Anonymous Referee #1
Received and published: 3 July 2017

Referee #1 General Comments: *Jayarathne et al. present emissions factors for various particle and gas-phase compounds emitted from combustion sources in Nepal. These sources include brick kilns, garbage burns, generators, water pumps, motorcycles, cooking stoves, and crop fires. These sources have not been well characterized in the past and significantly contribute to the air quality in this region. The study is very detailed and does a great job in comparing their values to previous work. EFs from this study are useful for source apportionment and regional air quality models. This study should be published in ACP after the authors address the below minor comments.*

Response to Referee #1 General Comments: We thank the reviewer for their careful review of the manuscript and their suggestions to improve it. We respond to each comment point-by-point below.

Minor Comments:

Referee #1 Comment 1: *Page 2, line 26: mobile instead of moveable*

Response to Referee #1 Comment 1: The field laboratory used in source characterization is most accurately described as "moveable", in our opinion, because the laboratory was stationary during sample collection. A mobile laboratory, on the other hand, implies that samples were collected while moving. For this reason and to maintain consistency with our companion paper from NAMaSTE (Stockwell et al., 2016b), we prefer to keep this described as a "moveable laboratory."

Referee #1 Comment 2: *Page 3, line 13: whereas the force-draught…*

Response to Referee #1 Comment 2: Following the reviewer's suggestion, we think it would be most clear to split this sentence in two, in order to first describe the zig-zag pattern and then the approach to forced-draught, both of which apply to the kiln that were characterized. The revised text reads, "Air moves in a zig-zag pattern through stacks of bricks and is vented through a central smoke stack. The forced-draught style employs a fan to generate air flow."

Referee #1 Comment 3: *Page 3, line 29: change quantity to amount*

Response to Referee #1 Comment 3: This change has been made as suggested.

Referee #1 Comment 4: *Page 4, line 6: the majority of which were not collected and were ultimately burned. . .*

Response to Referee #1 Comment 4: This change has been made as suggested.

Referee #1 Comment 5: *Page 4, line 11: The challenges in characterizing*

Response to Referee #1 Comment 5: This change has been made as suggested.

Referee #1 Comment 6: *Page 5, line 6: should be 1970s*

Response to Referee #1 Comment 6: This change has been made as suggested.

Referee #1 Comment 7: *Page 5, line 27: I do not think VOC has been defined yet.*

Response to Referee #1 Comment 7: We agree with the reviewer and have defined VOC here, which is its first appearance.

Referee #1 Comment 8: *Page 6, line 9: the phrase "particularly for women and children who are near to the source" is a bit confusing. Maybe just state that women and children spend more time indoors near the burning source?*

Response to Referee #1 Comment 8: We agree with the reviewer's suggestion and have revised the text to read: "particularly for women and children who spend more time indoors near the combustion source."

Referee #1 Comment 9: *Page 6, line 21: Please change the sentence starting with "High pollution" so it exhibits parallel structure.*

Response to Referee #1 Comment 9: As suggested by the reviewer, we have revised this sentence to exhibit parallel structure. The revised text reads: "High pollution levels in Kathmandu are a consequence of its growing population, rapidly expanding vehicular fleet (Shrestha et al., 2013), unpaved roadways, insufficient electrical power, widespread use of solid fuels for household energy needs (Smith et al., 2013), and common practice of burning garbage (Wiedinmyer et al., 2014)."

Referee #1 Comment 10: *Page 6, line 25: "Further, its valley topography traps pollutants, and. . ."*

Response to Referee #1 Comment 10: We have added a comma following "pollutants" as suggested.

Referee #1 Comment 11: *Page 6, line 27: " The Tarai. . ." sentence is awkward.*

Response to Referee #1 Comment 11: We have revised this sentence to read: "The Tarai, a predominantly agricultural region of southern Nepal, provided access to diesel groundwater pumps, agricultural residue burning, garbage burning, and additional samples of household biofuel combustion."

Referee #1 Comment 12: *Page 7, line 25: 2.5 m length for a sample inlet is quite long. What are the particle and semi-volatile losses for this inlet? Also Teflon coated filter holders may have significant particle losses. Has this been characterized?*

Response to Referee #1 Comment 12: The inlet was designed to allow sampling of smoke at a safe distance from combustion sources, and 2.5 m is not considered to be particularly long for combustion smoke sampling. Particle losses in a 2.5 m length if tubing with a 6.35 mm diameter at a flow rate of 7.5 lpm were estimated using the Particle Loss Calculator (PLC) (von der Weiden et al., 2009), allowing for losses due to diffusion, sedimentation, and turbulent inertial deposition. The model assumed unit density and a particle shape factor of 1. The estimated particle losses were less than 1% for particles in the size range of 50 nm – 2.5 μm that encompasses the vast majority of particle mass. Thus, sampling line losses are considered to be minor and are well within the estimated uncertainties. Semi-volatile losses were not considered, as semi-volatile species were neither gas nor particle phase analytes. Loss to the Teflon-coated filter holder was not characterized; however this is expected to be small because the filter holder is made of a non-sticking material and the particles are in contact with the filter holder for a very short time. Further, the filter holder manufacturer is not aware of any particle losses on the surface and expects any particle loss to the filter holder to be negligible (personal communication).

Referee #1 Comment 13: *Page 7, line 30: what temperature was the sample at when it was sampled?*

Response to Referee #1 Comment 13: Samples were collected at ambient temperatures (page 7 line 27), which ranged 12.3-28.6 ^{o}C in the Kathmandu valley, averaging 18.8 ^{o}C. To clarify this point, these temperatures have been added to page 7 line 27.

Referee #1 Comment 14: *Page 9, line 12: Can old be better quantified?*

Response to Referee #1 Comment 14: We have added the age of each generator at the time of sampling to the method description on page 9. The revised text reads: "Emissions from one petrol (4 kVA, 3 years old) and one diesel (5 kVA, 4 years old) generators were evaluated, using equipment rented in Kathmandu."

Referee #1 Comment 15: *Page 9, line 25: burning emissions sampled*

Response to Referee #1 Comment 15: We agree with the reviewer and have implemented this change.

Referee #1 Comment 16: *Page 10, line 1: Can the authors be a bit more specific about types of plastic? Predominately poly styrene? Or PVC? Or polypropylene?*

Response to Referee #1 Comment 16: We have clarified this point by adding "that were predominately made of polyethylene" on page 10 at line 2.

Referee #1 Comment 17: *Page 13, line 4: The sentence with "EFs for other particle. . ." is confusing? I could not quickly figure out the equations the authors were getting at.*

Response to Referee #1 Comment 17: We have clarified this sentence to read: "EFs for PM components were calculated as the product of $EF_{PM2.5}$ and the component's mass fraction in $PM_{2.5}$." With this clarification, we removed the example EF_{OC} calculation from the following line.

Referee #1 Comment 18: *Page 14, line 16: The sentence starting as "Another positive aspect. . ." is confusing. How does high concentrations (high concentrations of what? PM?) mean lower amounts of SVOCS? (SVOCs also has not been defined) I would think SVOCs would also be high concentration if PM is high concentration.*

Response to Referee #1 Comment 18: We have clarified this sentence to address the reviewer's questions. The revised text reads: "Also, sampling filters at high PM concentrations provides a better measure of total carbon (including SVOC and PM) since the capability to measure the evaporated SVOC in the gas phase is uncommon."

Referee #1 Comment 19: *Page 17, line 11: what does "processed in the same way" entail?*

Response to Referee #1 Comment 19: We have clarified this by describing the specific process that we were referring to. The revised text reads: "…with the sum of EF for OC, EC, metals and ions (excluding sulfate), which ranged 0.67-1.33 g kg^{-1} for the zig-zag kiln."

Referee #1 Comment 20: *Page 19, line 9: what does damp mean? How damp? Can this be quantified more than just damp?*

Response to Referee #1 Comment 20: While we do not have a specific measure of dampness, we have added discussion of the state of the garbage at page 19 on line 12: "For these samples, garbage had been dampened by rainfall the previous evening, making it difficult to ignite (requiring newspaper) and causing it require re-ignition on occasion (Stockwell et al., 2016b)."

Referee #1 Comment 21: *Page 19, line 26: Could it also be overestimated? Variability implies over and under estimation.*

Response to Referee #1 Comment 21: We agree with the reviewer and have revised this sentence to say "may be either over- or underestimated."

Referee #1 Comment 22: *Page 22, line 18: What does Measured organic species include? All those that are resolved in the GCMS? Or those that are positively identified with standards?*

Response to Referee #1 Comment 22: In light of this comment, we have clarified this point, by rewording this sentence to begin: "Organic species quantified by GCMS…"

Referee #1 Comment 23: *Page 22, line 28: What defines steady state operation? The engine reaches a certain temperature? How long did it take to reach steady state?*

Response to Referee #1 Comment 23: Collection of gas-phase measurements from agricultural diesel pumps commenced during the cold start-up phase and also encompassed regular continuous operation conditions (Stockwell et al., 2016b). Because collection of PM samples did not include the cold start phase, we wanted to make this clear to the reader. In order to align our terminology to our companion paper, we have revised "steady-state operation" to be "continuous operating conditions approximately 8 minutes after a successful start-up"

Referee #1 Comment 24: *Page 23, line 4: Why is EFPM2.5 being compared to EFPM1? The authors comment that these values are comparable but they should not as the particle cutoffs are different. There should be less PM1 than PM2.5 by mass.*

Response to Referee #1 Comment 24: We agree with the reviewer's comment. We compared to co-located measurements by Goetz et al. (in preparation) in an attempt to validate our observations. The similarities in EF_{PM1} and $EF_{PM2.5}$ suggest that the majority of the PM mass emitted from the groundwater pumps was less than 1 μm. However, since this manuscript remains in preparation at this time, we will remove the reference here and instead compare to the EPA emission factor (AP 42) of 6.0 g kg^{-1} (EPA, 1996).

Referee #1 Comment 25: *Page 25, line 6: Rephrase sentence starting with "The comparison. . ."*

Response to Referee #1 Comment 25: As suggested by the reviewer, we have rephrased this sentence. It now begins: "Changes to motorcycle EF before and after…"

Referee #1 Comment 26: *Page 30, line 23: should be e.g. instead of i.e*.

Response to Referee #1 Comment 26: We agree with the reviewer and have applied this correction in the revised manuscript.

Referee #1 Comment 27: *Figure 1: Why does this figure not include EC? Section 3.1 also does not include EC. Is there a reason it was not measured for these samples?*

Response to Referee #1 Comment 27: EC was below the detection limit for these samples. To clarify this point, we have added the following text to the Figure 1 captions: "EC was not detected in brick kiln emissions; optical measurements of BC from Stockwell et al. (2016) are provided in Table 1."

Referee #1 Comment 28: *Figure 2: These colors are difficult to distinguished from each other. Black for EC looks like smushed Metals contributions. The chloride and Nitrate blues look the same. Smushed green for ammonia looks blue-ish too.*

Response to Referee #1 Comment 28: We thank the reviewer for pointing this out. To improve the readability of these figures, we replaced the thick black lines in this figure with thin lines, altered the colors to provide more contrast, and widened the bars. We have applied these changes to the other similar figures in the text, to improve their readability as well.

Referee #1 Comment 29: *Figure 6: Why are biomarkers reported in different units than EF?*

Response to Referee #1 Comment 29: Biomarkers are normalized to the OC concentration, because this accounts for the variation in EF_{OC} (and thus $EF_{biomarkers}$) across different burns. To clarify this point, we have added the following text to the figure caption: "Normalization to OC accounts for the large changes in EF_{OC} observed across different combustion scenarios and demonstrates consistency in the molecular marker-to-OC ratios for common fuels."

Anonymous Referee #2
Received and published: 23 July 2017

Referee #2 Summary: *This paper describes particulate matter and its composition from several sources in the Kathmandu Valley of Nepal. In this paper, "composition" includes elemental and organic carbon, water-soluble inorganic ions and metals, and single organic species useful for speciation. The information presented here is relevant for atmospheric chemistry and will serve to provide inputs to atmospheric models. The measurement methods are competent and consistent with the state of the science. The paper is well written and the organization is clear. I support publication of this work after attention to some of the issues raised here.*

Response to Referee #2 Summary: We thank the reviewer for the careful review of this manuscript and the suggestions to improve its content and presentation. Responses to specific comments are provided point-by-point below.

Major issues

Referee #2 Comment 1: *The authors have gathered a lot of information in this measurement campaign, NAMASTE. They have chosen to publish several papers and to organize those papers by measurement type (gas phase in a different paper also published in Stockwell et al 2016, ACP- 16-11043-2016; particles in this paper, other papers promised.) This arrangement seems unavoidable or at least I can't think of a better one. I understand the need to divide the information into multiple presentations for tractability. However, I find that it leads to a somewhat haphazard feel and some repetition as each paper walks through a number of different emission sources and yet doesn't provide an overall integrated understanding of any single emission source. In this review I have some comments on the integration of this paper with the earlier paper. Although the earlier paper is already published, I hope that these comments can be useful to frame this and future publications.*

Response to Referee #2 Comment 1: We appreciate the reviewer's perspective on the integration of results from multiple platforms. As this is the second paper in a series of four, two of which are in preparation, we are not yet in a position to provide an integrated overview of all of the source measurements. Specific suggestions to improve the integration of this work and our previously published paper (Stockwell et al., 2016b) are addressed in the following responses.

Referee #2 Comment 2: *Another major issue is the very small number of samples for each source and the implication, given in the rationale, that these are representative of South Asia. If sources are different worldwide, then one might not expect sources in Kathmandu to be similar to those in India—why should they be? I understand that source testing always provides data from a small number of sources, relative to the total population. But the sample size and representativeness has to be discussed in the context of natural variability within the source population, and (if known) the causes of that variability. The paper has a good discussion of why the source categories were chosen, but hardly any discussion of why the individual units were selected or what they represent. This discussion could be improved. For example:*

- *two generators (one gasoline, one diesel?) are described only as "old" and "a size that is commonly used"—what does this designation entail? What power output and capacity factor? I assume these are four stroke engines but it's not stated.*
- *two diesel groundwater pumps: again what size? How were they operated? (This information is given later in the paper; should be in the Methods)*
- *motorcycles are said to be different in Kathmandu because of "steep gradients, congested traffic, low vehicle speeds, high altitude, and frequent re-starting" yet these motorcycles were measured at idle, capturing only the altitude—why? Are these two stroke or four stroke engines?*
- *biofuel stoves "brought a pot of water to boil"; is this Water Boiling Test with hot start, cold start simmer or is it a different sequence?*

Response to Referee #2 Comment 2: In response to this comment, we have removed the brief details of the sources from the introduction section of this study, consolidated the experimental details of the sources in the methods section, and added experimental details in the methods section as suggested.

Specifically, the following text was removed from page 4 line 31: "…these gasoline and diesel generators were described as "old" and were of a size that is commonly used at the household or small to medium commercial scales."

The description of the generators now reads: Emissions from petrol (4 kVA, 3 years old) and diesel (5 kVA, 4 years old) generators were evaluated, using equipment rented in Kathmandu. Both generators had four-stroke engines and were of a size that is commonly used at household or small to medium commercial scales. Generators were run without any electrical load (i.e. idling) and we estimate that they were running at approximately 20% capacity based on other idling generator tests performed in a follow-up study. Filter sampling occurred when the generator was under continuous operating conditions (i.e. not during start up). Diesel sold by the Nepal Oil Corporation specifies that sulfur content be less than 350 mg kg^{-1}, while the diesel sold in 2015 (for which data is available) ranged 165-337 mg kg^{-1} and averaged 240 mg kg^{-1}.

The revised description of the groundwater pumps now reads: "In the Tarai region, emissions from two diesel groundwater pumps. Pump 1 (4.6 kVA) was approximately 3 years old, while pump 2 (5 kVA) had been in use for less than 3 months. The pumps failed shortly after start-up on several occasions and were subsequently restarted. Filter samples were collected after the groundwater pumps had reached continuous operating conditions at approximately 8 minutes after a successful start-up. Consequently, the filter samples do not include the initial start-up phase, which was captured by real-time monitoring of gases and light-absorbing carbon (Stockwell et al., 2016), during which the pumps were visually observed to emit puffs of black smoke."

The motorcycles all had four-stroke engines as indicated on page 9, line 21. We have also elaborated upon the additional information about the motorcycles available in our companion paper: "The motorcycles had four-stroke engines, were powered by gasoline, and spanned four models (Honda Hero CBZ, Honda Hero Splendor, Honda Aviator, Bajaj Pulsar) that ranged in age from 3-15 years; details of their mileage at last service, total vehicle mileage, and age since purchase are provided by Stockwell et al. (2016; see Table S1)."

For garbage burning, we have added a reference to our companion paper that includes additional information about garbage composition and sampling details on page 10 at line 10: "Details of the garbage composition and sampling details are provided by Stockwell et al. (2016, see Table S2)."

For the cooking tests, the revised description reads: "Laboratory tests were used to study emissions from various stoves as they brought a pot of water to boil from a cold start (i.e. room temperature) to simulate cooking. These tests die not strictly follow a controlled protocol (e.g., the Water Boiling Test), such that stove efficiency was not determined. The studied stoves included traditional mud stoves, chimney stove, natural-draught rocket stove, induced-draught stove, bhuse chulo (insulated vertical combustion chamber), forced-draught biobriquette stove with an electrical charger, and biogas burner."

Also, we clarified the cooking conditions of the *in situ* tests: "The *in situ* testing of cooking fires in Tarai homes and a restaurant operated out of a personal kitchen provided real-world emissions samples from traditional mud stoves of the 1- or 2-pot design that were fired with hardwood, twigs, dung, or a mixture of dung and hardwood while normal cooking operations occurred. In sampling emissions from the in situ cooking fires, the inlets were positioned in a corner of the home to sample well-mixed integrated emissions."

In regards to the representativeness of the studied samples, we recognize that we have studied a small sample of a diverse population of combustion sources. We mention the need for further research to understand the diversity and variability of emissions regionally at the introduction (page 7, line 14) and the second paragraph of the conclusion section (page 31 line 5). In light of this comment, we have emphasized this at the beginning of the method section "2.1.2 Combustion Sources" by adding the following text: "The sources studied in NAMaSTE represent a small sample of a diverse population of combustion sources in Nepal and South Asia. The experiment was designed to characterize previously uncharacterized or under-sampled sources recognized as important to the region with a high degree of

chemical detail. The relatively small number of samples collected within each source category limits our understanding of the emissions variability within a source category and the representativeness of the studied samples of the broader population."

Referee #2 Comment 3: *In most of the descriptions no mention is made of the operating conditions: power levels, acceleration or steady state, fuel quality, analysis, moisture content (for solid fuel), sulfur content. For some sources, emissions vary during the course of operation, such as garbage burning, field burning, kiln operation. It's not stated whether the emissions were measured from beginning to end—probably not—or whether a fraction of the time was measured, which fraction was selected and why. The exception is "5 hours" for the brick kiln. How long were the samples? These factors affect emissions, representativeness, and comparability. When papers are given on individual sources this information would be expected. The presentation of just a small number of many different sources doesn't relieve this responsibility. The information may be in the earlier paper, but I would rather see repetition of this important data, rather than a repetition of the reasons for sampling.*

Response to Referee #2 Comment 3: As suggested by the reviewer, we added the duration of filter sample collection to manuscript, specifically in Table S1. This information is now mentioned in the text at the start of section 2.1.2: "The combustion sources analyzed are summarized in Table S1 (with the utilized fuels, location and duration of sampling, and average PM mass concentrations)."

The availability of chemical analysis of the coal fired in brick kilns and of the bricks is now mentioned: "Chemical analysis of the coal burned and bricks produced by each kiln are provided by Stockwell et al. (2016, see Table S3)." We were unable to analyze the chemical composition and moisture content of biofuels, due to the limitations on exporting these materials.

We have added information on the diesel fuel sulfur content to section 2.1.2: "Diesel sold by the Nepal Oil Corporation specifies that sulfur content be less than 350 mg kg^{-1}, while the diesel sold in 2015 (for which data is available) ranged 165-337 mg kg^{-1} and averaged 240 mg kg^{-1}."

Referee #2 Comment 4: *Finally, there could be more comparison with constraints. For example authors have both EC measurement (thermal optical) and BC measurement (PAX), but these are never compared except when one of them does not yield a result. Since both are employed to infer model inputs, this comparison should be discussed. There is a good discussion of this only for the pump results, and that one is rather inconclusive. Likewise, there are measurements of both SO2 and SO4, which should be possible to compare with fuel sulfur content.*

Response to Referee #2 Comment 4: Regarding the comparison of BC by PAX and EC by thermal-optical analysis we note that there is a forthcoming third set of related measurements from an aethalometer. All three methods produced useful information, but did not have perfect spatial and temporal overlap. The lack of perfect overlap effectively increases the amount of sampling, but also complicates comparisons. Thus we prefer to synthesize data more in the upcoming paper with access to all three sets of results.

In regards to comparing SO_2 and sulfate emissions with fuel sulfur content, we agree with the reviewer that this would be a very relevant comparison to make. However, in order to perform this comparison in a rigorous way, we would need additional information that is unknown to us, particularly the mass ratios of coal to biomass co-fired in the brick kilns and the sulfur content of the biofuels (if non-negligible). Because of these data limitations, we have not included this comparison in the revision.

