i-Propylnitrate (C ₃ H ₇ ONO ₂)	1.90 (1.61) x 10 ⁻⁴	2.40 (4.92) x 10 ⁻⁴	4.10 (8.38) x 10 ⁻⁴	5.90 (12.1) x 10 ⁻³	nm	nm

n-Propylnitrate (C ₃ H ₇ ONO ₂)	6.32 (5.23) x 10 ⁻⁵	9.01 (14.1) x 10 ⁻⁵	1.44 (3.25) x 10 ⁻⁴	1.82 (4.35) x 10 ⁻³	nm	nm
2-Butylnitrate (C ₄ H ₉ ONO ₂)	2.69 (2.14) x 10 ⁻⁴	1.05 (1.13) x 10 ⁻⁴	7.10 (20.3) x 10 ⁻⁴	2.45 (4.09) x 10 ⁻³	nm	nm
3-Pentylnitrate (C ₅ H ₁₁ ONO ₂)	4.75 (1.61) x 10 ⁻⁵	2.29 (2.08) x 10 ⁻⁵	3.13 (1.97) x 10 ⁻⁵	1.94 (4.06) x 10 ⁻⁴	nm	nm
2-Pentylnitrate (C ₅ H ₁₁ ONO ₂)	2.37 (2.10) x 10 ⁻⁵	1.63 (2.46) x 10 ⁻⁵	1.25 (1.26) x 10 ⁻⁵	1.82 (4.54) x 10 ⁻⁴	nm	nm
Alkanes						
Ethane (C ₂ H ₆)	0.717 (0.193)	0.380 (0.247)	0.422 (0.096)	2.06 (0.69)	1.08 (0.30)	0.160 (0.122)
Propane (C ₃ H ₈)	0.211 (0.073)	9.48 (8.41) x 10 ⁻²	0.116 (0.032)	0.819 (0.157)	0.457 (0.137)	0.202 (0.140)
i-Butane (C ₄ H ₁₀)	1.73 (0.71) x 10 ⁻²	4.60 (4.86) x 10 ⁻³	9.51 (2.75) x 10 ⁻³	7.27 (1.54) x 10 ⁻²	0.215 (0.126)	0.406 (0.478)
n-Butane (C ₄ H ₁₀)	4.71 (1.88) x 10 ⁻²	1.57 (1.67) x 10 ⁻²	2.68 (0.88) x 10 ⁻²	0.215 (0.047)	0.29 (0.09)	1.11 (1.48)
n-Pentane (C ₅ H ₁₂)	2.01 (0.98) x 10 ⁻²	4.44 (4.08) x 10 ⁻³	9.12 (3.71) x 10 ⁻³	6.80 (2.95) x 10 ⁻²	0.190 (0.254)	2.18 (1.73) x 10 ⁻²
n-Hexane (C ₆ H ₁₄)	1.03 (0.47) x 10 ⁻²	1.96 (1.58) x 10 ⁻³	5.31 (1.87) x 10 ⁻³	4.93 (1.10) x 10 ⁻²	0.291 (0.248)	1.85 x 10 ⁻² (-)
n-Heptane (C ₇ H ₁₆)	7.21 (3.43) x 10 ⁻³	9.23 (6.94) x 10 ⁻⁴	3.92 (1.23) x 10 ⁻³	3.17 (0.85) x 10 ⁻²	0.114 (0.069)	1.01 (1.35) x 10 ⁻²
2-Methylpentane (C ₆ H ₁₄)	6.21 (2.81) x 10 ⁻³	1.23 (0.99) x 10 ⁻³	2.57 (1.61) x 10 ⁻³	2.29 (1.67) x 10 ⁻²	0.231 (0.192)	9.93 (12.9) x 10 ⁻³
3-Methylpentane (C ₆ H ₁₄)	3.71 (1.70) x 10 ⁻³	1.21 (1.01) x 10 ⁻³	1.57 (0.76) x 10 ⁻³	7.54 (4.30) x 10 ⁻³	0.155 (0.137)	6.79 (6.63) x 10 ⁻³
Alkenes						
Ethene (C ₂ H ₄)	1.86 (0.48)	0.626 (0.284)	1.13 (0.38)	1.77 (0.35)	4.23 (1.39)	2.70 (1.17)
Propene (C ₃ H ₆)	0.807 (0.235)	0.286 (0.202)	0.417 (0.091)	1.61 (0.33)	1.47 (0.58)	0.576 (0.195)
1-Butene (C ₄ H ₈)	0.158 (0.047)	6.32 (4.59) x 10 ⁻²	8.38 (1.83) x 10 ⁻²	0.366 (0.096)	0.399 (0.331)	0.726 (0.904)
i-Butene (C ₄ H ₈)	0.133 (0.057)	3.46 (2.50) x 10 ⁻²	6.40 (1.86) x 10 ⁻²	0.353 (0.158)	0.281 (0.091)	0.846 (1.113)
trans-2-Butene (C ₄ H ₈)	4.45 (1.60) x 10 ⁻²	2.00 (1.27) x 10 ⁻²	2.38 (0.70) x 10 ⁻²	0.151 (0.055)	0.151 (0.010)	6.78 (5.98) x 10 ⁻²
cis-2-Butene (C ₄ H ₈)	3.38 (1.19) x 10 ⁻²	1.51 (0.95) x 10 ⁻²	1.80 (0.52) x 10 ⁻²	0.107 (0.047)	0.102 (0.016)	5.51 (4.76) x 10 ⁻²
3-Methyl-1- butene (C ₅ H ₁₀)	1.46 (0.48) x 10 ⁻²	5.74 (4.49) x 10 ⁻³	7.30 (1.94) x 10 ⁻³	3.82 (0.88) x 10 ⁻²	5.58 (3.50) x 10 ⁻²	7.43 (5.79) x 10 ⁻³
2-Methyl-1- butene (C ₅ H ₁₀)	2.71 (1.28) x 10 ⁻²	9.96 (10.9) x 10 ⁻³	1.19 (0.42) x 10 ⁻²	7.70 (3.99) x 10 ⁻²	nm	nm
2-Methyl-2- butene (C ₅ H ₁₀)	2.51 (1.26) x 10 ⁻²	6.40 (4.78) x 10 ⁻³	1.10 (0.47) x 10 ⁻²	9.17 (4.70) x 10 ⁻²	nm	nm
1-Pentene (C ₅ H ₁₀)	4.17 (1.59) x 10 ⁻²	9.65 (6.55) x 10 ⁻³	2.13 (0.60) x 10 ⁻²	0.122 (0.033)	0.168 (0.086)	1.43 (0.94) x 10 ⁻²

