

Anonymous Referee #1

Referee #1 General Comments: *I thank the authors for responding to my comments. I have read through the revised manuscript and still believe this paper should be accepted and published in ACP. Below are a few comments and thoughts I had while reviewing the paper.*

Response to Referee #1 General Comments: We thank the reviewer for the careful review of the resubmitted manuscript and appreciate their suggestions to improve it further. Specific comments are addressed point-by-point below.

Minor Comments:

Referee #1 Comment 1: *Page 8, line 24: "These tests die" die should be did*

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

Referee #1 Comment 2: *Section 2.2.3: A sub sample of QFF were analyzed for WSOC. Was the backup QFF also analyzed using this method to determine the positive gas-phase absorption artifact? (Kristensen et al., 2016) has shown that considerable amounts of organic acids absorb onto QFF which would show up in WSOC analysis.*

Response to Referee #1 Comment 2: To clarify this point, we have added the following text to line 2.2.3: "Water-soluble and water-insoluble fractions of OC were calculated relative to the total OC on the front filter and scaled to the artifact-corrected OC assuming equivalent sorption of water-soluble and water-insoluble gases." Our rationale for this approach is that a subset of backup QFF was analyzed for WSOC, but the observed concentrations were relatively low (generally within a factor of two of the field blank concentrations). Consequently, the backup filters were deemed unreliable for a robust artifact correction, leading us to make the above-stated assumption.

Referee #1 Comment 3: *Page 13, line 23: no comma after blank subtracted*

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

Referee #1 Comment 4: *Page 14, line 21: Sentence beginning with "Notably..." is a run-on sentence and confusing.*

Response to Referee #1 Comment 4: We have revised this sentence to read: "Notably, the filter-integrated MCE values reported herein correspond to the average MCE over the duration of filter sample collection. These values differ slightly from those reported by Stockwell et al. (2016) who analyzed the same source over different time periods."

Referee #1 Comment 5: *Page 19, line 3: The authors state that the uncertainty could be from absorption of semi-volatile compounds onto the filter (I assume they mean QFF). However, I was under the impression that the OC:EC measurements were subtracted by the OC:EC amounts detected on the back filter. (1) This subtraction approach seems imperfect as gaseous compounds could break through both filters and/or be absorbed by different amounts on the front filter compared to the back filter. But I do admit, this method is likely the best way to "correct" for gas-phase artifacts. What I am more confused about is why the authors claim the 11% difference could be caused by SVOC absorption when they supposedly corrected for it? It would be less confusing if the authors just pointed out the inherent errors in correcting for SVOC absorption.*

Response to Referee #1 Comment 5: We agree with the reviewer and in response to this comment, we have added the following sentence to section 2.2.2: "Potential biases in this approach derive from differential sorption on the front filter compared to the back filter, leading to additional uncharacterized measurement uncertainties." We have also revised the text at page 19 line 3 to read: "The sum of OC,

BC, and measured inorganic ions exceeded the measured PM_{2.5} mass by an average of 11%, which is within the propagated uncertainty of the analytical measurements.”

Referee #1 Comment 6: *Page 21, line 3: “...of PM2.5 reply...” should be rely*

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

Referee #1 Comment 7: *Page 21, line 20: the semi-colon should be a comma*

Response to Referee #1 Comment 7: This change has been made as suggested.

Referee #1 Comment 8: *Page 23, line 24: “nor sulfate was” should be were*

Response to Referee #1 Comment 8: This change has been made as suggested.

Referee #1 Comment 9: *Page 28, line 5: “contribute to the greater emissions...” greater seems awkward*

Response to Referee #1 Comment 9: We have revised this sentence to read: “As the EF_{PM_{2.5}} for hardwood decreases, the MCE increases (Table S2) consistent with smoldering conditions emitting more PM_{2.5}.”

Referee #1 Comment 10: *Page 28, line 13: perhaps consider adding reduced between the worlds the and global*

Response to Referee #1 Comment 10: This change has been made as suggested.

