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Key Points:
- Particulate matter emissions decreased marginally for intervention stoves and seasonality in emissions was observed for traditional stoves
- Intervention stoves increased particle-specific light absorption and are likely not a good way to mitigate climate warming by particles
- Two-season measurements suggest that there was strong seasonality in the linkage between emissions and indoor concentration and exposure

Supporting Information:
- Supporting Information S1
- Table S1

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Emission factors of health- and climate-relevant pollutants measured in home during a carbon-finance-approved cookstove intervention in rural India

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Abstract We present results of an emission characterization effort, completed as part of a larger intervention trial, of a carbon-finance-approved program replacing traditional cookstoves with "rocket"-style natural draft stoves. The 100 emission tests were conducted across 31 households in control and intervention groups, with repeated tests in most households during preintervention and postintervention periods. While mean fine particulate matter ($PM_{2.5}$) emission factor for intervention stoves was significantly lower than for traditional stoves in baseline measurements, they were only marginally lower than traditional stoves during follow-up. Intervention stove $PM_{2.5}$ emissions had a larger contribution from light-absorbing (elemental) carbon than traditional stoves. Repeated measurements in control households provide evidence for strong seasonality, likely due to differences in fuel moisture/types, in traditional stove emissions, with important implications for study design. Seasonality observed in control household emission factors (baseline $>$ follow-up) was in the opposite direction as that observed in indoor $PM_{2.5}$ concentrations (baseline $<$ follow-up), highlighting that seasonally varying conditions (e.g., ventilation rates) may modify the link between emissions and exposures. Emission factor differences in paired (pre/post) tests from the same households were similar to differences in the medians of entire groups, suggesting variability is dominated by test-to-test variation. Emission reductions from intervention stoves were significantly smaller than laboratory performance would suggest or that are required to strongly reduce exposures. Field emissions assessment like that presented here should be prioritized early in technology assessment and development to provide rigorous estimates of the benefits reasonably expected from interventions with the potential for substantial benefits to human health and the environment.

1. Introduction

Approximately 3 billion people, mostly in low- and middle-income countries [*Legros et al.*, 2009], rely on combustion of solid fuels (e.g., woody biomass or dung) in rudimentary stoves or open fires to meet their household energy needs for cooking, heating, and lighting. This practice has enormous health impacts on those exposed to household air pollutants (HAPs) emitted by these fires. An estimated 2.9 million deaths in 2015 have been attributed to HAP exposure [*Forouzanfar et al.*, 2016]; approximately a quarter of these are in India [*Institute for Health Metrics and Evaluation*, 2017]. HAPs consist of products of incomplete combustion (PIC) including CO, fine particulate matter ($PM_{2.5}$), and various other species associated with diseases such as childhood pneumonia, chronic obstructive pulmonary disease, and lung cancer [*World Health Organization*, 2014]. HAP exposures in indoor environments with rudimentary solid fuel stoves are often many times greater than guidelines established by the World Health Organization [*World Health Organization*, 2014]. Exposures are typically greater for women, who spend more time engaged in cooking, and the young children who are often near or on their mothers. The contribution from the large number of these relatively small sources to ambient air pollution can also be considerable. For example, an estimated 26% of 2010 population-weighted ambient $PM_{2.5}$ in South Asia was attributed to household cooking with solid fuels [*Chafe et al.*, 2014].

Climate impacts from these sources are substantial as well, as combustion is a source of long-lived greenhouse gases (GHG; e.g., CO_2 and methane) and a range of short-lived species with strong climate forcing. Household solid fuel combustion is estimated to contribute ~25% of global emissions of black carbon (BC), a component of fine PM composed of light-absorbing graphitic carbon, often also classified as elemental

carbon (EC) when quantified via thermooptical analysis [*Andreae and Gelencsér*, 2006]. BC is thought to be the second-leading contributor to current warming (after CO_2) [*Bond et al.*, 2013] and reducing BC emissions has been proposed as a way to mitigate short-term warming while providing air quality "cobenefits" [*Venkataraman et al.*, 2005; *Bond*, 2007; *Grieshop et al.*, 2009, 2011; *Sanford and Burney*, 2015; *Ramanathan et al.*, 2016]. The other major aerosol component in solid fuel combustion emissions is organic carbon (OC), which is typically considered to scatter short-wave solar radiation and thus offset some BC warming. However, OC from biomass combustion also includes brown carbon, which absorbs light at shorter visible and UV wavelengths and may make a considerable contribution to warming [*Saleh et al.*, 2014]. While the magnitude of the net aerosol climate effect of current biofuel combustion is uncertain, it is likely climate warming [*Kodros et al.*, 2015; *Lacey and Henze*, 2015]. Also considering GHG emissions, especially methane and CO_2 from nonrenewable use of biomass [*Bailis et al.*, 2015], and other climate warming substances associated with emissions (e.g., ozone), traditional biofuel use is likely warming [*Unger et al.*, 2010; *Grieshop et al.*, 2011]. As a result, household biofuel use has gained attention in recent years as a potential means to mitigate climate change while also addressing other health and environmental impacts. Carbon finance, in which offset GHG (and possibly BC) emissions are sold to subsidize technology provision, has been proposed as a means by which to fund these efforts.

The myriad negative effects of unimproved solid fuel combustion and the opportunity for health, climate, environmental, and other cobenefits have led to the development of variety of alternative technologies and programs to disseminate them over the past several decades [*Grieshop et al.*, 2011]. Measurements of pollutant emission factors (EFs) serve a variety of important purposes in assessing such efforts. First, they provide a means by which to benchmark and compare technology options, more recently against performance standards [*International Organization for Standardization/International Workshop Agreement*, 2012] established to move toward indoor air quality guidelines [*World Health Organization*, 2014]. Second, emissions are a first stage in the linkage between activity/energy service (e.g., cooking) and the associated exposures/impacts. Therefore, assessing the benefits associated with alternative technologies depends on accurate quantifications of emissions and other stove/fuel performance metrics [*Smith and Haigler*, 2008; *Grieshop et al.*, 2011; *Johnson et al.*, 2011b; *Johnson and Chiang*, 2015]. Finally, EFs are important inputs to emission inventories used to estimate the larger-scale impacts of traditional and alternative technologies on regional air quality and climate change. Numerous stove types [*Smith et al.*, 2000; *Zhang et al.*, 2000; *MacCarty et al.*, 2010; *Jetter et al.*, 2012; *Just et al.*, 2013; *Arora et al.*, 2014; *Preble et al.*, 2014; *Arora and Jain*, 2015] have undergone laboratory testing using standard protocols like the Water Boiling Test (WBT) [*Bailis et al.*, 2007]. Such tests are important to probe the potential of a stove and assess its relative performance under closely controlled conditions, thus serving the first purpose listed above. However, to meet the latter two objectives, it is more important to quantify how stoves perform in real-world settings during in-home use with the available fuels, as impacts/benefits will depend on these and not emissions under ideal conditions/operation. Therefore, in-field measurements of stove performance are essential to accurately assess baseline impacts and potential benefits at all scales.