trans-2-Pentene (C ₅ H ₁₀)	1.74 (0.65) x 10 ⁻²	8.89 (5.77) x 10 ⁻³	8.69 (2.22) x 10 ⁻³	5.14 (2.70) x 10 ⁻²	0.115 (0.035)	1.05 (0.83) x 10 ⁻²
cis-2-Pentene (C ₅ H ₁₀)	1.00 (0.36) x 10 ⁻²	5.55 (3.62) x 10 ⁻³	4.98 (1.26) x 10 ⁻³	2.50 (1.28) x 10 ⁻²	5.14 (0.76) x 10 ⁻²	8.69 x 10 ⁻³ (-)
1-Hexene (C ₆ H ₁₂)	6.10 (2.46) x 10 ⁻²	1.26 (0.73) x 10 ⁻²	3.09 (0.91) x 10 ⁻²	0.167 (0.050)	nm	nm
1,2-Propadiene (C ₃ H ₄)	3.76 (1.69) x 10 ⁻²	1.31 (0.62) x 10 ⁻²	2.32 (0.86) x 10 ⁻²	1.80 (0.923) x 10 ⁻²	7.15 (6.76) x 10 ⁻²	2.33 (1.07) x 10 ⁻²
1,2-Butadiene (C ₄ H ₆)	5.54 (1.68) x 10 ⁻³	2.82 (1.81) x 10 ⁻³	3.10 (1.06) x 10 ⁻³	4.33 (1.59) x 10 ⁻³	nm	nm
1,3-Butadiene (C ₄ H ₆)	0.203 (0.071)	7.44 (3.99) x 10 ⁻²	0.108 (0.061)	0.263 (0.082)	0.409 (0.306)	0.204 (0.144)
Isoprene (C ₅ H ₈)	8.94 (5.80) x 10 ⁻²	1.98 (1.48) x 10 ⁻²	3.03 (2.39) x 10 ⁻²	0.188 (0.143)	0.325 (0.443)	4.16 (2.23) x 10 ⁻²
1,3-Pentadiene (C ₅ H ₈)	1.96 (1.05) x 10 ⁻²	9.17 (4.79) x 10 ⁻³	9.39 (6.43) x 10 ⁻³	5.66 (2.94) x 10 ⁻²	nm	nm
Alkynes						
Ethyne	1.13 (0.42)	0.467 (0.160)	0.890 (0.323)	0.325 (0.238)	0.593 (0.443)	0.764 (0.363)
1-Propyne	9.42 (3.46) x 10 ⁻²	3.82 (1.76) x 10 ⁻²	5.99 (2.22) x 10 ⁻²	5.20 (2.83) x 10 ⁻²	nm	nm
1-Buten-3-yne (C ₄ H ₄)	5.04 (1.72) x 10 ⁻²	1.86 (0.90) x 10 ⁻²	3.46 (1.53) x 10 ⁻²	1.74 (1.26) x 10 ⁻²	nm	nm
1-Butyne (C ₄ H ₆)	7.72 (2.29) x 10 ⁻³	4.07 (2.24) x 10 ⁻³	4.48 (1.41) x 10 ⁻³	5.97 (1.93) x 10 ⁻³	2.29 (1.38) x 10 ⁻²	1.28 (0.47) x 10 ⁻²
2-Butyne (C ₄ H ₆)	4.31 (1.15) x 10 ⁻³	2.55 (1.44) x 10 ⁻³	2.47 (0.70) x 10 ⁻³	4.52 (1.40) x 10 ⁻³	1.86 (0.91) x 10 ⁻²	1.02 (0.66) x 10 ⁻²
1,3-Butadiyne (C ₄ H ₂)	6.07 (2.66) x 10 ⁻³	2.71 (1.21) x 10 ⁻³	5.43 (2.01) x 10 ⁻³	1.53 (1.31) x 10 ⁻³	nm	nm
Aromatics						
Benzene (C ₆ H ₆)	1.03 (0.33)	0.373 (0.149)	0.723 (0.218)	0.769 (0.175)	1.96 (0.45)	1.05 (0.19)
Toluene (C ₇ H ₈)	0.483 (0.273)	0.221 (0.085)	0.297 (0.077)	0.860 (0.167)	1.26 (0.05)	0.241 (0.160)
Ethylbenzene (C ₈ H ₁₀)	3.41 (0.791) x 10 ⁻²	1.25 (1.20) x 10 ⁻²	1.97 (0.40) x 10 ⁻²	9.78 (1.66) x 10 ⁻²	0.366 (0.085)	4.19 (4.25) x 10 ⁻²
m/p-Xylene (C ₈ H ₁₀)	6.36 (1.26) x 10 ⁻²	2.78 (1.56) x 10 ⁻²	4.03 (0.98) x 10 ⁻²	0.148 (0.030)	0.601 (0.294)	9.57 (7.99) x 10 ⁻²
o-Xylene (C ₈ H ₁₀)	2.38 (0.76) x 10 ⁻²	8.37 (5.78) x 10 ⁻³	1.44 (0.41) x 10 ⁻²	7.96 (1.91) x 10 ⁻²	0.228 (0.083)	3.93 (4.31) x 10 ⁻²
Styrene (C ₈ H ₈)	5.88 (1.58) x 10 ⁻²	2.28 (1.50) x 10 ⁻²	3.40 (1.90) x 10 ⁻²	8.63 (5.96) x 10 ⁻²	0.255 (0.091)	8.71 (6.69) x 10 ⁻²
i-Propylbenzene (C ₉ H ₁₂)	2.91 (0.77) x 10 ⁻³	1.20 (1.11) x 10 ⁻³	1.69 (0.45) x 10 ⁻³	9.30 (4.90) x 10 ⁻³	1.87 (1.40) x 10 ⁻²	1.70 (1.67) x 10 ⁻²
n-Propylbenzene (C ₉ H ₁₂)	6.48 (2.59) x 10 ⁻³	1.84 (1.65) x 10 ⁻³	4.02 (1.59) x 10 ⁻³	3.95 (2.69) x 10 ⁻²	3.10 (1.45) x 10 ⁻²	1.78 (1.58) x 10 ⁻²
3-Ethyltoluene (C ₉ H ₁₂)	1.44 (0.48) x 10 ⁻²	5.46 (4.40) x 10 ⁻³	8.59 (3.26) x 10 ⁻³	7.14 (4.13) x 10 ⁻²	5.61 (2.38) x 10 ⁻²	2.62 (0.54) x 10 ⁻²