Specific comments

Referee #2 Comment 5: *Page 7 Sample collection. How were the capture points of the probes aligned and how were they chosen? How was it ensured that a representative portion of the plume was captured?*

Response to Referee #2 Comment 5: We have clarified that the two inlets were connected to one another in section 2.1.1: "Smoke was drawn through two side-by-side sample inlets that were mounted on a ~2.5 m long pole…" To clarify the placement of the inlets, the following sentence is now included in section

2.1.1: “The pole upon which the inlets were mounted was positioned manually to sample the plume where the plume of smoke was well-mixed and had cooled to near-ambient temperatures.” Additional details have been added to the method descriptions of the cooking stoves as detailed in response to Referee #2 Comment 2.

Referee #2 Comment 6: *Page 10 Elemental and organic carbon "adjusted for positive sampling artifacts." How was this done? Denuders or subtracting loading of quartz filters behind Teflon filters?*

Response to Referee #2 Comment 6: We have clarified in section 2.2.2 the make of types and positions of filters used for the artifact correction and have added a reference for this approach: “The fraction of OC on quartz fiber backup filters relative to the front quartz fiber filters was used to estimate positive sampling artifacts from gas adsorption and was subtracted from the front filters (Kirchstetter et al., 2001; Roden et al., 2006).”

Referee #2 Comment 7: *Page 10-11 Field blanks were subtracted, but there isn't a mention of how large the field blank correction was. Is it significant relevant to average concentrations—especially for individual organic species? This could be a measure of contamination under the challenging field conditions.*

Response to Referee #2 Comment 7: We have a brief statement about the magnitude of the field blank correction to each of the method descriptions in section 2.2. In section 2.2.1: “There was no detectable increase in field blank filter masses and thus no field blank subtraction was applied.” In section 2.2.2: “A field blank subtraction was applied for OC and the amount of OC on field blanks was < 18% of the OC on sampled filters. EC was not detected on field blanks such that no EC field blank subtraction was applied.” In section 2.2.3: “The amount of WSOC recovered from field blanks was small in comparison to source samples that contained appreciable amounts of WSOC, (e.g., < 20% for biofuel emissions and mixed garbage burning), but larger for samples with primarily water-insoluble OC (e.g., approximately 60% for fossil fuel).” In section 2.2.4: “Results are reported only for ions whose concentrations are greater than the sum of either the mean field blank levels or the method detection limit (Jayarathne et al., 2014), whichever is larger, and three times the standard deviation of the field blank.” In section 2.2.5: “Results are reported only for metals for which the concentrations are greater than the sum of mean field blank levels and three times the standard deviation of the field blank.” In section 2.2.6: “Field blank concentrations were low in relation to those in source samples for most molecular markers, averaging < 10% for 3-ring PAH, < 1% for 4-ring or greater PAH, < 5% for hopanes in fossil fuel emissions samples (except for the zig-zag kiln in which was at < 45%), < 1% for levoglucosan in biofuel emission samples, and <10% for stigmasterol in dung burning emission samples. n-Alkane concentrations in field blanks averaged 50% of the concentrations measured in source emissions, which is reflected in many corresponding EF being below detection limits and having large relative uncertainties.”

The application of the above described criteria for reporting ions and metals resulted in adjustments to numerical values displayed in the tables and figures. Notably, the metals concentrations decreased and are no longer displayed in the figures.

Referee #2 Comment 8: *I don’t have many comments on the chemical measurement methods. They seem competent. The carbon-balance method is commonly used for sources where plumes are hard to capture.*

Response to Referee #2 Comment 8: No changes were made to the manuscript as a result of this comment.

Referee #2 Comment 9: *Zig Zag Kiln, Comparison with previous measurement (Weyant and Christian): This seems important because these are the only 3 measurements existing, to the best of my knowledge, and the results seem very different in this study. This paper states that “measurements were sampled within the stack at higher temperatures” but the Weyant paper described dilution to cool the sample stream before measuring, not measuring at stack temperature. Does the reasoning still apply? If the other measurements were cooled and diluted (but not diluted to ambient concentration), then in fact the gas-to-particle partitioning (without chemical conversion) would favor higher emission factors from the Weyant*

measurements, wouldn't it? "2-3 m downwind" (page 7) doesn't allow a lot of time for cooling. Authors finally compare the total carbon measurements as similar (for all measurements: Christian, Weyant and these) and attribute the difference in this study to sulfur and bound water. So it seems that there is a second hypothesis, difference is likely due to the SO2 conversion to SO4.

Response to Referee #2 Comment 9: We agree with the reviewer that the previous work by Weyant et al. (2014) was cooled and diluted by a factor of 1.5-4.2 prior to sampling. Because both studies sampled cooled emissions, temperature alone cannot explain the difference in EF_{PM}. We have removed the following text from the discussion: "Notably, measurements by Weyant at al. (2014) were sampled within the stack at higher temperatures, compared to 1-2 m downwind at ambient temperature. Consequently, the PM samples herein reflect more gas-to-particle partitioning that occurs as the smoke is cooled as well as chemical processing that occurs quickly post-emission (e.g., conversion of SO_2 to sulfate), both of which would contribute to higher measurements of PM mass." The main difference between our sampling methods and this prior study is that Weyant et al. would not capture chemistry or other evolution that could occur in the stack above their sample point and that our emissions were cooled and diluted naturally rather than in a forced manner. In its place, we have inserted the following text: "Notably, measurements by Weyant at al. (2014) were sampled from the stack and then diluted, compared to natural dilution that occurred 1-2 m downwind... Because the kiln emissions in this study were sampled downwind of the stack after they had cooled and diluted naturally, rather than pulled from it, our PM samples are likely to have undergone chemical evolution that occurs above the sampling port and/or quickly post-emission (e.g., conversion of SO_2 to sulfate), which could contribute to higher measurements of PM mass."

Referee #2 Comment 10: *Zig Zag Kiln, Sulfate and Water: The sulfur appears very important in the emission factor. Is the emitted sulfur (gas and particle) consistent with the amount of sulfur in the fuel? Also, is reporting bound water consistent with other measures of PM emission? For use in modeling, the report of bound water would seem to give a high bias for atmospheric PM concentrations since the models also account for water uptake.*

Response to Referee #2 Comment 10: As discussed in response to Referee #2 Comment 4, we do not have fuel composition data and thus are not able to compare the gas and particle sulfur emissions to the fuel. As noted at section 3.1 (end of the first paragraph), gravimetrically determined mass includes particle-bound water as well as hygroscopic water that is taken up at the relative humidity of the measurement. For a reader interested in the EF_{PM} without water, we have reported the "lower limit of $EF_{PM2.5}$ (that excludes the maximum possible amount of particle-bound water) to be 6.4 g kg^{-1}." As a result of this comment, we have added a footnote to Table 1 and S1 with the numerical value of EF_{PM} for zig-zag kiln that states "This value is expected to include hygroscopic water, see section 3.1 for the estimated value that excludes water."

Referee #2 Comment 11: *Zig Zag Kiln, "EC was not detected by thermal-optical analysis, and thus the optically determined EFBC at 0.112 g kg-1 for this source (Stockwell et al., 2016) is recommended to estimate the soot component of the smoke." This statement seems arbitrary, unless authors can show that this level of EC would be undetectable with this method.*

Response to Referee #2 Comment 11: Synthesis of the PAX, aethalometer and filters is complex and examined in more detail in another manuscript in preparation. Here we provide the PAX value to provide some context for understanding the light-absorbing carbon component of these PM emissions and thus maintain the suggestion of using EF_{BC} in the case EF_{EC} is not detected.

Referee #2 Comment 12: *Clamp Kiln, composition and closure: Since the clamp kiln PM had 20% sulfate, why is the particle bound water not also contributing to an underestimate of total PM when reconstructing total from the species? It seems this PM behaves quite different from the zig zag emitted PM*.

Response to Referee #2 Comment 12: We have added a sentence to section 3.2 to explain this difference: "Unlike the zig-zag kiln, there was no evidence of hygroscopic water contributions to PM mass; this is because in the clamp kiln emissions, the sulfate was fully neutralized by ammonium (possibly from

the biomass) to form ammonium sulfate, which deliquesces at 79-80% RH (Martin, 2000), well above the RH during gravimetric mass measurements."

Referee #2 Comment 13: *Low EF of levoglucosan (page 16) Is the fraction of PM (not absolute emission factor) also compared to wood?*

Response to Referee #2 Comment 13: We have added a comparison of the levoglucosan-to-PM mass ratio in the discussion of the zig-zag kiln emissions. The revised text in section 3.1 now reads: "This EF is markedly lower than those reported for open biomass fires (Christian et al., 2010) or cooking stoves (Sheesley et al., 2003) reported previously and in this work (section 3.7 and Table S3). Likewise, the levoglucosan contribution to PM mass is < 0.02%, compared to an average of 9% from the biomass-fueled cooking stoves in this study (Table S3). The small EF and mass fractions of levoglucosan reflects the relatively small amount of wood burned in this zig-zag kiln relative to coal."

Referee #2 Comment 14: *Garbage burning: Comparison between EC emission factor and PAX based BC emission factor. They seem very different. 7.4 g/kg (PM) x 2.6% EC = 0.19 g/kg. Whereas PAX BC is 0.56 g/kg (wet) or 6 g/kg (dry). This should be explained.*

Response to Referee #2 Comment 14: As noted in response to Reviewer #2 Comment 4, three methods were used to study light-absorbing carbon in NAMaSTE: PAX (Stockwell et al., 2016), thermal-optical analysis of EC on filters (this study), and an aethalometer (Goetz et al., in preparation), which all produced useful information but did not have perfect spatial and temporal overlap. The lack of perfect overlap effectively increases the amount of sampling, but also complicates comparisons. Thus we prefer to synthesize data more in the upcoming paper with access to all three sets of results. We believe that it will be more straightforward to omit this comparison here and the text in question has been removed from the revision.

Referee #2 Comment 15: *Garbage burning: emissions of PAH. Again totals are given in mg/kg, and it would also be useful to identify whether the PAH/PM ratio is relatively high.*

Response to Referee #2 Comment 15: We have clarified this point by the addition of the following sentence to section: "Although the absolute EFPAH were high, PAH accounted for < 0.2% of PM2.5 mass, consistent with the other non-fossil fuel combustion sources in this study (Table S3)." Accordingly, we have updated the abstract: "Garbage burning emissions contained triphenylbenzene and relatively high concentrations of heavy metals (Cu, Pb, Sb), making these useful markers of this source."

Referee #2 Comment 16: *Diesel generator: EFPM are compared with US EPA, but "generator" is not a unique class. There is a lot of speculation in this discussion, e.g. that generators would have lower emissions if better maintained.*

Response to Referee #2 Comment 16: To specify the source of the data within the AP-42 Compilation, we have added to the text that these are the "EPA Emission Factors (AP 42) for uncontrolled gasoline and diesel industrial engines." We have also added to the citation that the data derives from "Chapter 3: Stationary Internal Combustion Sources" with the specific data coming from Table 3.3-2.

We have also revised this section to avoid speculation by removing the following sentence and phrase: "A professionally-maintained diesel generator on the ICIMOD campus in Nepal was observed to have a high MCE (0.998) (Stockwell et al., 2016a) and likely a lower $EF_{PM2.5}$ than the rented diesel generator from which our filter sample was collected…" and "…suggest that well-maintained generators have lower PM emissions."

Referee #2 Comment 17: *Diesel generator, composition: I am surprised by the low EC content, particularly since there appears to be little oil contribution to the PM. In that case where does the OC come from? Authors cite another study that finds mostly OC in emissions for a high-sulfur fuel. Was no sulfur detected in this PM? Pumps OC-BC split discussion. This discussion is not strong and leads to question about the work presented. Information is presented from AMS data which haven't been*

published yet. A method is used to divide the PM that is not discussed in the methods. Authors point out that the different measurement methods were measuring at different times and over different conditions (e.g. another method included start-up and high black smoke emission while this one does not.) They then propose applying composition from other measurements to these emission factors after having just explained that the emissions were different. One gets the impression that the other measurements are better and these shouldn't even have been reported. Perhaps this impression could be improved with a better presentation.

Response to Referee #2 Comment 17: In regards to the diesel generator, we, too, were surprised at the low EC values. Unfortunately the PAX and aethalometers were not operational during these tests and thus we do not have an opportunity to cross-check these results. Regarding the OC, we have clarified what we have learned about its sources from the organic speciation: "The observed species reflect both combustion (i.e. tailpipe emissions) and engine oil evaporation (Schauer et al., 1999)." We have also clarified the following: "although neither sulfur dioxide (Stockwell et al. 2016) nor sulfate was detected in these emissions."

Regarding the groundwater pumps, in light of the reviewer's comment, we have removed the discussion about the OC and EC and instead refer the reader to a forthcoming manuscript for a further discussion of the comparison across measurement techniques. The revised text reads: "Chemical measurements indicated that the $PM_{2.5}$ was largely carbonaceous in nature (Table 1). Filter-based measurements indicated that the average contributions to PM mass for OC and EC were 77 and 3.4%, respectively, and that OC was primarily water insoluble ($\geq$ 88%). Further discussion on the light absorbing carbon fraction of diesel pump emissions and a comparison of measurement methods is provided elsewhere Goetz et al. (in preparation-a)."

Referee #2 Comment 18: *Motorcycles: Although the motorcycles were measured only during idle, interesting results about the change in PM emission and composition with servicing are presented here. It is stated that results are compared with start-up emissions. Why is that condition comparable with idle emission?*

Response to Referee #2 Comment 18: We have added the rationale for this comparison: "Instead, we compare ratios of EFPM2.5 to EFCO determined herein to those from prior studies of vehicles under start-up, which is more comparable than EF for driving conditions (i.e., highway or street driving)."

Referee #2 Comment 19: *However, it seems unlikely that the emissions profile here is representative, since only idle was included.*

Response to Referee #2 Comment 19: We agree with the reviewer and to clarify this point have added the following text at the beginning of section 3.6: "Because of the limited scope of the motorcycle emissions testing, both in terms of drive cycle and number of samples, the following data are neither representative of the diverse Kathmandu vehicle fleet nor their integrated emissions. Instead, we focus on the controlled variable in these tests, which is changes in emissions during idle as a result of servicing."

Referee #2 Comment 20: *Biofuels: Authors find that (1) field samples EF are higher than previous reported EF, (2) PM2.5, OC, and EC were not significantly different between the field and laboratory samples. Does this mean their laboratory EF are higher than previous lab EF? Is the finding of no significant difference between lab and field the same for the other reported species?*

Response to Referee #2 Comment 20: We have added the following sentence to the comparison of field and laboratory EF for biofuels: "In comparison of the laboratory EF_{PM} to the literature, the reported values are elevated with respect to some previously reported values (Akagi et al., 2011; Venkataraman and Rao, 2001), but lower than other cases (Keene et al., 2006)." We note that the "EF for $PM_{2.5}$, OC, and EC, however, were not significantly different across the field and laboratory samples ($p > 0.05$)…", but refrain from comparing other elements (e.g., chloride, potassium), since the biofuel chemical composition is not known and thus its role in changes to the composition of the emitted PM cannot be controlled..

Referee #2 Comment 21: *Although authors have discussed some previous literature, they have missed comparison with some other studies. The following 3 studies measured many types of household stoves in laboratory setting. Are the relative comparisons in this study (which stoves are better) similar?*

> Smith, K. R., et al. (2000), Greenhouse implications of household stoves: An analysis for India, *Ann. Rev. Energy Environ.*, *25*, 741-763.
>
> Jetter, J. J., and P. Kariher (2009), Solid-fuel household cook stoves: Characterization of performance and emissions, *Biomass & Bioenergy*, *33*(2), 294-305, 10.1016/j.biombioe.2008.05.014.
>
> Jetter, J., et al. (2012), Pollutant Emissions and Energy Efficiency under Controlled Conditions for Household Biomass Cookstoves and Implications for Metrics Useful in Setting International Test Standards, *Environ. Sci. Technol.*, *46*(19), 10827-10834, 10.1021/es301693f.

The following 2 studies compared laboratory and field emissions. Are the comparisons similar to those found here?

> Johnson, M., et al. In-field greenhouse gas emissions from cookstoves in rural Mexican households, *Atmos. Env.*, *42*, 1206-1222.
>
> Roden, C., et al. (2009), Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves, *Atmos. Env.*, *43*, 1170-1181.

Response to Referee #2 Comment 21: As suggested by the reviewer, we have expanded our comparison of stove types to include a discussion of the recommended literature. Specifically we have added the following text to section 3.7: “The observed trends across stove types are consistent with prior studies of cooking stoves. Here and in prior studies, biogas holds advantages over traditional cooking stoves in terms of the global warming potential of emissions and provides a viable and cleaner-emissions alterative to the direct combustion of dung as fuel (Smith et al., 2000). Several prior studies have also documented that vented, natural-draught, and forced-draught stoves provide lower PM emissions (Jetter et al., 2012; Jetter and Kariher, 2009; Roden et al., 2009; Smith et al., 2000).”

In regards to comparing field and laboratory emissions, we have added the following text: “The decrease in combustion efficiency in the field compared to the laboratory has been previously reported for cooking stoves, particularly in the case of open fires, and is attributed to operator skill (Jetter and Kariher, 2009; Johnson et al., 2008; Roden et al., 2009). EF for $PM_{2.5}$, OC, and EC, however, were not significantly different across the field and laboratory samples ($p > 0.05$), although significant increases in PM emissions for stoves in the field compared to the laboratory have been demonstrated in larger cooking stove studies (Johnson et al., 2008; Roden et al., 2009).”

Referee #2 Comment 22: *Heating fire Page 29: it is stated that this single source profile (one measurement) is "representative of open co-burning of dung and fuel wood under smoldering conditions in the Tarai." This seems like a strong statement without any support. It is also stated that "the high OC:EC ratio (~150) is also characteristic of smoldering combustion conditions" – it seems that a large number of high OC:EC ratios could be "characteristic" of smoldering conditions. I think that authors should be more careful of using the words "representative" and "characteristic" without being able to support the use of those words.*

Response to Referee #2 Comment 22: We agree with the reviewer and have replaced “characteristic” with “indicates” and “representative” with “provides insight to…”

Referee #2 Comment 23: *Uncertainties: Throughout the paper uncertainties are given. However I began to wonder what these uncertainties represent. Are they uncertainties in method, obtained from field blanks, and do they also represent natural variability among members of a source class? I began to*

suspect that the natural variability was not represented when an uncertainty was also given for the single heating fire. This should be clarified.

Response to Referee #2 Comment 23: The meaning of the uncertainties was described in the first paragraph of section 3: “The best estimates of source emissions were determined as the mean of available replicate measurements of a source category, or the most representative (or only available) sample from a source. For sources represented by a single sample, errors were propagated from analytical uncertainties. For sources represented by replicate samples, errors were calculated as one standard deviation of the mean.” Because of the importance of understanding the meaning of the reported uncertainties, we have added this description to the caption of Tables 1-4 and S3 in order to clarify and reinforce this meaning throughout the paper.

Referee #2 Comment 24: *Conclusion: This paper is rather long and it is sometimes difficult to extract the authors' contributions to the field. The paper concludes with a statement that the measurements will be useful. I think it would be very helpful to the reader for the authors to make a short list of the specific new information. A few examples: (1) I don't know other measurements of groundwater pump emissions, and so they could indicate that these are some of the first measurements. (2) In other cases the authors added to the database of emission factors for total PM, e.g. for kilns, or stoves. They could say that this is an addition, whether the measurements are higher, or lower. (3) There were several discussions of source markers; some were identified as unique markers and some were dismissed. These could be summarized.*

Response to Referee #2 Comment 24: We agree with the reviewer’s suggestion to clarify the contribution of this work to the field and have added the following text to the conclusion section: “These data expand the understanding of combustion emissions in a number of ways. First, we provide the first EFPM for diesel groundwater pumps that are prevalent in South Asia. Second, we add to the body of literature on PM emissions for brick kilns, garbage burning, generators, cooking stoves, and open biomass fires, in many cases expanding the chemical detail that is known about PM composition. Third, we confirm that molecular and elemental tracers identified in previous studies are applicable to South Asian combustion sources, namely Sb and TPB for garbage burning and coprostanol and cholestenol for dung burning, which are useful in source identification and apportionment. Fourth, through the study of motorcycle emissions before and after servicing, we demonstrate that significant PM reductions may be achieved by servicing. Fifth, our data suggests that burning of wet garbage substantially increases PM emissions relative to dry garbage, which warrants further investigation. Finally, NAMaSTE is the first to provide a detailed chemical characterization of in situ combustion emissions from within Nepal, providing locally- and regionally-specific emissions data.”

Editorial comments

Referee #2 Comment 25: *"Terai" is misspelled throughout*

Response to Referee #2 Comment 25: In publishing our companion paper, by Stockwell et al. (2016) we have learned that the journal-preferred spelling is “Tarai” and thus we have use this form throughout this manuscript.

Referee #2 Comment 26: *page 19 "a bit damp" is not professional language*

Response to Referee #2 Comment 26: As suggested we have revised this text, it now reads: “the mixture of organic and inorganic waste creates damp conditions, under which the fires smolder…”

Referee #2 Comment 27: *Title 3.7 "of" should be removed*

Response to Referee #2 Comment 27: We agree with the reviewer and have implemented this change.

