ABSTRACT

WATHORE, ROSHAN. In-use Characterization of Emissions and Fuel-use of Traditional, Natural- and Forced-Draft Cookstoves in Rural Malawi (Under the direction of Dr. Andrew Grieshop).

Emissions from traditional cooking practices in developing countries are a major concern due to detrimental health and climate effects. Adoption of alternative solid biomass cookstove technologies is seen as a potential solution to provide health and climate "co-benefits". To assess the performance of technologies currently being disseminated, in-home emission measurements of a range of traditional and alternative cookstoves were completed in Malawi, where more than 90% of the population is dependent on wood for cooking purposes. In this work we measured emissions and fuel use associated with in-home use of alternative stove technologies ranging from locally manufactured low-cost ceramic models to two state-of-the-art fan-driven gasifiers (FDCS; Philips and ACE-1), large institutional stoves and traditional cooking arrangements. Real-time measurements included CO_2, CO, and aerosol light absorption and scattering; integrated samples were collected to quantify fine particulate matter ($PM_{2.5}$) and elemental and organic carbon (EC/OC). Fuel based emission factors (EF) in grams of pollutant per kilogram of wood were derived; traditional stoves had the highest mean $PM_{2.5}$ EF of 7.8 ± 2.9 g kg^{-1} with intervention stoves leading to reductions ranging from 13 % for the locally manufactured stove to 47 % for the Philips. Despite the Philips showing a statistically significant reduction in the field, emissions were much greater than laboratory results, likely due to non-ideal operation of these stoves. Field based emissions factors were similar to those measured in other field studies and exhibited high variability. Real-time optical properties and EC measurements show that particles from all 'alternative' stoves are more absorbing than those from traditional stoves, indicating greater specific warming. EFs and fuel use measurements were combined to estimate average emission rates (ER); the intervention stoves had reduced emission rates for most species, but at best only met intermediate targets to mitigate intake and climate impacts. Estimated global warming commitments (equivalent CO_2 including short-and long-lived species) were evaluated, with EC exhibiting the highest non-CO_2 contribution to short-term warming. Estimates of exposure associated with measured performance of FDCS in the field is similar in range as improved wood burning cookstoves in the laboratory and two orders of magnitude higher than LPG stoves. Visual observation of operation of FDCS indicated that over-fueling of stoves likely contributed to high pollutant emission rates, suggesting that fuel homogenization (e.g. pelletizing) may be required for more consistent performance. These results are further evidence that current laboratory emission protocols do not represent real-world performance. Further, they indicate that even the most advanced current solid biomass stoves may not provide the health and climate benefits indicated by standard lab testing protocols; a vastly improved and robust stove/fuel system is required to consistently reach this potential.

In-use Characterization of Emissions and Fuel-use of Traditional, Natural- and Forced-Draft Cookstoves in Rural Malawi

by
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DEDICATION

To Dr. Tami Bond. Genius

BIOGRAPHY

Roshan Wathore has done his elementary schooling in Saudi Arabia and spent his middle and high school years in his hometown of Nagpur, India. He has a Bachelor of Technology degree in Chemical Engineering from Indian Institute of Technology Madras and a Master of Technology degree in Environmental Engineering and Management from Indian Institute of Technology Kanpur. Upon graduation, he started graduate school at NC State under the advising of Dr. Andrew Grieshop. He is thankful for the opportunity and learning experience he has had at NC State. After graduation, he looks forward to getting a career or whatever else life will bring him

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CHAPTER 1 BACKGROUND AND MOTIVATION