4-Ethyltoluene (C ₉ H ₁₂)	6.35 (2.36) x 10 ⁻³	2.54 (1.81) x 10 ⁻³	4.18 (1.96) x 10 ⁻³	3.71 (2.30) x 10 ⁻²	3.57 (1.74) x 10 ⁻²	2.07 (1.19) x 10 ⁻²
2-Ethyltoluene (C ₉ H ₁₂)	6.89 (2.50) x 10 ⁻³	2.70 (1.68) x 10 ⁻³	4.63 (2.07) x 10 ⁻³	3.76 (2.69) x 10 ⁻²	3.39 (1.34) x 10 ⁻²	2.10 (1.16) x 10 ⁻²
1,3,5- Trimethylbenzene (C ₉ H ₁₂)	3.87 (1.71) x 10 ⁻³	1.63 (1.22) x 10 ⁻³	2.65 (1.43) x 10 ⁻³	2.23 (1.60) x 10 ⁻²	1.79 (0.83) x 10 ⁻²	2.14 x 10 ⁻² (-)
1,2,4- Trimethylbenzene (C ₉ H ₁₂)	1.04 (0.46) x 10 ⁻²	4.25 (2.69) x 10 ⁻³	7.52 (4.28) x 10 ⁻³	6.23 (5.18) x 10 ⁻²	3.91 (1.65) x 10 ⁻²	1.74 (2.35) x 10 ⁻²
1,2,3- Trimethylbenzene (C ₉ H ₁₂)	4.76 (2.59) x 10 ⁻³	1.16 (0.81) x 10 ⁻³	3.84 (2.69) x 10 ⁻³	3.01 (3.16) x 10 ⁻²	2.34 (0.43) x 10 ⁻²	2.16 x 10 ⁻² (-)
Terpenes						
alpha-Pinene (C ₁₀ H ₁₆)	8.30 (5.40) x 10 ⁻⁴	5.38 (6.94) x 10 ⁻⁴	7.82 (6.32) x 10 ⁻⁴	2.26 (2.53) x 10 ⁻³	0.35 (0.49)	2.02 (2.33) x 10 ⁻²
beta-Pinene (C ₁₀ H ₁₆)	2.27 (1.49) x 10 ⁻³	1.37 (0.91) x 10 ⁻³	2.76 (3.15) x 10 ⁻³	2.89 (3.56) x 10 ⁻³	0.471 (-)	4.67 x 10 ⁻² (-)
Oxygenates						
Acetaldehyde (C ₂ H ₄ O)	0.805 (0.279)	0.334 (0.199)	0.447 (0.119)	1.70 (0.75)	1.88 (1.63)	0.541 (0.362)
Butanal (C ₄ H ₈ O)	4.28 (1.50) x 10 ⁻²	1.90 (1.29) x 10 ⁻²	2.68 (1.05) x 10 ⁻²	0.108 (0.047)	5.40 (2.19) x 10 ⁻²	8.28 (6.27) x 10 ⁻³
Acetone (C ₃ H ₆ O)	0.705 (0.219)	0.365 (0.226)	0.416 (0.108)	2.05 (0.52)	1.63 (0.38)	0.524 (0.256)
2-Butanone (C ₄ H ₈ O)	0.172 (0.057)	8.00 (6.18) x 10 ⁻²	0.103 (0.038)	0.498 (0.151)	0.262 (0.109)	0.232 (0.286)
2-Propenal (C ₃ H ₄ O)	0.186 (0.060)	0.127 (0.069)	0.127 (0.059)	0.295 (0.245)	nm	nm
MVK (C ₄ H ₆ O)	0.129 (0.040)	6.59 (4.56) x 10 ⁻²	6.31 (2.76) x 10 ⁻²	0.280 (0.147)	nm	nm
Furan (C ₄ H ₄ O)	0.109 (0.041)	5.98 (3.37) x 10 ⁻²	6.81 (2.19) x 10 ⁻²	0.379 (0.093)	0.534 (0.209)	0.241 (0.024)
2-Methylfuran (C ₅ H ₆ O)	0.117 (0.051)	5.92 (4.77) x 10 ⁻²	6.92 (2.83) x 10 ⁻²	0.488 (0.227)	nm	nm
Furfural (C ₅ H ₄ O ₂)	8.55 (6.05) x 10 ⁻²	4.28 (5.51) x 10 ⁻²	8.22 (5.09) x 10 ⁻²	0.316 (0.133)	nm	nm
Methanol (CH ₃ OH)	2.09 (1.14)	2.03 (2.01)	1.18 (0.40)	4.23 (3.40)	2.38 (0.90)	1.92 (0.61)
Ethanol (CH ₃ OH)	4.08 (5.93) x 10 ⁻²	2.18 (2.00) x 10 ⁻²	5.63 (6.69) x 10 ⁻²	7.62 (9.08) x 10 ⁻²	0.563 (0.589)	0.128 (0.017)