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Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE): Emissions of particulate matter from wood and dung cooking fires, garbage and crop residue burning, brick kilns, and other sources

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Abstract.

The Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE) characterized widespread and under-sampled combustion sources common to South Asia, including brick kilns, garbage burning, diesel and gasoline generators, diesel groundwater pumps, idling motorcycles, traditional and modern cooking stoves and fires, crop residue burning, and a heating fire. Fuel-based emission factors (EF; with units of pollutant mass emitted per kg of fuel combusted) were determined for fine particulate matter ($PM_{2.5}$), organic carbon (OC), elemental carbon (EC), inorganic ions, trace metals, and organic species. For the forced draught zig-zag brick kiln, $EF_{PM2.5}$ ranged 12-19 g kg^{-1} with major contributions from OC (7%), sulfate expected to be in the form of sulfuric acid (31.9%), and other chemicals not measured (e.g., particle bound water). For the clamp kiln, $EF_{PM2.5}$ ranged 8-13 g kg^{-1}, with major contributions from OC (63.2%), sulfate (~~20.8~~23.4%), and ammonium (~~14.2~~16%). Our brick kiln $EF_{PM2.5}$ values may exceed those previously reported, partly because we sampled emissions at ambient temperature after emission from the stack or kiln allowing some particle-phase OC and sulfate to form from gaseous precursors. The combustion of mixed household garbage under dry conditions had an $EF_{PM2.5}$ of 7.4 ± 1.2 g kg^{-1}, whereas damp conditions generated the highest $EF_{PM2.5}$ of all combustion sources in this study, reaching up to 125 ± 23 g kg^{-1}. Garbage burning emissions contained triphenylbenzene and relatively high concentrations of ~~polycyclic aromatic compounds (PAHs), triphenylbenzene, and~~ heavy metals (Cu, Pb, Sb), making these useful markers of this source. A variety of cooking stoves and fires fueled with dung, hardwood, twigs, and/or other biofuels were studied. The use of dung for cooking and heating produced higher $EF_{PM2.5}$ than

other biofuel sources and consistently emitted more $PM_{2.5}$ and OC than burning hardwood and/or twigs; this trend was consistent across traditional mud stoves, chimney stoves, and 3-stone cooking fires. The comparisons of different cooking stoves and cooking fires revealed the highest PM emissions from 3-stone cooking fires (7.6-73 g kg^{-1}), followed by traditional mud stoves (5.3-19.7 g kg^{-1}), mud stoves with a chimney for exhaust (3.0-6.8 g kg^{-1}), rocket stoves (1.5-7.2 g kg^{-1}), induced-draught stoves (1.2-5.7 g kg^{-1}), and the bhuse chulo stove (3.2 g kg^{-1}), while biogas had no detectable PM emissions. Idling motorcycle emissions were evaluated before and after routine servicing at a local shop, which decreased $EF_{PM2.5}$ from 8.8 ± 1.3 g kg^{-1} to 0.71 ± 0.45 g kg^{-1} when averaged across five motorcycles. Organic species analysis indicated that this reduction in $PM_{2.5}$ was largely due to a decrease in emission of motor oil, probably from the crankcase. The EF and chemical emissions profiles developed in this study may be used for source apportionment and to update regional emission inventories.

Keywords: source profile, aerosol, groundwater pump, motorcycles, PAH, Nepal, Indo-Gangetic Plains, Hindu Kush Himalaya, South Asia.

1 Introduction

Insufficient knowledge of air pollution sources in South Asia hinders the development of pollution mitigation strategies to protect public health (Gurung and Bell, 2013) and model representation of air quality and climate on local to global scales (Adhikary et al., 2007; Bond et al., 2013). Prevalent, but under-characterized combustion emission sources in South Asia include traffic, brick kilns, garbage burning, cooking stoves, and the open burning of biomass. To begin to address this gap, the Nepal Ambient Monitoring and Source Testing Experiment (NAMaSTE) was conducted to: i) characterize the emissions of gas and particle species produced by the many important combustion sources in Nepal as a model for South Asia, ii) develop emission factors and detailed emissions profiles for these sources to support revisions to regional emissions inventories, and iii) apply knowledge of source emissions to improve source apportionment of ambient air pollution. During April 2015, a moveable laboratory was deployed in Nepal to characterize *in situ* emissions from brick kilns, garbage burning, diesel and gasoline generators, diesel groundwater pumps, motorcycles, traditional and modern cooking stoves, and agricultural residue burning. Additional source emission tests were planned, but cancelled in response to the Ghorka earthquake that struck on April 25. Emissions of major gases (carbon dioxide [CO_2], carbon monoxide [CO], methane [CH_4], ammonia [NH_3], hydrochloric acid [HCl]), non-methane organic gases, and light-absorbing carbon (brown carbon [BrC] and black carbon [BC]) for these sources are reported by Stockwell et al. (2016). Further characterization of size-resolved particulate matter (PM) emissions by aerosol mass spectrometry (AMS)

is underway (Goetz et al., in preparation-a, b). In this paper, PM emission factors and chemical composition for these combustion sources are reported.

Across the Indo-Gangetic Plains (IGP), brick kilns generate a substantial amount of building materials. Bricks are dried and kilns are fired during the dry winter season, generally spanning from October to March in the IGP. The Kathmandu Valley in Nepal is home to more than 110 brick kilns (FNBI, 2016) and the greater Dhaka region is home to 1000 kilns (Guttikunda et al., 2013). Kilns vary in design, with some producing bricks in batches and others continuously; some have chimneys and others ventilate through gaps; some are forced-draught and others are natural-draught. Descriptions of common kiln types are provided elsewhere (Weyant et al., 2014; UNEP, 2014a). In NAMaSTE, emissions from two types of brick kilns were examined: zig-zag and clamp kilns. The zig-zag kiln is a continuous, moving fire kiln that is capable of producing 1-10 million bricks during a firing season. Air moves in a zig-zag pattern through stacks of bricks and is vented through a central smoke stack. ~~and t~~The forced-draught style employs a fan to generate air flow. The zig-zag configuration provides more even heating of bricks and yields a higher quality product (UNEP, 2014a), while consuming less energy per brick fired than the straight-line configuration used in the most common fixed chimney bull's trench kilns around South Asia. The clamp kiln is a smaller batch-style kiln that produces 10,000-200,000 bricks per batch (and less than 1 million bricks per season) (UNEP, 2014b). Unfired ("green") bricks are stacked in the center with fired bricks surrounding these; fuel—typically coal and biomass—is interspersed with the green bricks and ignited. There is no chimney and smoke escapes from the cracks in the top of the kiln. Some clamp kilns have been phased out in more industrialized areas in favor of continuous kilns that afford better efficiency, but this kiln type remains common in rural areas. Brick kilns are often fueled by low-quality coal, which is often supplemented with biomass (sawdust, briquettes, bagasse, etc.) or scrap tires (Maithel et al., 2012). Plumes of smoke are visible when kilns are in operation. Studies of several types of South Asian brick kilns have revealed that the bulk chemical composition of the PM is dominated by organic and elemental carbon (Weyant et al., 2014). Meanwhile studies in Mexico reveal that the PM also contains chloride and trace metals (Christian et al., 2010). Occupational exposure to brick kiln emissions can cause significantly reduced lung function (Zuskin et al., 1998) and oxidative stress (Kaushik et al., 2012). Because of the prevalence of brick kilns in South Asia, and their potential for significant local and regional influence on air quality, it is important to evaluate the ~~quantity~~ amount and chemical composition of particulate matter emitted, to further support source attribution, emissions inventories, and air quality modeling.

Globally, 2400 billion tons of domestic solid waste are estimated to be generated yearly, of which an estimated 41% is disposed through open burning, making garbage burning a significant source of air pollution (Wiedinmyer et al., 2014). In countries that lack programs for waste collection and disposal and/or with a large rural population, the extent of garbage burning is greater. For example, in Nepal, it was estimated that 1.1 million tons of waste were generated in 2013, the majority of which ~~was~~ were not collected (>84%) and ~~was~~ were ultimately burned at residential or dump sites (60%) (Wiedinmyer et al., 2014). In Kathmandu, much of the open waste burning occurs either in large trash piles accumulated on river banks or in small piles on streets and sidewalks. Although recognized as an important source of air pollution, the regional and global air quality impact of garbage burning remains highly uncertain due to limited data on the amount of waste burned and the quantity of pollutants emitted for different types of waste and burn conditions (Wiedinmyer et al., 2014). The challenges ~~to~~ in characterizing emissions from the open-burning of garbage include the fuel's inherent heterogeneity, various and often low-technology practices for burning garbage, and the range of scales on which it occurs, from residential point sources to municipal-scale dump sites (Bond et al., 2004). PM emitted from garbage burning contains significant amounts of organic and elemental carbon, with additional contributions from polycyclic aromatic hydrocarbons (PAH), polychlorinated dioxins and furans, and trace metals (e.g. Sb, Cu, Zn, Zb, Pb, V, As) (Woodall et al., 2012; Christian et al., 2010; Simoneit et al., 2005). Given the hazardous nature of garbage burning emissions and the widespread practice of burning garbage, it is important to evaluate the emissions from this source under real-world open-burning conditions.

Generators, powered by gasoline or diesel, are used in South Asia for electrical power generation, particularly in the absence of electricity provided by utilities through grid-based networks. In the Kathmandu Valley, generators are widely used for back-up power during power outages that were frequent and wide-spread until November 2016. Load shedding cut power to households upwards of 40 hours per week in Kathmandu, particularly during the dry winter season when water levels in rivers that provide hydroelectric power were low. Generator PM emissions vary greatly with generator model and manufacturer, fuel, engine size, engine load, running time, unit age, and maintenance (Zhu et al., 2009; Lin et al., 2006; Shah et al., 2006a). PM emissions from diesel engines are primarily elemental carbon and organic matter that result from combustion and/or evaporation of fuel or engine oil (Liang et al., 2005; Schauer et al., 1999, 2002). Although sharing many similarities, emissions from generators that operate under near to steady-state conditions vary from those of on-road vehicle engines that operate under transient conditions (Shah et al., 2006a). Within this study, emissions from gasoline and ~~two~~diesel generators were characterized to gain further insight to this widely-used combustion source. ~~; these gasoline and~~

~~diesel generators were described as "old" and were of a size that is commonly used at the household or small to medium commercial scales.~~

Groundwater pumps are widely used in South Asia as a means of accessing a consistent source of irrigation water, strengthening agrarian communities, and improving food security among growing populations; particularly in arid regions. Groundwater pump use has greatly expanded since emerging in the 1970~~'~~s, with nearly 20 million pumps in use in India, Bangladesh, Pakistan, and the plains in southern Nepal known as the Tarai in 2000 (Shah, 2009), although the number and location of such pumps are not well documented (Rawat and Mukherji, 2014). Pumps may be powered by either electricity or diesel, with the choice largely determined by energy prices and supply (Shah et al., 2006b). Diesel is the predominant fuel used (> 84%) in the IGP, including the Nepal Tarai (Shah, 2009; Shah et al., 2006b), while electricity and diesel have comparable market shares in India (Mukherji, 2008). Diesel PM is recognized by the International Agency for Research on Cancer as a group 1 carcinogen (IARC, 2013) and includes black carbon, a short-lived climate forcing agent (Ramanathan et al., 2005). In this study, we characterized the magnitude and chemical composition of PM emissions from two diesel groundwater pumps used in the Tarai region of Nepal.

Motorcycles are widely used for transportation in urban areas of Asia. In Nepal, they account for 80% of the vehicle fleet, consume 9% of the transport sector's fuel, and are undergoing the fastest growth of any vehicle sector (WECS, 2014; MoPIT, 2014). The motorcycles tested during NAMaSTE were 3-15 years old at the time of sampling and had 4-stroke engines (Stockwell et al., 2016), which is the most common engine type in Nepal (Shrestha et al., 2013). The motorcycles were manufactured in India and because four-stroke engines were not required to have catalytic converters until 2015 in India, it was assumed that the motorcycles tested did not have them (Stockwell et al., 2016). The absence of a catalytic converter leads to higher PM and PAH emissions, as do cold-starts when the catalyst is not fully operational (Spezzano et al., 2008). Emissions from vehicles in Kathmandu tend to be higher than in other parts of the world, due to steep gradients, congested traffic, low vehicle speeds, high altitude, and frequent re-starting (Shrestha et al., 2013); these conditions, despite their low engine stress, are responsible for high emissions of CO, volatile organic compounds (VOC), and PM (Oanh et al., 2012). In this study, the combined emissions from five motorcycles under idling conditions were evaluated before and after basic servicing. Although limited in scope, this study design provides insight to emissions reductions that may be achieved by servicing.

Biofuels are widely used in Asia as a source of energy for cooking and heating (Yevich and Logan, 2003). In the IGP, dung cake (formed by mixing cow dung and straw), fuelwood, and crop residue are major sources of household energy (Saud et al., 2011). Agricultural residues are also often burned in the fields at the end of the season to clear fields and return nutrients to the soil and this constitutes a major emission source (Yevich and Logan, 2003). Traditionally, cooking has involved the use of biofuels either in an open fire built between stones that support a pot (a.k.a. 3-stone fire) or in a closed fire in a mud structure (traditional mud stoves), which are located indoors and often do not have a chimney. Cooking indoors with high-emitting stoves produces a large fraction of regional emissions (Ramanathan and Carmichael, 2008) and the poor ventilation leads to high exposures to CO, other toxic gases, and PM, particularly for women and children who spend more time indoors near the combustion ~~are near to the~~ source (Davidson et al., 1986; Smith et al., 2013). Exposures are associated with myriad negative health outcomes including respiratory infections and low birthweight (Pope et al., 2010) that lead to premature mortality (Fullerton et al., 2008). To mitigate this risk, recent research efforts have focused on developing more efficient and less polluting cooking technologies (Kshirsagar and Kalamkar, 2014). Within this study, PM emissions from traditional and modern cooking technologies were evaluated using a variety of biofuels, with the goals of developing detailed chemical profiles of cooking stove emissions and assessing differences in emissions across fuel and stove types. In addition, *in situ* emissions from springtime agricultural residue burning in the field in the Tarai and from heating fires were also characterized.

The NAMaSTE campaign took place in two regions of Nepal: in and around Kathmandu and the Tarai, which provided access to numerous combustion sources of regional interest. Kathmandu, the capital of Nepal, suffers from high levels of fine particulate matter ($PM_{2.5}$) and gas-phase pollutants (Aryal et al., 2009). High pollution levels in Kathmandu are a consequence of its growing population, rapidly expanding vehicular fleet (Shrestha et al., 2013), unpaved roadways, insufficient electrical power, widespread use of solid fuels for household energy needs (Smith et al., 2013), ~~the frequent~~ and common practice of burning garbage (Wiedinmyer et al., 2014)~~, and unpaved roadways~~. Further, its valley topography that traps pollutants, and its long dry season are responsible for a daily pollution build-up (Panday et al., 2009). Kathmandu and its surroundings provided access to many targeted source types, including brick kilns, garbage burning, cooking stoves, motorcycles, and diesel generators. The Tarai, a predominantly agricultural region of southern Nepal, ~~Tarai, located in southern Nepal, is predominately agricultural and~~ provided access to diesel groundwater pumps, agricultural residue burning, garbage burning, and additional samples of household biofuel combustion.

EFs for combustion sources were determined by the carbon mass balance approach (Ward and Radke, 1993; Yokelson et al., 1999; Yokelson et al., 1996). Chemical profiles of $PM_{2.5}$ were developed by quantifying PM mass, organic and elemental carbon (OC and EC), water-soluble/insoluble organic carbon (WSOC/WIOC), water-soluble inorganic ions, metals, and organic species. Reported herein are the first detailed chemical profiles for many sources in South Asia, including clamp kilns, garbage burning, and diesel groundwater pumps. These particulate phase measurements, in combination with gas-phase, optical, and additional PM measurements reported in our companion papers by Stockwell et al. (2016) and Goetz et al. (in preparation-a) provide a detailed chemical description of these source emissions. These new emissions data can be used when expanding and updating emissions inventories, as source profiles in receptor-based source apportionment modeling, or to model exposures to air pollutants. More broadly, these data can provide a more accurate representation of the sources of air pollutants in Nepal and the rest of South Asia, and thus support evaluating air pollution impacts on climate and health as well as guiding mitigation strategies. NAMaSTE provides new insights into South Asian combustion emissions, but further research is needed to achieve a full understanding of the diversity, variability, and abundance of these emissions sources on a regional scale.

2 Methods

2.1 Field study of combustion emissions

NAMaSTE took place in and around Kathmandu Valley and in the Tarai region of southern Nepal from April 11-25, 2015. Because of the magnitude 7.9 Gorkha earthquake in Nepal on 25 April 2015, the study ended earlier than planned, before additional sources could be sampled.

2.1.1 Sample Collection

$PM_{2.5}$ was collected using a custom-built, dual-channel PM sampler. Smoke was drawn through two side-by-side sample inlets that were mounted on a ~2.5 m long pole, to allow post-emission sampling of the smoke from a safe distance, typically 2-3 m downwind of the stack or combustion source. The pole upon which the inlets were mounted ~~inlet~~ was positioned manually to sample the plume ~~at the point~~ where the plume of smoke was well-mixed and had cooled to near-ambient temperature~~s prior to sample collection~~. During the period of sample collection, ambient temperatures ranged 12-29 ^{o}C (on a five-minute basis) in the Kathmandu Valley and averaged 19 ^{o}C. Air was drawn through copper tubing to 2.5 µm sharp-cut cyclones (URG Corp.) followed by two Teflon

coated filter holders (Cole-Parmer). PM was collected on both 47 mm quartz fiber filters (QFF, Whatman) and 47 mm Teflon filters (PALL, Life Sciences). Air flow was maintained at a constant flowrate of 7.5 lpm through each channel and was logged continuously by flow meters (APEX, Inc.). The sampled air volume was calculated as the product of the average air flow rate through the filter and total sampling time. The filtered air was then passed to the land-based Fourier transform infrared (LA-FTIR) spectrometer multi-pass cell for the measurement of gas phase species as described by Stockwell et al. (2016).

Prior to sample collection, QFF were pre-baked at 550 °C for 18 hours to remove contaminants and stored in aluminum foil-lined petri dishes. For some samples, a second (backup) QFF filter was placed in series behind the first (front) QFF filter in order to assess gas adsorption to the front filter. Teflon filters were pre-weighed as described in section 2.2.1 and stored in plastic petri dishes. All petri dishes were sealed with Teflon tape before and after sampling. Field blanks were collected for every fifth sample. Filters were stored in a freezer at -20 °C before and after sample collection and were shipped frozen to the University of Iowa for chemical analysis. Reported values are corrected for positive sampling artifacts and were field blank subtracted.

2.1.2 Combustion sources

The combustion sources analyzed are summarized in Table S1 (with the utilized fuels, location and duration of sampling, and average PM mass concentrations). ~~and are partly described above and also in some detail by Stockwell et al. (2016), so only brief additional details are provided here.~~ The sources studied in NAMaSTE represent a small sample of a diverse population of combustion sources in Nepal and South Asia. The experiment was designed to characterize previously uncharacterized or under-sampled sources recognized as important to the region, with a high degree of chemical detail. The relatively small number of samples collected within each source category limits our understanding of the emissions variability within a source category and the representativeness of the studied samples of the broader population. Descriptions of each source are provided below, with reference to our companion paper (Stockwell et al., 2016) for additional information when available.

Emissions from seven cooking technologies were examined at the Renewable Energy Testing Station (RETS) in Kathmandu. Laboratory tests were used to study emissions from various stoves as they brought a pot of water to boil from a cold start (i.e. room temperature) to simulate cooking. These tests die not strictly follow a controlled protocol (e.g., the Water Boiling Test), such that stove efficiency was not determined. The studied stoves included~~:~~ traditional mud stoves, chimney stove, natural-draught rocket stove, induced-draught stove, bhuse

chulo (insulated vertical combustion chamber), forced-draught biobriquette stove with an electrical charger, and biogas burner. Emissions from 3-stone fires were also examined, but not under cooking conditions (i.e. no water was boiled), consequently this source is referred to as a "cooking fire" rather than a "cooking stove." The fires at RETS were fueled with hardwood, dung, twigs, mixtures thereof, sawdust, biobriquettes, or biogas (Table S1). Our data analysis emphasizes differences across fuels and technologies. A summary of the types of cooking stoves and fires studied at RETS is provided in Table S2 with a brief description of their typical operation and photograph for most stove types. The *in situ* testing of cooking fires in Tarai homes and a restaurant operated out of a personal kitchen provided real-world emissions samples from traditional mud stoves of the 1- or 2-pot design that were fired with hardwood, twigs, dung, or a mixture of dung and hardwood while normal cooking operations occurred. In sampling emissions from cooking fires, the inlets were positioned in a corner of the home to sample well-mixed integrated emissions.

Samples from all other sources were collected in the field. Agricultural waste burning was sampled in the Tarai and the filter samples were of co-burned rice, wheat, mustard, lentil, and grasses residues. A heating fire was sampled in Tarai, in which dung and twigs were openly burned to generate heat.