Referee #1 Comment 11: *Page 29, line 9: see comment I wrote above about gas absorption. Was gas absorption not corrected for? The methods indicate that it was.*

Response to Referee #1 Comment 11: We have clarified this sentence to read: “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 the measured OC exceeded the measured PM_{2.5} mass by a factor of three. All of these sources had relatively low PM_{2.5} emissions in comparison to other stove types. The results suggest that the measured OC was overestimated, despite the correction for gas adsorption. Because organic gas adsorption affects QFF but not Teflon filters, the EF_{PM_{2.5}} measurement for these stove types is considered valid.”

Referee #1 Comment 12: *Page 32, line 5: It is a bit to add a sentence that summarizes a point from a paper that is in preparation and was not talked about in detail in this paper. It would be more logical to remove this sentence.*

Response to Referee #1 Comment 12: In response to this comment we have moved the sentence in question to page 32 line 10 and revised the verb tense to future. This sentence now reads: “Co-located, size-resolved emissions measurements of these sources by AMS will provide further chemical insight into aerosol composition (Goetz et al., in preparation-a, b).”

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_{PM_{2.5}} 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_{PM_{2.5}} ranged 8-13 g kg⁻¹, with major contributions from OC (63.2%), sulfate (23.4%), and ammonium (16%). Our brick kiln EF_{PM_{2.5}} 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_{PM_{2.5}} of 7.4 ± 1.2 g kg⁻¹, whereas damp conditions generated the highest EF_{PM_{2.5}} 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 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_{PM_{2.5}} 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_{PM_{2.5}} 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.

15 **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 [CO₂], carbon monoxide [CO], methane [CH₄], ammonia [NH₃], 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.

5 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, 10 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. 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- 15 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 20 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 25 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 amount and chemical composition of particulate matter emitted, to further support source attribution, emissions inventories, and air 30 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 were not collected (>84%) and 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 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 diesel generators were characterized to gain further insight to this widely-used combustion source.

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 1970s, 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
5 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 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
10 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
15 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
20 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). 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,
25 garbage burning, cooking stoves, motorcycles, and diesel generators. 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.

EFs for combustion sources were determined by the carbon mass balance approach (Ward and Radke, 1993;
30 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 was positioned manually to sample the plume where the plume of smoke was well-mixed and had cooled to near-ambient temperatures. During the period of sample collection, ambient temperatures ranged 12-29 °C (on a five-minute basis) in the Kathmandu Valley and averaged 19 °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). 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 ~~do~~did 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. Three filter samples were collected from smoke escaping from the chimney, with each filter sampled only when the plume reached the sampling inlet. 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).

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.

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.

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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) 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). The studied motorcycles are among the most common models in Kathmandu (Shrestha et al., 2013).

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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 $EF_{PM_{2.5}}$ 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).

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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 °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 ($\mu\text{g m}^{-3}$) 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 cm² 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). Potential biases in this approach derive from differential sorption on the front filter compared to the back filter, leading to additional uncharacterized measurement uncertainties. 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 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 ($0.05 \mu\text{g cm}^{-2}$), 10% of the measured EC, and 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 cm² 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 M Ω 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). Water-soluble and water-insoluble fractions of OC were calculated relative to the total OC on the front filter and scaled to the artifact-corrected OC assuming equivalent sorption of water-soluble and water-insoluble gases.

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).. 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 ($\mu\text{g m}^{-3}$) 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, 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-D₁₀, benz(a)anthracene-D₁₂, cholestane-D₄, pentadecane-D₃₂, eicosane-D₄₂, tetracosane-D₅₀, triacontane-D₆₂, dotriacontane-D₆₆, hexatriacontane-D₇₄, levoglucosan-¹³C₆ and cholesterol D₆. Each sample was then extracted into 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 stigmaterol 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.

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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, CO₂, 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).

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

15 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: CO₂, CO, CH₄, EC, and OC. EFs for PM components were calculated as the product of EF_{PM2.5} and the component's mass fraction in PM_{2.5}. 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.

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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~~ These values differ slightly from those reported by Stockwell et al. (2016), ~~because they who analyzed the same sources over were typically collected over~~ different time periods, ~~although from the same source.~~

25

30 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 increase PM mass and OC emissions due to partitioning of SVOC to the particle phase (Lipsky and Robinson, 2006). Thus, EF_{PM2.5} and EF_{OC} depend on the dilution ratio and chemical composition of the source emissions, while EC is not affected (Lipsky and Robinson, 2006). 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, 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. On the other hand, source apportionment may be best based on ratios between low-volatility components.