*From Jayarathne et al. (2017), but part of same NAMaSTE study

nm indicates the species was not measured

(-) from Stockwell et al. (2016) indicates that the measurement was not above background.

Table 2. Emission factors (g VOC/ kg fuel C) for select compounds. The mean differences between dung/*angithi* and dung/*chulha* are shown and similarly for dung/*chulha* and brushwood/*chulha*. The significance between fuel or stove and EF is indicated with asterisks. Accompanying the mean differences is the average emission factor (g VOC/ kg fuel C) for dung cook fires and *chulha* cook fires, as well as the overall average for all performed cook fires.

Compound	Average EF for all cook fires (g/ kg fuel C)	<i>Angithi-chulha</i> average EF difference (g/ kg fuel C)	Average EF for dung fires (g/ kg fuel C)	Dung-brushwood average EF difference (g/ kg fuel C)	Average EF for <i>chulha</i> cook fires (g/ kg fuel C)
Ethane	2.47 (2.16)	4.18 ^{***}	3.70 (2.43)	1.19 ^{***}	1.60 (0.744)
Propane	0.827 (0.866)	1.88 ^{***}	1.32 (0.976)	0.397 ^{***}	0.448 (0.256)
n-Butane	0.200 (0.236)	0.52 ^{***}	0.331 (0.271)	0.0568 ^{***}	0.097 (0.063)
Ethene	4.17 (2.02)	N/A	5.64 (1.32)	4.05 ^{***}	3.88 (2.07)
Propene	2.24 (1.61)	2.50 ^{***}	3.38 (1.48)	1.72 ^{***}	1.63 (0.93)
1-Butene	0.473 (0.373)	0.644 ^{***}	0.718 (0.377)	0.213 ^{***}	0.327 (0.180)
Ethyne	2.32 (1.41)	-2.46 ^{***}	2.58 (1.63)	2.21 ^{***}	2.61 (1.37)
1-Propyne	0.196 (0.108)	-0.129 ^{**}	0.244 (0.116)	0.187 ^{***}	0.204 (0.112)
1-Butyne	1.74 x 10 ⁻² (7.74 x 10 ⁻³)	-0.101 ^{***}	0.219 (0.007)	0.105 ^{***}	0.017 (0.008)

* denotes p<0.05, ** p<0.01, *** p<0.001

Supporting Information

Emissions from village cookstoves in Haryana, India and their potential impacts on air quality

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Table S1. Average emission factors and standard deviation of PM_{2.5} and gas-phase species (g kg⁻¹ dry fuel carbon) for dung-*chulha*, brushwood-*chulha*, mixed-*chulha*, and dung-*angithi* cook fires. Sample size (n) was n=12 for dung-*chulha*, n=14 for brushwood-*chulha*, n=13 for mixed-*chulha*, n=10 for dung-*angithi*. SOAP values, k_{OH}, and MIR values used to calculate predicted SOAP-weighted potentials, OH reactivity, and ozone-forming potential are included for the quantified species. SOAP values were taken from Derwent et al. (2010), k_{OH} were found in the NIST Chemical Kinetics Database, and MIR values are found in Carter et al. 1994.