To date, in-home measurements of stove emission factors are relatively sparse and typically show that stoves perform significantly worse (in terms of efficiency and emissions) than during laboratory testing. For example, PM EFs measured in Honduras by *Roden et al.* [2009] of traditional and alterative stove models were a factor of 2–5 greater than they measured in lab WBTs. They also measured larger EC/TC (where TC = OC + EC is total carbon) values in laboratory tests, especially of traditional technologies, indicating greater particle-specific warming was implied in lab tests than was observed in the field. *Johnson et al.* [2008] made similar observations on several chimney stove models and cooking fires in Mexico, observing significant lower efficiency and EC/TC and higher PM and PIC emission rates during in-home tests than lab WBTs. Recent measurements by our group of several stoves in Malawi were broadly consistent with these as well [*Wathore et al.*, 2017]. In-home tests of traditional and alternative cookstoves in India have mostly focused on comparisons of indoor concentrations of PM, CO, and BC [*Kar et al.*, 2012; *Sambandam et al.*, 2014; *Muralidharan et al.*, 2015; *Patange et al.*, 2015], showing that alternative stoves do result in reduced indoor concentrations. However, to our knowledge, EFs measured during in-home use of traditional and alternative stoves in India have not been published.

To respond to this data gap, we completed a multiseason emissions sampling campaign as part of a larger research effort to evaluate India's first carbon-finance-approved cookstove replacement program. The intervention, study design, and background on the carbon-finance aspects are described in more detail in a

companion paper [*Aung et al.*, 2016]; here we focus on stove emissions performance. Specific objectives addressed in this paper are to (1) measure integrated emission factors of a number of health- and climate-relevant pollutants for traditional and intervention stoves; (2) evaluate variability in measured emissions across a number of dimensions: intertest, interhome, and interseason; (3) compare emission measurements with simultaneously collected indoor air quality data to give insight into linkages between the two, and their seasonal variation; and (4) analyze properties of real-time emissions to understand trends of use/performance relative to laboratory measurements.

2. Methods

2.1. Study Design

Emission tests were conducted in rural Indian homes in the village of Hire Waddarkal (HW) in the Koppal District of Karnataka State between September 2011 and August 2012. Testing was a component of an intervention trial developed around a Clean Development Mechanism (CDM)-approved cookstove replacement program (the first thus approved in India) being carried out by a regional non-governmental organization (NGO) [*Aung et al.*, 2016]. HW was a pilot village in the cookstove program established by the NGO. The vast majority (~99%) of households in this district/region depend on traditional mud/clay stoves (chulhas) for cooking and water heating, either inside or in partially covered cooking enclosures. Most households use two chulhas per household, with one used for vegetables or heating water, and another used for rice or bread (roti). Photos of traditional and intervention stoves are shown in Figure S1 in the supporting information.

Details on the intervention and community characteristics are presented in detail elsewhere [*Aung et al.*, 2016] and are only briefly described here. Of the 300 households in the village, 187 participated in the stove program. For the research study, these households were randomly assigned to intervention (n = 96) or control (n = 91) groups. Air quality, health, and fuelwood-, time-, and stove-use-measurements, along with an extensive survey, were completed during a "baseline" measurement season (hereafter called "baseline"; September–December 2011). After baseline measurements, households assigned to the intervention groups received two "rocket style" natural draft stoves after having their chulhas removed from the hearth, which was then rebuilt (in most households) to accommodate the new stoves. An identical set of "postintervention" measurements was then completed (hereafter "follow-up"; March–August 2012) after approximately 4–5 months (average of 194 days between baseline and postintervention measurements). The separation between baseline and follow-up was to allow households to learn to use the new stove and to come to a new "normal" usage pattern [*Hankey et al.*, 2015]. The NGO indicated that control households would be offered the intervention stoves at the end of the study to complete the CDM rollout.

As part of the study, 100 individual emission tests were completed in a subset of 31 separate study households (20 control; 11 intervention), during both baseline and postintervention seasons. Both morning (AM) and evening (PM) cooking sessions were captured in many, but not all cases, so two to four tests were conducted per household during the study. Several households that were initially in the intervention group were not using the intervention stoves at all on the testing day and have been grouped with control tests for results and analysis here.

2.2. Emission Testing Approach

In-home emission measurements were collected with the Stove Emission Measurement System (STEMS), described in detail elsewhere [*Wathore et al.*, 2017]. STEMS is powered by a 12 V battery and was developed for unsupervised measurement of emissions from in-home stove use to minimize obstruction/interruptions to household activity. Often, other study measurements (e.g., indoor air quality, surveys, and blood pressure) were being conducted simultaneously. STEMS sampled via an integrating in-plume probe [*Roden et al.*, 2006] and a $PM_{2.5}$ sharp-cut cyclone (BGI, Inc.) at 10 L min^{-1}. Measured quantities include CO_2 and CO, PM light scattering, temperature, relative humidity at 0.5 Hz, and particle light absorption coefficient (B_{ap}) at three wavelengths (λ = 467, 530, and 660 nm) at 1 Hz with a Particulate Soot Absorption Spectrometer (PSAP). CO_2, CO, light scattering, and temperature and humidity data were measured by sensors incorporated into a Portable Emission Measurement System (PEMS; Aprovecho Research, Cottage Grove, OR) sensor board which includes a nondispersive infrared CO_2 sensor, an electrochemical CO sensor, and a 635 nm wavelength laser light-scattering sensor. The light-scattering sensor was factory calibrated with smoke from flaming and

smoldering wood against a nephelometer operating at an optical wavelength of 530 nm and was corrected to yield particle scattering coefficient (B_{sp}) at 660 nm (to correspond to the PSAP red B_{ap}) following the method of *Weyant et al.* [2014]. PEMS and PSAP data streams were logged on a flash memory card during testing, with no computer required for operation.

Integrated PM samples were collected via two parallel 47 mm diameter filter trains. A bare quartz fiber filter and quartz behind Teflon were collected in parallel for thermooptical organic and elemental carbon (OC/EC) analyses. The "backup" quartz filter placed after the Teflon filter provides an estimate of the gas-phase adsorption artifact [*Subramanian et al.*, 2004] and an indication of the partitioning of primary organic aerosol emissions [*May et al.*, 2013], while the Teflon filter was used for gravimetric $PM_{2.5}$ measurements.

Flow rates through the two filter trains (2.5 L min^{-1} each) and a dilution line mixing upstream of the PSAP inlet were monitored using mass flow sensors (Honeywell AWM Series) calibrated before and after the measurement campaign with a reference flowmeter. CO and CO_2 sensors were calibrated on a monthly basis during the study using zero air and span gas cylinders procured in India. In each measurement "season," three field blank filters were collected after passing through all field protocols except sampling and analyzed using standard analysis protocols (see below) as a data quality assurance measure. Quartz filters were stored in a freezer between field collection and analysis. Gravimetric PM concentration was determined via conditioning and preweighing and postweighing Teflon filters on a microbalance (Sartorius M3P) in a temperature- and humidity-controlled room at the University of British Columbia. Organic and Elemental Carbon (OC/EC) analyses on quartz filters were conducted on a Sunset Labs Thermooptical Transmittance/Reflectance OC/EC Analyzer using the NIOSH 5040 Protocol as is described elsewhere [*Wathore et al.*, 2017]. Particle OC was determined based on the difference between the bare and backup quartz filters to account for positive vapor artifact. Standard lab protocols for OC/EC analysis include daily instrument blanks, weekly runs with external carbon standards, and repeated analysis of every tenth filter. Standard quality assurance protocols for gravimetric analysis included weighing in triplicate, daily reweight of calibration weights and lab blanks and of every tenth filter.

