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New Emission Factors and Efficiencies from in-Field Measurements of Traditional and Improved Cookstoves and Their Potential Implications

Evan R. Coffey,*[,†] Didier Muvandimwe,[†] Yolanda Hagar,[‡] Christine Wiedinmyer,[§,¶] Ernest Kanyomse,[∥] Ricardo Piedrahita,[†] Katherine L. Dickinson,[§,⊥] Abraham Oduro,[∥] and Michael P. Hannigan[†]

[†]University of Colorado Boulder, Mechanical Engineering, 1111 Engineering Drive Boulder, Colorado 80309, United States

[‡]University of Colorado Boulder, Applied Mathematics, 1111 Engineering Drive Boulder, Colorado 80309, United States

[§]National Center for Atmospheric Research, 3090 Center Green Dr., Boulder, Colorado 80301, United States

[∥]Navrongo Health Research Centre, Navrongo Upper East, Ghana

[⊥]University of Colorado Boulder, Center for Science and Technology Policy Research, 1333 Grandview Avenue Boulder, Colorado 80309, United States

Supporting Information

ABSTRACT: Household cooking using solid biomass fuels is a major global health and environmental concern. As part of the Research on Emissions Air quality Climate and Cooking Technologies in Northern Ghana study, we conducted 75 in-field uncontrolled cooking tests designed to assess emissions and efficiency of the Gyapa woodstove, Philips HD4012, threestone fire and coalpot (local charcoal stove). Emission factors (EFs) were calculated for carbon monoxide (CO), carbon dioxide (CO_2), and particulate matter (PM). Moreover, modified combustion (MCE), heat transfer (HTE) and overall thermal efficiencies (OTE) were calculated across a variety of fuel, stove and meal type combinations. Mixed effect models suggest that compared to traditional stove/fuel combinations, the Philips burning wood or charcoal showed significant fuel and energy based EF differences for CO, but no significant PM changes with wood fuel. MCEs were significantly higher for Philips wood and charcoal-burning stoves compared to the threestone fire and coalpot. The Gyapa emitted significantly higher ratios of elemental to organic carbon. Fuel moisture, firepower and MCE fluctuation effects on stove performance were investigated with mixed findings. Results show agreement with other in-field findings and discrepancies with some lab-based findings, with important implications for estimated health and air quality impacts.

INTRODUCTION

More than one out of three people worldwide rely on the use of biofuels such as wood and charcoal to satisfy their domestic energy needs for both cooking and heating.[1] Often, biomass cooking involves traditional stoves with open fires; as energy conversion systems, these often have lower efficiencies and produce higher levels of air pollution, compared to improved technologies.[2] Potential health and climate effects from the emissions coming from this solid fuel combustion have been documented.[1−3] For example, the World Health Organization estimates that there are 4.3 million premature deaths annually caused by this particulate pollution source.[2] The climate effects of biomass cooking are also significant. Particulate carbon emitted from residential, power and industrial biofuel combustion is estimated to account for 39% of total global combustion particulate emissions, yet uncertainties are high especially for the residential sector.[4] Particulate black carbon (BC), a subclassification of carbonaceous aerosol with specific absorptive optical qualities, has been estimated to have a global warming effect (+0.17 to +2.1 W m^{-2}) and has been estimated to be the second largest global warming agent after CO_2.[5]

Improved cookstoves (ICS) offer a potential solution to the linked environmental and health challenges stemming from traditional biomass stoves. However, the success of ICS interventions depends in large part in how much cleaner these stoves actually are compared to traditional biomass stoves. Many studies have used laboratory-based tests to measure the performance of improved biomass stoves using metrics such as fuel use, firepower (the amount of fuel energy released per unit time), energy efficiency, and emissions.[6−14] Laboratory-based tests typically use a standardized water boiling test (WBT) in a well-controlled environment following a specified stove testing protocol. While these tests provide

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valuable information on certain metrics of stove performance, the tightly controlled conditions under which they are performed are not necessarily representative of actual cooking activities in a real home. Few studies have been completed in the field where the goal is to capture real-world use scenarios.[15−27] The advantage of field-based tests is that stove performance metrics can be measured for a specific cooking task under conditions of actual use, providing measurements that more accurately represent true cooking patterns.

In-field emissions monitoring is critical to assess the performance and impacts of traditional and ICS in real world conditions when local cooks prepare meals using local fuels. Here we present the results from a study in Sub-Saharan Africa during which in-field, real-time cooking emissions such as carbon monoxide (CO), carbon dioxide (CO_2) and both particulate organic carbon (OC) and particulate elemental carbon (EC) were measured for the first time in this region of the world. We also report modified combustion, heat transfer and overall thermal efficiencies. The performances of ICS are compared to traditional stoves using mixed effects models, based on their energy efficiency and pollutant emissions, to allow quantification of impacts potential stoves may have in future cookstove study programs in Sub-Saharan Africa.

■ MATERIALS AND METHODS

The stove testing described herein was completed as part of the REACCTING (Research on Emissions Air Quality Climate and Cooking Technologies in Northern Ghana) project; a 2-year, 200-home randomized control study in the Kassena-Nankana (K−N) districts of Northern Ghana.[28] Briefly, 200 rural households were randomly assigned to four different intervention arms of 50 households each: three receiving different combinations of two intervention stoves and a fourth that served as a control group. To obtain measurements from a variety of stove-fuel-food combinations, we recruited a subset of the intervention households, across the various groups receiving different types of ICS, and conducted emissions measurements. These visits began 4−6 weeks after the stoves were disseminated and lasted for 22 months. For each visit, we contacted the household ahead of time to schedule a date and time for testing. The household was only asked to use a specific stove/fuel type that evening and could prepare a meal of their choice. In addition to the stove operation training received at the start of REACCTING,[28] the cooks were instructed to prepare the meal as they normally would on that stove. Meal type preferences were most likely impacted by the stove/fuel specified.[28] Our analysis technique, described below, and sample sizing were designed to cover the main stove/fuel/food combinations through the seasons. A summary of sampling information can be found in the Supporting Information (SI) (Table S1).