Compound	Dung- <i>chulha</i> Average (SD)	Brushwood- <i>chulha</i> Average (SD)	Mixed- <i>chulha</i> Average (SD)	Dung- <i>angithi</i> Average (SD)	SOAP	k _{OH} (x 10 ¹²) (cm ³ molec ⁻¹ s ⁻¹)	MIR (g O ₃ / g VOC)
Carbon dioxide (CO ₂)	3028 (80)	3365 (148)	3171 (83)	2745 (144)	0 ^a	0	0
Carbon monoxide (CO)	301 (33)	143 (78)	243 (47)	385 (61)	0 ^a	0.15	0.056
Methane (CH ₄)	21.3 (4.0)	12.9 (5.4)	15.8 (2.7)	46.7 (8.2)	0 ^a	0.006	0.0144
Sulfur-containing							
Carbonyl sulfide (OCS)	0.382 (0.135)	3.89 (1.36) x 10 ⁻²	0.279 (0.083)	1.09 (0.67)	0 ^a	0.002	0
DMS (C ₂ H ₆ S)	2.98 (1.55) x 10 ⁻²	3.68 (3.36) x 10 ⁻³	1.57 (0.74) x 10 ⁻²	0.134 (0.096)	0 ^a	5.4	0
Halogen-containing							
Dichloromethane (CH ₂ Cl ₂)	1.37 (1.48) x 10 ⁻³	5.79 (8.21) x 10 ⁻⁴	1.35 (2.20) x 10 ⁻³	1.41 (0.85) x 10 ⁻³	0 ^a	0.123	0.041
Chloromethane (CH ₃ Cl)	5.48 (2.41)	0.751 (0.393)	3.34 (1.37)	14.2 (5.8)	0 ^a	0.369	0.038
Bromomethane (CH ₃ Br)	2.02 (0.96) x 10 ⁻²	2.14 (0.57) x 10 ⁻³	1.42 (0.59) x 10 ⁻²	4.42 (1.75) x 10 ⁻²	0 ^a	0.0288	0.0187
Iodomethane (CH ₃ I)	1.88 (0.40) x 10 ⁻³	2.61 (0.61) x 10 ⁻⁴	7.87 (2.08) x 10 ⁻⁴	2.73 (0.51) x 10 ⁻³	0 ^a	0.0723	-0.56
Ethyl chloride (C ₂ H ₅ Cl)	7.83 (3.93) x 10 ⁻³	1.13 (0.95) x 10 ⁻³	5.23 (2.25) x 10 ⁻³	2.82 (1.08) x 10 ⁻²	0 ^a	0.423	0.29
Dichloroethane (C ₂ H ₄ Cl ₂)	2.71 (1.00) x 10 ⁻³	6.86 (5.78) x 10 ⁻⁴	4.04 (7.92) x 10 ⁻³	4.55 (2.82) x 10 ⁻³	0 ^a	0.023	0.21
Nitrates							
Methyl nitrate (CH ₃ ONO ₂)	5.62 (19.5) x 10 ⁻³	1.47 (4.00) x 10 ⁻²	2.19 (3.95) x 10 ⁻²	0.527 (1.048)	0 ^a	0.023	0
Ethyl nitrate (CH ₃ ONO ₂)	7.27 (14.5) x 10 ⁻⁴	1.50 (2.80) x 10 ⁻³	7.63 (21.77) x 10 ⁻³	0.140 (0.357)	0 ^a	0.178	0
i-Propylnitrate (C ₃ H ₇ ONO ₂)	5.85 (5.66) x 10 ⁻⁴	6.57 (13.64) x 10 ⁻⁴	1.37 (2.86) x 10 ⁻³	1.83 (3.75) x 10 ⁻²	0 ^a	0.287	0
n-Propylnitrate (C ₃ H ₇ ONO ₂)	1.94 (1.82) x 10 ⁻⁴	2.43 (3.83) x 10 ⁻⁴	4.81 (11.05) x 10 ⁻⁴	5.63 (13.44) x 10 ⁻³	0 ^a	0.731	0
2-Butylnitrate (C ₄ H ₉ ONO ₂)	8.27 (7.41) x 10 ⁻⁴	2.82 (3.07) x 10 ⁻⁴	2.39 (6.92) x 10 ⁻³	7.57 (15.15) x 10 ⁻³	0 ^a	0.93	0
3-Pentylnitrate (C ₅ H ₁₁ ONO ₂)	1.46 (0.49) x 10 ⁻⁴	6.15 (5.54) x 10 ⁻⁵	1.02 (0.62) x 10 ⁻⁴	6.01 (12.55) x 10 ⁻⁴	0 ^a	1	0
2-Pentylnitrate (C ₅ H ₁₁ ONO ₂)	7.27 (4.52) x 10 ⁻⁵	4.41 (6.71) x 10 ⁻⁵	4.05 (4.04) x 10 ⁻⁵	5.61 (14.00) x 10 ⁻⁴	0 ^a	1	0
Alkanes							
Ethane (C ₂ H ₆)	2.21 (0.60)	1.02 (0.64)	1.38 (0.30)	6.38 (2.14)	0.1	0.244	0.28