Emission measurements took place during normal household cooking activities, with STEMS and probe installation coordinated with household schedules to enable a short (5–15 min) "background" measurement period to establish ambient levels of measured species. The probe was placed approximately 1.5 m above the center of the hearth area, and so the probe sampled emissions from one or two stoves simultaneously. In households with chimney over the hearth area, one arm of the sampling probe was placed immediately above the top of the chimney exit above the roof [*Roden et al.*, 2006]. A full cooking session, with typical duration of 1–4 h, was captured during a test. In addition to emission measurements, fuel moisture content, fuel type (e.g., species), daily fuel use, other indoor sources (e.g., kerosene lamps), and stove descriptors (e.g., placement, condition, and proximity to ventilation) were measured and noted in field data logs. Fuel moisture was measured with an electronic moisture meter (BD-2100, Delmhorst Instrument Co.) from three locations on three pieces of wood; average values are reported in the spreadsheet table included in the supporting information. Response of the meter was adjusted using calibration factors for the four most common wood types using a gravimetric approach and is reported as wet basis. Fuels used in cooking sessions ranged widely and included wood from various tree species, *Albizzia amara*, *Azadirachta indica* (neem), and *Prosopis juliflora* (mesquite) being the most common, but also including pieces of cactus and agricultural residues (e.g., sunflower stalks).

The "plume probe" method for emission measurement has the advantages of being unobtrusive, quick to set up and giving a representative sample of gas and particle phase pollutants [*Roden et al.*, 2006; *Johnson et al.*, 2010]. However, it is not a "full capture" approach and so pollutant emission rates cannot be directly measured because emissions' dilution is not controlled. Fuel use on a per-cooking session basis was not collected to minimize intrusion and not disturb the households during other measurements (e.g., indoor air quality and blood pressure), so only data on household daily average fuel use are available and are published in the companion paper [*Aung et al.*, 2016].

Emission factors are calculated using the "carbon balance" approach, in which carbon from CO and CO_2 levels above background are attributed to fuel sources and a woodfuel carbon fraction of 50% is assumed, following reasoning described by *Roden et al.* [2006]. PSAP measurements include a correction for a recognized bias attributable to reduced response at increased filter loading [*Bond et al.*, 1999] and are dilution corrected based on the flows measured by the instrument's internal flowmeter and that used on the line which

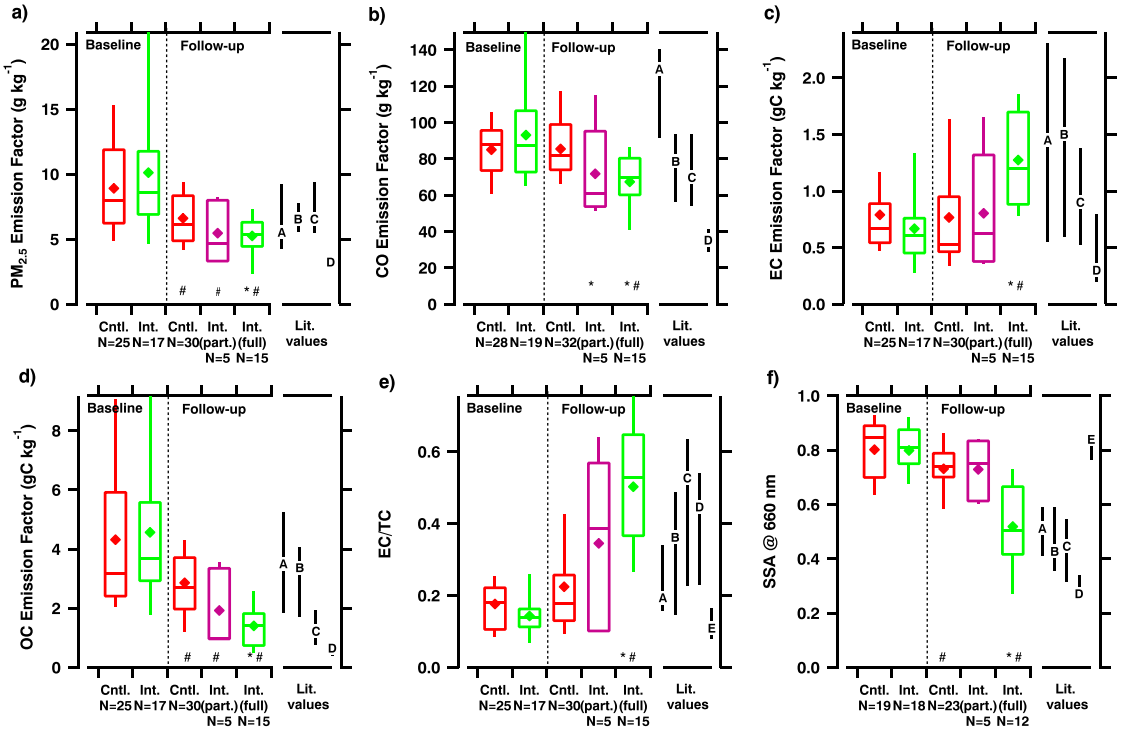

Figure 1. Box and whisker plots showing variability in quantities in baseline (before intervention; left two open bars) and follow-up (postintervention; right three open bars) measurements for control (red), intervention (green), and partial intervention (purple) groups. (a) $PM_{2.5}$ EF; (b) CO EF; (c) EC EF; (d) OC EF; (e) EC/TC; and (f) SSA. Boxes and whiskers indicate 25th to 75th and 10th and 90th percentiles, respectively; central lines indicate median and diamonds indicate group mean. Numbers on category axis indicate sample size; asterisk and number sign indicate that a group mean is significantly different (one-tailed Wilcoxon rank-sum test) than control group mean in same and previous seasons, respectively. Also shown with letters as markers are median and interquartile range for quantities from *Roden et al.* [2009] for several groups (test setting/stove type): (A) field/traditional, (B) field/improved no chimney, (C) field/improved with chimney, (D) lab/all tests, and (E) field/open biomass burning. Note that literature values for SSA are at λ = 532 nm and so SSA can only be compared in a relative sense between stove/combustion types due to the wavelength dependence of SSA [*Wathore et al.*, 2017].

metered filtered dilution air into the PSAP sampling line. Analysis also makes use of the commonly applied modified combustion efficiency (MCE = $\Delta CO_2/(\Delta CO_2 + \Delta CO)$, where Δ indicates background-subtracted molar mixing ratio) metric, used to estimate combustion efficiency. The optical properties of emitted particles are quantified using single scattering albedo (SSA = $B_{sp}/(B_{sp} + B_{ap})$, where a lower value indicates particles with greater relative absorption to overall particle light extinction. The wavelength dependence of aerosol light absorption is quantified using the Angstrom absorption exponent (AAE), approximated using the red and blue (λ = 660 and 467 nm, respectively) PSAP wavelengths and the power law dependence of absorption coefficient on wavelength ($B_{ap}(\lambda)\ \alpha\ \lambda^{-AAE}$) [*Andreae and Gelencsér*, 2006].

