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In-use emissions from biomass and LPG stoves measured during a large, multi-year cookstove intervention study in rural India

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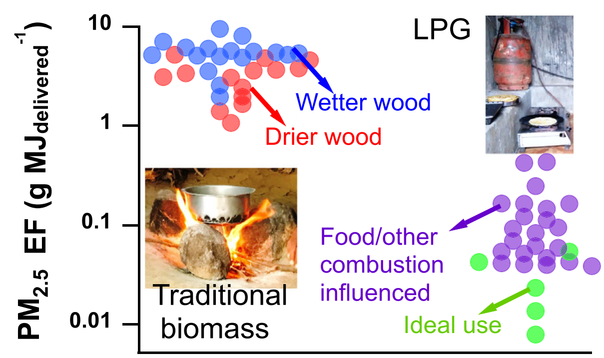
HIGHLIGHTS

- Measurements of real-world wood and LPG stove emissions of particulate and gaseous pollutants
- Quantification of emission factors and optical properties of emitted particles
- Fuelwood moisture and ambient relative humidity influence traditional stove emissions.
- In-home LPG emissions were substantially higher than those in lab.
- Food emissions can have large impact on observed LPG emissions.

GRAPHICAL ABSTRACT



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ABSTRACT

We conducted an emission measurement campaign as a part of a multiyear cookstove intervention trial in two rural locations in northern and southern India. 253 uncontrolled cooking tests measured emissions in control and intervention households during three ~3-month-long measurement periods in each location. We measured pollutants including fine particulate matter ($PM_{2.5}$), organic and elemental carbon (OC, EC), black carbon (BC) and carbon monoxide (CO) from stoves ranging from traditional solid fuel (TSF) to improved biomass stoves (rocket, gasifier) to liquefied petroleum gas (LPG) models. TSF stoves showed substantial variability in pollutant emission factors (EFs; g kg^{-1} wood) and optical properties across measurement periods. Multilinear regression modeling found that measurement period, fuel properties, relative humidity, and cooking duration are significant predictors of TSF EFs. A rocket stove showed moderate reductions relative to TSF. LPG stoves had the lowest pollutant EFs, with mean $PM_{2.5}$ and CO EFs (g $MJ^{-1}_{delivered}$) >90% lower than biomass stoves. However, in-home EFs of LPG were substantially higher than lab EFs, likely influenced by non-ideal combustion performance, emissions from food and possible influence from other combustion sources. In-home emission measurements may depict the actual exposure benefits associated with dissemination of LPG stoves in real world interventions.

1. Introduction

Globally over 3 billion people use solid fuel cookstoves as a source of household energy (World Health Organization, 2018). The resulting emissions lead to household air pollution (HAP) (Aung et al., 2016;

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Carter et al., 2016, 2017; Tagle et al., 2019) and ambient air pollution (Chowdhury et al., 2019; Conibear et al., 2018) associated with negative health impacts to those exposed. In 2017, 1.6 million deaths and 59 million disability adjusted life years (DALYs) were attributed to HAP exposure (State of Global Air, 2019). Cookstove emissions, comprised of long-lived green-house gases (e.g. CO_2, methane) and various short-lived species, also have myriad climate impacts (Grieshop et al., 2011; Huang et al., 2018; Kodros et al., 2015). Combustion of solid fuel emits black carbon (BC), a light absorbing component of aerosol that is estimated to have the 2nd largest global warming impact after CO_2 (Bond et al., 2013). Approximately 25% of annual BC worldwide was estimated to be emitted from residential solid fuel burning in 2012 (Bond et al., 2013). The other major component of solid fuel combustion aerosol is organic carbon (OC), which has light scattering properties and hence contributes to global cooling (Bond et al., 2004). However, brown carbon (BrC), a component of OC emitted from solid fuel combustion, absorbs light at shorter visible and UV wavelengths and may be moderately warming (Saleh et al., 2014).

Most impacts from cookstove emissions and HAP affect poorer residents of low and middle income countries. In 2017, 60% of India's population used solid fuels for cooking and 482,000 annual deaths were attributed to HAP (State of Global Air, 2019). India has a long history of national cookstove interventions (Ministry of Petroleum and Natural Gas, 2016; Singh et al., 2017; Venkataraman et al., 2010), from the National Program on Improved Chulha (NPIC; 1984 to 2002) to the National Biomass Cookstove Initiative (NBCI; 2009) to the most recent Pradhan Mantri Ujjwala Yojana (PMUY) program, launched in 2016 to reduce health risks among poor rural women and children by promoting access to liquefied petroleum gas (LPG) (Kar et al., 2019).

Considering the multiple impacts of solid fuel use, stove interventions can be evaluated from a number of different angles. Various studies have evaluated emissions in a laboratory setting (Arora et al., 2014; Jetter and Kariher, 2009; MacCarty et al., 2010) using a standard water boiling test (WBT) protocol ("The Water Boiling Test (WBT),", 2014). However, assessing the in-home performance of stoves is imperative as emissions may differ in laboratory versus in field conditions. For example, available evidence from multiple countries (Champion and Grieshop, 2019; Johnson et al., 2008, 2019; Mitchell et al., 2019; Shen et al., 2015; Weyant et al., 2019a) indicates that PM emissions are often greater (up to 2 to 5×) in-home than in-laboratory. Studies have focused on other aspects of interventions, including their effects on outdoor and indoor air quality (Aung et al., 2016; Chen et al., 2016; Chowdhury et al., 2013, 2019; Conibear et al., 2018; Kelp et al., 2018), exposure (Fitzgerald et al., 2012) and health (Clark et al., 2009; Das et al., 2018), and factors affecting stove adoption and long-term use (El Tayeb Muneer and Mukhtar Mohamed, 2003; Jan et al., 2017; Masera et al., 2005; Rhodes et al., 2014). Evidence from these studies suggests that intervention effectiveness depends on both stove adoption and in-home performance. Further, stove selection by households and long-term performance of both 'baseline' and 'intervention' technologies are both understudied. Therefore, a study integrating both the technical and socio-cultural aspects of intervention can provide valuable insights into this important problem (Simon et al., 2014).

Here we present results from a multi-year cookstove intervention trial encompassing socio-economic (stove adoption, fuel choice and use) and technical (emission, indoor air quality and exposure) aspects. During the intervention, study participants were given a choice of fuel-efficient biomass stoves as well as gas and electric options. Other analyses from this project report factors affecting stove adoption (Menghwani et al., 2019), LPG use (Kar et al., 2019), and biomass consumption (Singh et al., 2020). Here, we focus on emissions. The study aimed to measure in-home emissions from a variety of stoves including rocket-style biomass stoves, forced draft gasifiers, and LPG, with the aim of comparing variability in emissions between stove types while capturing seasonal and geographical variation. The data, collected over three measurement campaigns at each study site, represent the largest sample of in-home emission tests (N = 253) presented to date.