Nearly half of the world's population is dependent on solid fuels such as biomass, coal or dung for cooking purposes (Legros et al. 2009; Sumpter and Chandramohan 2013). Traditional methods of cooking leads to inefficient combustion of these fuels leads to emissions of air pollutants include greenhouse gases such as carbon-dioxide (CO_2) and methane (CH_4), and products of incomplete combustion (PIC) such as particulate matter (PM), carbon monoxide (CO), black carbon (BC, commonly known as soot) and polycyclic aromatic hydrocarbons (PAH); which have acknowledged health and climate effects. Estimates of premature death from the increased household air pollution caused by cookstoves are approaching roughly 4 million per year (Lim et al. 2012). Exposure is particularly high among women and children, who spend the most time in this environment. Climate effects include both short term and long term effects due to increased radiative forcing. BC is a dominant absorber of solar radiation in the atmosphere and regarded as the second highest contributor to global warming after CO_2 (Ramanathan and Carmichael 2008). It is estimated that 2% and 20% of global CO_2 and BC emissions come from biomass burning cookstoves, respectively (Smith et al. 2000), contributing to both local and regional climate change, effectively making it a globally relevant problem.

Solution to this global problem is the introduction and distribution of improved cookstove technologies; cookstoves that have improved thermal efficiency coupled with decreased pollutant emissions. Improved models range from rudimentary low-cost cookstoves often built from local materials to mass produced state-of-the-art forced draft cookstoves (FDCS) which rely on electrically-driven fans for more efficient combustion as compared to basic “improved” stove models. Small scale and large scale initiatives, such as those supported by the Global Alliance for Clean Cookstoves (GACC), are focused on distributing cleaner cookstoves with the aim of reducing these impacts to achieve the potential health and environmental benefits. The goals of the GACC include promoting the adoption of 100 million substantially improved stoves globally by 2020 with hopes of universal adoption by 2030 (Simon et al. 2014).

Quantifying health and climate benefits across different cookstove technologies has been a major focus across various fields of research from public health to climate modeling and forecasting. The designing and testing new cookstove technologies typically take place in the lab setting because it is easier to replicate testing conditions and less invasive that field testing. Specific testing protocols, like the Water Boiling Test (WBT), are used to measure the thermal efficiency and emission factors. Laboratory testing plays a critical role in understanding methods for reducing cookstove emissions and this data can be used as inputs for modeling purposes. For example, a cookstove emissions database can be utilized by climate modelers to identify the impacts of cookstove emissions on global, regional, and local climate.

However past research has shown that laboratory testing of cookstoves is not representative of actual operation of cookstoves in the real world in terms of emission quantities and characteristics. Laboratory testing severely underestimates emission factors of pollutants when compared to field studies (Roden et al. 2009a). This is attributed to factors such as local cooking practices, local conditions and the skill of the cook, which exhibit high variability in a real world setting.

The major focus of this work is to address the discrepancy between lab and field results by conducting a field campaign in Malawi: one of the poorest countries in the world, where more than 90% of the population is dependent on wood for cooking purposes. We measure emissions from in-home use of cookstoves in rural households in 2 districts of Malawi by collaborating with two pre-existing stove dissemination programs with the aim to characterize emissions and optical properties and assess health and climate impacts.

The subsequent text is the main section and the appendix, which is the body and the supporting information of a manuscript to be submitted to a journal in the near future.

CHAPTER 2: INTRODUCTION

Roughly 2.7 billion people depend on biomass burning cookstoves for cooking purposes (Foell et al. 2011). Among other environmental impacts (e.g. deforestation and forest degradation), biomass burning emits products of incomplete combustion (PIC) such as CO, particulate matter (PM), methane (CH_4), polycyclic aromatic hydrocarbons (PAHs) and black carbon (BC) with acknowledged health effects due to exposure of these pollutants including both short and long term effects such as cough and eye irritation, stroke, cardiovascular and chronic pulmonary disease and lung cancer (Lim et al. 2012; Rosenthal 2015). BC, commonly known as soot, is a component of fine particulate matter ($PM_{2.5}$, PM with aerodynamic diameter less than 2.5 μm) formed from combustion and estimated to have the second highest global climate warming impact after CO2 (Bond et al. 2013; Ramanathan and Carmichael 2008). Approximately 25 % of global annual BC emissions are from biomass burning provide domestic heating and cooking energy (Bond et al. 2013). Organic carbon (OC) which is co-emitted with BC is responsible for scattering of solar radiation which leads to a net cooling of the atmosphere (except for brown carbon, BrC which absorbs light), counteracting some of the warming effects of BC. BC mitigation efforts such as replacement of traditional cookstoves with alternative technologies have the potential to provide significant climate and health benefits by reducing exposure and emissions (Grieshop et al. 2009; Jeuland and Pattanayak 2012). For example, climate simulations by Shindell et al. (2012) emphasized the need to simultaneously mitigate emissions of CO_2 and other short lived species and estimated that decrease of BC and methane through the adoption of control measures could reduce projected global mean warming by about 0.5° by 2050.