3. Results and Discussion

3.1. Emission Test Characteristics

100 cooking sessions were captured in total during preintervention (baseline; *N*(control) = 28, *N*(intervention) = 19) and postintervention (follow-up; *N*(control) = 32, *N*(intervention) = 21) periods. In ~40% of total study households the adoption of the intervention was incomplete and only one of two intervention stoves was used [*Aung et al.*, 2016] alongside a traditional stove. A subset (*N* = 6) of postintervention (follow-up) tests are thus grouped separately as "partial" interventions.

3.2. Intergroup and Interseason Changes in Integrated Emission Factors

Figure 1 shows box and whisker plots of EFs and other relevant emission properties for the five groups discussed above. There is substantial overlap between emission characteristics in control and intervention groups in preintervention testing. In most cases, means were greater than medians, suggesting a nonnormal distribution; one-sided Wilcoxon rank-sum tests are used to test for significant differences ($p < 0.05$) between groups.

None of these quantities were significantly different between baseline control and intervention groups, suggesting that randomization was effective and that our sample is effectively characterizing variability in these quantities. EF and other values for all tests are included in a spreadsheet file in the supporting information.

For full intervention groups during follow-up, mean CO and OC EFs showed significant reductions relative to control, of 19 and 52%, respectively. Mean $PM_{2.5}$ EF was 20% lower in intervention tests ($p = 0.05$); the median EF was 14% lower than the control group median. Conversely, quantities related to EC and specific absorption all showed consistent changes indicating greater contributions to particle emissions from EC. Mean EC EF and EC/TC increased by 65 and 127%, respectively. A laboratory study of the same rocket stove (not shown in Figure 1) measured an EC/TC of 0.8, higher than the central tendency values of ~0.5 measured in the field, but it has been observed that laboratory tests typically have higher EC fractions than tests in the field [*Roden et al.*, 2009]. Higher EC/TC values indicate the increased prevalence of strongly light-absorbing carbon (EC) and are reflected in emissions from follow-up control and intervention stoves having an SSA of 0.72 and 0.53, respectively. Therefore, emissions from intervention stoves have significantly greater specific absorption, and since EC EFs are higher and fuel use unchanged, the net climate impact of the intervention is likely toward increased warming. Fuel use data are discussed in a companion paper [*Aung et al.*, 2016] and show no significant change in household daily use; possible reasons for the lack of observed fuel use reduction include worse-than-expected stove performance and increase in cooking activity to meet "suppressed demand" for energy. In general, properties of emissions from "partial intervention" households are, as expected, intermediate to the control and full intervention group and are highly variable, partially due to the small sample size.

For several quantities (PM EF, OC EF, and SSA), significant differences were observed between control group tests in the two seasons, suggesting that season can play an important role in the quantity and properties of emissions. For example, mean PM EF in controls was reduced by 26% from baseline to follow-up, similar to the in-season reduction in PM EF from control to full intervention households (in follow-up) of 20%. Therefore, a change in season had a similar *relative* impact on PM EF as the intervention. This change is likely due to baseline measurements encompassing the end of the monsoon season and the weather during follow-up being significantly hotter and drier [*Aung et al.*, 2016]. Fuelwood moisture content was significantly higher in baseline than follow-up (mean of 10% versus 7%; $p < 0.001$). However, variation in the distributions of fuel types and cooking practices may also influence emissions performance. We cannot make a quantitative determination on these latter points, but note that similar fuel types were used across seasons and no substantial change was noted in diet. Interestingly, though CO and PM EFs are correlated (discussed below and shown in Figure 3), only PM EF showed significant seasonality, indicating that CO and PM formation mechanisms may be affected differently by seasonal changes.

3.3. Comparison of Emissions and Indoor Concentration Data

Changes in pollutant quantity and properties measured during emission tests and indoor air quality measurements [*Aung et al.*, 2016] are broadly consistent and suggest two things about HAP in this setting: (1) there was substantial seasonal variation in the link between emissions and indoor concentration (and by extension, exposure) and (2) stove emissions are the dominant contributor to PM concentrations in these households. Significant seasonal differences in indoor concentrations in control households were also observed [*Aung et al.*, 2016] but in the opposite direction as that observed in PM EFs (median indoor $PM_{2.5}$ was 65% *higher* in control households in follow-up). This is a surprising finding, as one would expect, all else equal, for reduced emission factors to be associated with lower indoor concentrations. This association *is* observed within the follow-up emissions and indoor air quality measurements: the differences in intervention versus control EFs measured during follow-up are consistent with the reductions in indoor $PM_{2.5}$ concentrations (median intervention household concentration was 26% lower than that in the median control household). This implies that there is a consistent variation in the ventilation/air exchange conditions in these households between seasons, as a similar *reduction* in fuelwood use from baseline to follow-up was observed in both intervention and control households (median daily use reduced by 14%). Based on lower fuel use and PM EF values, we can estimate that daily PM emissions were *reduced* by ~40% in control households from baseline to follow-up. However, a ~60% *increase* in median 24 h $PM_{2.5}$ concentration was observed [*Aung et al.*, 2016]. Contributions from other air pollution sources are unlikely to be contributors to the observed seasonality. Other contributors (e.g., agricultural burning, vehicles, and indoor lighting) were observed to be rare or nonexistent during the study [*Aung et al.*, 2016]. For example, the use of kerosene for lighting or cooking was

observed in less than 5% of tests (see test-specific notes in supporting information spreadsheet). Overall, this suggests a substantial change in the linkage between emissions and indoor concentrations across seasons, which should be considered in future research.

Similarity in PM optical properties observed during emissions and indoor concentration measurements can also be used to address source contributions to HAP and examine interseasonal variation in the emission/concentration relationship. Direct comparison of absorption coefficient from the two measurements are complicated by the different methods (PSAP light absorption at $\lambda = 660$ nm in emissions and filter-based reflectance measurements of indoor 24 h filters) applied, though absorption and reflectance values have been shown to be well correlated [*Taha et al.*, 2007; *Larson et al.*, 2009]. However, absorption/PM ratios give insight into the sources of PM, as sources other than stove combustion (e.g., dust, regional aerosol, and incense) are expected to have much lower ratios than a cookstove source. Figure S2 in the supporting information shows the B_{ap}/PM ratio (units of $m^2\ g^{-1}$) for the baseline and follow-up emission measurements. The mean ratio for intervention stove tests (follow-up full intervention) is approximately twice (91% greater; $p < 0.05$) that for follow-up control tests, consistent with expectations from EC/TC and SSA data shown in Figures 1e and 1f. This increase is also consistent with a sharp (~70%) and significant increase in the mean ratio for indoor measurements discussed by *Aung et al.* [2016]. The seasonal change in the ratio for the control group was in the same direction for emission and indoor samples but less strong (13% and 36% increase, respectively). Therefore, the observed changes in the ratios of absorbance to $PM_{2.5}$ are broadly consistent between the two measurement sets, suggesting that cookstove emissions are driving indoor concentrations and there is a distinct seasonality in the link between emissions and indoor concentrations in this setting.