Household choice of a stove from a range of options was central to the overall study. During the intervention, the vast majority of study participants chose LPG over biomass stoves; as a result, our sample includes many measurements in LPG households. While not what we expected in designing the study, this represents a unique opportunity and a substantial contribution to the literature because previous studies of emissions from LPG stoves were conducted in lab conditions (Habib et al., 2008; MacCarty et al., 2010; Shen et al., 2017, 2018; Smith et al., 2000; Zhang et al., 2000; Zhang and Smith, 1999). Although a field study (Weyant et al., 2019b) conducted a small-scale LPG emission measurement campaign (six emission tests) in Nepal, to our knowledge, this is the first large-scale study of in-home emissions of LPG stoves.

This paper has six objectives: (a) present in-home measurements of pollutant emission factors from traditional and alternative biomass cookstoves; (b) evaluate in-home emissions from LPG stoves; (c) compare emissions across different stove types: traditional vs. alternative biomass stoves, chimney vs. non-chimney stoves, and alternate biomass stoves vs. LPG; (d) explore variability over time and between locations, (e) quantify which aspects of individual tests and households correlate with emissions, and (f) analyze the optical properties of emitted particles.

2. Materials and methods

2.1. Site description and study design

The study was conducted in Kullu district in Himachal Pradesh (HP) in northern India and Koppal district in Karnataka (KA) in southern India (study details in section S1 and locations in Fig. S1) (Menghwani et al., 2019). Rural households in Koppal initially cooked almost exclusively with traditional mud/clay stoves (chulhas). Kullu is located in the Himalayan foothills and many families also use a combined cooking and heating chimney stove called a Tandoor in winter months; there was also a much higher baseline prevalence of LPG stoves (~60%). Four communities in each district were included in the study, and 50 intervention and 10 control households were randomly selected in each community (480 households in total). Intervention households were allowed to choose from a wide range of alternate biomass and modern fuel stoves (Table S1). We varied stove pricing (free versus subsidized stove) and participants' ability to periodically exchange stoves at the community level (more details are found elsewhere (Menghwani et al., 2019)). In-home emission measurements were conducted before and after stove selection. Emissions were measured in ~10% of households during three measurement periods (baseline: BL, follow-up-1: F1, follow-up-2: F2) in both locations (six measurement campaign in total) (Table 1). The time between measurement campaigns allowed households to habituate to the new stoves (Hankey et al., 2015).

2.2. Emission testing approach

Emission measurements used the Stove Emission Measurement System (STEMS), a portable, battery-powered instrument package designed for unsupervised measurement of in-home cookstove emissions (Fig. S2). STEMS includes: an electrochemical carbon monoxide (CO) sensor, a nondispersive infrared carbon dioxide (CO_2) sensor, a 635 nm wavelength laser light scattering PM sensor, and a temperature/RH sensor. A LabJack data acquisition unit collects sensor outputs at 1 Hz. DAQFactory (Azeotech) data acquisition and control software are installed in a built-in PC. Two parallel 47 mm filter trains collect integrated PM samples: one contains a bare quartz fiber filter; the other contains a quartz filter behind a Teflon filter; the latter is used to correct for gas phase absorption artifacts (Subramanian et al., 2004). Quartz filters are used for thermo-optical organic and

Table 1
Summary of test numbers[a] for each stove type in each measurement period/location.

Location	Stove/fuel	Stove name	Baseline[b]	Follow-up 1[b]	Follow-up 2[b]	Total
Kullu	Biomass	Traditional	12 (11)	9 (8)	15 (15)	36 (34)
(HP)		Traditional Tandoor	20 (18)	6 (4)	10 (8)	36 (30)
		Himanshu Tandoor	5 (4)	6 (6)	8 (8)	19 (18)
		Prakti	–	4 (3)	–	4 (3)
		Teri	–	3 (3)	–	3 (3)
	LPG	LPG	5 (3)	6 (6)	8 (8)	19 (17)
Koppal	Biomass	Traditional	47 (45)	25 (22)	29 (24)	101 (91)
(KA)		Envirofit	–	6 (6)	–	6 (6)
	LPG	LPG	–	11 (11)	18 (15)	29 (26)
Study totals						**253 (228)**

[a] In each entry, the main number is the number of tests attempted; the number in parenthesis is the number of tests included in later analysis. Reasons for exclusion include data quality concerns or short duration (< 25 min for biomass stoves) of measurement. See Table S1 for information on stove options.
[b] Dates for HP: Baseline (March 2015 – May 2015), Follow up 1 (March 2016 – May 2016), Follow up 2 (May 2017 – June 2017). Dates for KA: Baseline (November 2015 – January 2016), Follow up 1 (September 2016 – November 2016), Follow up 2 (August 2017 – November 2017).

elemental carbon (OC/EC) analyses and Teflon filters used for gravimetric $PM_{2.5}$ analyses, described elsewhere (Wathore et al., 2017). The STEMS also incorporates an AE-51 MicroAeth (AethLabs) to measure real time PM light absorption at 880 nm wavelength. MicroAeth filter loading artifacts are corrected following Park et al. (2010) as described by Wathore et al. (2017). Table S2 summarizes STEMS configuration and changes during the study.

We applied the ‘plume probe’ method, using a six armed stainless steel probe to collect a representative sample of emissions from the plume 1–1.5 m above the cookstove (Champion and Grieshop, 2019; Roden et al., 2006; Wathore et al., 2017) (Fig. S2). In households with a chimney, the probe sampled immediately above the chimney's exit (Fig. S3b). We typically conducted two tests in households per day, during morning and evening cooking events. Testing included emission measurement during a cooking event (96 $\pm$ 58 min) with ~10 min of background measurement before and after the event. Note that emission measurements were during uncontrolled cooking activities (sometimes termed uncontrolled cooking tests) and often unsupervised to minimize disturbances to normal activities. For each test, we collected data about the stove, presence of a chimney, probe placement, and foods cooked during the session (Supplementary Spreadsheet in the SI).

