Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-510-AC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "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" *by* Thilina Jayarathne et al.

Thilina Jayarathne et al.

betsy-stone@uiowa.edu

Received and published: 14 September 2017

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., 2016) 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 SO2 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 $\sim$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 con-

centration), 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 EFPM. 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 SO2 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 SO2 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 EFPM without water, we have reported the "lower limit of EFPM2.5 (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 EFPM 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 EFBC in the case EFEC 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 un-

derestimate 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., 2016) and likely a lower EFPM2.5 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 PM2.5 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 EFPM 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 PM2.5, 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 PM2.5, 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 ($\sim$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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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-510, 2017.