3.4. Comparison With Other Field and Laboratory Emission Measurements

Figure 1 includes emission characteristics for field and lab measurements from *Roden et al.* [2009], the most extensive comparison of field emission data available in the literature (though others are discussed below). The range of PM EFs for traditional stoves (bar "A") from Roden et al. is quite consistent with the follow-up control group, but below the range of values measured at baseline. CO EFs for traditional stoves were higher in the Roden et al. study, but other quantities are in close agreement. Intervention stove emission quantities (follow-up full intervention) are also generally in good agreement with the "Improved stoves without chimneys" (bar "B") taken from *Roden et al.* [2009], though the EC/TC values were more closely overlapping with their "chimney stove" tests. Finally, a clear contrast is seen between EC/TC and SSA for the cookstove tests and those typically measured in open biomass burning plumes (bar "E"), which tend to have far less EC and greater SSA than biomass stove emissions measured in this study and from *Roden et al.* [2009].

The two field data sets also both clearly show that PM and CO EFs from field measurements are significantly higher and more variable than those measured in standard laboratory tests. Laboratory test EFs from *Roden et al.* [2009] (marker "D" in Figure 1) are far lower and less variable than those they measured in the field. For a more specific comparison, WBT fuel-based EFs for rocket stoves (similar to the intervention stove, which was also tested by *Just et al.* [2013]) from *Jetter et al.* [2012] range from 21–48 (median 35) g kg^{-1} and 1.4–3.9 (median 2.2) g kg^{-1} for CO and PM, respectively, depending on test phase, stove model, and fuel moisture level. Intervention stove CO and PM EFs in our field tests were greater by a factor of 2 or more, ranging from 34–115 (median 66) g kg^{-1} and 1.8–8.2 (median 5.3) g kg^{-1}, respectively. Traditional stove tests show a similar difference; CO and PM EFs for three stone fires from Jetter et al. ranged between 29–74 (median 44) and 1.8–8.8 (median 3.3) g kg^{-1} relative to of 38–152 (median 83) and 2.5–24.4 (median 8) g kg^{-1} for all traditional stove tests from the current study, for CO and PM EFs, respectively. Mean CO EFs measured during laboratory controlled cooking tests (CCTs) of traditional and natural draft improved stoves in India (converted from an energy basis using the measured wood energy density of 21 MJ kg^{-1}) were 44 and 34 g kg^{-1}, respectively [*Arora et al.*, 2014]. A related study measured PM, OC, and EC EFs broadly consistent with the laboratory measurements shown in Figure 1 but cannot be directly compared as they were of total particulate matter not $PM_{2.5}$ [*Arora and Jain*, 2015]. These strong contrasts between laboratory and field performance are consistent with differences observed in a range of other field measurements [*Johnson et al.*, 2008; *Wathore et al.*, 2017].

3.5. Paired Comparisons of Household Tests

Changes in EFs between seasons are also examined by comparing baseline to follow-up differences (pre-post) in individual households EFs across groups. This approach helps to control for consistent

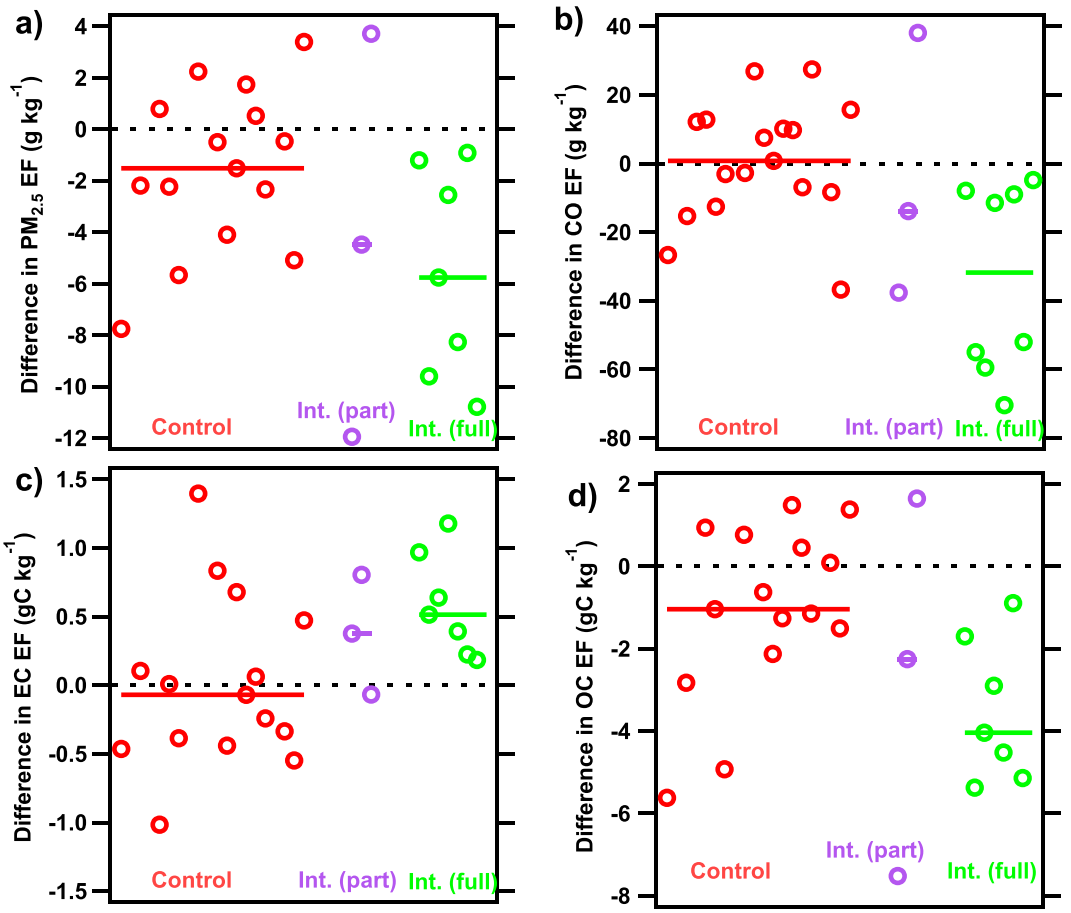

Figure 2. Plots showing the difference (post-pre) between EFs measured in the same households, separated by group, for (a) $PM_{2.5}$, (b) CO, (c) EC, and (d) OC. Horizontal spread in points is only to separate individual households; line shows median difference for the group.