We apply the carbon balance approach, as widely used in previous cookstove field studies (Coffey et al., 2017; Eilenberg et al., 2018; Roden et al., 2006; Wathore et al., 2017), to calculate fuel based emission factors (EFs). This assumes 50% of dry weight of biomass is carbon (82% for LPG), and that CO_2 and CO concentrations measured above background serve as a proxy for the fuel carbon. Other carbonaceous species (methane and non-methane hydrocarbons) make small contributions (< 5% of total carbon) to cookstove emissions (Johnson et al., 2008; Shen et al., 2013; Zhang et al., 1999, 2000) and are neglected in this analysis. Other metrics calculated include modified combustion efficiency ($MCE = \Delta CO_2/(\Delta CO_2 + \Delta CO)$; where Δ indicates a background corrected mixing ratio), and single scattering albedo ($SSA = b_{sp}/(b_{sp} + b_{ap})$, where b_{sp} and b_{ap} are particle scattering and absorption coefficients respectively). A higher MCE represents more efficient combustion and therefore lower formation of products of incomplete combustion; higher value of SSA indicates greater contribution of scattering to total light extinction by particles.

2.3. Household survey and kitchen performance tests

We conducted a household survey during each measurement period to collect information about the primary and secondary stoves in each household, location of the stoves (e.g. kitchen), dimension of the rooms with stoves including door and window information, types of food cooked, and household activity or other indoor sources (e.g. kerosene lamps) of emission. We conducted kitchen performance tests (KPT) in each measurement period in all emission test households. The KPT measures daily fuel consumption as well as moisture content (BD-2100, Delmhorst Instrument Co), size and species of wood, and number of people served (Summary Spreadsheet in the SI) (Singh et al., 2020). KPTs did not necessarily occur on the same day as the emissions test. Therefore, data is not available for each test day and we treat the KPT data as indicative of household fuel characteristics and consumption during that season.

2.4. Statistical analysis

Our data allows us to examine factors associated with variation in stove emissions. To do this, we apply non-parametric Wilcoxon rank sum test at 5% significance level to compare emission factors and properties between stove types, season and locations, and assess differences in fuel properties (e.g. fuel moisture, fuel use). We applied multilinear regression (MLR) to provide information on factors influencing EF variability and to quantify explainable variance in EFs; we implemented the MLR using IBM-SPSS software (“SPSS Software,” 2020). Covariates explored are measurement periods, fuel moisture, fuel consumption, RH, temperature, cooking duration and their interaction terms. Emission parameters were then added to the model to examine if variability in one pollutant is associated with other EFs (e.g. CO EF as a predictor for $PM_{2.5}$ EF model). Emission optical properties proxies (SSA and EC/TC) were also modeled using the same predictor variables (details on MLR approaches are in section S2).

3. Results and discussion

3.1. Emission test and household characteristics

We conducted 253 emission tests and exclude ~10% (N = 25) from the analyses because of instrument malfunction or minimal cooking duration (< 25 min for biomass stoves). Table 1 shows a summary of test numbers for each stove type, including number attempted (Table 1, main entry) and number analyzed (Table 1, in parentheses). Prakti, TERI, and Envirofit stoves were only measured in F1 because households initially selecting those stoves exchanged them for LPG in F2. We had a large number of tests (N = 125) for traditional solid fuel stoves (including three stone fires and simple mud/clay chulhas -TSF hereafter). We conducted 48 tests (~21% of total) on traditional chimney (Traditional tandoor, TT) and improved chimney (Himanshu tandoor, HT) stoves in HP. This study also offers a rich in-home emission measurement data set (N = 43, ~19% of total) for LPG stoves.

Household surveys implemented in parallel with each emissions testing campaign indicate that stove/fuel “stacking” was common (i.e., use of multiple fuel- or stove-types in a household) (Ruiz-Mercado and Masera, 2015). Primary stoves were mostly (72 and 89% in HP and KA, respectively) operated in the kitchen. However, some households used stoves in other locations (e.g., bedroom, veranda, attic, sitting room, outside). The volume of cooking spaces varied, and tended to be larger in KA (37 ± 16 m^3) than HP (25 ± 14 m^3). Most KA households (62%) had built-in chimneys over the hearth (Fig. S1-a). In addition, cooking practices differed between HP and KA. In HP, the most common foods included roti (Indian flat bread), vegetable, dal, rice and tea and during winter months, stoves were used for space heating. In KA, stoves were used to prepare rice, sambar (a spicy south Indian broth), tea/milk, roti, and vegetables and for water heating.

3.2. Emission factors of biomass stoves

Fig. 1a-e shows the distributions of pollutant EFs in HP. We observe substantial variation, but no consistent trend in $PM_{2.5}$ EF among