Brick kilns were studied near the Kathmandu Valley. For the zig-zag kiln emissions were examined over the course of five hours, which captured several fuel feeding cycles in which coal and bagasse were added to the kiln. ~~,~~ ~~t~~Three filter samples were collected from smoke escaping from the chimney, with each filter sampled only when the plume reached the sampling inlet. ~~over five hours, which captured several fuel feeding cycles in which coal and bagasse were added to the kiln.~~ Emissions from the clamp kiln were also collected in triplicate. The clamp kiln was fueled primarily with coal and was co-fired with hard wood, although most of the hardwood was likely consumed before we sampled this kiln late in its 18-day firing cycle. Chemical analysis of the coal burned and bricks produced by each kiln are provided by Stockwell et al. (2016, see Table S3). ~~The clamp kiln was fueled primarily with coal and was co-fired with hard wood, although most of the hardwood was likely consumed before we sampled this kiln late in its 18-day firing cycle.~~

Emissions from ~~one~~ petrol (4 kVA, 3 years old) and ~~one~~ diesel (5 kVA, 4 years old) generators were evaluated, using equipment rented in Kathmandu. Both generators had four-stroke engines ~~were qualitatively described as old~~and were of a size that is commonly used at household or small to medium commercial scales. Generators were run without any electrical load (i.e. idling) and we estimate that they were running at approximately 20%

capacity based on other idling generator tests performed in a follow-up study. Filter sampling occurred when the generator was under continuous operating conditions (i.e. not during start-up). Diesel sold by the Nepal Oil Corporation specifies that sulfur content be less than 350 mg kg^{-1}, while the diesel sold in 2015 (for which data is available) ranged 165-337 mg kg^{-1} and averaged 240 mg kg^{-1}.

In the Tarai region, emissions from two diesel groundwater pumps. Pump 1 (4.6 kVA) was approximately 3 years old, while pump 2 (5 kVA) had been in use for less than 3 months. The pumps failed shortly after start-up on several occasions and were subsequently restarted. Filter samples were collected after the groundwater pumps had reached continuous operating conditions, which occurred approximately 8 minutes after a successful start-up. Consequently, the filter samples do not include the initial start-up phase, which was captured by real-time gas phase emissions monitoring (Stockwell et al., 2016), during which the pumps were visually observed to emit puffs of black smoke.

Emissions from five motorcycles while idling were evaluated before and after servicing, which involved an oil change, cleaning air filters and spark plugs, and adjusting the carburetor. Filter samples were collected as a composite of emissions from 5 vehicles, each sampled one-by-one, for approximately 10 minutes each, onto the same filter. The motorcycles had four-stroke engines, were powered by gasoline, and spanned four models (Honda Hero CBZ, Honda Hero Splendor, Honda Aviator, Bajaj Pulsar) ~~and~~ that ranged in age from 3-15 years; details of their mileage at last service, total vehicle mileage, and age since purchase are provided by Stockwell et al. (2016; see Table S1). ~~(Stockwell et al., 2016).~~ The studied motorcycles are among the most common models in Kathmandu (Shrestha et al., 2013).

Emissions from garbage burning were studied for mixed garbage (n=3) and sorted trash that isolated foil-lined bags (n=1) and mostly plastic burning (n=1). Fires were ignited shortly before sample collection. Two distinct conditions were studied: damp conditions in Kathmandu and dry conditions in Tarai. Garbage burning under dry conditions is assumed to prevail and used in the best estimate of $EF_{PM2.5}$ as discussed in section 3.3. The garbage burning emissions sampled in the Tarai was collected from a mixture of typical domestic waste that included cardboard and chip bags. Four additional samples of PM from garbage burning were collected in Kathmandu in which the material was damp from rainfall the previous night and the fire was rekindled with newspaper on occasion (Stockwell et al., 2016); these samples are more representative of conditions where inorganic waste and damp organic waste are burned together at a dump site. The mixed garbage sample in Kathmandu included food

waste, paper, plastic bags, cloth, diapers, and rubber shoes and was sampled twice, whereas other garbage burning emissions were sampled only once. Some garbage was sorted to gain insight into emissions from specific garbage components. One such sample of plastic mostly consisted of heavy clear plastic, some plastic cups, and food bags that were predominantly made of polyethylene. Another such sample of foil wrappers included chip bags, candy wrappers, and aluminum foil-lined bags. Details of the garbage composition and sampling details are provided by Stockwell et al. (2016, see Table S2).

2.2 Chemical analysis of particulate matter

2.2.1 Measurement of $PM_{2.5}$ mass

Before and after sample collection, Teflon filters were conditioned for 48 hours in a desiccator and weighed using an analytical microbalance (Mettler Toledo XP26) in a temperature (22.0 ± 0.5 ^{o}C) and humidity (34 ± 12 %) controlled room. PM mass was calculated as the difference of pre-and post-sampling filter weights, each determined in triplicate. Field-blank subtracted filter masses were converted to mass concentrations (μg m^{-3}) by dividing by the sampled air volume. There was no detectable increase in field blank filter masses and thus no field blank subtraction was applied. The relative error in the PM mass measurements was propagated from the standard deviation of field blank filter masses (an estimate of method precision) and 15% of the measured value (to account for potential background influences, described in section 3).

2.2.2 Elemental and organic carbon

Organic carbon and elemental carbon were determined following the NIOSH 5040 method (NIOSH, 2003) on 1.0 cm^2 punches of QFF (Sunset OC-EC Aerosol Analyzer, Sunset Laboratories, Tigard, OR). All OC measurements were field blank subtracted and adjusted for positive sampling artifacts. The fraction of OC on quartz fiber backup filters relative to the front quartz fiber filters was used to estimate positive sampling artifacts from gas adsorption and was subtracted from the front filters (Kirchstetter et al., 2001). EC was not detected on any backup filters, indicating that PM collection of the front filter was sufficiently high that breakthrough was negligible. A field blank subtraction was applied for OC and the amount of OC on field blanks was < 18% of the OC on sampled filters. EC was not detected on field blanks such that no EC field blank subtraction was applied. Uncertainty in OC measurements was propagated from the standard deviation of the field blank OC levels and ~~5~~10% of the OC concentration, a conservative estimate of the precision error in replicate sample analysis

(NIOSH, 2003). Uncertainty in EC measurements was propagated from the instrumental uncertainty (0.05 µg cm^{-2}), ~~5~~10% of the measured EC, and ~~5~~10% of pyrolyzed carbon, which refers to OC that charred during analysis.

2.2.3 Water-soluble organic carbon

A sub-sample of QFF filter (taken with a machined 1.053 cm^2 punch) was analyzed for water soluble organic carbon (WSOC) using a total OC analyzer (GE, Sievers 5310 C) following methodology described elsewhere (Budisulistiorini et al., 2015). WSOC was extracted into 15.0 mL of >18.2 MΩ resistivity ultra-pure water (Thermo, Barnstead Easypure II) using acid washed (10% nitric acid) and pre-baked (550 °C for 5.5 hours) glassware. Inorganic carbon was removed with an inorganic carbon remover (GE, Sievers ICR). WSOC was quantified using a standard calibration curves prepared from potassium hydrogen phthalate (Ultra Scientific). The amount of WSOC recovered from field blanks was small in comparison to source samples that contained appreciable amounts of WSOC, (e.g., < 20% for biofuel emissions and mixed garbage burning), but larger for samples with primarily water-insoluble OC (e.g., approximately 60% for fossil fuel).

2.2.4 Measurement of inorganic ions by ion chromatography

Inorganic ions were quantified in aqueous extracts of filter samples by ion exchange chromatography with conductivity detection (Dionex-ICS 5000). Sample preparation, analysis, and instrument detection limits followed Jayarathne et al. (2014). ~~(Jayarathne et al., 2014)~~. The uncertainty was propagated using the average field blank, the standard deviation of the field blanks, and 10% of the metal concentration. Results are reported only for ions whose concentrations are greater than the sum of either the mean field blank levels or the method detection limit (Jayarathne et al., 2014), whichever was larger, and three times the standard deviation of the field blank.

2.2.5 Quantification of metals by inductively coupled plasma mass spectrometry

Total metals were dissolved following a procedure based on US EPA Method 3052 (USEPA, 1995). In brief, Teflon filters were cut in half using ceramic blades and then digested in a 2:1 mixture of concentrated nitric and hydrochloric acid (TraceMetal Grade, Fisher Chemical) using a MARS 6 microwave assisted digestion system (CEM Corporation, Matthews, NC) at 200 °C for 13 minutes. Extracts were filtered (0.45 µm PTFE) and analyzed for metals using a Thermo X-Series II quadrupole ICP-MS instrument (Thermo Fisher Scientific Inc., Waltham, MA, USA). The instrument was calibrated against IV-ICPMS-71A ICP-MS standard (Inorganic Ventures) at concentrations ranging from 0.1 - 50 ppb. The reported data is field blank subtracted and converted

to metal concentrations (µg m^{-3}) using total filter area, extraction volume, and sampled air volume. The uncertainty was propagated using the average field blank, the standard deviation of the field blanks, ~~measurements~~ and 10% of the metal concentration. Results are reported only for metals for which the concentrations are greater than the sum of mean field blank levels and three times the standard deviation of the field blank.

2.2.6 Organic species by gas chromatography mass spectrometry

All glassware used in preparing filter extracts was prewashed and baked at 500 ˚C. Source sample filters were sub-sampled prior to organic species characterization. Filter sub-samples were spiked with a suite of isotopically labelled internal standards which were used in quantification: pyrene-D_{10}, benz(a)anthracene-D_{12}, cholestane-D_4, pentadecane-D_{32}, eicosane-D_{42}, tetracosane-D_{50}, triacontane-D_{62}, dotriacontane-D_{66}, hexatriacontane-D_{74} , levoglucosan-$^{13}C_6$ and cholesterol D_6. Each sample was then extracted in to a hexane : acetone (1:1) mixture as described in Al-Naiema et al. (2015). The solvent extracts were subsequently concentrated to a final volume of 100 µL using a Turbovap (Caliper Life Sciences, Turbo Vap LV Evaporator) and minivap (Thermo Scientific, Reacti-Vap™ Evaporator) under high-purity nitrogen (PRAXAIR Inc.). Each analysis batch contained ten source samples and quality control samples containing two field blanks, one lab blank, and one spike recovery sample. These extracted samples were stored at - 20 °C until analysis.

Hydroxyl-bearing analytes were analyzed following trimethylsilyl (TMS) derivatization, as described in Stone et al. (2012), which converts active hydrogen atoms to TMS groups, thus eliminating their ability to hydrogen bond (Nolte et al., 2002). Briefly, 10 µL of the extract was blown down to complete dryness, reconstituted in 10 µL of pyridine (Burdick & Jackson, Anhydrous), and then 20 µL of the silylation agent N,O-bis-(trimethylsilyl)trifluoroacetamide (Fluka Analytical, 99%) was added. The mixture was heated at 70 ˚C for 3 h before instrumental analysis.

Filter extracts were analyzed for organic species using gas chromatography (GC; Agilent Technologies 7890A) coupled to mass spectrometry (MS; Agilent Technologies 5975). The GC-MS was equipped with an Agilent DB-5 column (30 m length × 0.25 mm inner diameter × 0.25 µm film thickness) and electron ionization (EI) source. Helium served as the carrier gas (PRAXAIR Inc.). An aliquot of 3 µL was injected operating in the splitless mode following the temperature program described in Stone et al. (2012). Responses of analytes were normalized to the corresponding isotopically-labeled internal standard and five-point linear calibration curves (with

correlation coefficients, $R^2 \geq 0.995$) were utilized for the quantification of organic species. Compounds that were not in the standards were measured by assessing the response curve from the compound that is most analogous in structure and retention time. All reported species concentrations were field blank subtracted, and had spike recoveries in the range of ± 20% of the expected concentration. Field blank concentrations were low in relation to those in source samples for most molecular markers, averaging < 10% for 3-ring PAH, < 1% for 4-ring or greater PAH, < 5% for hopanes in fossil fuel emissions samples (except for the zig-zag kiln in which was at < 45%), < 1% for levoglucosan in biofuel emission samples, and <10% for stigmasterol in dung burning emission samples. n-Alkane concentrations in field blanks averaged 50% of the concentrations measured in source emissions, which is reflected in many corresponding EF being below detection limits and having large relative uncertainties. The analytical uncertainties for the measured species were propagated from the standard deviation of the field blanks and 20% of the measured concentration.

2.3 Emission factor calculation

A field-deployable, Fourier transform infrared (FTIR) spectrometer and whole air sampling with gas chromatography were used to quantify mixing ratios of up to 80 gases, including CO, CO_2, acid gases (HCl, HF, etc.) and volatile organic compounds as described by Stockwell et al. (2016). The carbon mass balance approach was used to determine fuel-based EFs for gases, in units of mass of pollutant per kilogram of fuel burned (g kg^{-1}) (Stockwell et al., 2016). EF for CO (EF_{CO}) were converted to EF for fine particle mass ($EF_{PM2.5}$) by the ratio of filtered PM mass (M_{PM}) and the corresponding mass of CO (M_{CO}) drawn through the filter that was measured in series by FTIR, following Eq. (1).

$$EF_{PM2.5} = \frac{M_{PM}}{M_{CO}} \times EF_{CO} \quad (1)$$

The EF_{CO} used in this calculation were calculated to coincide with filter sampling times and thus may differ slightly from those reported by Stockwell et al. (2016). These EF_{CO} were calculated using major carbon-containing species in the mass balance equation: CO_2, CO, CH_4, EC, and OC. EFs for ~~other particle phase species~~PM components were calculated ~~in the same way~~as the product of $EF_{PM2.5}$ and the component's mass fraction in $PM_{2.5}$~~, using their mass ratio to PM mass~~. ~~For example, EF_{OC} was calculated as the product of $EF_{PM2.5}$ and the OC-to-PM ratio for each source.~~ Uncertainties in EFs were propagated from the relative error in EF_{CO}, conservatively estimated at 5% (Stockwell et al., 2016) and the analytical uncertainty of the particle phase species.

2.4 Modified combustion efficiency

The modified combustion efficiency (MCE), calculated as $MCE = \Delta CO_2/(\Delta CO + \Delta CO_2)$, was used as an indicator of the relative amount of flaming combustion (MCE > 0.98-0.99) to smoldering combustion (~0.75-0.85) (McMeeking et al., 2009). Notably, the filter-integrated MCE values reported herein correspond to the average MCE over the duration of filter sample collection and they differ slightly from those reported by Stockwell et al. (2016), because they were typically collected over different time periods, although from the same source.

3 Results and discussion

The 41 source samples reported herein are summarized in Table S1 by source category, specific emission source, fuels, and fire numbers. EFs for particle-phase species, including $PM_{2.5}$, OC, EC, 8 inorganic ions, 12 metals (for 28 of 41 samples), and 68 organic species are reported in Table S3. For each source category, Tables 1-2 summarize the best estimate of $EF_{PM2.5}$ and $PM_{2.5}$ composition, including OC, EC, water-soluble inorganic ions, and metals as mass fractions for fossil/waste-fueled and bio-fueled combustion sources, respectively. Tables 3-4 summarize the best estimates of organic species emissions normalized to OC for fossil/waste-fueled and bio-fueled combustion sources, respectively. The best estimates of source emissions were determined as the mean of available replicate measurements of a source category, or the most representative (or only available) sample from a source. For sources represented by a single sample, errors were propagated from analytical uncertainties. For sources represented by replicate samples, errors were calculated as one standard deviation of the mean. In cases when components were not detected in all replicate samples, $PM_{2.5}$- or OC-normalized concentrations were averaged among the available data. This calculation reflects that species go undetected due to low filter loadings, rather than differences in species mass fractions within a source category.

The reported EFs reflect partially-diluted emissions, as plumes were sampled several meters downwind of the source after cooling to ambient temperature. The average $PM_{2.5}$ mass concentrations measured in source samples (Table S1) ranged from 45 – 82,600 μg m^{-3} and averaged 10,900 μg m^{-3}. High PM concentrations were required to capture source signatures *in situ*; however, the combination of high PM levels with large emissions of semi-volatile OC (SVOC) can ~~overestimates~~increase ~~of~~ PM mass and OC emissions due to partitioning of ~~semi-volatile organic species~~SVOC to the particle phase (Lipsky and Robinson, 2006). ~~The~~ Thus, $EF_{PM2.5}$ and EF_{OC} depend on the dilution ratio and ~~the~~ chemical composition of the source emissions ~~(e.g., semi-volatile OC is affected~~, while EC is not affected~~)~~ (Lipsky and Robinson, 2006). ~~Because of this dependence, $EF_{PM2.5}$ and EF_{OC}~~

~~depend on the sampling conditions.~~ The partitioning effect may add some uncertainty to EF comparisons between sources in this study and between studies in the literature in general, since sampling systems cannot be designed to sample all sources at the same concentration and concentrations are often not reported with EF. We document the sample concentrations in Table S1 in part to help remedy this. Furthermore, different concentrations may be relevant for different study objectives. For instance, near-source high concentrations may be preferred for cooking fire exposure assessment. Also, ~~Another positive aspect of~~ sampling filters at high PM concentrations ~~is obtaining~~provides a better measure of total carbon (including SVOC and PM) since the capability to measure the evaporated SVOC in the gas phase is uncommon. On the other hand, source apportionment may be best based on ratios between low-volatility components.

To estimate the potential influence of background PM on the source emissions, the sampled concentrations of PM and OC were compared to background levels. The $PM_{2.5}$ concentrations in source plumes (Table S1) were compared to the average $PM_{2.5}$ concentration measured in Kathmandu at a suburban site, named Bode (27.689° N, 85.395° E), in the westerly outflow of Kathmandu city (Sarkar et al., 2016) where, during NAMaSTE, the ambient $PM_{2.5}$ concentration at Bode ranged 30-95 μg m^{-3} and averaged (±standard deviation) 62±19 μg m^{-3}. Using this method, we estimate that in 90% of the studied plumes, background PM contributed <8% of the collected PM. And in 65% of the studied plumes, background contributed <4% of the collected PM. For some sources with low PM emissions, background PM was more influential, contributing 10-20% for emissions from biobriquettes burned in a forced-draught stove with an electrical charger and hardwood burned in a forced-draught cooking stoves and 30% for motorcycles after servicing. The gasoline generator emissions were sufficiently close to ambient PM concentrations, such that source emissions could not be defined. In addition, the sampled OC concentrations were compared to background OC levels estimated from OA measured by AMS (Goetz et al., in preparation-a) for all sources excluding generators and the background was estimated to contribute 0.02-2.8% (averaging 0.7%) of the OC collected.

Particle-phase EF are complementary to those reported by Stockwell et al. (2016) for organic and inorganic gases and aerosol optical properties. A comparison of the EF reported herein to the size- and chemically-resolved emission factors by AMS is provided by Goetz et al. (in preparation-a). Together, these datasets provide a more thorough and in some cases initial characterization of gas and particle emissions from many important combustion sources in South Asia. EF and PM composition are discussed in the following sub-sections by source category, followed by a description of their potential applications.

3.1 Zig-zag kiln

The induced-draught zig-zag kiln, fueled primarily by coal with some bagasse, had a mean fuel-based $EF_{PM2.5}$ of 15.1 ± 3.7 g kg^{-1} across three replicate samples. The corresponding MCE was very high at 0.994, indicative of flaming and relatively complete combustion. Major components contributing to PM mass included OC (ranging 4-11%, averaging 7%) and sulfate (ranging 27-35%, averaging 32%) (Table 1; Figure 1a), where sulfate was expected to be primarily in the form of sulfuric acid as described below. The majority of the $PM_{2.5}$ mass was not explained by the species measured. ~~Only trace levels of m~~Metals associated with clay were not detected—aluminum ~~(0.014%)~~, iron ~~(0.011%)~~, and titanium ~~(0.010%)~~ —indicating that brick dust was not a major part of the unexplained $PM_{2.5}$ mass. Other water-soluble ions had minor mean contributions to $PM_{2.5}$ mass: ammonium (0.29%), ~~sodium (0.016%),~~ fluoride (0.011%), chloride (0.065%), and nitrate (0.14%). The deficit of cationic counterions for sulfate (corresponding to < 4% neutralization of sulfate), suggests that the majority of sulfate was in the form of sulfuric acid, although these two species are indistinguishable by the extraction and ion chromatography methods applied. Sulfuric acid is a very hygroscopic compound that spontaneously uptakes water at low relative humidity near 0% (Jacobson, 2005). Because sulfuric acid is prone to hydration at the relative humidity conditions of our gravimetric analysis (34 ± 12 %, section 2.2.1) and the condensation of water droplets on Teflon filters was visually observed for samples from this source, it is expected that particle-bound water accounts for some of the unexplained $PM_{2.5}$ mass. Since the gravimetric methods utilized for determination of $EF_{PM2.5}$ include particle-bound water (Tsyro, 2005), we use the sum of the measured $PM_{2.5}$ components and assume an OC to organic matter conversion factor of 1.4 to estimate the lower limit of $EF_{PM2.5}$ (that excludes the maximum possible amount of hygroscopc water) to be 6.3 g kg^{-1}.