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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.

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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.

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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. Metals associated with clay were not detected—aluminum, iron, and titanium—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%), fluoride (0.011%), chloride (0.065%), and

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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 ($34 \pm 12\%$, 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 $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$ (that excludes the maximum possible amount of hygroscopic 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 ($EF_{SO_4} 4.9 \text{ g kg}^{-1}$), while the majority of sulfur emissions were gaseous SO_2 ($EF_{SO_2} 12.7 \text{ g kg}^{-1}$; 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 ($0.98 \text{ m}^2 \text{ g}^{-1}$) 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 reflects the relatively small amount of wood burned in this zig-zag kiln relative to coal.

5 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.

10 Significant differences in emissions were found from the induced-draught zig-zag kiln compared to prior studies (Table 5). First, the mean $EF_{PM_{2.5}}$ for the induced-draught zig-zag kiln ($15.1 \pm 3.7 \text{ g kg}^{-1}$) was considerably higher than $EF_{PM_{2.5}}$ reported by Weyant et al. (2014) for induced-draught zig-zag kilns fueled with coal in India ($0.6 - 1.2 \text{ g kg}^{-1}$). Notably, measurements by Weyant et al. (2014) were sampled within the stack and then diluted, compared to natural dilution that occurred 1-2 m downwind. Because the kiln emissions in this study were
15 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
20 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} . Thus, the difference in $EF_{PM_{2.5}}$ 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
25 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 ($0.63-1.26 \text{ g kg}^{-1}$) and those by Weyant et al. ($0.08-0.67 \text{ g kg}^{-1}$) and Christian et al. ($0.669-1.783 \text{ g kg}^{-1}$), the main reasons for the increased $EF_{PM_{2.5}}$ 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.

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3.2 Clamp kiln

The clamp kiln studied produced a mean $EF_{PM_{2.5}}$ of $10.7 \pm 2.7 \text{ g kg}^{-1}$ 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 (23.4%), ammonium (16.0%), chloride (5.7%), and nitrate (2.0%) (Table 1; Figure 1b). Minor components included BC (0.2%), and potassium (0.2%). The sum of OC, BC, and measured inorganic ions exceeded the measured $PM_{2.5}$ mass by an average of 11%. ~~which. 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. 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-methylchrysene. 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 $7.4 \pm 1.2 \text{ g kg}^{-1}$ 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 $9.8 \pm 5.7 \text{ g kg}^{-1}$ (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 creates damp conditions, under which the 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 $124 \pm 23 \text{ g kg}^{-1}$ (MCE 0.889) and $82 \pm 13 \text{ g kg}^{-1}$ (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 $50 \pm 9 \text{ g kg}^{-1}$ (MCE 0.973), while plastic burning had an EF_{PM2.5} of $84 \pm 13 \text{ g kg}^{-1}$ (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. $EF_{PM_{2.5}}$ 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.

5 The wide range of $EF_{PM_{2.5}}$ 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 $PM_{2.5}$ rely 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.

15 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, 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. 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.

30 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 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_{PM_{2.5}} 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_{PM_{2.5}} for US military diesel generators that exhibited an average (± standard deviation) of 1.2 ± 0.6 g kg⁻¹ (Zhu et al., 2009). Although limited to one sample, the rented diesel generator studied in Nepal had a high EF_{PM_{2.5}} value and comparisons to other studies.

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-were~~ 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.

Organic species quantified by GCMS accounted for 12% of the OC emitted from the diesel generator, inclusive of n-alkanes (11%), 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 engine oil evaporation (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. 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). $EF_{PM_{2.5}}$ for the groundwater pumps were $8.7 \pm 0.7 \text{ g kg}^{-1}$ for pump 1 (4.6 kVA model) and $5.5 \pm 0.5 \text{ g kg}^{-1}$ for pump 2 (5 kVA model) (Figure 3b; Table S3). The higher $EF_{PM_{2.5}}$ 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 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). 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). 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.