Cooking Systems: Stoves, Fuels, and Food. *Stoves.* In-field stove performance testing was conducted on two types of traditional stoves commonly used in the K−N districts as well as two ICS introduced by the REACCTING project. Threestone fires (TSF) and locally crafted charcoal stoves (coalpots) were tested. TSF, using wood and crop residue fuels, are the primary stoves at rural households with most using coalpots as a secondary stove.[28] The REACCTING intervention employed two improved biomass stoves: the domestically made Gyapa rocket woodstove and the imported Philips HD4012-LS forced-draft, gasifier stove. Of the intervention groups, one group received two Gyapa stoves, one received two Philips stoves and one received one Gyapa and one Philips. The Gyapa woodstove is designed with a ceramic combustion chamber to retain heat for cooking, while the Philips design represents a transformative technological increase in efficiency and cleaner combustion using a battery powered fan to provide primary and secondary air for combustion. It is important to note the Philips, designed for solid biomass fuel, was frequently fueled with charcoal while Gyapas and TSF strictly burn wood/crop biomass in this region. SI Figure S1 shows the four stove types.

Fuels. Cooking fuels in the K−N districts are comprised of woody biomass, charcoal, and occasionally dung. Woody biomass is sourced primarily from neem (*Azadirachta indica*), sheanut (*Vitellaria paradoxa*), and mango (*Mangifera indica*) trees. Charcoal is made locally in earth-mound kilns. Carbon content of tropical trees in Ghana ranges between 45.8 and 49.8%.[29] A woody biomass carbon content of 49(±2)% was assumed in this analysis, which agrees closely with similar measurement campaigns.[18,30] Charcoal carbon content was assumed to be 72(±3)%.[30,31] Fuel moisture estimates (dry basis) were made in the field using a hand-held meter (General MMD5NP). Calorific values of a subset of wood (n = 9) and charcoal (n = 1) samples were measured using a bomb calorimeter (IKA C200). Higher heating values (HHV) for woody samples ranged from 18.8 to 19.5 MJ kg^{-1} with a median value of 19.1 MJ kg^{-1} (SD, 0.2 MJ kg^{-1}). The median HHV for charcoal was 32.0 MJ kg^{-1} (0.1 MJ kg^{-1}).

Food. All emission field measurements were taken during supper hours (15:00−20:00 local time). The most typical meal prepared in the districts, Tuo Zaafi (TZ), is a thick millet flour porridge, which is prepared by boiling water, adding ground millet flour, then simmering and stirring vigorously until a dense smooth porridge results. This meal typically takes 20−50 min to boil and the remaining time simmering, depending on the amount of food prepared. This starchy staple is usually eaten with vegetable soup made either alongside or prior to the TZ. A variety of local rice and bean dishes were prepared as well. Most dishes were cooked in local round-bottomed cast-aluminum pots varying in estimated size (small: <2 L, medium: 2−4 L, and large: > 4 L).

Stove Performance Metrics and Real-Time Cooking Measurements. In-field stove performance was assessed using a modified uncontrolled cooking test (UCT) procedure measuring gas phase and particulate emissions.[32] The UCT employs similar principles as the controlled cooking test (CCT) but aims to better reflect real performance in a given setting.[33] Unlike the CCT, the UCT allows the cook to prepare a local dish of their size and type using local fuels. The cook's preferences inherently add to the test variability. Gas phase and particulate emissions measurements were taken using an emission pod (EPOD) similar to the portable emission measurement system (PEMS) developed by Aprovecho.[34] The EPOD is powered by a standalone 12VDC battery and incorporates a hood[35] to capture emissions. The hood was assembled and placed over each test stove while the EPOD sensors stabilized. Three sides of the adjustable hood were lowered to ensure the highest level of emission capture while the side facing the cook was partially rolled up for unobstructed cooking. Emission sampling began directly before ignition, which was typically done using matches and crop stalks. Emissions were pulled into a 12-in. diameter duct with a large fan creating a well-mixed flow. A magnehelic pressure gauge (Dwyer Instruments, Inc.) was connected to a pitot tube inside

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the duct to measure air velocity used in the calculation of total diluted flow (4.8 ± 0.2 m^3min^{-1}) leaving the hood. The sample stream (2.00 ± 0.030 L min^{-1}) was drawn from the total flow through a PTFE tube to the sensor box through a 90 mm quartz fiber particulate matter (PM) filter (PallFlex, Tissuquartz 2500 QAT-UP). The sensor box, made of inert materials, housed a series of gas phase sensors. The sensor array measured CO (CO-B4 electrochemical, Alphasense, LTD), CO_2 (S-200 NDIR, ELT, Corp), temperature, and relative humidity. Subminute, logged data were stored on a modified UPOD platform (mobilesensingtechnology.com). A schematic of the apparatus is depicted in SI Figure S2.

Gas-phase sensors were calibrated via normalization in a laboratory setting using API CO-300 and Li-COR 840a CO_2 and H_20 gas analyzers prior to sampling. Additionally, multiple calibrations were performed throughout the entire sampling period from January 2014 to August 2016. Calibrations details and results are available in the Supporting Information (Figures S3, S4).

Total integrated PM, with no size cut, was collected on quartz filters for subsequent analysis of EC and OC using a Sunset Laboratory analyzer instrument following NIOSH 5040.[36] Research has shown that the size distributions of aerosol emissions from biomass combustion are typically unimodal with a peak in the range of 0.26−0.38 μm.[23] As such, we expect the PM collected to be approximated to PM with aerodynamic diameters ≤2.5 μm ($PM_{2.5}$).[12] This analysis did not correct for gas-phase artifacts on the filters.