The combination of particle-phase ion measurements and gas-phase measurements by Stockwell et al. (2016) provides a means of determining gas-particle distributions of some elements. On a molar basis, less than 1% of the measured F and Cl were detected in the particle phase, with > 99% in the gas phase as HF and HCl, respectively; this signals very fresh emissions as discussed in Stockwell et al. (2014). The F emitted is likely to have originated in the clay material used to make the bricks (EPA, 1996). On a molar basis, 20% of sulfur was emitted in the particle phase as sulfate (EF_{SO4} 4.9 g kg^{-1}), while the majority of sulfur emissions were gaseous SO_2 (EF_{SO2} 12.7 g kg^{-1}; Stockwell et al., 2016), indicating that within 1-2 meters of the stack, a substantial fraction of SO_2 had been oxidized to form sulfate.

OC comprised an appreciable fraction of PM mass and EF_{OC} averaged 1.0 g kg^{-1}. The EF_{OC} was within 10% of the EF for OA reported as "brown carbon" (EF_{BrC}), estimated by PAX (Stockwell et al., 2016), suggesting that the mass absorption coefficient they used (0.98 m^2 g^{-1}) was reasonably appropriate for this source and that there was not a substantial positive artifact due to the adsorption of semi-volatile organic compounds in the filter-based OC measurement. EC was not detected by thermal-optical analysis, and thus the optically-determined EF_{BC} at 0.112 g kg^{-1} for this source (Stockwell et al., 2016) is recommended to estimate the soot component of the smoke. The BC-to-total carbon (TC) ratio is therefore 0.10, indicating predominantly organic emissions.

The carbon component of the organic species measured by GCMS accounted for an average of 0.58% of OC. The most abundant individual species measured was levoglucosan, a well-established tracer of biomass burning (Simoneit et al., 1999), for which the mean EF was 1.69 mg kg^{-1}. This EF is markedly lower than those reported for open biomass fires (Christian et al., 2010) or cooking stoves (Sheesley et al., 2003) reported previously and in this work (section 3.7 and Table S3~~),~~). Likewise, the levoglucosan contribution to PM mass is < 0.02%, compared to an average of 9% from the biomass-fueled cooking stoves in this study (Table S3). The small EF and mass fractions of levoglucosan ~~which~~ reflects the relatively small amount of wood burned in this zig-zag kiln relative to coal. Very low levels of hopanes and low-molecular weight PAHs with 3 rings were observed (Table 3), while higher-molecular weight PAHs, including picene, a proposed tracer of coal combustion (Oros and Simoneit, 2000), were not detected. Low levels of organic species are consistent with the high MCE value and reflect relatively complete combustion of the coal.

Significant differences in emissions were found from the induced-draught zig-zag kiln compared to prior studies (Table 5). First, the mean $EF_{PM2.5}$ for the induced-draught zig-zag kiln (15.1 ± 3.7 g kg^{-1}) was considerably higher than $EF_{PM2.5}$ reported by Weyant at al. (2014) for induced-draught zig-zag kilns fueled with coal in India (0.6 – 1.2 g kg^{-1}). Notably, measurements by Weyant at al. (2014) were sampled within the stack ~~at higher temperatures~~and then diluted, compared to natural dilution that occurred 1-2 m downwind ~~at ambient temperature~~. ~~Consequently, the PM samples herein reflect more gas-to-particle partitioning that occurs as the smoke is cooled as well as chemical processing that occurs quickly post-emission (e.g., conversion of SO_2 to sulfate), both of which would contribute to higher measurements of PM mass.~~ Because the kiln emissions in this study were sampled downwind of the stack after they had cooled and diluted naturally, rather than pulled from it, our PM samples are likely to have undergone chemical evolution that occurs above the sampling port and/or quickly post-emission (e.g., conversion of SO_2 to sulfate), which could contribute to higher measurements of PM

mass. Christian et al. (2010) used similar sampling methods to this study and estimated $PM_{2.5}$ mass from the sum of the particle-phase measurements of OC, EC, metals and ions (but not sulfate) for two batch-style brick kilns fueled primarily by biomass in Mexico; their reconstructed $PM_{2.5}$ mass totaled 1.24 and 1.96 g kg^{-1} and are in good agreement with the sum of EF for OC, EC, metals and ions (excluding sulfate) for the zig-zag kiln, which ranged 0.67-1.33 g kg^{-1} ~~data from this study processed in the same way (0.90-1.82 g kg^{-1})~~. Thus, the difference in $EF_{PM2.5}$ is expected to be due to sulfate and hygroscopic water. Second, the observed EC:TC ratios are much lower than the range of values from 0.75-0.90 reported previously for induced-draught zig-zag kilns in South Asia (Weyant et al., 2014) and from 0.84-0.89 for two batch-style kilns in Mexico (Christian et al., 2010). In comparison, the smoke emitted from the zig-zag kiln in this study was qualitatively described as white, with puffs of black smoke emitted only when fuel was added. With total carbon emissions comparable across this study (0.63-1.26 g kg^{-1}) and those by Weyant et al. (0.08-0.67 g kg^{-1}) and Christian et al. (0.669-1.783 g kg^{-1}), the main reasons for the increased $EF_{PM2.5}$ from the induced-draught zig-zag kiln in Nepal are the high emissions of sulfate (likely in the form of sulfuric acid) and hygroscopic water ~~when collecting samples at ambient temperature~~.

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3.2 Clamp kiln

The clamp kiln studied produced a mean $EF_{PM2.5}$ of 10.7 ± 2.7 g kg^{-1} across three replicate tests. The average MCE was 0.952, reflecting less complete combustion than the induced-draught zig-zag kiln (Stockwell et al., 2016). On average, the $PM_{2.5}$ emitted from the clamp kiln included the following major components: OC (63.2%), sulfate (~~20.8~~23.4%), ammonium (~~14.2~~16.0%), chloride (5.~~1~~7%), and nitrate (~~1.8~~2.0%) (Table 1; Figure 1b). Minor components included BC (0.2%), ~~sodium (0.7%),~~ and potassium (0.2%)~~, and calcium (0.3%)~~. The sum of OC, BC, and measured inorganic ions exceeded the measured $PM_{2.5}$ mass by an average of ~~7~~11%. This is within the propagated uncertainty of the analytical measurements, but likely reflects adsorption of semi-volatile gases to the filter and over-estimation of OC mass. Unlike the zig-zag kiln, there was no evidence of hygroscopic water contributions to PM mass; this is because in the clamp kiln emissions, the sulfate was fully neutralized by ammonium (possibly from the biomass) to form ammonium sulfate, which deliquesces at 79-80% RH (Martin, 2000), well above the RH during gravimetric mass measurements. ~~Metals associated with clay were detected in clamp kiln emissions at levels an order of magnitude greater than for the zig-zag kiln (Table 1), suggesting some incorporation of clay dust into the emitted PM.~~ Neither particulate fluoride nor gas phase HF were detected from the clamp kiln. Chloride, however, was a significant component of PM, but gaseous HCl was below the FTIR detection limit and other chlorinated organic gases (e.g. CH_3Cl) were not greater than background levels (Stockwell et al., 2016).

Emissions of carbonaceous aerosol were the greatest contributor to $PM_{2.5}$ mass, with an average EF_{OC} of 6.77 g kg^{-1}. The OC was an average of 95% water insoluble, characteristic of fresh emissions from fossil fuel combustion. As with the zig-zag kiln emissions, EC was not detected by thermal-optical analysis. Consequently optically-determined BC, averaging 0.0172 g kg^{-1} (Stockwell et al., 2016) provides an estimate of the soot component of the smoke and yielded a BC-to-TC ratio of 0.0025. The BrC measurement by the PAX yielded an estimated OA (using the same average MAC as above) that was only 26% of our OC, suggesting that the MAC for these emissions was actually lower than average as expected for the low BC/TC ratio (Saleh et al., 2014).

The measured organic species accounted for an average of 9.1% of the OC. The dominant class of compounds detected was *n*-alkanes, which had an EF of 638 mg kg^{-1} for carbon numbers ranging from 18-35. The EF for 22 measured PAHs with three to six aromatic rings averaged 18.7 mg kg^{-1}, with the most abundant PAHs being chrysene, benz(a)anthracene, benzo(e)pyrene, and 1-methylcrysene. Picene—a molecular marker for coal combustion (Oros and Simoneit, 2000; Zhang et al., 2008)—was detected in all three clamp kiln samples, with an average EF of 0.53 mg kg^{-1}. In addition, hopanes that are present in coal and other fossil fuels (Oros and Simoneit, 2000; Zhang et al., 2008) were also detected (Table 3). The low emissions of levoglucosan (1.67 mg kg^{-1}) suggest that most of the hardwood had been consumed in the kiln before our sampling began.

In comparison to the batch-style kiln studied by Christian et al. (2010), the clamp kiln had substantially higher emissions of OC and lower MCE, both consistent with less complete combustion (Table 5). Like the zig-zag kiln, OC dominated EC in clamp kiln emissions. Clamp kilns were not studied by Weyant et al. (2014), although our $EF_{PM2.5}$ exceeded those from all seven kiln designs they studied, likely due to higher emissions of OC and sulfate as described in section 3.1.

3.3 Garbage burning

Emissions from five different garbage burning fires were characterized (Figure 2). The sample of waste burning at the household level under dry conditions (see Section 2.1.2) had an $EF_{PM2.5}$ of 7.4 ± 1.2 g kg^{-1} and an MCE value of 0.980 that indicated primarily flaming combustion. This $EF_{PM2.5}$ is similar to prior studies of garbage burning, including: i) waste burning in municipal landfills near Mexico City of 9.8 ± 5.7 g kg^{-1} (Akagi et al., 2011), ii) the open burning of military waste that had an average $EF_{PM2.5}$ of 19.4 g kg^{-1} (Woodall et al., 2012), assuming that 45% of the garbage was composed of carbon, following the recommendation of Wiedinmyer et al.

(2014), iii) household waste burning in a burn barrel with average $EF_{PM2.5}$ of 5.3 and 17.5 g kg^{-1} for avid recyclers and non-recyclers, respectively (Lemieux et al., 2000) and iv) the EF for total suspended particulate of 8 g kg^{-1} (Gerstle and Kemnitz, 1967) for open burning of municipal refuse in the U.S. EPA's Compilation of Air Pollutant Emission Factors (EPA, 1996). Because of the good agreement of this $EF_{PM2.5}$ with prior studies, this value is recommended as the emission factor for this source over the results from other garbage burning samples in this study (Table 1).

Much higher $EF_{PM2.5}$ were observed for garbage burning under damp conditions, which is not the typical case, but can be encountered at dump sites where the mixture of organic and inorganic waste ~~causes~~ creates damp conditions, under which the ~~garbage to be a bit damp and~~ fires smolder for a long time. For these samples, garbage had been dampened by rainfall the previous evening, making it difficult to ignite (requiring newspaper) and causing it to require re-ignition on occasion (Stockwell et al., 2016). Two samples from the same mixed waste fire produced $EF_{PM2.5}$ values of 124 ± 23 g kg^{-1} (MCE 0.889) and 82 ± 13 g kg^{-1} (MCE 0.926). The variation among these samples collected from the same fire is attributed to differences in the fire cycle (i.e. the extent of smoldering versus flaming). Aluminum foil-lined bags, burned under the same damp conditions, had $EF_{PM2.5}$ of 50 ± 9 g kg^{-1} (MCE 0.973), while plastic burning had an $EF_{PM2.5}$ of 84 ± 13 g kg^{-1} (MCE 0.951). These data demonstrate that emissions vary substantially with fuel composition, as shown by the variations between the mixed garbage and sorted trash burns as well as prior studies. $EF_{PM2.5}$ from garbage burning samples under damp conditions exceeds those burned under dry conditions by factors of 2.5-25. Because of the potential to decrease garbage burning emissions substantially by avoiding burning damp garbage, this trend should be further investigated.

The wide range of $EF_{PM2.5}$ observed herein, as evidenced by a relative standard deviation of 63% across the five garbage burning samples, suggests a high degree of variability across fires, which translates to large uncertainties in estimating emissions from this source. Because global garbage burning estimates of $PM_{2.5}$ reply upon the EF reported by Akagi et al. (2011) and the U.S. EPA compilation (EPA, 1996) to estimate the global impact of trash burning (Wiedinmyer et al., 2014), variability in $PM_{2.5}$ emissions is not well-represented and consequently emissions from this source may be either over- or underestimated. Further constraining the impact of garbage burning on ambient PM on national, regional, or global scales requires a better understanding of the amount of garbage burning in addition to the variability in EF for different fuel composition, moisture content, and burn conditions.

The major element present in $PM_{2.5}$ emitted from garbage burning was carbon, primarily in the form of OC. The chemical profile of $PM_{2.5}$ (Table 1; Figure 2) was estimated from the average emissions of the three mixed household garbage burning samples spanning samples collected under dry conditions (n=1) and wet conditions (n=2) and was 77% OC, 2.6% EC, and 1.5% chloride, with minor contributions (< 1%) from ammonium, ~~sodium,~~ potassium, fluoride, nitrate, and sulfate, and no detectable contributions from sodium, calcium, or magnesium (Table 1). OC:EC ratios for mixed garbage burning under damp conditions were 50 and 15 (EC was below detection limits in the sample burned under dry conditions), and overlapped the range for this ratio reported by Christian et al. (2010) for garbage burning in Mexico. ~~PAX based EF_{BC} were available for mixed garbage burning in Kathmandu under wet conditions (0.56 g kg^{-1}) and Tarai under dry conditions (6.04 g kg^{-1}), suggesting high variability in BC emissions, with the latter strongly BC dominated.~~ Chlorine in garbage burning is primarily emitted as HCl and results to a large degree from polyvinylchloride (PVC) plastics (Lemieux et al., 2000; Christian et al., 2010). In agreement with these prior studies; the majority of chlorine emitted from trash burning was initially in the gas phase as HCl (Stockwell et al., 2016), with 30% in the particle phase for mixed garbage burning under damp conditions and < 3% in the particle phase for mixed garbage burning under dry conditions. The bulk chemical signatures of burning foil wrappers and plastic were similar to mixed garbage in their dominance of OC, although they had higher mass fractions of EC.

Prior work has demonstrated that garbage burning has a unique signature of metals, making them useful in source identification and apportionment. For combustion sources in and around the Mexico City Metropolitan Area, Christian et al. (2010) reported antimony (Sb) in garbage burning at levels 555 times greater than biomass burning. For garbage burning emissions in Nepal, Sb was detected above field blank levels and method detection limits only in garbage burning emissions (Table 1) and ~~one~~ the traditional mud stove cooking fire, in which plastic was used for ignition. These results indicate that this element is unique to garbage burning, particularly plastic. In addition to Sb, mixed garbage burning emitted Cu, Pb, and other trace elements.

1,3,5-Triphenylbenzene (TPB) is proposed as a tracer of garbage burning emissions, due to its specificity to this source, high concentration in source emissions relative to other species, and detection in urban areas where garbage burning occurs (Simoneit et al., 2005). TBP was detected in all five garbage burning samples, with EF_{TPB} of 0.38-1.87 mg kg^{-1} for mixed waste burning, 0.27 mg kg^{-1} for foil wrappers, and 0.55 mg kg^{-1} for plastic bags. Meanwhile, TPB was not detected in any other combustion samples in this study, further emphasizing its

specificity to garbage burning. Mass normalized emissions of TPB were 12-51 μg gPM^{-1} for mixed waste, 5.3 μg gPM^{-1} for foil wrappers, and 6.5 μg gPM^{-1} for plastic burning. These values fall in the middle of the range of those reported by Simoneit et al. (2005) that were 0.2 μg gPM^{-1} for new polyethylene bags in the US and 57-208 μg gPM^{-1} for new plastic bags, roadside litter, and landfill trash in Chile. These comparisons demonstrate that TPB mass fractions can span three orders of magnitude, but may cover a much narrower range when measured in a single region. Thus, in using this tracer for source apportionment, it is recommended to use *in situ* emission factors developed within the region of study and that Sb and TPB be used in concert to provide inorganic and organic constraints to estimates of emissions from garbage burning.

The carbon fraction of the organic species measured in emissions from mixed garbage burning accounted for an average of 12% of the observed OC, with the largest contributions from levoglucosan (9.8%) marking the inclusion of cellulosic materials in the garbage, n-alkanes (1.8%), PAHs (0.2%), sterols (0.1%) and hopanes (<0.01%). The dominance of n-alkanes in garbage burning emissions is consistent with prior work by Simoneit et al. (2005) in Chile. The even-carbon preference characteristic of *n*-alkanes in polyethylene was lost during combustion due to thermal cracking (Simoneit et al., 2005), yielding carbon preference index (CPI) values in the range of 0.6-1.1.

EF for the 23 measured PAHs across the five garbage burns ranged from 15-152 mg kg^{-1}, with the minimum corresponding to mixed waste burning in Tarai and the maximum corresponding to plastic waste burning. Emissions of particle phase PAH from garbage burning are notably high from garbage burned under damp conditions in comparison to other sources (Ravindra et al., 2008), with maximum levels exceeding 1- or 2-pot traditional stoves in this study (38-56 mg kg^{-1}; Table S3) and the open burning of scrap tires, 56 mg kg^{-1} (Downard et al., 2015). Although the absolute EF_{PAH} were high, PAH accounted for < 0.2% of $PM_{2.5}$ mass, consistent with the other non-fossil fuel combustion sources in this study (Table S3). The combination of high PAH emissions and the health impacts of these compounds (e.g. carcinogenicity, teratogenicity) highlight the health risks associated with garbage burning. A number of other toxic, carcinogenic, and mutagenic chemicals associated with garbage burning that were not measured here, such as polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans (Lemieux et al., 2000), and nitro-PAH (Lee et al., 1995) also contribute to the hazards associated with exposure to garbage burning emissions.

3.4 Diesel and petrol generators

$EF_{PM2.5}$ was 9.2 ± 1.5 g kg^{-1} for the diesel generator and 0.8 ± 1.8 g kg^{-1} for the petrol powered generator (Figure 3a; Table S3). $PM_{2.5}$ concentrations in the sampled smoke plume from the petrol generator were not significantly greater than background PM levels, resulting in a high uncertainty. The observed EFs are near to the average values reported in the EPA Emission Factors (AP 42) for uncontrolled gasoline and diesel industrial engines of 6.0 g kg^{-1} and 2.0 g kg^{-1}, respectively (EPA, 1996). Recent studies have shown consistently lower $EF_{PM2.5}$ for US military diesel generators that exhibited an average (± standard deviation) of 1.2 ± 0.6 g kg^{-1} (Zhu et al., 2009). ~~A professionally maintained diesel generator on the ICIMOD campus in Nepal was observed to have a high MCE (0.998) (Stockwell et al., 2016) and likely a lower $EF_{PM2.5}$ than the rented diesel generator from which our filter sample was collected.~~ Although limited to one sample, the rented diesel generator studied in Nepal had a~~-~~ high $EF_{PM2.5}$ value and comparisons to other studies~~ suggest that well maintained generators have lower PM emissions~~.

Chemically, OC and EC accounted for the greatest fraction of $PM_{2.5}$ mass (Figure 3a). For the diesel generator, $PM_{2.5}$ was 80% OC and 6% EC. The predominance of OC and EC in diesel generator emissions is consistent with prior studies that showed their mass contributions in excess of 83% (Liu et al., 2005; Zhu et al., 2009). The diesel generator OC-to-EC ratio of 12.7 is in the range previously observed for a diesel generator running on high sulfur diesel at a relatively low load (0-25 kW) (Liu et al., 2005), although neither sulfur dioxide (Stockwell et al. 2016) nor sulfate was detected in these emissions. For the petrol generator, EC was not detected and the measured OC mass (after correction for gas adsorption to the filter) was 118% of $PM_{2.5}$ mass, which implies OC is the dominant chemical component, but indicates that positive artifacts remain despite the correction. In both diesel and petrol generators, OC was mostly insoluble in water (>73%), consistent with fresh combustion emissions and fuel and oil evaporation.

~~Measured o~~Organic species quantified by GCMS accounted for 12% of the OC emitted from the diesel generator, inclusive of n-alkanes (11%), ~~and~~ PAH (0.96%), and hopanes and steranes (0.13%). The n-alkanes with 22-23 carbons contributed the most to OC in diesel generator PM, compared to n-alkanes with 13-17 carbons dominating in diesel fuel (Liang et al., 2005). The observed species reflect both combustion (i.e. tailpipe emissions) and ~~hopanes and steranes together account for 0.13% of OC and reflect a small contribution of~~ engine oil evaporation ~~to OC emissions~~ (Schauer et al., 1999). For the petrol generator, only 3.8% of OC was attributed to organic species, primarily n-alkanes (0.6%). Meanwhile, EF of metals were very similar between the two

generator types, indicating that their emissions were independent of fuel type and probably were due to background PM and/or abrasion.