5

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
10 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. $EF_{PM_{2.5}}$ was $8.81 \pm 1.33 \text{ g kg}^{-1}$ before servicing and dropped considerably to $0.71 \pm 0.45 \text{ g kg}^{-1}$ 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
15 (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).
20 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).

25 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. Instead, we compare ratios of $EF_{PM_{2.5}}$ 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
30 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).

5 Changes to motorcycle EF before and after servicing indicate 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
10 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.

15

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
20 from traditional mud stoves in this study. EF_{PM2.5} determined from these field tests were $10.7 \pm 1.6 \text{ g kg}^{-1}$ for hardwood, $5.3 \pm 0.8 \text{ g kg}^{-1}$ for twigs, $14.5 \pm 2.2 \text{ g kg}^{-1}$ for dung (all in a 1-pot stove) and $15.0 \pm 2.3 \text{ g kg}^{-1}$ 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
25 are greater than values compiled by Akagi et al. (2011) for EF_{PM2.5} from open cooking that averaged $6.73 \pm 1.61 \text{ g kg}^{-1}$, 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
30 (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 $EF_{PM_{2.5}}$ 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. 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 $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$ from MCE using relationships developed in the laboratory would overestimate $EF_{PM_{2.5}}$ in the field.

The use of dung, or a mixture of dung and wood, consistently gave higher $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$ 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_{PM_{2.5}} for hardwood decreases, the MCE increases (Table S2) ~~suggesting consistent with that the smoldering conditions contribute to the greater emissions of emitting more~~ PM_{2.5}. When dung was used as fuel, the 3-stone cooking fire again generated the highest EF_{PM_{2.5}} (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 reduced global warming potential of emissions and provides a viable and cleaner-emissions alternative 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 (Smith et al., 2000; Jetter and Kariher, 2009; Jetter et al., 2012).

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, 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 ($28\pm 7\%$ by mass) and sulfate ($21\pm 6\%$ 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. ~~All of these sources had relatively low $PM_{2.5}$ emissions in comparison to other stove types. The results suggest~~ing that the measured OC was overestimated, ~~perhaps despite the correction for~~ ~~due to~~ gas adsorption. Because organic gas adsorption affects QFF but not Teflon filters, the $EF_{PM_{2.5}}$ 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. $EF_{PM_{2.5}}$ was $11.5 \pm 2.2 \text{ g kg}^{-1}$. 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 (8.6%), 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. $EF_{PM_{2.5}}$ was $20.0 \pm 1.4 \text{ g kg}^{-1}$. Two factors are likely to contribute to this relatively high $EF_{PM_{2.5}}$: 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 0.40%; the high OC:EC ratio (~ 150) also indicates 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 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.

5 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.

10 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

15 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 $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$ or EF_{OC} , as this would be redundant.

25 **4 Conclusions**

We report $EF_{PM_{2.5}}$ 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

30 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 cholesterol for dung burning, which are useful in source identification and apportionment.

5 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. ~~Co-located, size-resolved emissions measurements of these~~
10 ~~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. ~~Co-located, size-resolved emissions measurements of these sources by AMS provides~~ will provide
15 further chemical insight into aerosol composition (Goetz et al., in preparation-a, b).

Field Code Changed

Field Code Changed

With a focus on detailed characterization of under-studied source sectors, NAMaSTE 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.