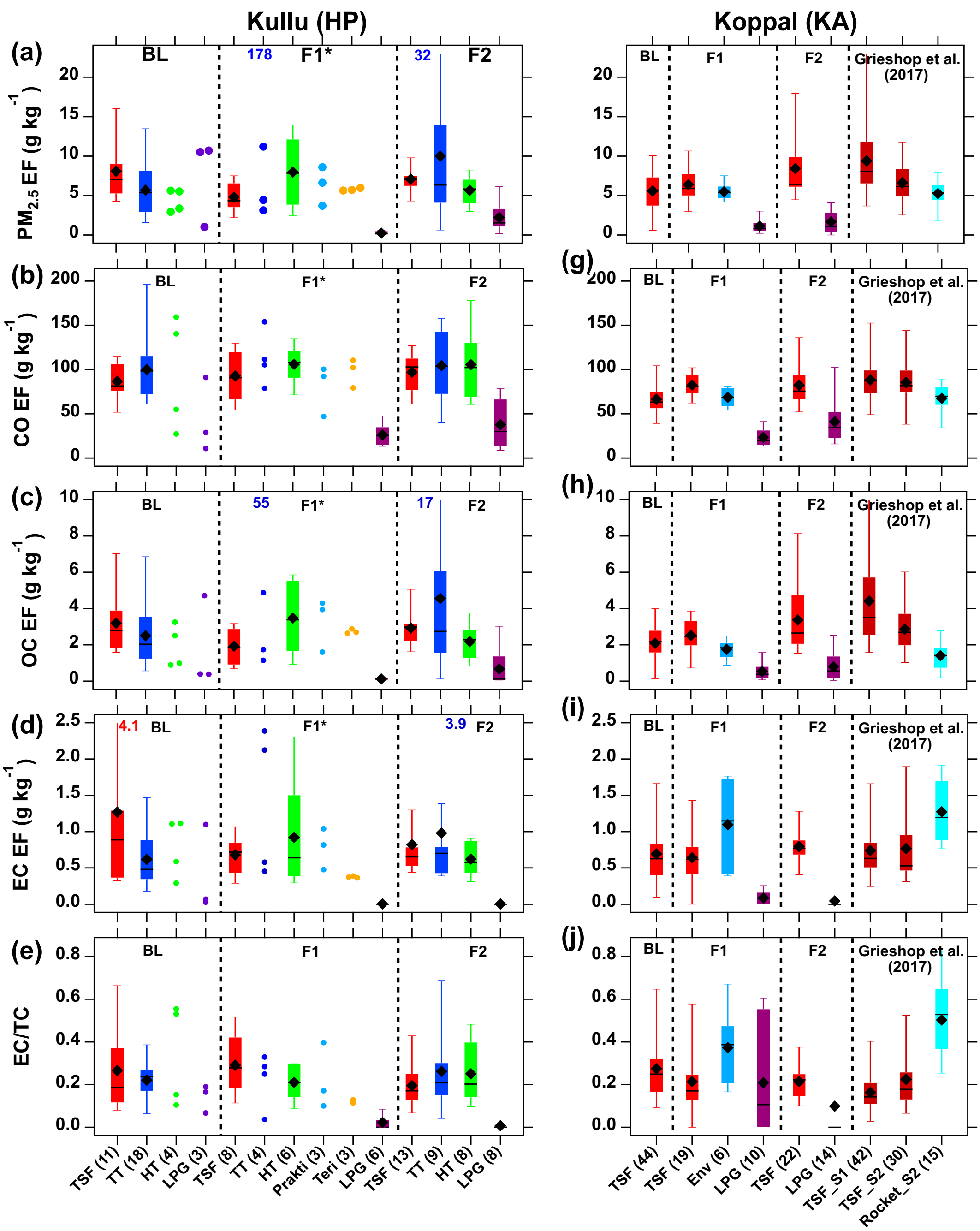

Fig. 1. Box and whisker plots of pollutant EFs and emission properties for the different stoves used in HP (a-e) and KA (f-j): $PM_{2.5}$ EF (a, f); CO EF (b, g); OC EF (c, h); EC EF (d, i) and EC/TC (e, j). Three sections in each panel represent three measurement periods: BL, F1 and F2 respectively (left to right). In KA plots, the rightmost section shows the results of a previous study conducted in the same district (Grieshop et al., 2017). Labels on category axes indicate stove types, and boxes are colored to differentiate them (red: TSF, green: TT, orange: HT, cyan: Prakti, orange: Teri, blue: Envirofit and violet: LPG). 'TSF_S1', 'TSF_S2' and 'Rocket_S2' labels in the 'Grieshop et al. (2017)' panel (in KA plots) indicates TSF stoves measured in season 1 and 2 and rocket stoves during season 2, respectively. Number of tests for each stove is in parentheses on the category axes. The boxes in this paper represent the interquartile range; horizontal line and diamond inside the box indicate median and mean, respectively. Whiskers extend to indicate extreme data points (≤1.5* inter-quartile range). The upper whiskers for some stove types are out of scale on the y-axis (outliers) and are thus shown with numbers. An asterisk after F1 (in HP plots) indicates greater uncertainty in EF values in that measurement period because of a CO sensor issue (discussed in section S3). We used points (circles) instead of box and whiskers for any groups with less than 5 tests. Note that results from all tests from both sites are listed in the Summary Spreadsheet in the SI.

biomass stoves across measurement periods (Fig. 1a). Among the biomass stoves, TSF stoves had the highest $PM_{2.5}$ EF of 8.1 ± 4.4 g kg^{-1} and 7.1 ± 1.4 g kg^{-1} (mean ± SD) during BL and F2 respectively. OC EF tracked $PM_{2.5}$ closely (Fig. 1a and c), consistent with OC-dominated PM (OC/PM: 0.4 ± 0.1). CO EF showed slightly less stove-to-stove variability across measurement periods (Fig. 1b). Note that mean relative uncertainties, estimated via error propagation from measurement uncertainties (Section S3), in $PM_{2.5}$, OC and CO EFs for biomass (LPG) stove tests are 24% (29%), 29 (35%) and 36% (37%) respectively, smaller than the variability in EFs within groups (e.g. coefficients of variation of $PM_{2.5}$ EF are 41% and 128% for TSF and LPG stoves, respectively).

We tested traditional and improved tandoors in HP. Mean $PM_{2.5}$, CO and OC EFs of TT were within 4–17% of traditional plancha (chimney) stoves of a recent field study (Eilenberg et al., 2018). EFs of HT were not significantly different from TT, possibly due to lack of difference in combustion performance or the influence of different operation. HT tests showed higher variability in $PM_{2.5,}$ OC and EC EF (Fig. 1a, c, d) during F1 than BL and F2 ($PM_{2.5}$ EF range: 2.7, 11.5 and 5.3 g kg^{-1} respectively at BL, F1 and F2). Furthermore, mean $PM_{2.5}$ EF of HT was 46% and 20% lower than that of TSF at BL and F2 respectively ($p \le 0.05$), though the trend was opposite (66% higher) in F1 ($p = 0.07$). OC shows a similar trend to $PM_{2.5}$ EF but other pollutant EFs (CO, EC) for these two stoves were not significantly different.

Tests included three of a forced-draft stove (TERI). EFs and other emission properties had minimal variability (COVs ranged between 0.03 and 0.17), which is surprising as even previous lab tests have sometimes showed much higher variability (COV > 1.0) (Jetter et al., 2012; Wang et al., 2014). However, the small sample size of TERI stove tests (N = 3) means these results need to be interpreted with caution. The $PM_{2.5}$ EFs of TERI and Prakti (5.8 ± 0.2 and 6.3 ± 2.5 g kg^{-1}) were comparable to those of rocket and gasifier stoves in a previous field study (Wathore et al., 2017) and not significantly lower than the TSFs, consistent with previous studies (Coffey et al., 2017; Johnson et al., 2011; Rose Eilenberg et al., 2018).

Fig. 1f-j show distributions of EFs from KA tests. $PM_{2.5}$ and CO EFs of Envirofit were 5.3 ± 1.1 g kg^{-1} and 68.5 ± 10.5 g kg^{-1}, respectively, comparable to a different rocket stove in Grieshop et al. (2017). Envirofit mean CO and OC EFs were 17% and 31% less, respectively, than those for TSF ($p < 0.05$) during F1, also similar to the Grieshop et al. study (Grieshop et al., 2017). However, pollutant EFs of TSF stoves in these two studies were significantly different in some measurement periods. For example, $PM_{2.5}$ EFs at baseline measured in the previous study (Grieshop et al., 2017) were 68% and 47% higher than our BL and F1 measurements, respectively. Similarly, CO EFs at baseline and follow-up of the previous study were 33% and 29% higher respectively than BL here. Note that TSF EFs varied significantly across measurement periods in this study (discussed in Section 3.5).