Gas-phase and particle measurements were corrected for background concentrations using pre- and postcooking sampling periods, to reflect the emissions associated with cooking only. The partial capture carbon balance method (CBM)[18,20,23,25,26,37−39] was used to calculate emission factors (EFs), mass of a pollutant emitted per mass of fuel used, for CO and CO_2 (eq 1) and carbonaceous particulates (eq 2; TC = EC+OC). Total $PM_{2.5}$ was estimated using the sum of organic matter (OM) and EC, where OM/OC is estimated to be 1.9,[18] although we are aware this could be an underestimate of the total $PM_{2.5}$ mass. EFs were calculated per mass fuel burned (eqs 1 and 2), utilizing carbon mass ratios and therefore requiring conversions of average test volumetric concentrations of CO and CO_2 above ambient levels (ΔCO + CO_2), using the ideal gas law, to mass concentrations. The volume sampled is the product of sample flow rate and cooking duration.

$$\mathrm{EF}\left(\frac{\mathrm{mass_{species}}}{\mathrm{mass_{fuel}}}\right) = [\mathrm{species}]\left(\mathrm{in}\frac{\mathrm{mg_{species}}}{\mathrm{m}^3}\right) \times \left(\frac{1}{\Delta\mathrm{CO} + \Delta\mathrm{CO_2}}\right) \times \left(\frac{\mathrm{m^3_{CO2}}}{0.4905\mathrm{kg_C}}\right) \times \left(\frac{0.49\mathrm{kg_C}\mathrm{or}0.72\mathrm{kg_C}}{\mathrm{kg_{fuel}}}\right) \quad (1)$$

$$\mathrm{EF}\left(\frac{\mathrm{mass_{PM}}}{\mathrm{mass_{fuel}}}\right) = \left(\frac{\mathrm{PM_{filter-mass}}}{\mathrm{Volume_{sampled}}}\right) \times \left(\frac{1}{\Delta\mathrm{CO} + \Delta\mathrm{CO_2}}\right) \times \left(\frac{\mathrm{m^3_{CO2}}}{0.4905\mathrm{kg_C}}\right) \times \left(\frac{0.49\mathrm{kg_C}\mathrm{or}0.72\mathrm{kg_C}}{\mathrm{kg_{fuel}}}\right) \quad (2)$$

The CBM uses average molar concentrations of CO and CO_2 for an entire cooking event to calculate modified combustion efficiencies (MCE, eq 3), which we then use to approximate combustion efficiency (CE, eq 3). This approximation is reasonable as the majority of fuel carbon is emitted as CO and CO_2.[40] Other research has shown that ignoring byproducts such as methane, nonmethane hydrocarbons and other carbonaceous aerosols contributes negligibly,[41] causing a small bias from 1 to 2%.[7,39]

$$\mathrm{CE} = \frac{\text{energy released}}{\text{energy in fuel}} \sim \mathrm{MCE} = \left(\frac{\mathrm{CO_2}}{\mathrm{CO} + \mathrm{CO_2}}\right)(\text{molar basis}) \quad (3)$$

$$\mathrm{OTE} = \frac{\text{useable energy}}{\text{energy in fuel}} = \frac{\text{sensible heat} + \text{latent heat}}{\text{chemical potential energy}} = \left(\frac{m_{fc} \times C_{pw} \times (T_{bw} - T_{iw}) + (m_{w,evap} \times H_l)}{H_f \times \left(\frac{m_{f,u}}{M+1}\right) - (H_c \times \Delta C_c)}\right) \quad (4)$$

m_{fc} = mass of food cooked (kg),

C_{pw} = specific heat of water (4.186kJ $kg^{-1}K^{-1}$)

T_{bw} = boiling temperature (K),

T_{iw} = initial water temperature (K)

$m_{w,evap}$ = mass of evaporated water (kg),

H_l = latent heat of vaporization (2257kJ kg^{-1})

H_f = calorific value of fuel (kJ kg^{-1}),

$m_{f,u}$ = mass of fuel used (kg, wet)

M = fuel moisture content (dry basis),

H_c = calorific value of char (kJ kg^{-1}),

ΔC_c = mass of char (kg)

Overall thermal efficiency (OTE, eq 4) was calculated for each sample and defines how efficient the stove is converting fuel carbon to useable heat. The latent heat of vaporization[42] was determined using local boiling temperature (372 K) and pressure (10^5Pa). Calorific values of the fuel were calculated by subtracting 1320 kJ kg^{-1} and 760 kJ kg^{-1} from the measured HHV for wood and charcoal fuel, respectively.[10] Meals were weighed in the field using digital (Escali P115C) and analog (Taylor 3880) food scales. Due to difficulties of making temperature measurements of meals, a 75° temperature change was assumed for sensible heat calculations. Uncertainty associated with this assumption and other calculations is discussed in Section S1 of the SI. In a subsample of UCTs (n = 6), individual ingredients were weighed; water was identified, in several meals, as the overwhelming majority of food mass and consistent percentages of evaporated water mass to total meal mass were found resulting in 35% (SD, ± 3%) of useable energy as sensible heat, despite the widespread use of lids. The ratio of sensible heat to latent heat was consistent across stove/fuel types preparing the most common meals. Therefore, latent

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heat was assumed to be 2.86 times the sensible heat calculation for all samples.