Propane (C ₃ H ₈)	0.650 (0.219)	0.253 (0.211)	0.378 (0.106)	2.53 (0.49)	0	1.1	0.49
i-Butane (C ₄ H ₁₀)	5.31 (2.14) x 10 ⁻²	1.23 (1.22) x 10 ⁻²	3.12 (0.92) x 10 ⁻²	0.225 (0.048)	0	2.12	1.23
n-Butane (C ₄ H ₁₀)	0.145 (0.058)	4.20 (4.22) x 10 ⁻²	8.83 (3.06) x 10 ⁻²	0.665 (0.148)	0.3	2.4	1.15
n-Pentane (C ₅ H ₁₂)	6.18 (2.46) x 10 ⁻²	1.19 (1.03) x 10 ⁻²	2.99 (1.24) x 10 ⁻²	0.210 (0.091)	0.3	3.8	1.31
n-Hexane (C ₆ H ₁₄)	3.16 (1.30) x 10 ⁻²	5.25 (4.05) x 10 ⁻³	1.74 (0.63) x 10 ⁻²	0.153 (0.034)	0.1	5.2	1.24
n-Heptane (C ₇ H ₁₆)	2.22 (1.02) x 10 ⁻²	2.49 (1.86) x 10 ⁻³	1.29 (0.41) x 10 ⁻²	9.79 (2.63) x 10 ⁻²	0.1	6.8	1.07
2-Methylpentane (C ₆ H ₁₄)	1.91 (0.75) x 10 ⁻²	3.28 (2.51) x 10 ⁻³	8.44 (5.36) x 10 ⁻³	7.07 (5.18) x 10 ⁻²	0	5.2	1.5
3-Methylpentane (C ₆ H ₁₄)	1.14 (0.53) x 10 ⁻²	3.23 (2.64) x 10 ⁻³	5.12 (2.44) x 10 ⁻³	2.33 (1.33) x 10 ⁻²	0.2	5.2	1.8
Alkenes							
Ethene (C ₂ H ₄)	5.73 (1.36)	1.68 (0.73)	3.68 (1.15)	5.47 (1.09)	1.3	8.51	9
Propene (C ₃ H ₆)	2.48 (0.60)	0.765 (0.506)	1.36 (0.28)	4.98 (1.04)	1.6	30.1	11.66
1-Butene (C ₄ H ₈)	0.488 (0.140)	0.169 (0.115)	0.274 (0.058)	1.13 (0.30)	1.2	31	9.73
i-Butene (C ₄ H ₈)	0.411 (0.125)	9.25 (6.25) x 10 ⁻²	0.209 (0.060)	1.09 (0.49)	0.6	51	1.23
trans-2-Butene (C ₄ H ₈)	0.137 (0.047)	5.36 (3.30) x 10 ⁻²	7.79 (2.22) x 10 ⁻²	0.466 (0.170)	4.0	64	15.16
cis-2-Butene (C ₄ H ₈)	0.104 (0.037)	4.04 (2.45) x 10 ⁻²	5.88 (1.69) x 10 ⁻²	0.331 (0.145)	3.6	56	14.24
3-Methyl-1-butene (C ₅ H ₁₀)	4.50 (1.44) x 10 ⁻²	1.53 (1.13) x 10 ⁻²	2.39 (0.62) x 10 ⁻²	0.118 (0.027)	0.6	32	6.99
2-Methyl-1-butene (C ₅ H ₁₀)	8.33 (2.97) x 10 ⁻²	2.65 (2.72) x 10 ⁻²	3.92 (1.38) x 10 ⁻²	0.238 (0.124)	0.9	61	6.4
2-Methyl-2-butene (C ₅ H ₁₀)	7.72 (3.14) x 10 ⁻²	1.71 (1.24) x 10 ⁻²	3.61 (1.53) x 10 ⁻²	0.284 (0.146)	1.9	87	14.08
1-Pentene (C ₅ H ₁₀)	0.128 (0.048)	2.58 (1.64) x 10 ⁻²	6.99 (1.98) x 10 ⁻²	0.378 (0.102)	0	27.4	7.21
trans-2-Pentene (C ₅ H ₁₀)	5.34 (1.68) x 10 ⁻²	2.38 (1.49) x 10 ⁻²	7.79 (2.22) x 10 ⁻²	0.159 (0.064)	3.1	67	10.56
cis-2-Pentene (C ₅ H ₁₀)	3.08 (0.90) x 10 ⁻²	1.49 (0.94) x 10 ⁻²	5.88 (1.69) x 10 ⁻²	7.72 (3.98) x 10 ⁻²	3.1	65	10.38
1-Hexene (C ₆ H ₁₂)	0.188 (0.072)	3.38 (1.83) x 10 ⁻²	0.101 (0.030)	0.517 (0.157)	0	37	5.49
1,2-Propadiene (C ₃ H ₄)	0.116 (0.033)	3.51 (1.57) x 10 ⁻²	7.55 (2.72) x 10 ⁻²	5.56 (2.84) x 10 ⁻²	1.8 ^b	0.45	8.45
1,2-Butadiene (C ₄ H ₆)	1.70 (0.48) x 10 ⁻²	7.54 (4.69) x 10 ⁻³	1.01 (0.33) x 10 ⁻²	1.34 (0.49) x 10 ⁻²	1.8 ^b	27	9.35
1,3-Butadiene (C ₄ H ₆)	0.626 (0.221)	0.201 (0.105)	0.352 (0.195)	0.813 (0.254)	1.8	67	12.61
Isoprene (C ₅ H ₈)	0.275 (0.151)	5.31 (3.88) x 10 ⁻²	9.89 (7.79) x 10 ⁻²	0.582 (0.444)	1.9	100	10.61
1,3-Pentadiene (C ₅ H ₈)	6.02 (3.37) x 10 ⁻²	2.47 (1.26) x 10 ⁻²	3.08 (2.11) x 10 ⁻²	0.175 (0.091)	1.8 ^c	117	12.5
Alkynes							
Ethyne (C ₂ H ₂)	3.46 (1.21)	1.25 (0.39)	2.89 (1.00)	1.01 (0.74)	0.1	0.9	0.95
1-Propyne (C ₃ H ₄)	0.290 (0.084)	0.103 (0.045)	0.195 (0.070)	0.161 (0.087)	0 ^a	3.1	6.72