3.3. In-home emission of LPG stoves

LPG stoves had the lowest EFs for all pollutants across the study. Mean $PM_{2.5}$ EF (1.8 ± 2.4 g kg^{-1}) and CO EF (34.3 ± 23.1 g kg^{-1}) of all LPG tests were 76% and 60% lower respectively than biomass stove tests (Fig. 1). In addition, LPG has a higher calorific value and the LPG stoves have better thermal efficiency than most biomass stoves. EFs on a 'delivered-energy' basis, calculated using calorific values and stove efficiencies from other studies (Table S3), show much greater differences between LPG and biomass stoves. For LPG, estimated mean emissions of $PM_{2.5}$ (CO) per energy delivered are 0.07 (1.38) g $MJ^{-1}_{delivered}$ respectively, 94–98% (90–97%) lower than for biomass stoves (range depends on assumed thermal efficiencies).

Fig. 2a-b show $PM_{2.5}$ and CO EFs for all LPG tests. Notably, in-home LPG EFs have variability similar to biomass stoves, with higher variability in F2 than F1. Mean CO and $PM_{2.5}$ EFs were also higher in F2 than F1, and the EF distributions were significantly different in some cases ($p = 0.001$ for $PM_{2.5}$ EF in HP and $p = 0.02$ for CO EF in KA). Some LPG tests measured extremely high CO and $PM_{2.5}$ EFs (Fig. 2). Fig. S5a plots CO EF against $PM_{2.5}$ EF, and shows that tests with high $PM_{2.5}$ EF are generally associated with low CO:$PM_{2.5}$ ratio. One possible explanation for this behavior is that other sources (apart from fuel combustion) may have contributed to $PM_{2.5}$ measured above the stoves, since CO is co-emitted with PM during fuel combustion. Fig. S5a also shows the range of CO:$PM_{2.5}$ ratio observed during lab tests in which PM and CO are solely from fuel combustion (Shen et al., 2018). This range hypothetically marks the 'fuel combustion emission zone'. Many high $PM_{2.5}$ emitting tests are outside this zone, implying the possible contribution of other sources (e.g. food emissions). Due to the multi-faceted study design, emission measurements were often unsupervised, making it difficult to conclude what exactly occurred during the testing period. However, household surveys revealed that incense or lamp burning, and simultaneous use of other biomass stoves (shown by orange and red circles respectively in Fig. 2a-b) possibly affected some measurements. Since frying food is a common source of PM (Buonanno et al., 2009; Torkmahalleh et al., 2012, 2017; Weyant et al., 2019b), we divided the LPG tests (excluding red and orange points in Fig. 2a-b) into tests with and without oil used during cooking to explore the effect of food types on LPG EFs. EF distributions of 'oil' events have higher variability than those without oil (Figs. 2b-c, S5c-d). Mean EFs of $PM_{2.5}$, CO and OC of were also higher, indicating that frying food might be a factor contributing to mean emissions and variability. Excluding LPG tests impacted by biomass stoves, lamp/incense and frying, mean

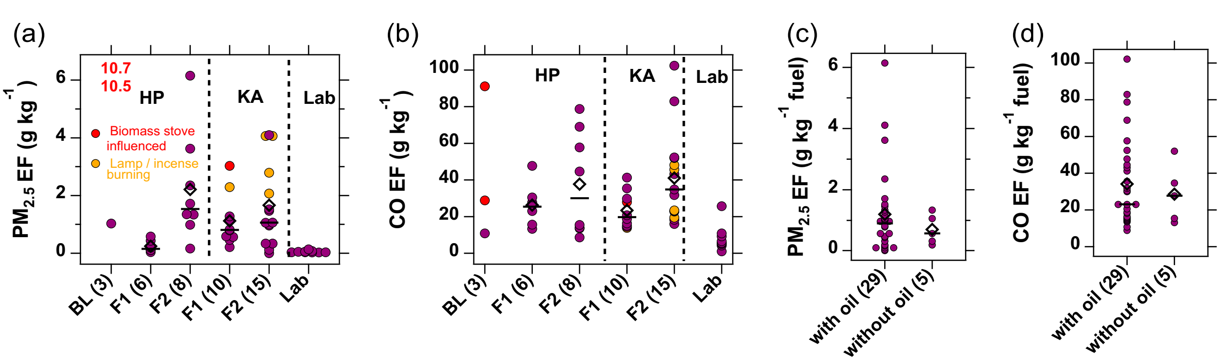

Fig. 2. Jitter plots showing $PM_{2.5}$ EF (a) and CO EF (b) from all LPG tests. The panel sections represent measurement locations: Kullu (HP), Karnataka (KA) and lab respectively from left to right. The lab results are adapted from Shen et al. (2018) Label and number on category axis indicate measurement periods (BL, F1 and F2) and number of LPG tests in each period respectively. Black diamond and horizontal bar indicate mean and median of each distribution. Red and orange circles represent the LPG tests possibly influenced by 'other biomass stoves' and 'lamp/incense', respectively. (c) and (d) are the jitter plots of $PM_{2.5}$ EF and CO EF respectively of LPG tests excluding the red and orange points in (a) and (b), showing the distributions of EFs when food was cooked with (left distribution) and without oil (right).

$PM_{2.5}$ and CO EF are 0.7 ± 0.5 g kg^{-1} and 28.6 ± 15.7 g kg^{-1} respectively, which are comparable to an earlier lab study (Smith et al., 2000). However, these mean $PM_{2.5}$ (CO) EFs are ~14 (1.5) times higher than those measured in a recent lab study (Shen et al., 2018) that included badly worn-out, poorly operated LPG stoves (right-hand sections of Fig. 2a-b), suggesting these lab results may underestimate 'real-world' emissions from LPG use.

Though we cannot completely exclude the influence of sources other than the primary fuel combustion in some of these measurements, all sources are relevant in terms of their influence on HAP exposures. One way to examine this is to view emission rates relative to International Organization for Standardization (ISO) voluntary performance targets (Tiers) (International Organization for Standardization (ISO), 2018), established for controlled lab test results based on WHO targets for HAP levels (which are not only affected by combustion emissions) (World Health Organization (WHO), 2014). Viewed in this light, only 12% of all LPG tests carried out meet ISO Tier-5 criteria for $PM_{2.5}$ and CO (Fig. S5b). Most of the tests (57%) would classify as Tier-4 while three LPG tests fall in Tier-2; some of the Tier-2 and 3 zones are the impacted tests (biomass stove, lamp) as shown in Fig. S5b. However, it must be noted that gravimetric filter masses for all Tier-5 and 80% of Tier-4 tests were below our limit of detection (SI Sec. S3 and Fig. S6a), highlighting the difficulty of measuring these very clean sources under field conditions. Note that ISO Tiers are not intended for field test results and presented here for comparison purposes only. Our results demonstrate that while in-home emissions from ideal operation of LPG stoves approach those observed in lab testing, even exclusive use of a 'clean fuel' does not guarantee emissions will meet stringent guidelines aiming to drastically cut HAP exposures.