Stove Performance Mixed Modeling. The benefits of conducting uncontrolled tests is that they capture real-world performance of stove systems including fuels, cooking vessels and users. The downside is that these tests introduce a wide range of influential factors that vary across tests such as individual cooking behaviors, kitchen geometries, and environmental conditions, making it more difficult to isolate the effects of interest (e.g., variation due to stove type, fuel type, or fuel moisture). To address these factors, mixed effects models were employed to quantify differences among the improved and traditional stove performance metrics. Most studies examine field stove performance using Student's t tests (e.g.,[38]). To our knowledge, no study has used mixed effects models to analyze EFs and efficiencies from stoves operated in the field by end users, although many have compared kitchen pollutant concentrations, exposures and health outcomes using such methods.[24,43−46] Stove/fuel combination categories were created for this analysis to compare with other published results.[7,38] To satisfy the normality assumptions required to use regression modeling, logarithmic transformations of EFs and efficiencies (γ) were fit using eq 5 with slight modifications. Differences among stove performance factors were assessed using the modeled coefficients (β) along with error (ε) and random household effects (α) from repeated measurements. Correlation coefficients among model parameters were found to be very weak to moderate.

$$\log(\gamma) \sim \beta_0 + \beta_1(\text{stovefuel}) + \beta_2(\text{firepower} \times \text{stovefuel}) + \beta_3(\text{MCE\_std} \times \text{stovefuel}) + \beta_4(\text{moisturelevel}) + \beta_5(\text{foodproductionrate}) + \alpha + \epsilon \qquad (5)$$

Firepower (kW) fluctuates constantly during combustion. Mean firepower was used in the modeling process. Higher firepower can decrease overall cooking time, often in exchange for lower thermal efficiencies. Laboratory stove testing using time-resolved PM measurements have shown large "spikes" of incomplete combustion products linked to fueling events,[47] which may not be captured well by average firepower. In an effort to explain variation linked to this phenomenon, the standard deviation of minute-averaged MCE calculations (MCE_std) were incorporated into the models. Food production rate (g min^{-1}) is the ratio of the mass of food cooked to cooking time. This metric allows us to account for stove/fuel combinations that cook variable amounts of food for different durations. Biomass fuel moisture effects on stove performance are not well understood with some research finding nonlinear relationships with PM and CO emissions and fuel use during lab tests,[48] with others pointing to decreased combustion efficiency leading to higher pollutant emissions,[13,49,50] and still others finding no significant effects.[18] Moisture measurements were categorized as low (≤10% dry basis), medium (11−20%), and high (≥20%), to shed light on these discrepancies in the literature. Moisture levels for charcoal were assumed low and therefore not included in modeling. This is a safe assumption as charcoal is commonly kept dry in small sacks in covered spaces. Model interactions between stove/fuel type and firepower and MCE_std offer additional information on whether spikes in these measurements have a better or worse effect on stove performance. In other words, firepower or variations of MCE during cooking events may be significant factors explaining pollutant emissions or efficiency differences between ICSs and TSFs.

■ RESULTS AND DISCUSSION

Seventy-five UCTs (TSF: $n = 21$, Gyapa: $n = 18$, Philips wood: $n = 12$, Philips charcoal: $n = 14$, coalpot: $n = 10$) were completed between November 2013 and August 2015 (SI Table S2). The values from the model comparisons, reported below, represent mean percent differences (transformed back to linear scale) from traditional cooking counterparts (Gyapa and Philips wood compared to TSF, and Philips charcoal to coalpot) indicating 95% confidence intervals (CI) and p values (see SI Table S3 and Figure S5). Statistical significance is assessed at the 5% level.

Emission inventories often include EFs as a mass of species emitted per unit mass of dry fuel consumed. Figure 1 shows

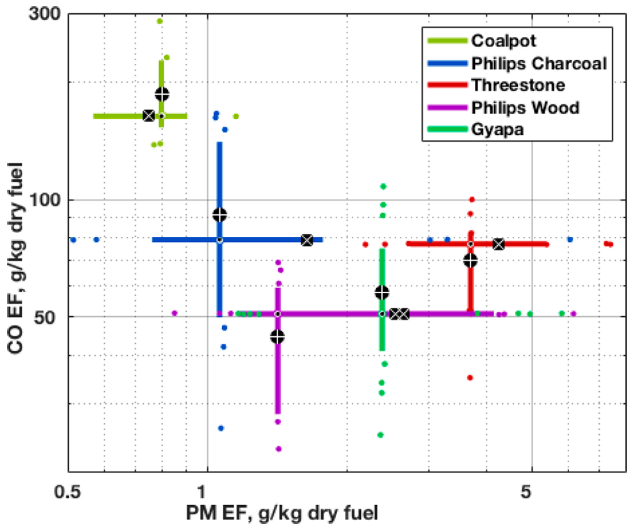

Figure 1. Vertical and horizontal boxplots showing distributions of dry fuel basis EFs of the five stove/fuel categories indicating medians (white bullseyes) and interquartile range, CO means (circle with "+") and PM means (square with "x"). Dots are samples outside the interquartile range.

distributions of CO and PM EFs with mean and median values for each stove/fuel group on logarithmic axes. On average, the Philips burning wood emitted 46% (−65, −18; lower and upper % confidence interval, $p < 0.01$) *less* CO and 13% (0, 28, $p = 0.04$) *more* CO_2 than the TSF per kg of dry fuel. These significant differences are a result of the Philips' combustion environment; jets of air allow thorough mixing while the light, ceramic combustion chamber facilitates higher combustion temperatures. The Gyapa rocket stove, on the other hand, had no significant CO or CO_2 EF (dry fuel basis) differences from the TSF. Philips MCE variations were associated with 20% (4, 37, $p = 0.01$) *higher* CO EFs, suggesting a highly variable combusting Philips, perhaps due to variable fuel preparation, fuel loading or fan control, could have higher CO emissions than a poorly operated TSF. Neither Philips nor Gyapa PM emissions were significantly different from the TSF. Although not significant, the magnitude of the medium (+) and high (−) moisture level effects on PM emissions relative to low moisture were opposite, suggesting nonlinear relationships between PM emissions and fuel moisture.[48,51,52] Compared to the coalpot, the Philips burning charcoal emitted 77% (−92, −34, $p < 0.01$) *less* CO and 58% (−90, 81, $p = 0.04$) *less* PM on average.