interhousehold differences in EFs. Figure 2 shows a plot of the change in EFs for paired tests; most points represent the average of two tests per season in a given household. In general, the median differences in post-pre EFs are consistent with the difference in group median EFs (Figure 1), suggesting that full-group comparisons capture interhousehold variability. For example, the median reduction (follow-up–baseline) in household PM EF (Figure 2a) in control (full intervention) groups was 2.2 (5.8) g kg^{-1}, while the difference in median PM EFs (Figure 1a) was 2.3 (3.4) g kg^{-1}. Median changes across all EFs are in the same direction (decrease in CO, PM, and OC, minimal change or increase in EC) and of similar magnitude to differences in group medians, confirming that interhousehold variability is well characterized in our sample. A notable observation in Figure 2 is the bifurcation in EF difference observed among the full intervention group, especially noticeable for PM, CO, and OC. Individual household reductions in PM and CO EFs are well correlated (R^2 = 0.75) for the full intervention group, indicating that there is a strong and relatively consistent difference in household stove operation/performance. This suggests that while all households received training in operation of the same stoves, household variation in stove usage or the type or condition of fuel used may lead to consistent differences in emissions performance. However, the sample size for these paired test households is small and the larger differences are both due to large values for preintervention or small values for postintervention EFs, and so no firm conclusions about what drives these differences can be reached. Further, due to the lack of real-time observations during testing, we cannot comment on operational practices in specific households.

An association between CO and $PM_{2.5}$ EFs also holds in the complete data set (Figure 3), though the correlation is only moderately strong (R^2 = 0.52). The linear relationship across stoves is quite similar to that found for field measurements by *Roden et al.* [2009], despite the fact that their tests were in Honduras on a different set

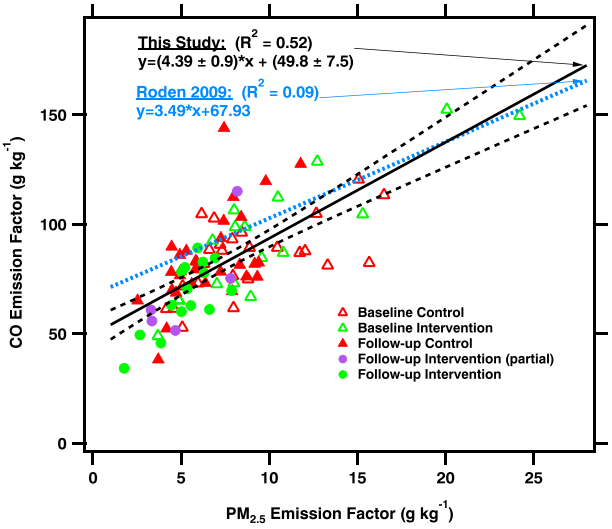

Figure 3. Scatterplot of all CO versus PM emission factors measured during campaign along with linear regression fit of all data and 95% confidence interval on the regression. Also shown (blue dotted line) is the regression fit for field emissions data from *Roden et al.* [2009].

of stoves with different fuel types. While there is substantial scatter in data from this study and that of Roden et al., this consistency does suggest that CO, which is substantially easier to measure with portable equipment than PM, may be a useful proxy for the central tendency in PM EFs but that high variability results in relatively wide confidence intervals on this relationship. Figure S3 in the supporting information shows the same data with regression fits for the individual test groups. The slope is somewhat greater for the follow-up measurements (and especially the intervention stoves) than for baseline measurements. However, the variability in regression parameters for the three groups with traditional stoves (control group baseline and follow-up and intervention group baseline) is of similar magnitude to the variability across stove types. Therefore, this suggests that the relationship may vary by season and technology, but high variability is pervasive and precludes a firm statement as to their relationship.

3.6. Emission Optical Properties

Test-average B_{sp} correlated strongly ($R^2 = 0.76$; supporting information Figure S4) with gravimetric $PM_{2.5}$ concentration, suggesting that optical scattering data is an effective proxy for PM mass. This correlation is stronger within individual groups, and the slope varies with season and stove technology. Linear regression indicates a mass scattering coefficient (MSC) of 3.2 $m^2\ g^{-1}$ across all tests. Figure S4 shows the correlation and the range of MSC values for groups, with lower MSC values for groups with higher EC/TC (Figure 1e). This observation cautions against using light-scattering measurements as direct proxies for PM mass across differing conditions and/or technologies but does indicate that within these groups it may be used as an indicator of time-varying PM concentrations.

Figure 1f shows that there were significant differences in the SSA of emissions across seasons and between stove technologies, with both traditional and intervention stove emissions in follow-up showing greater specific absorption (lower SSA) than baseline tests. This is consistent with the increased EC/TC ratio observed in these tests; SSA and EC/TC show a reasonably strong correlation ($R^2 = 0.49$; supporting information Figure S5) that is generally consistent with, though less steep than, that observed for emissions for laboratory burning for range of wildland fuels [*Pokhrel et al.*, 2016]. Minimal association was evident between MCE with SSA (supporting information Figure S6), though fairly robust relationships between these quantities have been observed in open burning of wildland fuels [*Liu et al.*, 2014; *McMeeking et al.*, 2014; *Pokhrel et al.*, 2016].

Analysis of the multiwavelength absorption data from the PSAP shows a wavelength dependence of absorption (Figure S7 in the supporting information) not substantially higher than that expected from pure BC (AAE ~1) and lower than values often seen in open biomass burning emissions [*Andreae and Gelencsér*, 2006; *Saleh et al.*, 2014]. Intervention stove emissions had significantly lower AAE (group mean ~0.9) than those from traditional stoves (group means ranged from 1.2 to 1.3), which were not significantly different across seasons and groups. Preble et al. also observed lower AAE values for another rocket stove model than for a three stone fire in laboratory testing [*Preble et al.*, 2014]; the rocket stove emissions in that study also had higher EC/PM ratios. These AAE values suggest that brown carbon likely makes a relatively small contribution to absorption for most emissions from cookstoves, and especially those from alternative stoves. This is not surprising, as test-average EC/TC ratios (Figure 1e) are above the range (~0.1) where substantial non-BC absorption is expected [*Saleh et al.*, 2014].