3.4. Optical properties

We observed moderate correlations ($r^2 = 0.31$) between test average b_{sp} and gravimetric $PM_{2.5}$ concentrations (Fig. S7a). Correlation varied across stove technologies (Table S4) with TSF stoves showing the lowest correlation ($r^2 \sim 0.34$). Chimney stoves (TT, HT) showed better correlation than non-chimney stoves (Table S4). Fig. S8 shows distributions of mass scattering cross-section (MSC - the ratio of scattering coefficient (b_{sp}) to gravimetric $PM_{2.5}$ concentration) categorized by stove type in both locations. MSC of TT was significantly higher than TSF in BL and F2. Like $PM_{2.5}$ EF, MSC from TSF stoves showed inter-period variability. These observations suggest that stove technologies and seasonal/environmental conditions should be considered before using light scattering measurement as a direct proxy for PM concentrations.

Figs. 1(e, j) and S8 show distributions of EC/TC and SSA across different seasons, locations and stove types. In KA, Envirofit stove emissions had 31% lower SSA ($p = 0.01$) and 76% higher EC/TC ($p = 0.02$) than those from TSF stoves in F1. In HP, HT SSA was 39% and 54% lower than TT during BL and F2, respectively ($p = 0.05$–0.07). This is notable considering that no significant difference in EFs was found between these two stoves types. We observed inter-site and inter-period variability in TSF SSA values (Fig. S9). In HP, SSA was significantly lower in F1 than BL and F2 ($p < 0.005$), a trend similar to $PM_{2.5}$ EF (Fig. 1a). No significant difference in EC EF between the seasons and similar trends in OC and $PM_{2.5}$ EF likely explain the similarity between $PM_{2.5}$ EF and SSA trends, with SSA moderated by (mostly scattering) OC contributions. In KA, traditional stove SSA during F1 was significantly higher than that during F2, opposite to what we saw for $PM_{2.5}$ EF (Fig. 1f), likely driven by increased EC EF during F2 relative to F1. Across the six measurement periods, F1 in HP had significantly lower SSA than the others. In contrast, mean EC/TC of TSF was higher in F1 in HP relative to other periods, although the differences between measurement periods were not always statistically significant. A scatter plot of SSA versus EC/TC (Fig. S11a) shows a negative correlation; however, correlations were not good for all stove types. Correlation was reasonably strong ($r^2 = 0.54$) for TT stoves. Correlation between SSA and EC/TC was in a similar range ($r^2 = 0.47$ to 0.57) to that observed cookstove studies conducted in India and Malawi (Grieshop et al., 2017; Wathore et al., 2017), but different than observed for open biomass burning of various fuels (Pokhrel et al., 2016). No association was found between SSA and MCE (Fig. S11b), a finding similar to another field study (Grieshop et al., 2017).

3.5. Sources of variability in TSF stove emissions

Here we explore sources of variability across our large set of TSF emission tests. In KA, TSF stoves were the only biomass stoves to display strong inter-period variability in emissions. Fig. S12a shows significant ($p < 0.05$) increases in mean $PM_{2.5}$ EFs from BL to both F1 and F2. Likewise, EC, OC and CO EFs increased significantly ($p < 0.05$) (Fig. 1). In contrast, EC/TC decreased from BL to F1 and F2 ($p = 0.03$ and 0.06 for F1 and F2 respectively). We conducted F1 and F2 measurements during a similar time of year and observed smaller differences between F1 and F2 than between BL and either follow-up. Analysis of household-level paired tests shows differences between measurement periods in TSF EFs in individual households is consistent with the change in group EFs. For example, paired $PM_{2.5}$ EFs of TSF stoves between BL and F2, and F1 and F2 indicate higher EFs during F2 than during BL and F1 suggesting inter-period variability is not simply due to inter-household variability (detail in Section S5 and Fig. S13). We also observed inter-period variability in TSF emissions in HP (Fig. S12a). $PM_{2.5}$ EF in F1 was ~30% lower than both BL and F2 ($p = 0.008$). OC also shows a similar trend. Notably, BL and F1 were both conducted between March and May. In contrast to KA, CO and EC from TSF stoves did not show inter-period variability in HP.

To examine inter-site variability in TSF stove emissions, we compared data pooled by site. Fig. S14a and S14b show the $PM_{2.5}$ and CO EFs of TSF stoves from this study with those from two other locations in India from separate studies (Eilenberg et al., 2018; Fleming et al., 2018). No significant difference was observed in the pooled $PM_{2.5}$ EF between our two study sites. However, $PM_{2.5}$ EF (9.1 ± 4.7 g kg^{-1}) measured in the Haryana study (Fleming et al., 2018) was significantly higher than those in other three locations. For example, mean $PM_{2.5}$ EFs in Haryana was 33, 39 and 78% higher than those in HP, KA and Chennai (Eilenberg et al., 2018) respectively ($p = 0.009$, 0.02 and 0.03). CO EF varied across all sites. For example, mean CO EFs at HP were 24% higher ($p = 0.0001$) than KA. Further, both HP and KA had 46 and 17% higher CO EF than Haryana. OC and EC EFs did not exhibit inter-location variability.

To understand factors associated with variability in TSF EFs, we examine EF data in the context of information on fuel properties (moisture content and species), cooking activity (duration and fuel use) and ambient conditions (relative humidity and temperature). Fuel moisture content (MC) has been shown to affect stove EFs in previous studies (Coffey et al., 2017; Grieshop et al., 2017; Van Zyl et al., 2019a). When we stratify TSF $PM_{2.5}$ EFs in KA into two MC (dry basis) categories based on categories used by van Zyl et al. (2019b): low (≤15%) and high (>15%), the higher MC group has a 35% higher mean $PM_{2.5}$ EF ($p = 0.004$) (Fig. S15a). The observed similar variations across measurement periods and locations of $PM_{2.5}$ EF and MC distributions (Fig. S12a, c) also suggests an association between MC and EFs.