1-Buten-3-yne (C ₄ H ₄)	0.155 (0.048)	5.01 (2.27) x 10 ⁻²	0.113 (0.048)	5.37 (3.87) x 10 ⁻²	0 ^a	20	10.48
1-Butyne (C ₄ H ₆)	2.37 (0.69) x 10 ⁻²	1.09 (0.57) x 10 ⁻²	1.46 (0.44) x 10 ⁻²	1.85 (0.60) x 10 ⁻²	0 ^a	8	6.11
2-Butyne (C ₄ H ₆)	1.33 (0.48) x 10 ⁻²	6.84 (3.70) x 10 ⁻³	8.06 (2.13) x 10 ⁻³	1.40 (0.44) x 10 ⁻²	0 ^a	8	16.32
1,3-Butadiene (C ₄ H ₂)	1.87 (0.75) x 10 ⁻²	7.27 (3.00) x 10 ⁻³	1.77 (0.62) x 10 ⁻²	4.74 (4.07) x 10 ⁻³	0 ^a	16	5.76
Aromatics							
Benzene (C ₆ H ₆)	3.18 (1.09)	1.00 (0.37)	2.36 (0.68)	2.38 (0.55)	92.9	1.2	0.72
Toluene (C ₇ H ₈)	1.49 (1.03)	0.597 (0.218)	0.972 (0.250)	2.66 (0.52)	100.0	6.2	4
Ethylbenzene (C ₈ H ₁₀)	0.105 (0.027)	3.34 (3.03) x 10 ⁻²	6.43 (1.20) x 10 ⁻²	0.303 (0.052)	111.6	7.5	3.04
m/p-Xylene (C ₈ H ₁₀)	0.196 (0.044)	7.45 (3.89) x 10 ⁻²	0.131 (0.030)	0.458 (0.093)	75.8	19	2.51
o-Xylene (C ₈ H ₁₀)	7.31 (2.44) x 10 ⁻²	2.24 (1.46) x 10 ⁻²	4.68 (1.21) x 10 ⁻²	0.246 (0.060)	95.5	14	2.51
Styrene (C ₈ H ₈)	5.88 (1.81) x 10 ⁻²	2.28 (1.50) x 10 ⁻²	3.40 (1.90) x 10 ⁻²	8.64 (5.96) x 10 ⁻²	212.3	43	1.53
i-Propylbenzene (C ₉ H ₁₂)	8.97 (2.73) x 10 ⁻³	3.21 (2.78) x 10 ⁻³	5.51 (1.31) x 10 ⁻³	2.88 (1.51) x 10 ⁻²	95.5	6.6	6.23
n-Propylbenzene (C ₉ H ₁₂)	2.00 (0.87) x 10 ⁻²	4.90 (4.14) x 10 ⁻³	1.30 (0.47) x 10 ⁻²	0.122 (0.083)	109.7	5.7	6.23
3-Ethyltoluene (C ₉ H ₁₂)	4.44 (1.67) x 10 ⁻²	1.46 (1.14) x 10 ⁻²	2.79 (0.98) x 10 ⁻²	0.221 (0.128)	94.8	22.4	7.39
4-Ethyltoluene (C ₉ H ₁₂)	1.96 (0.79) x 10 ⁻²	6.78 (4.65) x 10 ⁻³	1.35 (0.59) x 10 ⁻²	0.115 (0.071)	69.7	13.6	4.44
2-Ethyltoluene (C ₉ H ₁₂)	2.12 (0.87) x 10 ⁻²	7.22 (4.33) x 10 ⁻³	1.50 (0.62) x 10 ⁻²	0.116 (0.083)	94.8	13.2	5.59
1,3,5-Trimethylbenzene (C ₉ H ₁₂)	1.19 (0.55) x 10 ⁻²	4.37 (3.18) x 10 ⁻³	8.59 (4.28) x 10 ⁻³	6.89 (4.93) x 10 ⁻²	13.5	60	11.76
1,2,4-Trimethylbenzene (C ₉ H ₁₂)	3.22 (1.59) x 10 ⁻²	1.14 (0.70) x 10 ⁻²	2.43 (1.28) x 10 ⁻²	0.193 (0.160)	20.6	32	8.87
1,2,3-Trimethylbenzene (C ₉ H ₁₂)	1.47 (0.78) x 10 ⁻²	3.11 (2.08) x 10 ⁻³	1.24 (0.82) x 10 ⁻²	9.30 (9.75) x 10 ⁻²	43.9	29	11.97
Terpenes							
alpha-Pinene (C ₁₀ H ₁₆)	2.55 (1.92) x 10 ⁻³	1.44 (1.81) x 10 ⁻³	2.54 (2.08) x 10 ⁻³	6.97 (7.78) x 10 ⁻³	17.4	52	4.51
beta-Pinene (C ₁₀ H ₁₆)	6.98 (5.20) x 10 ⁻³	3.69 (2.46) x 10 ⁻³	8.98 (10.14) x 10 ⁻³	8.93 (10.96) x 10 ⁻³	18.1	74	3.52
Oxygenates							
Acetaldehyde (C ₂ H ₄ O)	2.48 (0.92)	0.897 (0.520)	1.46 (0.38)	5.26 (2.34)	0.6	16	6.54
Butanal (C ₄ H ₈ O)	0.132 (0.047)	5.09 (3.32) x 10 ⁻²	8.77 (3.50) x 10 ⁻²	0.334 (0.145)	0	24	5.97
Acetone (C ₃ H ₆ O)	2.17 (0.69)	0.977 (0.584)	1.36 (0.35)	6.34 (1.63)	0.3	0.19	0.36
2-Butanone (C ₄ H ₈ O)	0.530 (0.183)	0.214 (0.161)	0.338 (0.130)	1.54 (0.47)	0.6	1.2	1.48
2-Propenal (C ₃ H ₄ O)	0.573 (0.202)	0.342 (0.177)	0.414 (0.192)	0.914 (0.761)	1.0 ^a	20	7.45
MVK (C ₄ H ₆ O)	0.396 (0.121)	0.176 (0.117)	0.205 (0.087)	0.865 (0.458)	1.0 ^a	19	9.65
Furan (C ₄ H ₄ O)	0.336 (0.141)	0.160 (0.086)	0.223 (0.072)	1.17 (0.29)	1.0 ^a	40	9.15

2-Methylfuran (C ₅ H ₆ O)	0.360 (0.177)	0.158 (0.123)	0.226 (0.093)	1.51 (0.70)	1.0 ^a	62	8.3
Furfural (C ₅ H ₄ O ₂)	0.263 (0.218)	0.113 (0.143)	0.265 (0.161)	0.981 (0.414)	1.0 ^a	35	10
Methanol (CH ₃ OH)	6.43 (3.77)	5.38 (5.20)	3.84 (1.26)	13.1 (10.6)	0.3	0.9	0.67
Ethanol (CH ₅ OH)	0.126 (0.220)	5.80 (5.19) x 10 ⁻²	0.184 (0.220)	0.236 (0.281)	0.6	3.4	1.53

^agiven values of either 0 or 1, SOAP values not reported

^bvalues based on 1,3-butadiene

^cvalue based on (*E*)-1,3-Pentadiene

Table S2. Summary of aromatic compounds, oxygenates, halogen- and sulfur- containing compounds that do not have higher emissions when using *angithi* stove and/or dung fuel. Significant relationships between emissions and either stove or fuel are indicated by marking the type that gives the larger emissions. N/A indicates no relationship was found. P values are listed in addition to the fuel or stove type if the relationship is not clear.