20 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
25 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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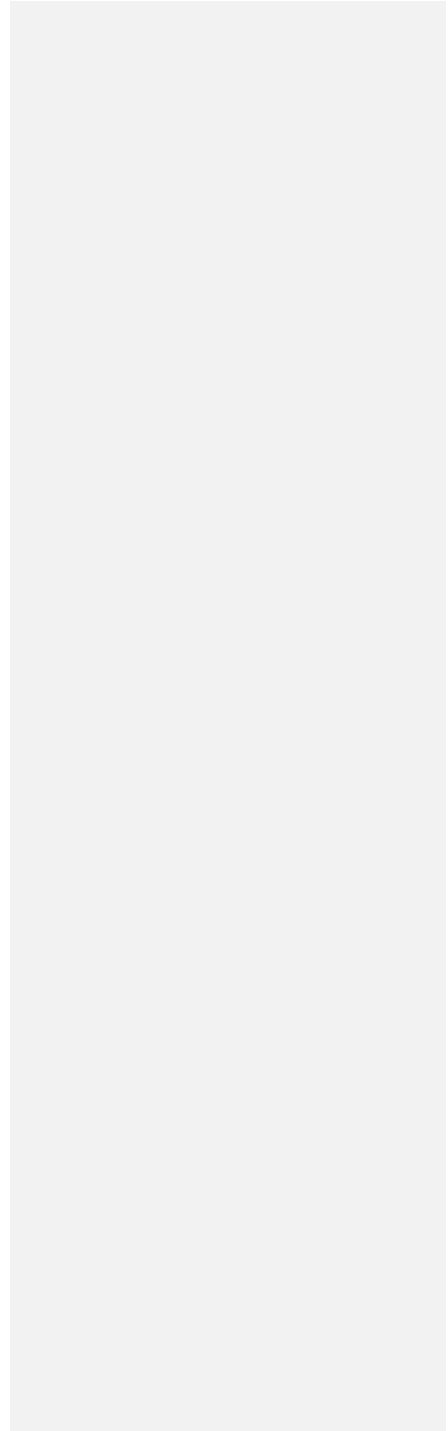


Table 1: Summary of emissions data for select combustion sources, including modified combustion efficiency (MCE), emission factors for PM_{2.5} (g kg⁻¹), and fine particle composition (as PM_{2.5} weight percent). Errors are shown in parenthesis; a description of their calculation is provided in section 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 ²	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)
<i>Water-soluble inorganic ions</i>								
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)					
<i>Metals</i>								
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)
<i>Water-soluble inorganic ions</i>								
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)
<i>Metals</i>								
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)
Benzo(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(GH)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																
17 α (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)
17 β (H)-21 α (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)
17 α (H)-21 β (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)		
$\alpha\beta\beta$ -20(R)-C27-Cholestane			0.07	(0.00)							0.05	(0.01)	0.05	(0.01)		
$\alpha\beta\beta$ -20(S)-C27-Cholestane											0.07	(0.02)	0.08	(0.02)		
$\alpha\alpha\alpha$ -20(S)-C27-Cholestane							0.06	(0.02)			0.04	(0.01)	0.11	(0.03)		
$\alpha\beta\beta$ -20(R)-C28-Ergostane							0.02	(0.01)			0.06	(0.01)	0.10	(0.02)	2.13	(0.55)
$\alpha\beta\beta$ -20(S)-C28-Ergostane							0.03	(0.01)			0.06	(0.01)	0.09	(0.02)	1.40	(0.39)
$\alpha\beta\beta$ -20(R)-C29-Sitostane							0.06	(0.01)			0.10	(0.09)	0.20	(0.05)	5.01	(1.24)
$\alpha\beta\beta$ -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)	
Trtriacontane	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)	
Tetracontane	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)		
Levogluosan	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	
<i>Polycyclic aromatic hydrocarbons</i>								
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)
Benzo(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(GH)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								
<i>Alkanes</i>								
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)
tettracontane	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)
<i>Sterols and Stanols</i>								
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)	<i>n</i>	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	<i>This study and Stockwell et al., 2016</i>
Induced-draught zig-zag (Nepal)	3	0.994	15.1 ± 3.7	1.0	0.11	<i>This study and Stockwell et al., 2016</i>
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: $EF_{PM_{2.5}}$ and $PM_{2.5}$ composition (as percent by mass) for forced draught zig-zag kilns (a) and clamp kilns (b). For the average $EF_{PM_{2.5}}$, 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: $EF_{PM_{2.5}}$ and $PM_{2.5}$ composition (as percent by mass) for garbage burning. $EF_{PM_{2.5}}$ 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: $EF_{PM_{2.5}}$ 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: $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$, 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 $EF_{PM_{2.5}}$ 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 $EF_{PM_{2.5}}$ 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