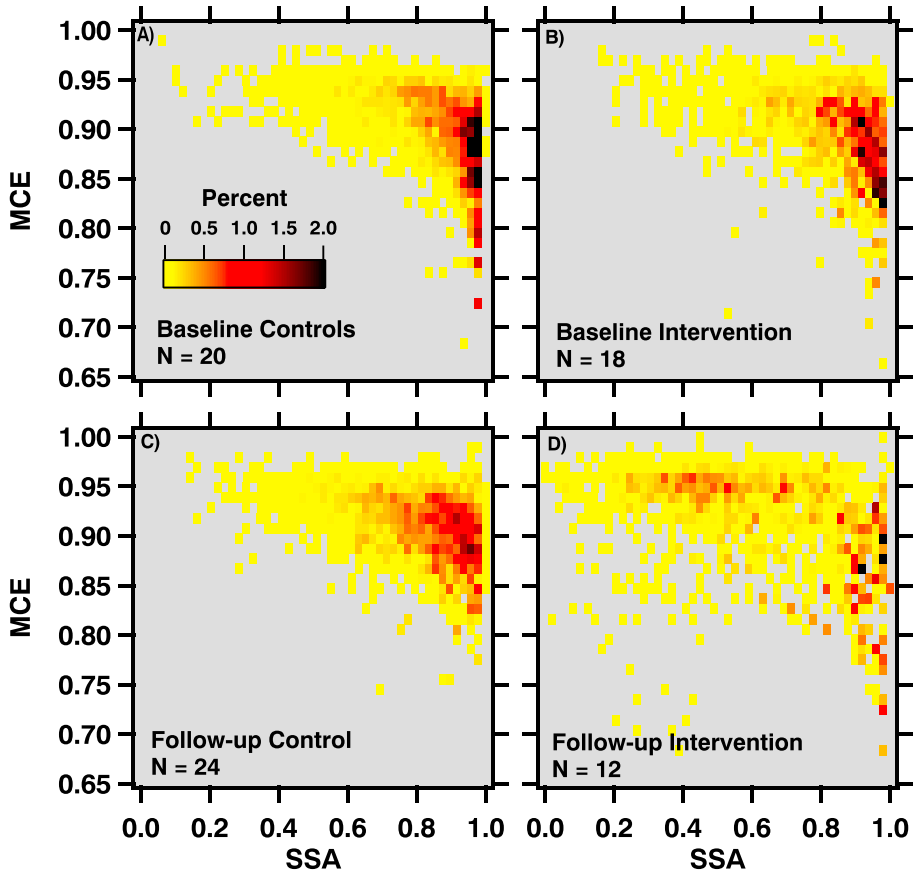

Figure 4. Bivariate histogram (PaRTED) plots [*Chen et al.*, 2012] showing the distribution of emitted PM (fraction of total aerosol light scattering) as a function of the MCE and SSA at the time of emission. (a) Baseline measurement control, (b) intervention groups, (c) follow-up measurement control, and (d) "full" intervention groups.

3.7. Real-Time Patterns in Emissions

Figure 4 shows "Patterns of Real-time Emission Data" (PaRTED) plots [*Chen et al.*, 2012] for the pooled data from the control and full intervention groups from baseline and follow-up measurements. The generation of these plots is discussed in *Chen et al.* [2012] and is briefly described here. PaRTED plots are bivariate histograms showing the distribution of total PM (as represented by real-time light scattering) emitted from a group of tests across the range of MCE and SSA values observed during individual combustion events (defined here as 1 min averages of real-time data). The histogram is weighted by the instantaneous scattering EF (IEF_{scat}; m^{-2} kg^{-1}), which is expected to be a reasonable proxy for PM emission factor given the strong correlation between B_{sp} and gravimetric PM (Figure S3), and then normalized. The histograms thus show the distribution of combustion (MCE) and particle optical properties (SSA) under which PM was emitted for a given test group. As is expected, the distributions for baseline control (Figure 4a) and intervention (Figure 4b) groups are very similar, with PM emissions dominated (81/76% for baseline control/intervention) by highly scattering (SSA $>$ 0.8) emissions emitted during combustion with 0.95 $>$ MCE $>$ 0.75. More absorbing emissions (SSA $<$ 0.7) make a smaller (10/14% for baseline control/intervention) contribution to PM emissions and are concentrated at MCE $>$ 0.9, indicating they are produced during more efficient, flaming combustion.

These distributions are distinct from those from follow-up tests for control (Figure 4c) and intervention (Figure 4d) groups, both of which show greater contribution from more absorbing particles. The control test distribution shows a similar shape to those from baseline tests but approximately double the contribution (19%) from events with SSA $<$ 0.7. This is consistent with the seasonality observed in baseline PM and OC EFs and SSA, all lower in follow-up (Figure 1). Even more striking is the difference between the intervention and control groups. The PaRTED plot for the follow-up intervention group shows a much

greater contribution from lower SSA events (45% at SSA < 0.7). It also has two clear "clusters," with one centered at MCE ~0.95 and SSA ~0.4 (~40% of total scattering but ~70% of was carbon emitted under these conditions) and the other with a similar location to the clusters observed in the control groups. This suggests that the intervention stoves may be operated in two distinct "modes": one in which they appear to operate similarly to baseline chulhas and emit more scattering particles at lower, more variable MCE; the other in which they are consistently operating at higher MCE and emit more absorbing particles.

This also suggests that a large fraction of the PM was emitted during relatively "rarer" events. PaRTED plots weighted by combustion event ($\Delta CO_2 + \Delta CO$), which serves as a proxy for fuel consumption rate, rather than IEF_{scat} (Figure S8) give insight into the relative prevalence (on a fuel-use basis) of events and thus indicate how much time/activity is spent in each mode. These show that for the intervention stove tests (Figures 4d/S8d) only 30% of the carbon is emitted at MCE < 0.9 and SSA > 0.7 but about 60% of PM is. Therefore, these "rarer" events contribute disproportionately to total PM emissions, suggesting perhaps that more variable/less controlled operation and/or starting emissions contribute to the difference between laboratory and field tests [*Roden et al.*, 2009]. The large contribution from starting emissions was also found during field measurements of more advanced stoves [*Wathore et al.*, 2017]. The distribution in Figure 4d is similar to that observed by Chen et al. for "improved nonchimney" stoves (similar to the rocket stove tested here), suggesting that these patterns are at least somewhat characteristic of operation of natural draft alternative stoves, even when operated in locations as distinct as Honduras and India. Lab tests of the same type of stove tend to show *only* the high-MCE/low-SSA "mode" in this distribution [*Chen et al.*, 2012; *Repoff*, 2015].

4. Conclusions and Implications

We present results of an emission characterization effort of a carbon-finance-approved cookstove intervention with a "rocket"-style natural draft cookstove including preintervention and postintervention emission tests in the same households. This study design allowed us to examine several important aspects that have not been studied during the field emission factor measurements completed to date. For example, we measured the same households across two seasons for both control and intervention groups, which gave us insights into the seasonal variation in emissions, the uptake of technologies across our study population and, because indoor air quality was also measured, linkages between emissions and indoor concentrations across seasons.

Mean CO, $PM_{2.5}$, and OC, EFs were significantly ($p \leq 0.05$) reduced (19, 20, and 52%, respectively), and EC EF increased (65%), in the intervention stove relative to traditional technologies in follow-up measurements. While mean intervention stove (follow-up) $PM_{2.5}$ EFs were significantly lower (~40%) than traditional stoves in baseline measurements, they were only 20% lower ($p = 0.05$) than traditional stoves in the follow-up period, which were also significantly lower than baseline traditional stove EFs. Therefore, while intervention stoves provided emission reductions, these reductions were significantly smaller than laboratory performance [*Jetter et al.*, 2012; *Just et al.*, 2013] would suggest or that are required to strongly reduce exposures [*Grieshop et al.*, 2011; *Johnson and Chiang*, 2015]. Relatively small changes in PM EF combined with the fact that fuel use was not significantly reduced in intervention households are consistent with the minor reductions (relative to controls) observed in mean 24 h indoor $PM_{2.5}$ concentrations in the full trial population [*Aung et al.*, 2016]. The strong seasonality, likely due to differences in fuel moisture and fuel types, in PM EFs (and other emission characteristics) is an important finding. For example, it means that an intervention trial without controls would have likely overestimated the emission reduction from the stoves (or conversely, underestimated reductions had the seasons been reversed).