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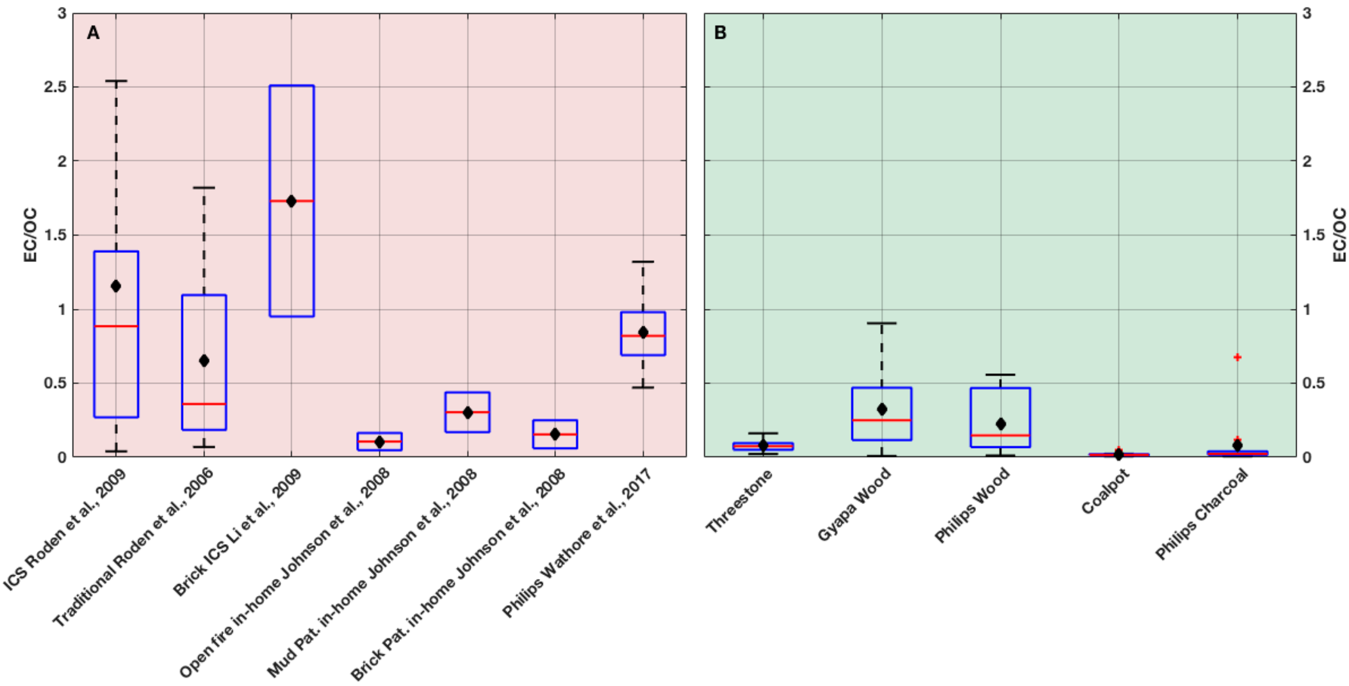

Figure 2. Boxplots of EC/OC values measured in field from literature (A) and contributions from this study (B) by stove/fuel type. Box is interquartile range, whiskers: first/99th percentiles, line: median, diamond: mean. Box with no whiskers are max and min. EC/TC values reported in literature modified to EC/OC where TC = EC+OC. Li et al.[23] reported BC/OC and used IMPROVE protocol.

Research points to the increasingly important contribution of start-up emissions to overall PM,[20,26] underscoring the importance of time-resolved PM measurements.

Gyapa EC EFs on a dry fuel basis (SI Table S3), were on average more than twice as high (122%, 9, 353, $p = 0.03$) as the TSF while the Philips (−22%, −71, 111, $p = 0.62$) burning wood was not statistically different. The Philips burning charcoal had no significant EC EF differences from the coalpot. The Gyapa·MCE_std and Philips·MCE_std interactions were significant factors in explaining 36% (19, 55, $p < 0.01$) and 52% (10, 109, $p = 0.01$) *more* EC than the TSF·MCE_std interaction effect. One postulation is that compared with the TSF, the ICSs' MCE fluctuations may result in more flaming where EC is preferentially formed. Field notes or visual documentation of flaming events/frequency could offer valuable information in future sampling. Wood fuel moisture effects on EC EFs show medium levels not significantly different than low levels, yet high moisture levels were a significant factor linked to EC reductions of 77% (−90, −47, $p < 0.01$). These results, although combustion temperatures were not directly measured in most samples, add evidence to the hypothesis of Zhang et al.[7] that relatively lower temperatures with high moisture fuel burning are not favorable for the formation of EC.

A growing body of research is finding increased ratios of EC to OC emissions from improved biomass stoves in the field[20,26,38,53,54] relative to traditional stoves. As such, it is critical to understand the ratio of EC to OC in the emissions from traditional and ICS in real-use scenarios.[55] EC/OC values were calculated using the quotient of EC to OC EFs (dry fuel basis). Figure 2A depicts the distributions of EC/OC data collected from other field campaigns and those sampled from this study by stove/fuel type (Figure 2B). From pane B, the ICSs exhibit more variation overall compared to traditional counterparts. This variation may be dominated by mixed user behavior (e.g., Philips fan speed, fuel (over)loading and inadequate fuel preparation). EC/OC values were on average 202% (63, 459, $p < 0.01$) *more* than the TSF for the Gyapa burning wood, and not significantly different for the Philips. Higher fuel moisture was linked to significant EC/OC reductions for medium (−49%, −71, −11, $p = 0.02$) and high (−69%, −85, −37, $p < 0.01$) levels relative to low levels (SI Table S3). EC/OC from both charcoal stoves had very low median values (<0.03) with the Philips showing high variability especially at varying firepower (111%, 21, 268, $p = 0.01$).