Compound	Stove	Fuel
Benzene	<i>Chulha</i>	Dung
Styrene	<i>Angithi</i> (p=0.0723)	Dung
Acrolein	<i>Angithi</i> (p=0.0787)	Dung
Methanol	<i>Angithi</i>	N/A
Ethanol	N/A	N/A
Furfural	<i>Angithi</i>	Dung (p=0.03)
Dichloromethane	N/A	N/A
1,2-dichloroethane	<i>Angithi</i>	N/A
Iodomethane	<i>Angithi</i> (p=0.0891)	Dung

Figure S1. Emission factors (g VOC/ kg fuel C) versus moisture content of the fuel. Red markers indicate dung fuel was used, while blue markers indicate brushwood fuel. Filled square markers indicate *chulha* stoves, while open circles indicate *angithi* stoves were used. If a significant correlation was observed ($r^2 > 0.70$) for a particular fuel-stove combination, the linear regression trendline is shown.

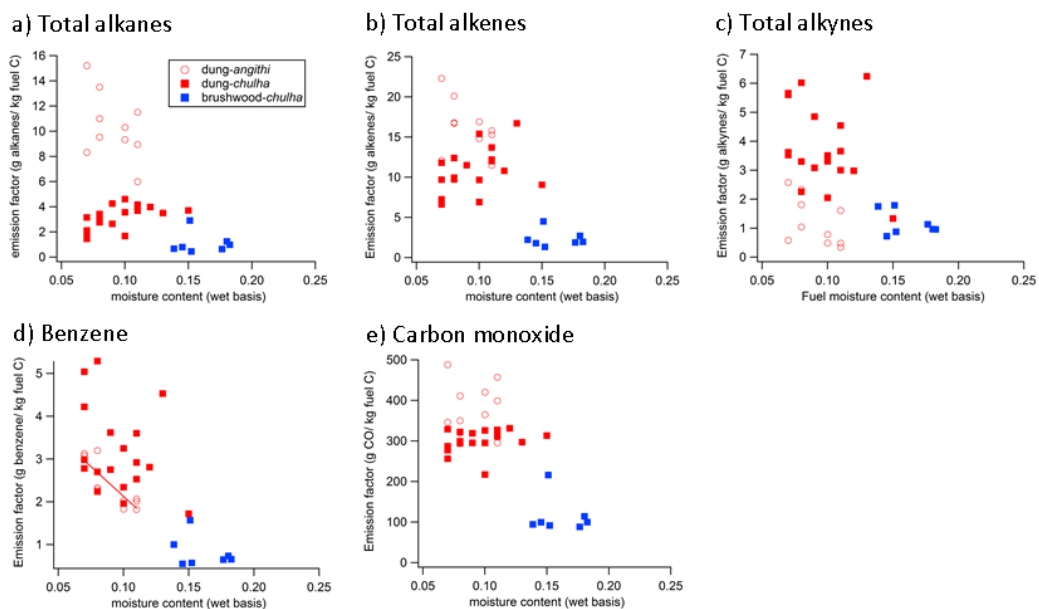
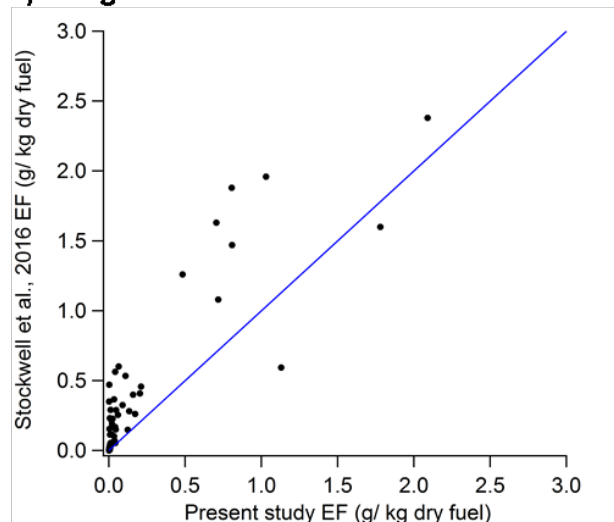


Figure S2.1 Comparison of EFs (g/ kg dry fuel) in the present study to those of Stockwell et al., 2016. The blue line indicates 1:1 agreement of the emission factors, with points above the blue line indicating higher EFs reported in Stockwell et al., 2016. Likewise, points below represent compounds that have a higher EF in the present study.

a) Dung



b) Brushwood

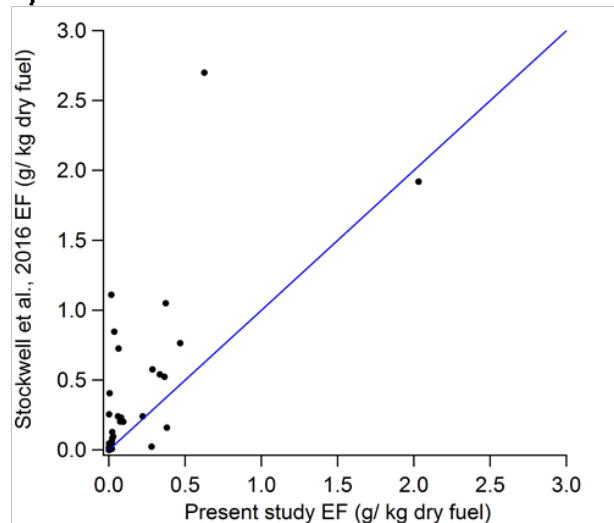


Figure S2.2 Emission factors (g/ kg fuel C) plotted as a function of modified combustion efficiency for select species. Open circles indicate cooking events conducted with *angithi* stoves, whereas filled squares indicate *chulha* stoves. Color indicates fuel, either brushwood (blue), dung (red), or mixed (purple). Crosses indicate measurements from Stockwell et al. 2016.