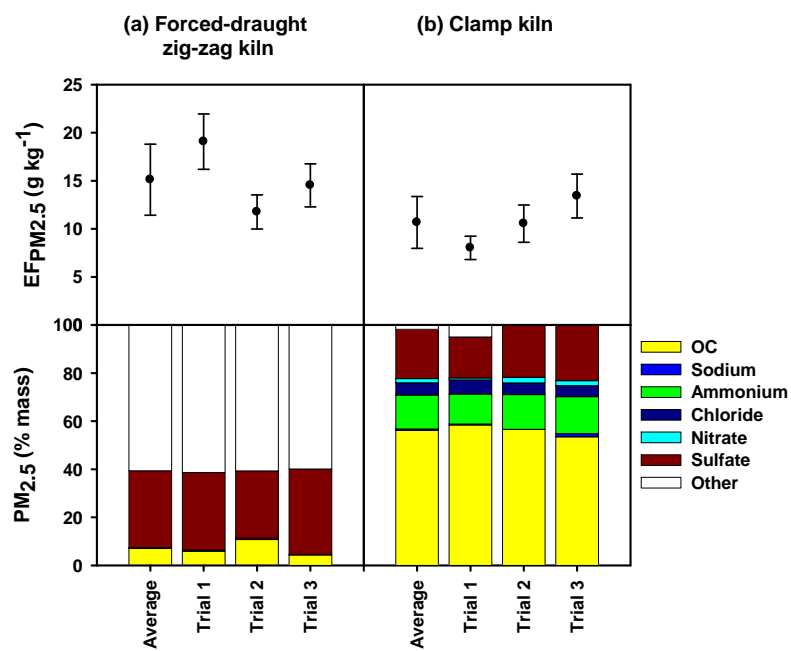


Figure 2

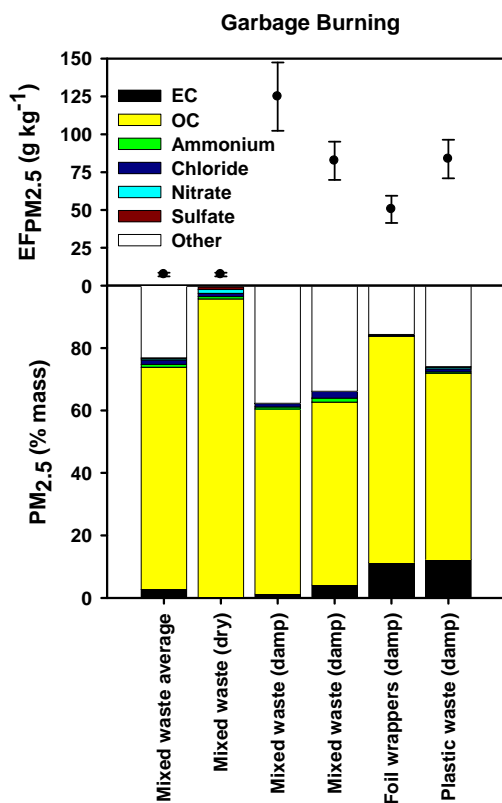


Figure 3

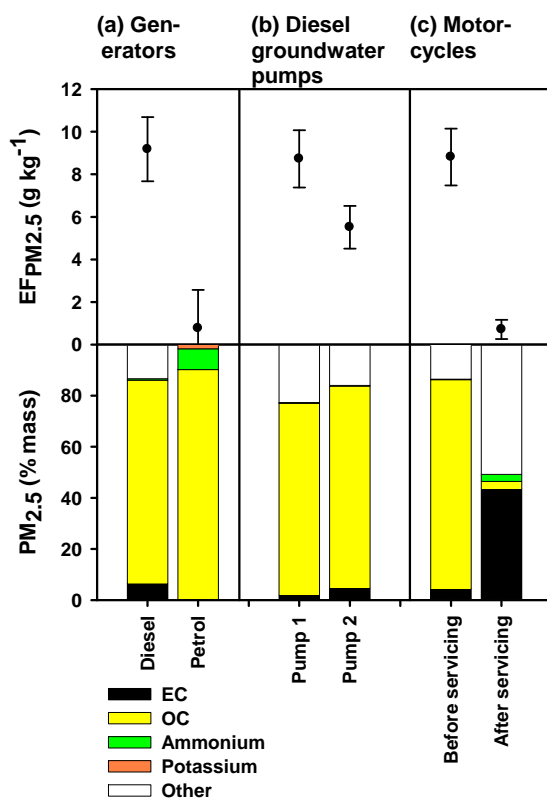