Interest in quantification and reduction of these impacts has spurred the development of a range of alternative cookstoves with varying configurations, levels of sophistication and performance. Models range from rudimentary low-cost cookstoves often built from local materials to mass produced state-of-the-art forced draft cookstoves (FDCS) which rely on electrically-driven fans for more efficient combustion as compared to basic "improved" stove models. Grieshop et al. (2011) estimated that, based on published data on the laboratory performance of cookstoves, health (quantified as daily intake of $PM_{2.5}$ for users) and climate impacts (quantified as global warming commitment or GWC) of various stove-fuel combinations can each span two orders of magnitude, with all wood burning stoves associated with significantly greater impacts than modern fuel burning stoves such as LPG and kerosene. Among wood burning stoves, fan and gasifier stoves displayed roughly 3 times lower climate impacts than traditional, assuming non-renewable biomass. Estimated exposures associated with gasifier and fan stoves were an order of magnitude less than those from traditional and basic "improved" stoves, but a factor of 4-10 higher than those estimated for LPG.

This variation among different stoves types and the need to specify performance standards to enable comparison across different technologies has led to the development of a performance tier framework by the International Workshop Agreements (IWA 2012). In this approach, stoves are placed into tiers based on emission performance and are rated based on indicators in four categories: fuel efficiency, total emissions, indoor emissions and safety (Johnson and Chiang 2015). Figure A1 in the Appendix illustrates the ratings for indoor emissions performance. Tier values range from 0 for the worst performing stoves (traditional stove or three stone fire) to 4 for the best performing stoves that are expected to produce emissions low enough to meet World Health Organization (WHO) indoor air quality guidelines (Johnson and Chiang 2015). Standardized laboratory emission tests, typically the Water Boiling Test (WBT) (Bailis et al. 2007) is used to determine emission tiers for a stove for the purposes of stove certification, but comparing to field testing data gives a means by which to compare expected and observed performance relative to recommended benchmarks. As they represent standards, such tests emphasize repeatability and consistency to benchmark cookstove performance under similar operating conditions and serves as a guideline for policy-makers, investors, manufacturers, consumers and others in the cookstoves community.

While standardized laboratory testing is necessary for such benchmarking, evidence from the field has shown that it typically greatly overestimates performance relative to typical in-home use. Estimates of health and climate benefits associated with different stove technologies, such as those discussed above, depend on accurate estimates of real-world EFs and fuel use (Johnson et al. 2008; Roden et al. 2009; Shen et al. 2013). One field and laboratory comparison by Roden et al. (2009) found that in-field emission factors of PM (in grams emitted per kilogram of wood consumed) were 2 - 4 times higher than those measured during lab WBT tests. Another study in China found that field emission factors of PM2.5 exceeded laboratory observations by over a factor of 5 (Guofeng et al. 2012). Chen et al.

(2012) showed that the distribution of real time modified combustion efficiency (MCE) and aerosol optical properties observed in field sampling was not well-represented by the WBT. Hence, understanding how local conditions (e.g., user behavior, fuel type, cooking style) influence real-world emissions and emission factors is essential for assessing the actual health and climate benefits associated with alternative technologies.