Measured EFs were highly variable, and our study design enabled us to partially control for interhousehold variability. The median interseason (baseline – follow-up) EF differences in paired tests from the same households were similar to differences in the medians of entire groups, suggesting that variability is dominated by test-to-test variation and that our sample captured this overall variability effectively. A subset of paired tests in intervention households showed much stronger decreases in CO and PM EF either due to high preintervention or low postintervention values. While this suggests that some households operate the stoves in consistently cleaner ways, we do not have observations that can point to specific behaviors/factors that lead to this outcome. Furthermore, relying on careful operation for stove performance is likely not advisable in such interventions, as an effective technology should be robust to widely varying user behavior.

Our results suggest that rocket stoves such as that tested here are not an effective means to reduce BC emissions and mitigate climate forcing from these short-lived species (though they may provide other benefits including fuel use reductions). Other lab [*MacCarty et al.*, 2008; *Preble et al.*, 2014; *Arora and Jain*, 2015] and field [*Roden et al.*, 2009; *Johnson et al.*, 2011a; *Kar et al.*, 2012] studies have also observed greater absolute or relative contributions from EC/BC to PM emissions from natural draft stoves, and so this finding should not be especially surprising at this point. Intervention stoves had significantly higher EC EFs, and EC contributions to PM, than traditional stoves, and thus light absorption from aerosols will be greater and the potential offsets from cooling OC lower. This is borne out by measurements of aerosol optical properties showing that the rocket stoves emitted particles with significantly lower SSA than those emitted by traditional stoves in either season. Analysis of real-time emission data shows that PM emissions from intervention stove tests appeared in two clusters, with more absorbing particles emitted largely at higher efficiencies and a substantial fraction of PM mass emitted in relatively rare events of more highly scattering particles. Such rare events likely represent start-up and fuel feeding [*Wathore et al.*, 2017], along with smoldering-phase emissions. This suggests that, as indicated by lab testing, "ideally" operating rocket stoves will have lower PM emissions but that specific absorption of the emitted particles will likely increase.

The results of this study are consistent with observations of indoor and ambient air quality also collected during this trial [*Aung et al.*, 2016]. The marginally significant reduction in PM EFs is consistent with the observed small reduction in indoor PM concentrations (and presumably exposures) and no observed decrease in village-center PM levels. Likewise, in both emissions and indoor observations, the ratio of absorption to PM was increased during follow-up measurements, and more strongly for households making full use of the intervention stove. This is consistent with the observed strong seasonality in emission factors and properties and the dominant contribution from stoves to indoor PM. Interestingly, the seasonality observed in PM emission factors in control households (follow-up $<$ baseline) was in the opposite direction as that observed in indoor PM concentrations (follow-up $>$ baseline), highlighting that seasonally varying conditions (e.g., meteorology and household ventilation/air exchange rates) may be important modifiers for the link between indoor emissions and concentrations and exposures. This is an important finding and highlights the importance of carefully designing evaluations to separate the influence of time-varying processes and properties.

While this intervention study provides valuable insights into the emissions performance of traditional stoves and a single alternative natural draft stove, a number of limitations should be considered in interpreting results or in planning future trials. First, our overall study design, in which multiple outcomes (e.g., emissions, air quality, and health) were considered, affected our ability to complete detailed observations of household activities that might have yielded important information about stove performance. For example, direct fuel-use measures on per-task/-meal basis or observations of activity that allow linkage between fuel type/activity and emissions were considered too obtrusive, and so this information is not available. Per-task fuel use activity would have given important insight into the potential (if not realized) fuel savings from the stove, and activity observations might have given information about specific fuels/practices associated with high (or low) emitting operation of stoves. Likewise, the use of a partial-capture sampling arrangement (again, to minimize disruption to households) did not allow us to directly measure emission rates. Further, unattended measurements with a single filter set meant that some tests were compromised by overloaded filters (N ~7; typically of duration $>$4 h) or other issues (N ~13; mostly with PSAP issues) that might have been caught had testing been supervised. However, such trade-offs may be worthwhile considering the insights (e.g., the seasonality in linkages between emissions and indoor concentrations discussed above) that these parallel measurements have generated. Finally, we studied a single stove intervention in one village, and so great caution must be taken before extrapolating our results to other locations or program types. For example, our observation of no change in fuel use is by no means universal, and a number of interventions have been accompanied by substantial fuel savings and thus air quality/climate benefits. [*Berrueta et al.*, 2008; *Johnson et al.*, 2013]. However, our study emphasizes that careful evaluation may be necessary in different locations in order to ensure that hoped for benefits (e.g., fuel use, emissions, or climate forcing reductions) are realized.

Biomass-fueled stoves will likely serve an important interim role in many of the poorest areas as households transition to inherently cleaner gaseous and liquid fuels or electrical appliances. While stoves using unprocessed biomass fuels have the potential to strongly reduce health- and climate-related impacts of traditional combustion approaches [*Grieshop et al.*, 2011], this potential is often not reached in field settings. Pollutant emission is the first step in the chain of processes leading to impacts (HAP exposure;

climate forcing) that stove interventions aim to mitigate. It thus makes sense to emphasize this type of evaluation very early in intervention planning, to ensure that technology performance can lead to the intended positive outcomes. For example, the stoves used in this carbon-finance-approved intervention were associated with significant (CO) to marginal decreases (PM) in some pollutants and increases in others (EC). Emission measurements discussed here are consistent with a number of the outcomes observed in our larger study [*Aung et al.*, 2016]. While collecting emission measurements is more technically complex than some other quantitative assessments of performance (e.g., kitchen performance tests (KPTs) for fuel use and integrated samples for indoor air quality), this study and others [*Johnson et al.*, 2008; *Roden et al.*, 2009; *Wathore et al.*, 2017] suggest that a relatively limited number (tens) of tests could be used to assess the field performance of a proposed technology during pilot-scale deployment. These pilot assessments can indicate not only whether a technology can meet targets for performance during initial deployment but also be used to gain insights into user behavior and preference and how technology and intervention design can be modified to maximize the utility and performance of a technology. The need for early field evaluation of emissions performance is recognized in protocols currently under development within the International Organization for Standardization process [*International Organization for Standardization*, 2017] and by the Gold Standard Foundation [*Gold Standard Foundation*, 2015] that emphasize the need for field assessment of stoves in planning or seeking carbon finance to support interventions. Field emissions assessment like that presented here should thus be prioritized early in technology assessment and development to provide rigorous, quantitative estimates of the benefits reasonably expected from proposed interventions.

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