Average modified combustion efficiencies (Figure 3) increased 6% (0.6, 12, p = 0.03) for the Philips burning wood relative to the TSF but no significant differences for the Gyapa. The Philips burning charcoal had MCE 35% (14, 59, p = 0.04) larger relative to the coalpot. No ICS had significantly different OTEs or HTEs relative to its traditional counterpart. Rather, the mean operational firepower and food production rate fixed effects significantly estimated efficiencies in the mixed effects models. Wood burning stoves operated at higher firepower were linked to HTE and OTE decreases of 16% (−19, −12, $p < 0.01$) and 15% (−20, −12, $p < 0.01$) respectively. Similarly, charcoal burning stoves operated at higher firepower were 23% (−29, −16, $p < 0.01$) and 24% (−30, −18, $p < 0.01$) *less* efficient transferring heat to the food and overall, respectively. This finding supports the conjecture that on average, traditional and improved stoves are less efficient when operated at higher firepower supporting the need for variable firepower testing.[37,56] Focusing efforts to improve HTE in the field could reduce overall emissions, perhaps more effectively than optimizing MCE alone. The quicker a given mass of food was prepared the higher the efficiencies, but not substantially (1.1%, 1, 1.3, $p < 0.01$). Pot size was not a significant factor for estimating HTE or OTE as found in the lab,[48] but this could be due to a lack of observations across the stove/fuel categories. No significant fuel moisture effects on

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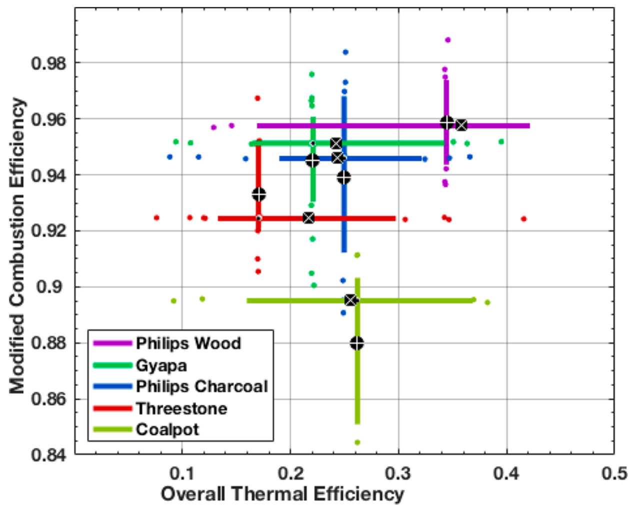

Figure 3. Vertical and horizontal boxplots showing distributions of overall thermal and modified combustion efficiencies for each stove/fuel category with median (white bullseyes) and interquartile range, MCE means (circle with "+") and OTE means (square with "x"). Dots are samples outside the interquartile range.

efficiencies were observed, contrary to other findings[9] suggesting decreased efficiency with higher fuel moisture content. SI Figure S6 shows distributions of efficiencies and EC/OC by stove/fuel type.

Emission factors based on useful energy delivered (Figure 4) provide the best metric for comparing performance across various stove/fuel types. Reductions of CO per MJ of energy delivered for the Philips burning wood were 51% (−69, −22, $p < 0.01$) on average compared to the TSF, with no significant difference for the Gyapa. The Philips burning charcoal emitted 91% (−97, −69, $p < 0.01$) *less* CO per MJ energy delivered than the coalpot. These substantial reductions are in large part due to improved combustion rather than heat transfer improvements. Firepower, again, was linked to 20% (6, 35, $p < 0.01$) and 39% (5, 83, p = 0.03) *higher* CO EF per MJ delivered for wood and charcoal stoves, respectively. PM emissions per MJ delivered were not significantly different for the ICS compared to traditional counterparts. The Philips·MCE_std interaction effect suggests that periods of high combustion variability may result in 22% (5, 42, $p = 0.01$) *more* CO than the TSF·MCE_std interaction on an energy delivered basis. There has been increasing discussion on how representative thermal efficiency (OTE) as a metric is of overall stove performance.[10,33,37] A stove that evaporates large quantities of water (whether or not it is considered useful to the user) would have a higher OTE than a stove that completed the same task using the same amount of fuel with less evaporation. To avoid biasing highly evaporative stoves, some prefer to report specific fuel consumption (SFC, ratio of dry fuel mass to food mass) as a more representative overall stove performance metric. Specific fuel consumption (SI Table S3) was 50% (−65, −27, $p < 0.01$) *less* for the Philips burning wood than the TSF while neither the Gyapa nor the Philips burning charcoal had significantly different performance. Simply stated, the Philips on average requires half the amount of dry fuelwood as a TSF to produce the same amount of food.

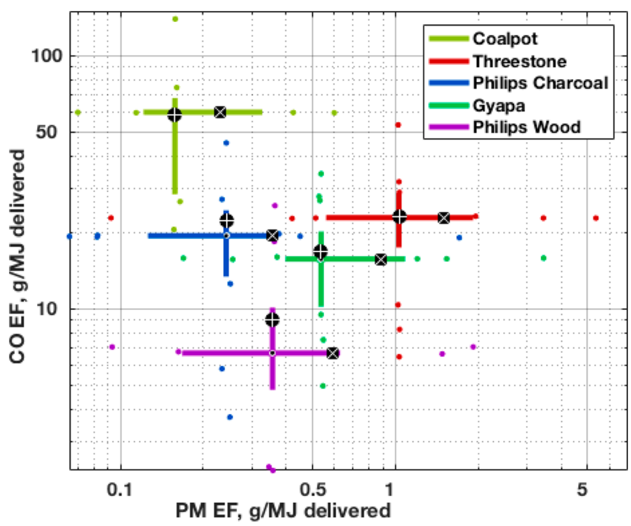

Figure 4. Vertical and horizontal boxplots showing distributions of useful energy basis EFs of each stove/fuel category with median (white bullseye) and interquartile range, CO means (circle with '+') and PM means (square with 'x'). Dots are samples outside the interquartile range.