3.5 Groundwater pumps

Filter samples from groundwater pumps were collected after the pump had been turned on and reached continuous operating conditions~~steady-state operation~~. Thus, the reported EF do not include the initial start-up phase during which the pump was visually observed to emit puffs of black smoke (Stockwell et al., 2016). $EF_{PM2.5}$ for the groundwater pumps were 8.7 ± 0.7 g kg^{-1} for pump 1 (4.6 kVA model) and 5.5 ± 0.5 g kg^{-1} for pump 2 (5 kVA model) (Figure 3b; Table S3). The higher $EF_{PM2.5}$ of pump 1 is likely related to its age (approximately 3 years) and lower MCE (0.986) compared to pump 2 that was newer (less than 3 months of use) and had a higher MCE (0.996), since combustion at lower efficiency generates more PM per mass fuel burned. The magnitude of PM emissions from diesel groundwater pumps ~~were in good agreement with EF_{PM1} values reported by Goetz et al. (in preparation-a) of 9.2 and 5.2 g kg^{-1} and~~ were similar to the diesel generator in this study (section 3.4) and the EPA emission factor (AP 42) of 6.0 g kg^{-1} (EPA, 1996).

Chemical measurements indicated that the $PM_{2.5}$ was largely carbonaceous in nature (Table 1). ~~While the various measurement methods employed during NAMaSTE agreed well on the magnitude of $EF_{PM2.5}$ and the carbonaceous nature of the emissions, different methods had varying results with respect to the split between OC and EC fractions.~~ Filter-based measurements indicated that the average contributions to PM mass for OC and EC were 77 and 3.4%, respectively, and that OC was primarily water insoluble (≥ 88%). Further discussion on the light absorbing carbon fraction of diesel pump emissions and a comparison of measurement methods is provided elsewhere Goetz et al. (in preparation-a). ~~Meanwhile, the EF_{BC} reported by Stockwell et al. (2016) for pumps 1 and 2 were 6.13 and 5.31 g kg^{-1}, respectively and were comparable to the estimates of $EF_{PM2.5}$, which suggested that the PM was mostly BC. However, the PAX at 870 nm that only responds to BC was not operational that day and the PAX EF_{BC} were based on absorption at 405 nm, which can have a contribution from BrC. A large BrC contribution seemed unlikely due to the very low single scattering albedo (SSA), but some BrC absorption could have occurred. Further, Goetz et al. (in preparation-a) reported that pump 1 had a larger OA fraction than BC (0.64:0.35) while pump 2 had a lower OA fraction than BC (0.08:0.92) based on the AMS and aethalometer. Differences across methods are also expected due in part to the different stages of operation captured by each technique. The higher BC EFs from the optical instruments included sampling during start-up when high BC was seen visually and the filters reflect only the subsequent steady-state conditions. The relative importance of these~~

~~stages in normal use probably varies and is not known to us. In addition, differences stem from the use of different instruments and methods, and exemplify the complexity in reconciling substrate deposited versus *in situ* aerosol and chemical versus optical detectors. Because filters are more prone to sampling artifacts and only captured steady-state conditions, we refer to the PAX data reported by Stockwell et al. (2016) to represent the split between scattering and absorbing aerosol emissions over the operation cycle of the groundwater pumps. Their average SSA at 405 nm of 0.405 ± 0.137 corresponds to the ratio of scattering to total extinction and indicates that the absorption fraction of total extinction is 0.595, which is consistent with the average AMS split of 0.64 BC to 0.36 OA.~~

The carbon fraction of the organic species measured by GCMS accounted for an average of 3.2% of the OC emitted from the diesel groundwater pumps. n-Alkanes contributed the most to the speciated OC mass at 2.4%, with maximum contributions from those with 22-23 carbons, similar to the diesel generator. Fuel evaporation was reflected by the presence of hopanes (0.11%) and combustion indicated by PAHs (0.4%). On a species level, the two groundwater pumps had different PAH profiles, with pump 2 emitting PAH primarily in the lower molecular weight range (with maxima for phenanthrene and fluoranthene) and pump 1 emitting PAH with higher molecular weights (with a maximum emission of benzo(ghi)fluoranthene) like the diesel generator (section 3.4). Metals EFs were similar across both groundwater pumps, and more generally were consistent with EF from gasoline and diesel generators. Accordingly, they did not provide a unique metal signature allowing for distinction between generators and groundwater pumps.

3.6 Motorcycles – before and after servicing

Emissions from five motorcycles were evaluated while idling before and after servicing, which involved an oil change, cleaning air filters and spark plugs, and adjusting the carburetor. Because of the limited scope of the motorcycle emissions testing, both in terms of drive cycle and number of samples, the following data are neither representative of the diverse Kathmandu vehicle fleet nor their integrated emissions. Instead, we focus on the controlled variable in these tests, which is changes in emissions during idle as a result of servicing. $EF_{PM2.5}$ was 8.81 ± 1.33 g kg^{-1} before servicing and dropped considerably to 0.71 ± 0.45 g kg^{-1} after servicing (Figure 3c). OC, the major chemical component of emissions before servicing, dropped from 7.21 g kg^{-1} to 0.02 g kg^{-1} after servicing. Simultaneous decreases in hopanes (25 to 1 mg kg^{-1}), steranes (5.4 to 0.25 mg kg^{-1}), and *n*-alkanes (86.7 to 8.1 mg kg^{-1}) indicate that the reductions in OC are largely due to decreasing emissions of motor oil. Prior studies of vehicle emissions indicate that motor oil emissions originate in the crankcase (Zielinska et al., 2008),

suggesting that the engine service reduces the crankcase emissions, perhaps by removing old oil and cleaning of the filters. Meanwhile, other emissions categories were largely unchanged before and after servicing, including the measured PAH species (11.2 and 6.8 mg kg^{-1}), EC (0.39 and 0.31 g kg^{-1}), and metals (Table S3). Consequently, the source profiles for motorcycles before and after servicing are significantly different from one another, particularly with respect to their OC:EC, PAH:OC, and metal:PM ratios. Similar to gasoline-powered vehicles recently-serviced, well-functioning motorcycles have a different emissions profile than motorcycles lacking service (Lough et al., 2007).

Prior studies of motorcycles report condition-based EF (as g km^{-1} or g $start^{-1}$), which demonstrate that emissions and fuel consumption change under different speeds and conditions (Oanh et al., 2012). Consequently, driving condition-based EF cannot be directly compared to fuel-based emission factors (in units of g kg^{-1}) from idling vehicles. ~~To reconcile the difference in units and driving conditions~~Instead, we compare ratios of $EF_{PM2.5}$ to EF_{CO} determined herein to those from prior studies of vehicles under start-up, which is more comparable than EF under driving conditions (i.e., highway or street driving). The ratio of $PM_{2.5}$: CO (wt/wt) was 11.4 ‰ before servicing and 0.89 ‰ after servicing. The before-servicing value is quite similar to the 12.7 ‰ and 10.4 ‰ reported for motorcycle start-up by Oanh et al. (2012) for Hanoi and Shrestha et al. (2013) for Kathmandu, respectively, both using adjusted International Vehicle Emissions (IVE) EF. In contrast, the post-servicing value observed in this study is remarkably low, due to servicing significantly reducing emissions of PM, but slightly increasing CO (Stockwell et al., 2016).

~~The comparison of emissions~~Changes to motorcycle EF before and after servicing indicate~~s~~ that major reductions in $PM_{2.5}$, OC, and motor oil constituents in particular, may be achieved by vehicle servicing. In addition, Stockwell et al. (2016) demonstrated that servicing also has the benefit of reducing gaseous emissions of NO_X and non-methane hydrocarbons, amid slight increases in CO emissions. Follow up studies of individual motorcycles in Nepal (rather than the combined emissions from 5 motorcycles presented herein) have indicated that the major PM reductions we reported here were probably due to the servicing of one high emitting motorcycle (ICIMOD, unpublished data), suggesting that efforts to reduce $PM_{2.5}$ emissions from motorcycles should initially focus on high emitters. This approach is supported by the work of Zhang et al. (1995) on CO emissions from vehicles in Kathmandu and elsewhere that have demonstrated that high emitting vehicles account for a large fraction of fleet emissions and that high emitting vehicles generally lack maintenance and repair.

3.7 Emissions from the combustion of biofuels in cooking stoves and 3-stone cooking fires

$EF_{PM2.5}$ for the combustion of various biofuels in cooking stoves and 3-stone cooking fires are shown in Figure 4, while MCE are provided in tabular format in Table S3. Our discussion emphasizes the four field tests conducted in traditional mud stoves, which are considered to be the best representation of real-world cooking emissions from traditional mud stoves in this study. $EF_{PM2.5}$ determined from these field tests were 10.7 ± 1.6 g kg^{-1} for hardwood, 5.3 ± 0.8 g kg^{-1} for twigs, 14.5 ± 2.2 g kg^{-1} for dung (all in a 1-pot stove) and 15.0 ± 2.3 g kg^{-1} for a mixture of dung and hardwood (in a 2-pot stove). The magnitude of these values were up to 3 times higher than EF reported for traditional mud stoves by Venkataraman and Rao (2001) that ranged 2.8-4.8 g kg^{-1} for wood, biofuel briquettes, and dung that were diluted before sampling. The observed $EF_{PM2.5}$ for traditional mud stoves are greater than values compiled by Akagi et al. (2011) for $EF_{PM2.5}$ from open cooking that averaged 6.73 ± 1.61 g kg^{-1}), but were lower than the particulate carbon emissions reported by Keene et al. (2006) for dung burning (22.9 g kg^{-1}). In addition to fuel type, variability in $EF_{PM2.5}$ in cooking stove emissions have been attributed to the extent of flaming or smoldering combustion, with peak PM emissions occurring during the latter stage (Arora et al., 2014); dilution prior to PM collection (as discussed at the onset of section 3); rate of fuel consumption (Venkataraman et al., 2005); air flow through the stove (e.g., natural or forced draught); pot size and material (Gupta et al., 1998; Kar et al., 2012). The fact that field tests gave average $EF_{PM2.5}$ at the upper range of previously reported values is significant with respect to estimations of regional emissions from this stove type.

The comparison of emissions from 1 or 2 pot traditional mud stoves studied in the laboratory to those in the field showed that MCE was lower in the field samples (averaging 0.925) than in the lab samples (averaging 0.958) at a statistically significant level ($p = 0.01$). This suggests that field fires normally burn with a lower degree of combustion efficiency than in controlled studies~~, but is limited by the small data set (n = 4 for field tests and n = 4 for laboratory tests)~~. The decrease in combustion efficiency in the field compared to the laboratory has been previously reported for cooking stoves, particularly in the case of open fires, and is attributed to operator skill (Johnson et al., 2008; Jetter and Kariher, 2009; Roden et al., 2009). EF for $PM_{2.5}$, OC, and EC, however, were not significantly different across the field and laboratory samples ($p > 0.05$), although significant increases in PM emissions for stoves in the field compared to the laboratory have been demonstrated in larger cooking stove studies (Johnson et al., 2008; Roden et al., 2009). In comparison of the laboratory EF_{PM} to the literature, the reported values are elevated with respect to some previously reported values (Akagi et al., 2011; Venkataraman and Rao, 2001), but lower than other cases (Keene et al., 2006). MCE was strongly correlated with $PM_{2.5}$ for the biofuel laboratory tests ($r = -0.959$; n=16; Figure 5), excluding charcoal and biogas fuels. When including the 3-

stone fire burning dung (with an exceptionally high $EF_{PM2.5}$ 72.7 g kg^{-1} and MCE of 0.863) this correlation increased slightly (r = -0.979). In contrast, EFs for PM were only weakly correlated with MCE in the four field-based tests (r=-0.394); this makes it difficult to determine how much of the difference between lab and field is due to differences in combustion state (smoldering versus flaming). For this dataset, simply estimating $EF_{PM2.5}$ from MCE using relationships developed in the laboratory would overestimate $EF_{PM2.5}$ in the field.

The use of dung, or a mixture of dung and wood, consistently gave higher $EF_{PM2.5}$ than burning wood alone for both field-based and laboratory studies (Figure 4). The higher EF_{PM} from dung compared to wood has been observed previously for fuel-based and energy-based EF (Venkataraman and Rao, 2001; Sheesley et al., 2003; Keene et al., 2006; Oanh et al., 1999; Saud et al., 2013). The induced-draught stove when burning charcoal emitted less PM than a mixture of hardwood and dung (Figure 4), consistent with prior studies that demonstrated that charcoal leads to relatively low PM emissions (Kshirsagar and Kalamkar, 2014). Likewise, biobriquettes have been shown to have lower EF_{PM} compared to wood and dung (Oanh et al., 1999; Sheesley et al., 2003). Among the cooking fuels we measured, biogas had the lowest $EF_{PM2.5}$ overall, but is not widely used. Together, results from this and prior studies demonstrate that on a per mass-of-fuel basis, dung is a high PM emitter, followed by wood, biobriquettes, and charcoal, with biogas providing the lowest PM emissions.

The control of fuel burned in the laboratory allows for comparison across different stove designs and 3-stone cooking fires. In the case of hardwood, the highest $PM_{2.5}$ emissions were observed for the 3-stone cooking fire (7.6 g kg^{-1}), followed by the 1-pot traditional mud stove (4.9 ± 0.9 g kg^{-1}), chimney stove (3.0 ± 0.5 g kg^{-1}), rocket stove (1.47± 0.4 g kg^{-1}), and the forced-draught stove (1.2 ± 0.5 g kg^{-1}). As the $EF_{PM2.5}$ for hardwood decreases, the MCE increases (Table S2) suggesting that the smoldering conditions contribute to the greater emissions of $PM_{2.5}$. When dung was used as fuel, the 3-stone cooking fire again generated the highest $EF_{PM2.5}$ (73 ± 11 g kg^{-1}) followed by the 1-pot traditional mud stove (20 ± 3 g kg^{-1}). More generally, and considering the breadth of the fuels studied, the comparisons of different cooking stoves and cooking fires revealed the highest PM emissions from 3-stone cooking fires (7.6-73 g kg^{-1}), followed by traditional mud stoves (5.3-19.7 g kg^{-1}), mud stoves with a chimney for exhaust (3.0-6.8 g kg^{-1}), and then rocket (1.5-7.2 g kg^{-1}), induced-draught stoves (1.2-5.7 g kg^{-1}), and bhuse chulo (3.2 g kg^{-1}), while biogas had no detectable PM emissions. The observed trends across stove types are consistent with prior studies of cooking stoves. Here and in prior studies, biogas holds advantages over traditional cooking stoves in terms of the global warming potential of emissions and provides a viable and cleaner-emissions alterative to the direct combustion of dung as fuel (Smith et al., 2000). Several prior

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studies have also documented that vented, natural-draught, and forced-draught stoves provide lower PM emissions (Smith et al., 2000; Jetter and Kariher, 2009; Jetter et al., 2012).

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The PM emitted from biofuel burning was primarily carbonaceous matter (Figure 4; Table 3). For the four field tests of traditional mud stoves, $PM_{2.5}$ mass was comprised of 49-68% OC and 3.3-18% EC (Table S2). On average, 34±3% of OC was water-soluble, with the majority being water insoluble. Ratios of OC:EC ranged from 2.8 to 21, with the greatest values corresponding to the use of dung as fuel. This range of OC:EC values and trend with maximum OC:EC occurring for dung cake are consistent with prior studies of similar fuel types in the IGP (Saud et al., 2013; Deka and Hoque, 2015). Major inorganic ions contributing to $PM_{2.5}$ mass include potassium (0.5-1.8%), ammonium (0.8-5.3%), and chloride (2.4-9.2%), with minor contributions (< 0.6%) from sodium, ~~calcium,~~ fluoride, nitrate, and sulfate. The largest mass fractions of ammonium and chloride in $PM_{2.5}$ were observed for fuels blends that included dung. Chlorides in $PM_{2.5}$ emitted from biofuel burning are primarily in the form of water-soluble salts (Keene et al., 2006; Sheesley et al., 2003). In emissions involving dung, ammonium is the dominant counter ion to chloride, while both ammonium and potassium contribute appreciably as counter ions to chloride in $PM_{2.5}$ emissions from wood. This difference in chloride salt composition is derived from dung having a significantly higher mass fraction of nitrogen compared to grasses and wood fuels (Keene et al., 2006). In addition, dung burning had higher mass contributions for chloride, while wood, twig, and agricultural residue burning had relatively more potassium. Charcoal burning PM was particularly enriched in potassium (~~31~~28±7% by mass) and sulfate (~~23~~21±6% by mass), in contrast to the other studied fuels that had lower mass fractions of these ions. For 19 of 24 biofuels, the sum of the measured PM components was less than the measured $PM_{2.5}$ mass and non-carbon elements associated with organic matter (i.e., hydrogen, oxygen, nitrogen) are expected to make up the majority of this difference. In the case of hardwood burning in the rocket stove, hardwood burning in the forced-draught stove, and biobriquettes in the forced-draught stove with an electrical charger under ignition and cooking conditions—all of which had relatively low $PM_{2.5}$ emissions in comparison to other stove types—the measured OC exceeded the measured $PM_{2.5}$ mass by a factor of three, suggesting that the measured OC was overestimated, perhaps due to gas adsorption. Because organic gas adsorption affects QFF but not Teflon filters, the $EF_{PM2.5}$ measurement for these stove types is considered valid.

Organic molecular markers provide additional means of chemically distinguishing between $PM_{2.5}$ emissions from different fuel types. Sheesley et al. (2003) found that cow dung burning uniquely emits three stanols—5β-stigmastanol, coprostanol, and cholestanol—that are characteristic of anaerobic microbial reduction that occurs

during digestion in higher animals. In this study, 5β-Stigmastanol, was detected in emissions from combustion of hardwood as well as twigs (Figure 6) indicating that either this molecule is not unique to dung burning or the GCMS measurement method used in this study were unable to distinguish between 5α– and 5β-stigmastanol, of which the former has been reported in wood smoke (Fine et al., 2001). Consequently, we do not consider 5β-stigmastanol to be a unique marker for dung burning. Coprostanol and cholestanol are diastereomers that co-eluted from the GC column and had identical mass spectra, so they were quantified together. Coprostanol and/or cholestanol were uniquely detected in $PM_{2.5}$ emitted from dung burning (Figure 6, Table 4), further supporting that these species are unique molecular markers of this source. As a mass fraction of OC, coprostanol and cholestanol emissions from traditional mud stoves ranged 0.15-0.27 mg gOC^{-1}; these values are one order of magnitude lower than those reported by Stone et al. (2010) for cow dung cake burning in a traditional mud stove and are nearly two orders of magnitude lower than those reported by Sheesley et al. (2003) for a catalyst-equipped wood stove. Meanwhile, levoglucosan—a biomass burning marker (Simoneit et al., 1999)—was emitted at comparable levels from all three studies, suggesting that stanol emissions are particularly sensitive to dung burning conditions in comparison to levoglucosan. Due to their specificity, coprostanol and cholestanol are recommended for use as molecular markers of dung combustion; however source apportionment will be sensitive to the dung burning profile used, due to the high variability in the marker-to-OC ratios, and thus sensitivity testing to the input dung burning profile is recommended.

3.8 Open burning of biomass: crop residue and heating fires

One sample was collected from the co-firing of several crop residue fuel types, including rice, wheat, mustard, lentils, and grasses during the pre-monsoon in the Tarai. $EF_{PM2.5}$ was 11.5 ± 2.2 g kg^{-1}. The corresponding gas-phase data for this mixed crop residue fire may be found in Stockwell et al. (2016; column B in their Supplemental Table S9). The majority of PM mass was explained as OC (55%), EC (8.~~5~~6%), chloride (10%), potassium (7.2%), ammonium (2.5%), and nitrate (2.5%) (Figure 4). A relatively high mass fraction of chloride was observed and, combined with the non-detection of HCl in the gas phase (Stockwell et al., 2016), this indicates that particle-phase chloride was the major form. In addition, higher concentrations of levoglucosan and other biomarkers were present in emissions from this source, although no unique marker species were identified among those reported in Table 3. These data expand both the number and chemical detail of prior emissions measurements of agricultural fires in the IGP (Rajput et al., 2014a; Rajput et al., 2014b; Singh et al., 2014).

Open burning was also examined in the form of a heating fire, in which dung and twigs were burned outdoors in a pile as a means of generating heat. $EF_{PM2.5}$ was 20.0 ± 1.4 g kg^{-1}. Two factors are likely to contribute to this relatively high $EF_{PM2.5}$: the inclusion of dung as fuel, which generates more PM than wood fuels (Section 3.7) and the low MCE value (0.861) that corresponds to relatively more smoldering. OC comprised 64.9% of $PM_{2.5}$, while EC contributed 0.~~43~~40%; the high OC:EC ratio (~150) ~~is~~ also ~~characteristic~~ indicates ~~of~~ smoldering combustion conditions. Additionally, this fire contained dung burning tracers coprostanol and cholestanol, lower amounts of levoglucosan relative to wood burning (but values on par with dung-fueled cooking), and a relatively high ratio of ammonium to potassium. This source profile ~~is considered to be representative of~~provides insight to open co-burning of dung and fuel wood under smoldering conditions in the Tarai.