Wood species can also influence EFs (Chen et al., 2007; Iinuma et al., 2007; McMeeking et al., 2009). Wood species data collected during KPTs allowed us to identify three fuelwood species in BL and F1 (not used in F2) at KA that were associated with high $PM_{2.5}$ and CO EFs. Local names of these species: Kalli (*Euphorbia tirucalli*), Kakki (*Cassis fistula*), and Kari jaali (*Acacia nilotica*) start with K and we abbreviate them as 'K-species' hereafter. Mean $PM_{2.5}$ (Fig. S15b) and CO EFs were 49% and 24% higher respectively ($p < 0.005$) for tests with K-species relative to those without, suggesting fuel species may contribute to variability in emissions. Mean fuelwood MC of the group with K-species was also 34% higher than in other tests, suggesting that MC alone or as influenced by fuelwood species may drive the difference between the two groups. We include both as covariates in multilinear regression (MLR) modeling, discussed in Sections 3.6 and S2.

Tests were also stratified by duration of cooking events (≤2.5 h and >2.5 h) and fuel use rates (≤4 kg/day and >4 kg/day) to examine links between fuel consumption, cooking duration and EFs. We only found significant differences for EC EFs between the fuel use and cooking duration groups. Mean EC EFs were 37% and 36% higher ($p < 0.005$) for the lower fuel use and shorter cooking duration group, respectively (Fig. S15c, d). Fuel use was also positively correlated ($r = 0.42$) with cooking duration (Fig. S16) implying that higher daily fuel use was associated with longer cooking event that may have contributed to longer smoldering times during cooking, possibly leading to lower overall EC EFs.

Fig. S15e shows a scatter plot of $PM_{2.5}$ EF and test-averaged relative humidity indicating a weak, positive correlation ($r^2 = 0.18$) and suggesting that ambient RH influenced EF. We also observed consistent trends in RH and $PM_{2.5}$ EFs of TSF across measurement periods and locations (Fig. S12a, b). The positive association between RH and $PM_{2.5}$ EF is not surprising considering the fact that fuelwood MC is a function of ambient RH (Glass and Zelinka, 2010; Van Zyl et al., 2019a) and we observed a positive association between MC and $PM_{2.5}$ EF above. However, no correlation was found between fuelwood MC and test-averaged RH. One possible reason is the absence of MC data for each test. Moreover, RH varies on an hourly or daily basis whereas MC content changes gradually and depends on cutting method and storage conditions.

3.6. Multilinear regression analysis

The univariate analyses discussed above show the effect of individual factors on the variability in pollutant EFs. Here, we apply multilinear regression to control for covariance, identify significant factors and quantify their relative contribution to explainable variance in EFs. Table 2 summarizes MLR results and shows significant predictors in each model (details in Tables S5 to S7).

MC and RH appeared as significant predictors in the $PM_{2.5}$ EF model, explaining 23% of total variability (Tables 2, S5 and S7). The positive coefficients are consistent with the univariate analysis where MC and RH were positively associated with $PM_{2.5}$ EF. Adding emission-associated parameters (e.g. CO EF and SSA) as covariates to the $PM_{2.5}$ model increased predictive power of the model substantially ($R^2 = 58\%$). We find $PM_{2.5}$ and CO EFs were positively associated, which was also

Table 2
Summary of stepwise (backward elimination[a]) 'multilinear regression analysis showing (in shaded cells) signficant predictors with standardized (by standard deviation) coefficients of each model with % variability explained in terms of adjusted R^2 (BL-HP is the reference).

Covariates	Dependent variables: Emission parameters					
	Emission factors				Optical properties	
	Log ($PM_{2.5}$ EF)	Log (CO EF)	Log (OC EF)	Log (EC EF)	Log (SSA)	Log (EC/TC)
	Standardized coefficients					
	Without measured emission parameters					
Adjusted R^2	23	16	19	6	56	14
Measurement periods		BL-KA (-0.41)			F1-HP (-0.72), F2-KA (-0.39)	F2-KA (0.34)
MC	0.3		0.26	0.26		
RH	0.32		0.31		0.44	-0.74
Temperature					0.66	-0.51
Fuel use rate					0.6	-0.31
Cooking duration				*	*	
	With measured emission parameters					
Adjusted R^2	58	26	53	8	71	44
Measurement periods		BL-KA (-0.31)			F1-HP (-0.44)	F1-HP (-0.34)
MC					0.29	
RH						
Temperature						
Fuel use rate						
Cooking duration					*	
Log ($PM_{2.5}$ EF)	NA	0.41	NA	NA	NA	NA
Log (CO EF)	0.32	NA	0.46			
Log (OC EF)	NA	NA	NA	NA	NA	NA
Log (EC EF)	NA	NA	NA	NA	NA	NA
Log (SSA)	0.64		0.5	-0.31	NA	-0.84
Log (EC/TC)	NA	NA	NA	NA	-0.51	NA

*Cooking duration appeared as a significant factor in forward selection and bidirectional stepwise models of these emission parameters (details in Table S7 and the supplementary spreadsheet in the SI).

[a] Outcome of other stepwise approaches (forward selection and bidirectional/hybrid) and the traditional approach (model having all covariates included) are shown in Table S7 and the supplementary spreadsheet in the SI. Stepwise approaches (forward, backward and bi-directional) show better performance (in terms of adjusted R^2 and Akaike information criterion-AIC) than the traditional regression, as expected. Stepwise approaches show the same results (significant predictors, adjusted R^2 and AIC) in eight out of twelve models. For the remaining four, two models have better performance (higher adjusted R2 and lower AIC) for 'backward selection' and two for 'forward selection' and 'bidirectional' stepwise approaches (details in Table S7 and the supplementary spreadsheet in the SI).

observed in previous studies in India (Grieshop et al., 2017) and Honduras (Roden et al., 2009) but was not consistently observed in indoor air quality studies (Carter et al., 2017). Changes in SSA had higher, than CO EF, influence on $PM_{2.5}$ EF (Table 2). However, the fact that 42% of $PM_{2.5}$ EF variability remained unexplained indicates that factors beyond those included here contributed to EF variability.

For the CO EF model, measurement season (BL-KA) was the only significant predictor, explaining 16% variability (Table 2). The negative coefficient for BL-KA (relative to BL-HP) is consistent with the univariate analysis showing mean CO EF in BL-KA was 23% lower than during BL-HP (Fig. 1b and g). $PM_{2.5}$ EF was a significant covariate when emission parameters were added to CO EF model, as expected. However, the increase in predictive power of the CO EF model with emission parameters is lower than in the $PM_{2.5}$ EF model. The OC EF model is consistent with that for $PM_{2.5}$ EF with and without the emission parameters. This is expected, because OC was the main component of $PM_{2.5}$. For EC, cooking duration and MC were significant covariates. The negative relationship between MC and EC EF is similar to that found in previous analyses in both lab (Van Zyl et al., 2019a) and field conditions (Coffey et al., 2017; Grieshop et al., 2017). The negative association with cooking duration is consistent with the univariate finding (Fig. S15c). Significant factors for the EC model changed from MC and cooking duration to SSA when we added emission parameters (CO EF and SSA); however, model performance remained the same.