Two campaigns employing similar methods have measured CO and PM EFs in the range of 28−143 (gCO kg fuel^{-1}) and 0.7−11.5 (gPM$_4$ kg fuel^{-1}) for a variety of traditional and improved wood stoves in Honduras.[18,20] These researchers measured increased EC/OC (Figure 2A) for the ICS compared to traditional open fires but results did depend on specific stove attributes (e.g., ICSs with and without chimneys). Wathore and colleagues[26] measured alternate cookstove models' performance in Malawi finding increased EC/OC (Figure 2A) from the Philips stove among traditional technologies. Johnson and co-workers[38] (Figure 2A) measured EC/OC emissions from improved mud-cement (EC/OC = 0.27) and brick (0.17) Patsari stoves that were on average three and two times the value of the traditional open fire (0.09), respectively. Several repeated tests on in-home open fires resulted in average CO_2 and CO EFs of 1532 ± 70 and 81 ± 14 (g kg fuel^{-1}), respectively, while the mud-cement Patsaris emitted 1558 ± 66 (gCO_2 kg fuel^{-1}) and 65 ± 12 (gCO kg fuel^{-1}) and the most carefully designed brick Patsari performing significantly better at 1617 ± 81 (gCO_2 kg fuel^{-1}) and 19 ± 26 (gCO kg fuel^{-1}).[38] Ludwig and co-workers[17] found average CO_2 and CO EFs of 1650 and 100 (g kg fuel^{-1}), respectively, from 94 wood-fueled stoves in Zimbabwe. Li and colleagues[23] performed modified field WBTs in four Chinese provinces reporting BC/OC for wood stoves in the range of 0.84−1.98, values that are substantially larger than our findings; however, this could partly be explained by a difference in analysis protocols. In general, findings from the literature for woody biomass emissions agree well with our measurement magnitudes. Twelve Controlled Cooking Tests (CCTs) using the Philips stove in Western India yielded significant kitchen level reductions in $PM_{2.5}$ (66%) and CO (55%) relative to the clay chula.[24] Moreover, the thermal efficiency of the Philips used in India was 25% on average, roughly 10 percentage points lower than our finding and with much less variation. Decreased variability is expected of more controlled CCTs completing a specific cooking task, in this case bringing a specified quantity of water to a boil. Differing test protocols, user behavior and fuel properties are responsible for efficiency discrepancies. The literature contains few in-field charcoal stove emission results. However, a campaign in Kibera Kenya measured 10 Jiko stoves finding

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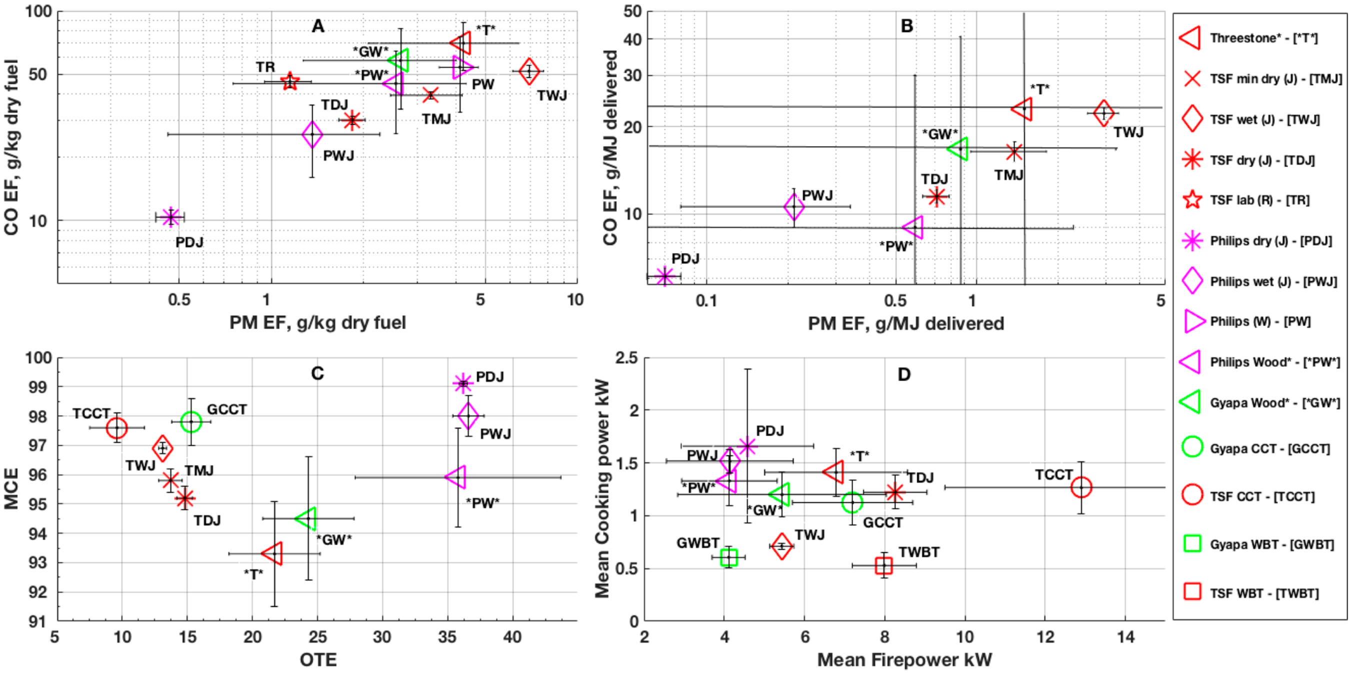

Figure 5. Lab and field test results of (A) CO and PM dry fuel basis EFs, (B) CO and PM per energy delivered basis EFs, (C) MCE (%) and OTE (%), and (D) mean cooking power and firepower levels from (*) this study, (J) Jetter et al.[10] WBTs, (R) Roden et al.[20] and (W) Wathore et al.[26] Results from 4 CCTs (v2.0) and 3 modifications of the WBT (Emissions and Performance Test Protocol) between the Gyapa (GWBT) and TSF (TWBT) were conducted outside at CU Boulder between 2014 and 2015. Error bars represent ±1 SD. Cold start values reported for WBTs.

average (±SD) CO_2, CO and PM mass emissions factors of 2130 ± 200, 393 ± 98 and 6.65 ± 5.2 g kg charcoal^{-1}, respectively.[25] Our CO_2 EFs have very high agreement, but CO and PM EFs found in this study are substantially higher than our findings. MCEs measured in that campaign averaged 84%, four percent lower than ours, explaining more products of incomplete combustion. Some of these discrepancies could also be due to varying charcoal carbon content and fuel moistures. Additional measurements from charcoal stoves in the field are recommended.