3.9 Potential applications of emission factors and source profiles

The fuel-based EFs generated in NAMaSTE (Tables 1-4, Table S3) have several potential applications. First, when combined with activity data (i.e. mass consumption of fuels), emissions inventories specific to Nepal and the IGP may be generated. The use of locally- and regionally-specific EFs are expected to provide a more accurate representation of sources and are expected to improve air quality and climate models for the region. Alternatively, emissions inventories using global average values can be based on more data. Energy-based EF (mass of pollutant per energy output) can be calculated from these EF (mass of PM per mass of fuel) and fuel energy densities (energy per mass of fuel). Second, detailed chemical profiles may be used in receptor-based source apportionment modeling following the chemical mass balance approach (Schauer et al., 1996; Stone et al., 2010). This model requires that the input source profiles represent sources likely to impact the receptor location. The source profiles presented herein depict *in situ* emissions from many important, and previously undercharacterized sources, and therefore are considered to be the most representative source profiles for many sources in Nepal and South Asia. When apportioning OC based on organic tracers, highly source specific tracers will be useful in the delineation of regionally-important sources (e.g. TPB and Sb from garbage burning, coprostanol and cholestanol for dung burning). Third, when combined with gas-phase emissions data from Stockwell et al. (2016), acute to chronic health risks may be assessed among the major gaseous and particle-phase species emitted. Through these intended applications, these emissions data can contribute to a better understanding of air quality, PM sources, and their impacts on human health.

Source-averaged $EF_{PM2.5}$ and composition data provided in Tables 1-4 are intended for use in the above-mentioned applications. Notably, the relative errors in $PM_{2.5}$ and OC mass have been incorporated into the errors reported for bulk chemical constituents and organic species shown as ratios, respectively. Use of these values should maintain the reported relative errors (in parenthesis in Tables 1-4) and should not be propagated to include errors in $EF_{PM2.5}$ or EF_{OC}, as this would be redundant.

4 Conclusions

We report $EF_{PM2.5}$ for a number of different widespread and under-sampled combustion sources in Nepal, including brick kilns, garbage burning, diesel and gasoline generators, diesel groundwater pumps, traditional and modern cooking stoves, crop residue burning, and open burning of biofuels. These data expand the understanding of combustion emissions in a number of ways. First, we provide the first EF_{PM} for diesel groundwater pumps that are prevalent in South Asia. Second, we add to the body of literature on PM emissions for brick kilns, garbage burning, generators, cooking stoves, and open biomass fires, in many cases expanding the chemical detail that is known about PM composition. Third, we confirm that molecular and elemental tracers identified in previous studies are applicable to South Asian combustion sources, namely Sb and TPB for garbage burning and coprostanol and cholestenol for dung burning, which are useful in source identification and apportionment. Fourth, through the study of motorcycle emissions before and after servicing, we demonstrate that significant PM reductions may be achieved by servicing. Fifth, our data suggests that burning of wet garbage substantially increases PM emissions relative to dry garbage, which warrants further investigation. Finally, NAMaSTE is the first to provide a detailed chemical characterization of *in situ* combustion emissions from within Nepal, providing locally- and regionally-specific emissions data. ~~PM composition measurements provide chemically-detailed profiles of major PM components (i.e. OC, EC, water-soluble inorganic ions) as well as trace elements and organic species. For brick kilns, garbage burning, diesel groundwater pumps, and biofuel combustion, which are widespread sources of air pollution in South Asia, we provide the first detailed chemical characterization of $PM_{2.5}$. For other sources (i.e. cooking stoves, agricultural residue burning), our detailed PM measurements extend what is known about composition for these sources.~~ Co-located, size-resolved emissions measurements of these sources by AMS provides further chemical insight into aerosol composition (Goetz et al., in preparation-a, b). In combination with co-located measurements reported by Stockwell et al. (2016) that include aerosol optical properties (EF for scattering and absorption, single scattering albedo, and absorption Ångström exponent) and EF

for ~80 important gases, a chemically and physically thorough analysis of the sampled combustion emissions is provided.

With a focus on detailed characterization of under-studied source sectors, NAMaSTE ~~likely~~ does not fully capture the broad diversity of combustion sources in the IGP and South Asia. This is partly because NAMaSTE was reduced in scope in response to the Gorkha earthquake, resulting in fewer replicates and numbers of sources studied. Analyses of rapidly-changing vehicle fleets, particularly under driving conditions found in the region, are needed to better constrain emissions from this source sector. For other source categories, further field-based studies are needed to better understand source variability and diversity. In particular, the inherent heterogeneity in garbage composition and apparent sensitivity of its emissions to combustion conditions such as moisture content warrants further inquiry. Likewise, moisture affects emissions from biomass, especially in open burning of wood and crop residues. The present and future improvements to understanding emissions in this region will provide a more accurate representation of air pollution sources within South Asia and can support updates to emissions inventories, improvements to regional air quality and climate models, and assessments of air quality impacts on health.

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Table 1: Summary of emissions data for select combustion sources, including modified combustion efficiency (MCE), emission factors for $PM_{2.5}$ (g kg^{-1}), and fine particle composition (as $PM_{2.5}$ weight percent). Errors are shown in parenthesis; a description of their calculation is provided in section 3. Missing values are below method detection limits. For sources represented by a single sample, errors were propagated from analytical uncertainties. For sources represented by replicate samples, errors were calculated as one standard deviation of the mean.

Combustion Source	**Induced-draught zig-zag brick kiln**		**Clamp brick kiln**		**Garbage burning**		**Generator**		**Generator**		**Groundwater pump**		**Motorcycles - before servicing[1]**		**Motorcycles- after servicing[1]**	
Fuel	Coal, bagasse		Coal, hardwood		Mixed waste		Diesel		Gasoline		Diesel		Gasoline		Gasoline	
Number of samples	3		3		3		1		1		2		1		1	
MCE	0.994		0.952		0.931		0.980		0.390		0.991		0.603		0.582	
EF $PM_{2.5}$ (g kg^{-1})	15.11	(3.69)	10.66	(2.70)	7.37	(1.22)	9.17	(1.51)	0.77	(1.80)	7.12	(2.27)	8.81	(1.33)	0.71	(0.33)
Fine particle composition (weight percent of $PM_{2.5}$)																
Elemental carbon (EC)	0.74[3]		0.16[3]		2.6	(2.0)	6.3	(1.3)			3.4	(1.9)	4.5	(0.8)	43.5	(27.8)
Organic carbon (OC)	7.0	(3.3)	63	(5)	77	(32)	80	(18)	118	(91)	77	(3)	82	(15)	3	(12)
Water-soluble inorganic ions																
Ammonium (NH_4^+)	0.294	(0.126)	16.0	(3.1)	0.958	(0.314)	0.583	(0.770)			0.269	(0.029)	0.188	(0.183)	2.71	(2.45)
Potassium (K^+)	0.0070	(0.0001)	0.38	(0.14)	0.156	(0.190)										
Fluoride (F^-)	0.011	(0.006)			0.139	(0.087)										
Chloride (Cl^-)	0.065	(0.045)	5.7	(0.3)	1.48	(0.61)										
Nitrate (NO_3^-)	0.143	(0.154)	2.0	(1.1)	0.78	(1.05)										
Sulfate (SO_4^{2-})	31.92	(3.79)	23.4	(5.5)	0.465	(0.532)										
Metals																
Nickel (Ni)	0.001	(0.004)														
Copper (Cu)					0.004	(0.001)										
Arsenic (As)	0.0007	(0.0004)	0.017	(0.013)	0.001	(0.001)										
Selenium (Se)	0.0055	(0.0001)			0.002	(0.001)										
Cadmium (Cd)	0.00004	(0.00002)	0.00016	(0.00012)	0.001	(0.002)										
Antimony (Sb)					0.025	(0.033)										
Lead (Pb)	0.003	(0.001)	0.005	(0.003)	0.057	(0.077)										

1) Combined emissions of five motorcycles; servicing included an oil change, cleaning air filters and spark plugs, and adjusting the carburetor

2) This value is expected to include hygroscopic water, see section 3.1 for the estimated value that excludes water.

3) Estimated from optical measurements of black carbon from Stockwell et al. (2016)

Table 2: Summary of emissions data for biofuel combustion sources, including modified combustion efficiency (MCE), emission factors for $PM_{2.5}$ (g kg^{-1}), and fine particle composition (as $PM_{2.5}$ weight percent). Errors are shown in parenthesis; a description of their calculation is provided in section 3. Missing values are below method detection limits. For sources represented by a single sample, errors were propagated from analytical uncertainties. For sources represented by replicate samples, errors were calculated as one standard deviation of the mean.

Combustion Source	**Traditional mud cooking stove**		**Traditional mud cooking stove**		**Agricultural fire**		**Open burning**	
Fuel	Wood		Wood, dung		Crop residues[1]		Dung, twigs	
Number of samples	2		2		1		1	
MCE	0.931		0.919		0.934		0.861	
EF $PM_{2.5}$ (g kg^{-1})	7.97	(3.80)	14.73	(0.33)	11.48	(1.92)	20.00	(3.06)
Fine particle composition (weight percent of $PM_{2.5}$)								
Elemental carbon (EC)	14	(5)	5.1	(2.3)	8.5	(1.94)	0.43	(0.13)
Organic carbon (OC)	52	(5)	61	(10)	55	(13)	65	(7)
Water-soluble inorganic ions								
Sodium (Na^+)	0.048	(0.066)	0.385	(0.350)				
Ammonium (NH_4^+)	1.12	(0.44)	4.46	(1.25)	2.54	(0.77)	1.854	(0.383)
Potassium (K^+)	1.78	(0.04)	0.520	(0.083)	7.22	(1.62)	0.804	(0.200)
Fluoride (F^-)	0.081	(0.016)	0.039	(0.009)			0.018	(0.022)
Chloride (Cl^-)	3.20	(1.07)	8.58	(0.86)	10.01	(2.17)	3.709	(0.679)
Nitrate (NO_3^-)	0.423	(0.125)	0.209	(0.216)	2.50	(0.62)	0.541	(0.140)
Sulfate (SO_4^{2-})	0.334	(0.194)	0.456	(0.040)	0.415	(0.818)	0.297	(0.269)
Metals								
Nickel (Ni)					0.017	(0.012)	0.005	(0.004)
Copper (Cu)	0.005	(0.004)	0.001	(0.001)				
Arsenic (As)	0.004	(0.002)	0.001	(0.000)				
Selenium (Se)	0.006	(0.004)						
Cadmium (Cd)	0.002	(0.002)	0.001	(0.000)	0.001	(0.000)	0.001	(0.000)
Antimony (Sb)	0.005[2]	(0.006)						
Lead (Pb)	0.007	(0.007)	0.004	(0.001)				

1) Rice, wheat, mustard, lentil, and grasses
2) Plastic was used to ignite this fire

Table 3: Summary of emissions data for select combustion sources with respect to organic species normalized to organic carbon mass (mg gOC^{-1}). Errors are shown in parenthesis; a description of their calculation is provided in section 3. Missing values are below method detection limits, which are provided sample-by-sample in Table S1.

Combustion Source	**Induced-draught zig-zag brick kiln**		**Clamp brick kiln**		**Garbage burning**		**Generator**		**Generator**		**Groundwater pump**		**Motorcycles - before servicing[1]**		**Motorcycles-after servicing[1]**	
Fuel	Coal, bagasse		Coal, hardwood		Mixed waste		Diesel		Gasoline		Diesel		Gasoline		Gasoline	
Number of samples	3		3		3		1		1		2		1		1	
Polycyclic aromatic hydrocarbons																
Phenanthrene	0.02	(0.00)	0.01	(0.00)	0.09	(0.06)	0.012	(0.005)	0.09	(0.04)	0.37	(0.43)	0.010	(0.003)	1.47	(0.45)
Anthracene			0.01	(0.00)	0.02	(0.01)	0.007	(0.002)			0.10	(0.11)	0.007	(0.002)	0.20	(0.12)
Fluoranthene	0.04	(0.01)	0.08	(0.03)	0.20	(0.13)	0.03	(0.01)	0.04	(0.02)	0.73	(0.55)	0.09	(0.02)	6.33	(1.54)
Pyrene	0.01	(0.00)	0.11	(0.06)	0.24	(0.16)	0.09	(0.02)	0.09	(0.02)	0.56	(0.14)	0.14	(0.03)	15.6	(3.8)
Methylfluoranthene			0.21	(0.11)	0.06	(0.01)					0.09	(0.00)				
9-Methylanthracene			0.02	(0.01)	0.03	(0.03)	0.04	(0.01)	0.08	(0.07)	0.05	(0.03)	0.004	(0.003)	0.76	(0.52)
Benzo(ghi)fluoranthene			0.13	(0.07)	0.21	(0.14)	2.62	(0.60)	0.19	(0.07)	0.38	(0.28)	0.30	(0.07)	76.1	(18.4)
Cyclopenta(cd)pyrene			0.09	(0.05)	0.09	(0.08)	0.17	(0.04)					0.26	(0.06)	42.7	(10.4)
Benz(a)anthracene			0.37	(0.23)	0.11	(0.07)	0.74	(0.17)			0.18	(0.18)	0.04	(0.01)	5.27	(1.31)
Chrysene			0.43	(0.10)	0.16	(0.08)	1.35	(0.31)	0.09	(0.04)	0.16	(0.15)	0.05	(0.01)	6.88	(1.68)
1-Methylchrysene			0.22	(0.04)			0.06	(0.01)							0.27	(0.24)
Retene	0.03	(0.02)	0.09	(0.01)	0.20	(0.26)							0.002	(0.004)		
Benzo(b)fluoranthene			0.18	(0.08)	0.12	(0.07)	1.14	(0.26)	0.03	(0.09)	0.24	(0.06)	0.05	(0.01)	11.8	(2.9)
Benzo(k)fluoranthene			0.14	(0.04)	0.10	(0.07)	1.04	(0.24)	0.01	(0.08)	0.20	(0.05)	0.04	(0.01)	8.81	(2.21)
Benzo(j)fluoranthene			0.03	(0.03)	0.04	(0.03)	0.05	(0.01)			0.04	(0.01)	0.01	(0.00)	0.44	(0.39)
Benzo(e)pyrene			0.23	(0.09)	0.10	(0.07)	0.98	(0.22)	0.08	(0.06)	0.25	(0.06)	0.07	(0.02)	23.5	(5.7)
Benzo(a)pyrene			0.15	(0.07)	0.10	(0.06)	0.25	(0.06)			0.13	(0.03)	0.06	(0.01)	16.5	(4.0)
Perylene			0.05	(0.03)	0.01	(0.01)	0.05	(0.01)			0.005	(0.004)	0.04	(0.01)	3.55	(0.91)
Indeno(1,2,3-cd)pyrene			0.07	(0.01)	0.08	(0.05)	0.66	(0.15)	0.14	(0.04)	0.29	(0.07)	0.09	(0.02)	23.8	(5.8)
Benzo(GHI)perylene			0.08	(0.03)	0.09	(0.06)	0.69	(0.16)	0.82	(0.20)	0.27	(0.06)	0.27	(0.06)	82.6	(20.0)
Dibenz(ah)anthracene			0.03	(0.01)	0.02	(0.02)	0.05	(0.01)			0.04	(0.01)			0.37	(0.66)
Picene			0.08	(0.03)	0.02	(0.03)	0.05	(0.01)								
Triphenylbenzene					0.030	(0.013)										
Tricyclic terpanes																
17α(H)-22,29,30-Trisnorhopane			1.00	(0.34)	0.01	(0.00)	0.13	(0.03)			0.09	(0.07)	0.22	(0.05)	3.57	(0.87)
17β(H)-21α(H)-30-Norhopane	0.02	(0.02)	1.14	(0.37)	0.04	(0.01)	0.29	(0.07)			0.21	(0.05)	0.70	(0.16)	7.58	(1.96)
17α(H)-21β(H)-Hopane	0.02	(0.01)	1.24	(0.42)	0.06	(0.04)	0.24	(0.06)	0.02	(0.10)	0.22	(0.01)	0.84	(0.19)	12.8	(3.3)
22(S)-Homohopane			0.42	(0.12)			0.17	(0.04)			0.11	(0.07)	0.42	(0.10)	8.30	(2.02)
22(R)-Homohopane			0.37	(0.12)			0.16	(0.04)			0.09	(0.08)	0.37	(0.08)	8.02	(1.95)
22(S)-Bishomohopane			0.29	(0.03)			0.11	(0.03)			0.06	(0.04)	0.32	(0.07)	7.88	(1.91)
22(R)-Bishomohopane			0.27	(0.05)			0.10	(0.02)			0.07	(0.04)	0.27	(0.06)	8.00	(1.94)
22(S)-Trishomohopane			0.12	(0.02)			0.05	(0.01)					0.19	(0.04)		
22(R)-Trishomohopane			0.08	(0.02)			0.04	(0.01)					0.15	(0.03)		
αββ-20(R)-C27-Cholestane			0.07	(0.00)							0.05	(0.01)	0.05	(0.01)		
αββ-20(S)-C27-Cholestane											0.07	(0.02)	0.08	(0.02)		
ααα-20(S)-C27-Cholestane							0.06	(0.02)			0.04	(0.01)	0.11	(0.03)		
αββ-20(R)-C28-Ergostane							0.02	(0.01)			0.06	(0.01)	0.10	(0.02)	2.13	(0.55)
αββ-20(S)-C28-Ergostane							0.03	(0.01)			0.06	(0.01)	0.09	(0.02)	1.40	(0.39)
αββ-20(R)-C29-Sitostane							0.06	(0.01)			0.10	(0.09)	0.20	(0.05)	5.01	(1.24)
αββ-20(S)-C29-Sitostane							0.04	(0.01)			0.07	(0.06)	0.12	(0.03)	3.52	(0.90)

Alkanes																
Pristane			0.17	(0.07)	0.38	(0.15)	1.01	(0.38)			1.85	(1.54)	0.14	(0.11)		
Norpristane	0.02	(0.07)	0.03	(0.02)	0.22	(0.20)	0.10	(0.09)			0.23	(0.21)	0.05	(0.04)		
Phytane	0.02	(0.15)	0.04	(0.03)	0.04	(0.02)	0.07	(0.14)	1.27	(1.57)	0.13	(0.04)	0.01	(0.05)		
Squalane	0.09	(0.03)	1.64	(0.15)	0.35	(0.20)	1.00	(0.38)	0.25	(2.82)	0.33	(0.45)	0.04	(0.10)	11.0	(20.0)
Octadecane	0.01	(0.05)	0.11	(0.16)	0.33	(0.10)	0.08	(0.08)			0.23	(0.19)	0.04	(0.03)	0.33	(5.33)
Nonadecane			0.18	(0.07)	0.38	(0.14)	1.02	(0.37)			1.87	(1.75)	0.15	(0.10)		
Eicosane			1.42	(0.17)	0.69	(0.12)	6.44	(1.59)	0.53	(3.13)	2.42	(1.07)	0.58	(0.20)	31.7	(24.5)
Heneicosane	0.06	(0.03)	3.36	(0.18)	0.68	(0.20)	18.55	(4.27)	0.93	(0.53)	4.02	(1.90)	0.79	(0.18)	23.4	(6.9)
Docosane			4.01	(0.29)	0.77	(0.03)	24.54	(5.92)			4.15	(1.75)	0.83	(0.41)	7.8	(60.8)
Tricosane	0.34	(0.14)	7.48	(0.15)	1.32	(0.19)	24.35	(5.72)	1.08	(3.91)	4.89	(0.68)	1.27	(0.36)	81.3	(37.5)
Tetracosane	0.32	(0.26)	8.65	(0.97)	1.80	(0.30)	19.30	(4.63)	7.43	(8.24)	3.10	(1.65)	1.37	(0.46)	29.7	(55.1)
Pentacosane	0.47	(0.11)	8.78	(1.31)	1.42	(0.63)	13.40	(3.46)			1.76	(0.92)	0.76	(0.48)	39.5	(80.4)
Hexacosane	0.32	(0.10)	6.96	(0.57)	1.59	(0.50)	6.71	(2.14)			0.74	(0.09)	0.61	(0.51)	40.8	(89.9)
Heptacosane	0.26	(0.08)	8.54	(0.54)	1.94	(0.98)	4.79	(1.84)	1.50	(13.64)	1.52	(1.95)	0.73	(0.55)	53.5	(96.9)
Octacosane	0.74	(0.24)	9.41	(0.54)	1.10	(0.81)	3.93	(1.33)	1.53	(8.71)	0.42	(0.88)	0.07	(0.32)	13.2	(60.1)
Nonacosane	0.63	(0.25)	9.16	(0.89)	1.66	(0.66)	2.25	(1.05)	2.88	(8.91)	0.22	(0.87)	0.44	(0.35)	20.9	(61.5)
Triacontane	0.45	(0.16)	6.68	(1.41)	1.38	(0.61)	1.06	(0.76)	0.60	(7.28)	0.09	(0.72)	0.46	(0.31)	11.3	(50.6)
Hentriacontane	0.35	(0.34)	7.10	(1.35)	1.05	(0.64)	0.78	(0.62)	2.14	(6.14)			0.53	(0.27)	0.13	(41.37)
Dotriacontane	0.27	(0.21)	4.69	(0.71)	1.04	(0.45)	0.40	(0.38)	1.41	(3.84)			0.09	(0.14)	9.9	(26.5)
Tritriacontane	0.18	(0.08)	3.90	(0.32)	1.31	(0.89)	0.23	(0.35)	1.37	(3.73)	0.06	(0.36)			5.6	(25.4)
Tetratriacontane	0.28	(0.13)	2.66	(0.18)	1.40	(0.76)	0.31	(0.19)	2.95	(2.05)			1.78	(0.42)	20.1	(14.1)
Pentatriacontane			1.60	(0.31)	1.17	(0.71)	0.26	(0.19)	2.19	(2.02)			1.52	(0.36)		
Levoglucosan	1.6	(1.3)	0.2	(0.1)	98.5	(49.2)	0.5	(0.4)	9.3	(4.6)	2.8	(1.4)	0.6	(0.2)	119	(41)
Sterols and Stanols																
Cholesterol			1.53	(0.21)	0.19	(0.00)										
Stigmasterol					0.15	(0.01)	0.21	(0.07)	1.83	(0.71)						
b-Sitosterol					0.71	(0.15)	0.56	(0.30)	2.65	(2.84)	0.50	(0.47)	0.12	(0.08)	26.5	(20.6)
Campesterol					0.15	(0.01)										
Cholestanol and coprostanol																
Stigmastanol					0.03	(0.25)										

1) Combined emissions of five motorcycles; servicing included an oil change, cleaning air filters and spark plugs, and adjusting the carburetor

Table 4: Summary of emissions data for biofuel combustion sources with respect to organic species normalized to organic carbon mass (mg gOC^{-1}). Tricyclic terpanes were not detected. Errors are shown in parenthesis; a description of their calculation is provided in section 3. Missing values are below method detection limits, which are provided sample-by-sample in Table S1.