Models for SSA explained the most variation ($R^2 = 71$ and 56% for models with and without emission parameters, respectively). The model without emission parameters showed that F1-HP was associated with the largest variation in SSA, followed by temperature, fuel use, RH and F2-KA (Table 2). EC/TC became a significant factor when emission parameters were included. The negative association between EC/TC and SSA was discussed in Section 3.4. The EC/TC model showed a poor fit with covariates. The significant covariates were similar to the SSA model but inversely associated, due to the negative association between EC/TC and SSA. When other emission parameters were included, the model fit improved, with season (F1-HP) and SSA as significant factors.

4. Implications

We find that EFs from a range of improved biomass stoves do not show systematic or unambiguous reductions relative to TSF, although we do observe improvements in some pollutants or during certain seasons. The initial goal of this study, to evaluate in-home emission performance of various alternative biomass and a non-biomass stoves (LPG), was challenged by the fact that so few households showed any interest in the biomass options. This by itself offers an important lesson, as emissions performance of an unused stove is not relevant. Our measurements of LPG stoves reveal several interesting facts. Although mean $PM_{2.5}$ and CO EFs (g $MJ_{delivered}^{-1}$) of LPG were over 90% lower than biomass stoves, some LPG tests had extremely high EFs compared to lab studies. Our test results suggest that food emissions or contributions from other activities lead to high 'effective' EFs in many cases and thus to net impacts on HAP that are much higher than those expected based on combustion emissions alone. In other cases, we could identify no such outside influence; we can only speculate that this could be due to LPG stove degradation or variable fuel quality. Hence an important take-away is that even a clean stove is not a silver bullet for HAP in the presence of potential emissions from the food itself or from other cooking and non-cooking sources (Piedrahita et al., 2020). Subsequent publications on our HAP measurements from this study will further explore this outcome and assess the effectiveness of the intervention by exploring the within-household variation in indoor air quality due to switching to LPG/alternative biomass stoves.

Observed inter-period variability in emissions from 'baseline' TSF stoves reinforces the need for baseline measurements to assess the effectiveness of interventions. Unlike KA, CO and EC EF did not show inter-period variability in HP. Interestingly, an intervention study conducted in Karnataka (Grieshop et al., 2017) several years ago found no inter-period variability in CO and EC EFs, although CO and $PM_{2.5}$ EF were correlated in that study as in this study. This suggests that PM and CO formation mechanisms may differ by time and locations even for the same stove type. In general, we found no evidence of consistent differences in emissions between our sites at nearly the opposite ends of India. However, comparison with other studies (Eilenberg et al., 2018; Fleming et al., 2018) conducted in two different locations in India indicates that TSF EFs exhibit substantial inter-site variability. These findings caution against using single EF values for a stove type across all locations, and suggest reporting the geographical location of measurements along with EFs while developing emission inventories.

The univariate and multivariate analyses of TSF emissions identified factors driving emission variability, with multiple implications. Factors in addition to stove type influence emission performance, indicating that these factors should be considered while comparing performance between different stove types. Further, these suggest real world measures that can be implemented to reduce emissions. For example, higher fuelwood MC and RH are associated with increased HAP emissions, thus highlighting the potential importance of fuel storage practices. Altogether, fuel properties, cooking practices, and ambient conditions explain ~50% of the variability in emissions performance in our tests. This implies that there are other factors driving variability (Bilsback et al., 2018); future research may explore these, or this may be the result of irreducible complexity in poorly-controlled combustion. These findings are derived from measurements of TSF stoves, and should not be directly applied to other stove types.

Our large data set also enables us to comment on the sample size required for evaluating the performance of stoves since it can be impractically large in real world conditions (Thompson et al., 2019). For our TSF data set, we find that quantifying CO EF requires the least number of tests; 3 tests achieved mean values within the 95% confidence interval of our full-group mean (Fig. S17). In contrast, EFs of $PM_{2.5}$ and its components are highly variable, requiring roughly 30 tests to achieve the same level of confidence, for the same stove type (TSF), measurement period (BL) and location (KA), comparable to the finding of others (Thompson et al., 2019). Therefore, this larger data set can also help inform the design of future interventions; the number of tests is crucial determinant of the technical complexity and costs associated with field testing of both baseline and alternative cooking technologies.

CRediT authorship contribution statement

Mohammad Maksimul Islam: Formal analysis, Software, Data curation, Writing - original draft, Writing - review & editing. **Roshan Wathore:** Data curation, Investigation, Writing - review & editing. **Hisham Zerriffi:** Conceptualization, Funding acquisition, Writing - review & editing. **Julian D. Marshall:** Conceptualization, Funding acquisition, Writing - review & editing. **Rob Bailis:** Conceptualization, Funding acquisition, Writing - review & editing, Project administration. **Andrew P. Grieshop:** Conceptualization, Supervision, Methodology, Writing - review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Additional details on study design, the multilinear regression analysis, measurement limit of detection and uncertainty analysis and a CO sensor issue, household paired analysis, mass absorption coefficient (MAC) comparisons, the sample size required to evaluate performances of stoves. SI figures show schematic of test sites and stoves, STEMS configurations in the field, LPG emission factors, $PM_{2.5}$ filter limit of detection comparisons, inter-site variability in TSF EFs, factors driving variability in TSF EFs, correlation between cooking duration and fuel use rate, distributions of MAC, MSC and SSA, scatter plots between SSA and EC/TC, SSA and MCE, absorption EF and EC EF, and B_{sp} and gravimetric $PM_{2.5}$ concentration, running average of EFs for increasing test numbers. SI tables report a brief description of the stoves involved and the sensors/instruments and associated measurement uncertainties, assumed stove parameters for energy-based-EF determination, correlations between gravimetric and optical comparisons of PM, additional details on outcomes of MLR analysis.

A Supplementary Spreadsheet lists information on all individual tests including EFs and associated uncertainty, stove used, foods cooked, time of measurement and presence of chimney. KPT results, details of MLR analysis and significance testing between groups are also tabulated. Supplementary data to this article can be found online at doi: https://doi.org/10.1016/j.scitotenv.2020.143698.

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