Figure 4

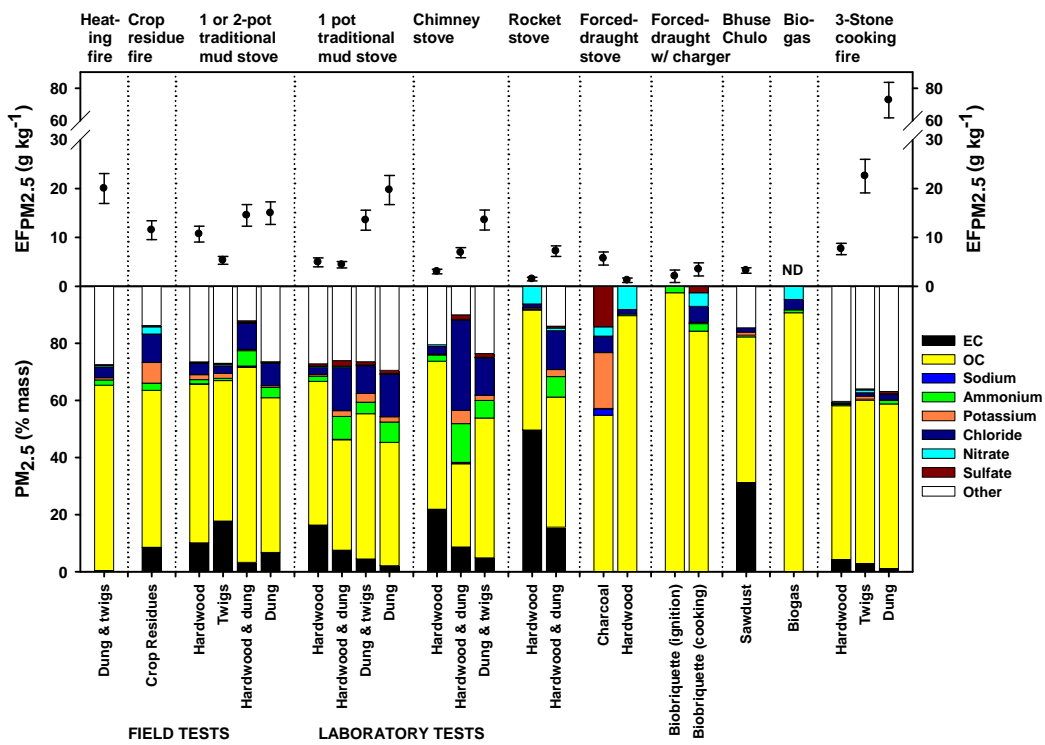


Figure 5

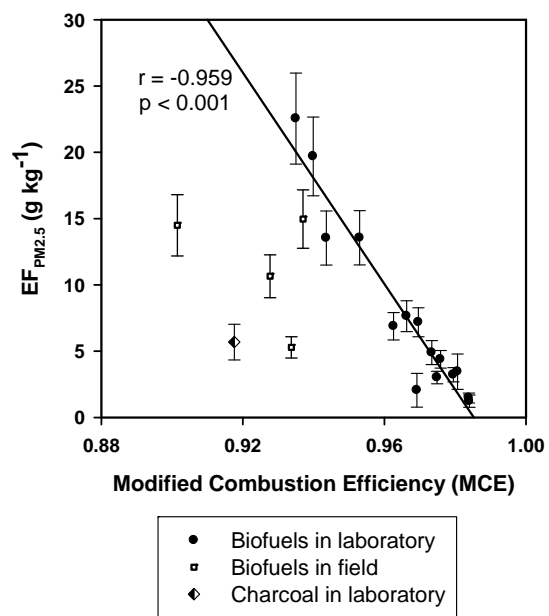


Figure 6