Emission factors (dry fuel and energy delivered bases) and efficiencies from lab tests[10,20] were compared to our measurements (Figure 5), comparing the "gap"[18,20,21,24,26,57] between field and lab measurements. Jetter et al.[10] performed TSF WBTs in triplicate at dry (9.5%) and wet (22.6%) wood (red oak) moisture levels, carefully and "minimally" (9.7% moisture) tending the burning process. The Philips HD4012, meeting the ISO (International Organization for Standardization) IWA (International Workshop Agreement) Tier 3 emissions ratings, was tested in triplicate at dry (8.7%) and wet (23.5%) fuel moisture levels. Cold start WBT results are presented for a more representative comparison, although limitations on these comparisons are discussed below. On average, the TSF and Philips operated in the field emitted 2−3 times more CO on a dry fuel mass basis than lab tests (Figure 5A). In fact, the Philips operated in the field has similar CO and PM dry fuel basis EF as TSF (Tier 1) tests in the lab. TSF PM dry fuel basis EF are well represented by the range of conditions tested in the lab, yet the Philips in the field emitted 2−4 times that seen in the lab (Figure 5A). The open fire tested in the lab by Roden and colleagues[20] emitted substantially less PM than we found while Wathore and co-workers measured similar in-field EF for the Philips. Although average MCE (Figure 5C) from TSF and Philips operated in the field were 2−4 and 2−3 percentage points lower than corresponding lab MCE, respectively, average TSF OTE operated in the field were about 2 times higher than lab values and comparable for the Philips. The reasons for the higher field TSF OTE are unknown; however, distinct testing protocols, pot types and fuel properties are contributing to this difference. For example, WBT cold starts expose water to much shorter periods of evaporation leading to less heat transferred in the form of latent heat. This is one major limitation of comparing OTE of UCTs and WBTs, namely not being able to account and control for varying ratios of sensible to latent heat. Contrary to suspicions that lab tests do not adequately cover the firepower range encountered in the field and therefore do not accurately reflect real-world performance metrics, there is good overlap of average lab and field firepower (Figure 5D). Without cook survey information, it is difficult to know preferences regarding firepower, but from anecdotal information, cooks prefer more firepower for faster cooking. Baldwin[58] advised incorporating "cooking process efficiency: so that as little energy as possible is used to cause the [physio-chemical] changes [occurring] in cooking food", into testing which in turn embodies "control efficiency" and "pot efficiency", the ability to modify firepower/cooking power and ability to transfer heat from the fuel and retain in the food, respectively. This begs the question, could focusing efforts to increase efficiencies over a range of larger turn-down ratios (maximum to minimum stove firepower output) and improving cooking process efficiency (e.g., pressure cookers[10]) in the field be more impactful than optimizing combustion of solid fuels? Yes, closing the seemingly small gap in MCE from the lab to the field can have significant impacts on overall emissions, yet based on the modeling results above and anecdotal evidence of the desire for more firepower from ICS, fuel properties and user behavior may be determining EF and efficiency more than stove design alone. That being said, average field CO and PM EF on an energy delivered basis map much closer to lab values (Figure 5B). This is a result of the relatively high efficiency these stoves attain in the field due in large part to the evaporation of water, a requisite for cooking most dishes. Four modified CCTs (version 2.0) preparing two cups of rice and three tests using modifications of the WBT (3.0), called the Emissions and Performance Test Protocol, were completed outdoors in Boulder (~10 °C) assessing the Gyapa and TSF burning wood (pine). The Emissions and Performance Test Protocol uses a foam lid to minimize evaporation and stipulates a 90 °C upper limit as opposed to boiling temp. This significantly reduces the amount of evaporated water and in turn reduces the total latent heat included as useable energy, partially explaining the lower efficiencies. MCE, OTE, firepower and cooking power from these tests are shown in Figure 5C and D to highlight the variability different testing protocols, environments and user operation can have on stove performance and resulting emissions. One possible implication of this is that OTE (or fuel use by a population) should be assessed over a range of meal types.

Differences between laboratory and field results have significant climate and health implications in regards to intended vs realized impacts of lab-tested stoves when certain performance indicators can be significantly different when operated by end-users in local context, as other have estimated.[26] Although these results represent a narrow slice of real-world performance for a small selection of stoves in a given setting, the cookstove research and health community along with atmospheric modelers can incorporate these findings into current and future health and atmospheric models. Likewise, international institutions, like many partners of the Global Alliance for Clean Cookstoves, can make more informed decisions choosing intervention technologies (e.g., Country Action Platforms) using real-world data collected in the field and have more realistic expectations of health and climate outcomes.

Emissions inventories depend on the quantification of emission factors from different stove and fuel combinations. This study highlights the variability in EFs measured in field cooking activities. The choice of performance metric (emission per fuel mass, emission per mass of food, emission per mass of energy delivered) and the statistical value of the EF (including mean and median values) could impact subsequent emission inventory development that use emission factors to drive emission estimates. For example, mean PM emission factors from the Philips burning wood were almost twice the median value. The results presented here provide emission factors that can be used to evaluate the sensitivity of inventories to these important input variations.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b02436.

> Uncertainty sources and estimates, stove pictures, EPOD schematic, stove/fuel performance summary and modeled comparisons results, sample characteristics and calibration information available (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: +1-303-735-8054; fax: +1-303-492-3498; e-mail: evan.coffey@colorado.edu.

ORCID

Evan R. Coffey: 0000-0001-5511-9111

Present Address

¶University of Colorado, CIRES, 216 UCB Boulder, CO 80309.

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Notes

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