Combustion Source	**Traditional mud cooking stove**		**Traditional mud cooking stove**		**Agricultural fire**		**Open burning**	
Fuel	Wood		Wood, dung		Crop residues[2]		Dung, twigs	
Polycyclic aromatic hydrocarbons								
Phenanthrene	0.14	(0.11)	0.18	(0.15)	0.03	(0.01)	0.04	(0.01)
Anthracene	0.06	(0.05)	0.11	(0.12)	0.017	(0.004)		
Fluoranthene	0.94	(0.03)	0.58	(0.19)	0.24	(0.06)	0.16	(0.04)
Pyrene	1.16	(0.07)	0.55	(0.32)	0.26	(0.06)	0.19	(0.04)
Methylfluoranthene	0.39	(0.09)	0.20	(0.03)	0.11	(0.03)	0.08	(0.02)
9-Methylanthracene	0.03	(0.01)	0.03	(0.01)	0.09	(0.02)	0.03	(0.01)
Benzo(ghi)fluoranthene	1.17	(0.59)	0.50	(0.05)	0.17	(0.04)	0.10	(0.02)
Cyclopenta(cd)pyrene	1.54	(0.86)	0.56	(0.25)	0.04	(0.01)	0.06	(0.01)
Benz(a)anthracene	1.02	(0.50)	0.48	(0.10)	0.13	(0.03)	0.14	(0.03)
Chrysene	0.76	(0.38)	0.30	(0.00)	0.11	(0.03)	0.13	(0.03)
1-Methylchrysene	0.12	(0.05)	0.06	(0.01)	0.03	(0.01)	0.03	(0.01)
Retene			0.03	(0.01)	0.04	(0.01)		
Benzo(b)fluoranthene	0.86	(0.25)	0.39	(0.11)	0.13	(0.03)	0.10	(0.02)
Benzo(k)fluoranthene	0.35	(0.27)	0.17	(0.02)	0.05	(0.01)	0.04	(0.01)
Benzo(j)fluoranthene	0.39	(0.21)	0.19	(0.11)	0.03	(0.01)	0.12	(0.03)
Benzo(e)pyrene	0.39	(0.18)	0.19	(0.05)	0.09	(0.02)	0.07	(0.02)
Benzo(a)pyrene	0.85	(0.48)	0.33	(0.07)	0.10	(0.02)	0.07	(0.02)
Perylene	0.18	(0.03)	0.08	(0.06)	0.002	(0.004)	0.003	(0.003)
Indeno(1,2,3-cd)pyrene	0.52	(0.39)	0.20	(0.09)	0.07	(0.02)	0.04	(0.01)
Benzo(GHI)perylene	0.49	(0.08)	0.30	(0.25)	0.06	(0.02)	0.03	(0.01)
Dibenz(ah)anthracene	0.10	(0.03)	0.06	(0.03)			0.02	(0.01)
Picene	0.25	(0.06)	0.13	(0.10)			0.01	(0.00)
Triphenylbenzene								
Alkanes								
Pristane	0.03	(0.16)	0.15	(0.03)	0.18	(0.27)		
Norpristane	0.01	(0.06)	0.05	(0.01)	0.06	(0.09)		
Phytane			0.02	(0.02)	0.02	(0.14)		
Squalane			0.16	(0.09)	0.10	(0.28)	0.43	(0.22)
Octadecane			0.05	(0.01)	0.02	(0.08)		
Nonadecane			0.16	(0.01)	0.22	(0.26)		
Eicosane	0.06	(0.07)	0.39	(0.14)	0.41	(0.34)	0.10	(0.21)
Heneicosane	0.13	(0.04)	0.43	(0.04)	0.36	(0.10)	0.27	(0.07)
Docosane	0.06	(0.58)	0.34	(0.19)	0.20	(0.86)	0.43	(0.60)
Tricosane	0.10	(0.02)	0.61	(0.13)	0.73	(0.45)	1.45	(0.47)
Tetracosane			0.47	(0.17)	0.20	(0.76)	1.66	(0.68)
Pentacosane			0.50	(0.23)	0.15	(1.10)	1.29	(0.84)
Hexacosane			0.21	(0.21)			0.92	(0.89)
Heptacosane			0.78	(0.45)	0.20	(1.32)	2.07	(1.08)
Octacosane	0.12	(0.57)	0.42	(0.16)			1.95	(0.79)
Nonacosane	0.26	(0.59)	1.79	(0.40)	2.00	(1.05)	4.47	(1.27)
Triacontane	0.11	(0.06)	1.01	(0.25)			2.83	(0.89)
Hentriacontane	0.19	(0.16)	2.06	(0.90)	0.15	(0.59)	6.71	(1.67)
Dotriacontane	0.11	(0.25)	0.56	(0.23)			2.53	(0.69)
Tritriacontane	0.17	(0.14)	1.07	(0.35)	0.11	(0.36)	4.94	(1.21)
Tetratriacontane	0.31	(0.15)	0.42	(0.16)	0.22	(0.19)	1.31	(0.34)
Pentatriacontane			0.26	(0.08)			1.06	(0.28)
Levoglucosan	115.1	(57.2)	48.2	(14.2)	291	(67)	33.7	(7.8)
Sterols and Stanols								
Cholesterol			0.28	(0.14)			0.52	(0.24)
Stigmasterol	0.66	(0.14)	0.69	(0.32)	3.68	(0.86)	0.82	(0.20)
b-Sitosterol	3.51	(0.21)	1.06	(0.33)	6.31	(1.55)	1.70	(0.47)
Campesterol	1.48	(0.36)	0.82	(0.36)	3.04	(0.70)	1.02	(0.24)
Cholestanol and coprostanol			0.21	(0.09)			0.72	(0.17)
Stigmastanol	0.31	(0.06)	0.56	(0.23)			1.54	(0.36)

1) Rice, wheat, mustard, lentil, and grasses

Table 5: Comparison of brick kiln emissions of $PM_{2.5}$, OC, and BC in this study to prior studies of similar kiln design.

Kiln type (location)	*n*	MCE	EF $PM_{2.5}$ (g kg^{-1})	EF OC (g kg^{-1})	EF BC (g kg^{-1})	Reference
Clamp (Nepal)	3	0.950	10.7 ± 1.6	6.74	0.02	*This study and Stockwell et al., 2016*
Induced-draught zig-zag (Nepal)	3	0.994	15.1 ± 3.7	1.0	0.11	*This study and Stockwell et al., 2016*
Induced-draught zig-zag (India)	3	0.987	0.6-1.2	0.01-0.7	0.07-0.5	Weyant et al. 2014
Batch-style (Mexico)	2	0.968	1.2-2.0[1]	0.07-2.8	0.6-1.5	Christian et al. 2010

1) Estimated from measurements of OC, EC, metals, and ions (but not sulfate)

Figure 1: $EF_{PM2.5}$ and $PM_{2.5}$ composition (as percent by mass) for forced draught zig-zag kilns (a) and clamp kilns (b). For the average $EF_{PM2.5}$, error bars for averages correspond to one standard deviation, while those for individual trials show the analytical uncertainty. EC was not detected in brick kiln emissions; optical measurements of BC from Stockwell et al. (2016) are provided in Table 1.

Figure 2: $EF_{PM2.5}$ and $PM_{2.5}$ composition (as percent by mass) for garbage burning. $EF_{PM2.5}$ from the combustion of mixed waste under dry conditions was substantially lower than mixed waste burned under damp conditions. The former was considered the best estimate of $PM_{2.5}$ emissions from this source and is shown as the mixed waste average. Error bars correspond to analytical uncertainties.

Figure 3: $EF_{PM2.5}$ and $PM_{2.5}$ composition (as percent by mass) for generators (a), diesel groundwater pumps (b), and motorcycles before and after servicing (c). Error bars correspond to analytical uncertainties.

Figure 4: $EF_{PM2.5}$ and $PM_{2.5}$ composition (as percent by mass) for various types of biomass burning, including open burning (heating and crop residue fires), cooking stoves, and 3-stone fires. Within a stove type, fuels are positioned with increasing dung content, revealing that burning or co-burning of dung yielded higher $PM_{2.5}$ emissions. Error bars correspond to analytical uncertainties.

Figure 5: A scatter plot of MCE versus $EF_{PM2.5}$, with the regression line applied only to the biofuel samples in the laboratory combustion tests. Excluded from this regression were charcoal burning, biogas, and the very high $EF_{PM2.5}$ for the 3-stone fire fueled with dung (see section 3.7). The field tests consistently fall below the regression line, indicating that biomass burning in measured in the field is lower in both MCE and $EF_{PM2.5}$ compared to the laboratory measurements.

Figure 6: Emission ratios of select organic species in field tests, normalized to OC (mg gOC^{-1}), for of open burning (crop residue and heating fires) and 1-2 pot traditional mud cooking stoves. Normalization to OC accounts for the large changes in EF_{OC} observed across different combustion scenarios and demonstrates consistency in the molecular marker-to-OC ratios for common fuels. Cholesterol, cholestanol, and coprostanol are observed only when dung is burned and are characteristic markers of this source.

Figure 1

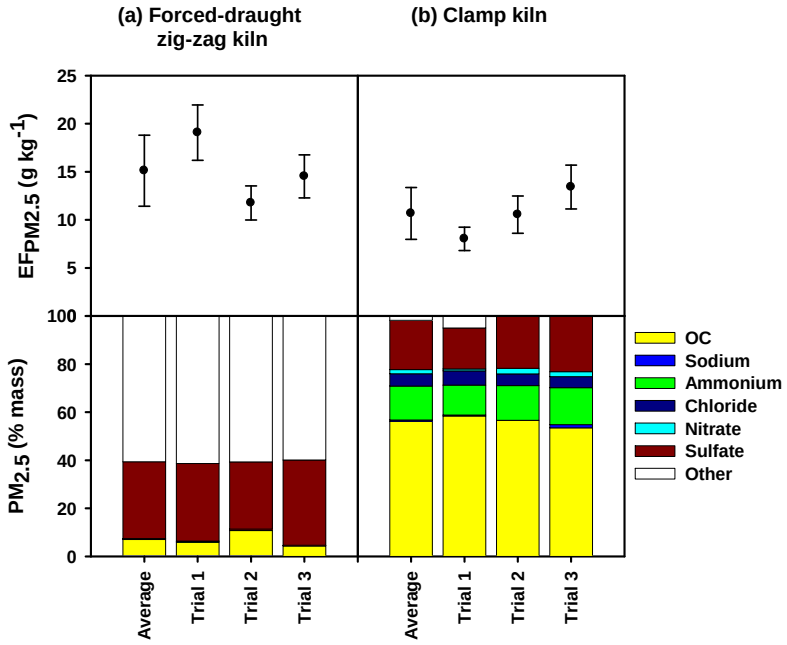

(a) Forced-draught zig-zag kiln
(b) Clamp kiln
EFPM2.5 (g kg-1)
PM2.5 (% mass)
25
20
15
10
5
100
80
60
40
20
0
Average
Trial 1
Trial 2
Trial 3
OC
Sodium
Ammonium
Chloride
Nitrate
Sulfate
Other

Figure 2

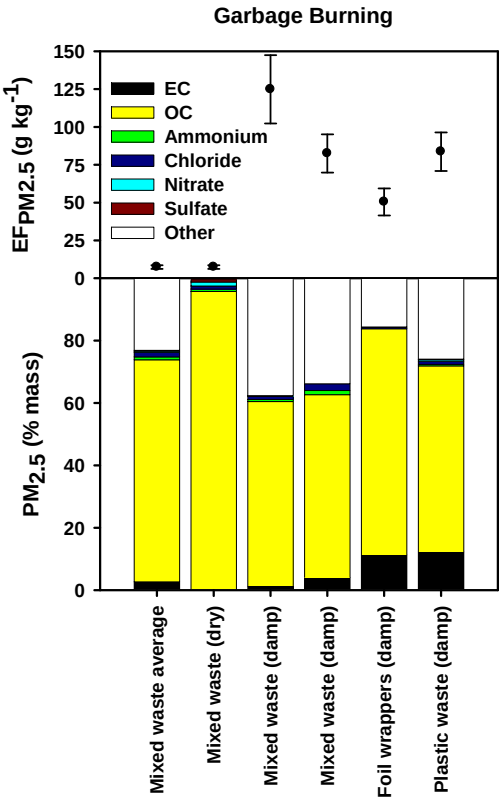

Garbage Burning
EC
OC
Ammonium
Chloride
Nitrate
Sulfate
Other
EF PM2.5 (g kg-1)
PM2.5 (% mass)
150
125
100
75
50
25
0
80
60
40
20
0
Mixed waste average
Mixed waste (dry)
Mixed waste (damp)
Mixed waste (damp)
Foil wrappers (damp)
Plastic waste (damp)

Figure 3

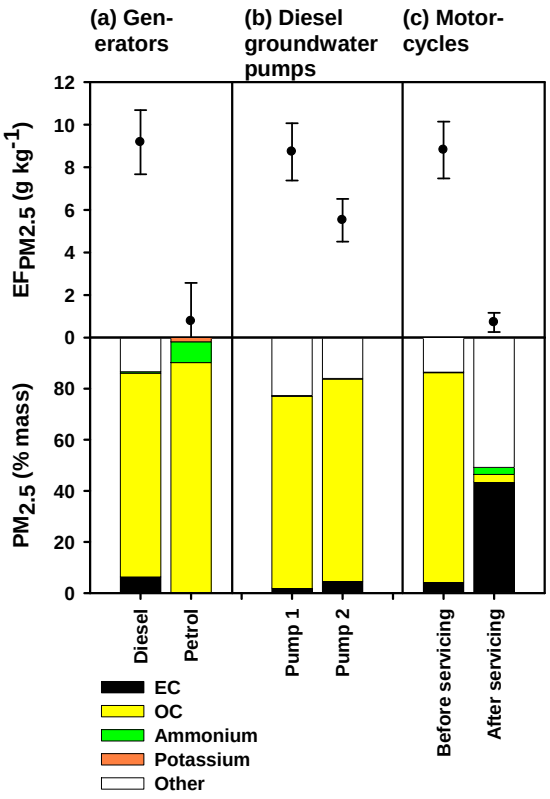

(a) Gen-erators
(b) Diesel groundwater pumps
(c) Motor-cycles
EF PM2.5 (g kg-1)
PM2.5 (% mass)
Diesel
Petrol
Pump 1
Pump 2
Before servicing
After servicing
EC
OC
Ammonium
Potassium
Other

Figure 4

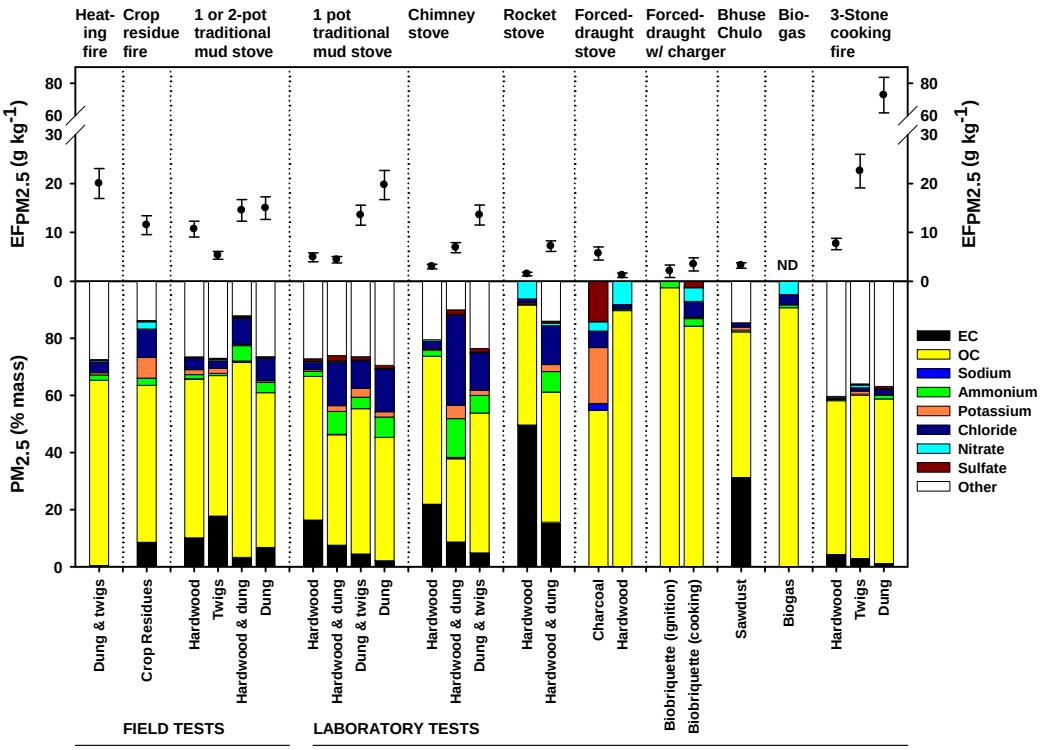

Heat-ing fire
Crop residue fire
1 or 2-pot traditional mud stove
1 pot traditional mud stove
Chimney stove
Rocket stove
Forced-draught stove
Forced-draught w/ charger
Bhuse Chulo
Bio-gas
3-Stone cooking fire
EFPM2.5 (g kg-1)
PM2.5 (% mass)
ND
EC
OC
Sodium
Ammonium
Potassium
Chloride
Nitrate
Sulfate
Other
Dung & twigs
Crop Residues
Hardwood
Twigs
Hardwood & dung
Dung
Dung & twigs
Charcoal
Biobriquette (ignition)
Biobriquette (cooking)
Sawdust
Biogas
FIELD TESTS
LABORATORY TESTS

Figure 5

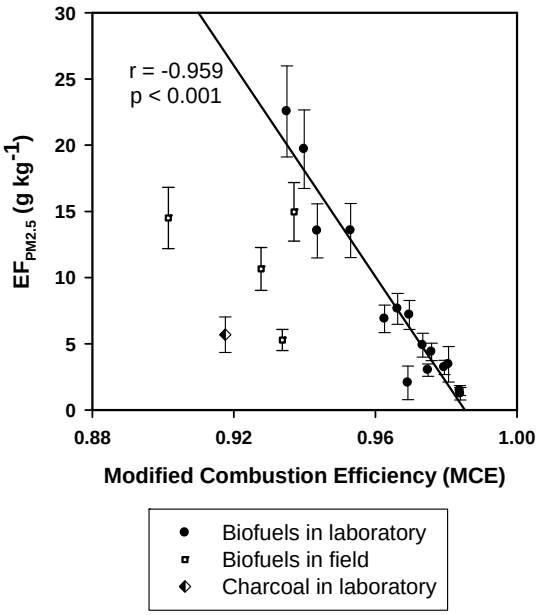
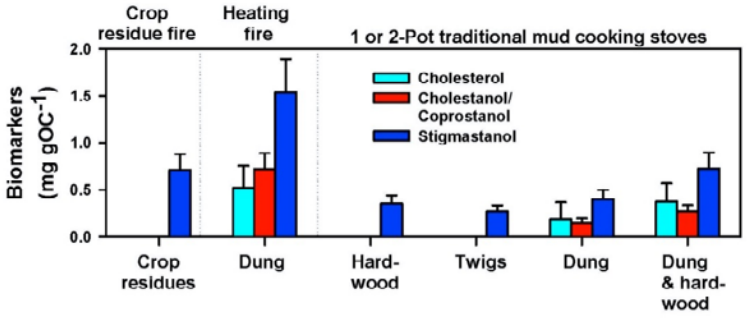

r = -0.959
p < 0.001
EF PM2.5 (g kg-1)
Modified Combustion Efficiency (MCE)
0
5
10
15
20
25
30
0.88
0.92
0.96
1.00
Biofuels in laboratory
Biofuels in field
Charcoal in laboratory

Figure 6


Crop residue fire
Heating fire
1 or 2-Pot traditional mud cooking stoves
Biomarkers (mg gOC-1)
2.0
1.5
1.0
0.5
0.0
Cholesterol
Cholestanol/ Coprostanol
Stigmastanol
Crop residues
Dung
Hard-wood
Twigs
Dung
Dung & hard-wood