FDCS have been held up as having great potential to greatly reduce PIC emissions (and impacts) via improved combustion efficiency. Emissions testing of FDCS has demonstrated emission reductions as high as 90% and 93% for $PM_{2.5}$ and EC EF's in laboratory settings when compared to traditional stoves (Jetter and Kariher 2009; MacCarty et al. 2008), putting them in the highest tiers (3 or 4) for indoor PM emissions. To date, in-field measurements of emissions from FDCS are limited. (Kar et al. 2012) reported that a FDCS reduced plume zone BC concentrations by a factor of 4 relative to traditional stoves during controlled cooking tests in simulated kitchens in the field. Patange et al. (2015) reported a statistically significant reduction of 40% in 24 hour BC concentrations when traditional stoves were replaced with FDCS in rural northern India. However, these studies only measured real-time BC mass concentrations and did not provide emission factors and other gas-phase (e.g, CO2) and aerosol species (e.g., organic carbon (OC)) which are essential for quantifying EFs (and better quantifying net climate impacts) were not measured.

Subsidizing clean cookstoves via carbon offset sales has been long held up as a means to enable access to improved technologies by poor households along with mitigating climate change (Smith and Haigler 2008). While the current state of the carbon markets call the near-term practicality of this into question (Sanford and Burney 2015), it remains a tantalizing possible source of finance. Current carbon finance methodologies include greenhouse gases (GHGs) but fail to account for the climate impact of PICs such as BC, CO, OC and non-methane hydrocarbons (NMHC), mainly because of the high uncertainty and variability in their emissions (Freeman and Zerriffi 2014; Sanford and Burney 2015) and the differing spatial and temporal scales of their impacts relative to GHGs (Bond 2007; Unger et al. 2010). BC dominates cookstove PIC climate impacts (Bond et al. 2004; Grieshop et al. 2011) and has become a focus for mitigation near-term climate change. In response, the Gold Standard Foundation recently proposed guidelines and methodology (www.goldstandard.org/articles/black-carbon-and-other-short-lived-climate-pollutants) for BC mitigation projects to monitor and quantify BC reductions as a way to incentivize and finance effective cookstove projects. This effort sidesteps some of the uncertainty and complication associated with quantifying BC climate impacts by assuming constant pre-defined values to quantify climate benefits via GWC calculations, thus making all BC reductions commensurate. An important missing piece in this effort is a rigorous understanding of in-situ emissions from current cookstove technologies and to what extent the emission reductions indicated by current lab results are actually achieved.

In response, we conducted a field emission measurement campaign in 2 districts of Malawi, Africa by collaborating with two pre-existing stove dissemination programs with the aim to characterize emissions and optical properties and assess health and climate impacts. The collaborating organizations had contrasting objectives which focused on implications of adoption of alternative cookstove technologies on exposure- and fuel-use reduction, respectively; and neither had undertaken cookstove emission measurements as part of their evaluation. In addition to testing in-home stoves, we also measured emissions from larger institutional stoves used in a school. The specific objectives of this work are to:

1. Measure fuel use and fuel and task-based emission factors from in-home use of alternative and baseline household and institutional cookstove technologies
2. Compare fuel based emission factors with relevant field and laboratory studies
3. Analyze real-time optical properties (absorption and scattering) of aerosols during in-home use.
4. Evaluate global warming commitments (GWC) associated with measured species as a proxy for climate impacts of in-home and institutional stoves
5. Quantify and compare health and climate impacts/benefits suggested by lab and field-based measurements

CHAPTER 3: METHODS

3.1 Study site details - Malawi

Emission testing on several stove models was conducted in Malawi, a small landlocked country in south-eastern Africa, where more than 90 % of the population uses biomass as their main source of domestic energy (Fullerton et al., 2011). It is one of the most densely populated countries in Sub-Saharan Africa and amongst the poorest nations in the world ranking 173 among 188 countries on the Human Development Index (Human Development Report 2015, UNDP). (Jary et al., 2014) concluded that a large cookstove intervention is feasible in Malawi. High poverty rates, high dependence on firewood and a predominant rural population makes Malawi an ideal case study country for studying potential impacts and co-benefits of introduction of improved stove technologies (Jary et al., 2014; Timko & Kozak, 2016).

Emission testing was done in two communities in different districts on stoves at the opposite end of the technology spectrum discussed above. In Chikhwawa, as a part of the Cooking and Pneumonia Study (CAPS; www.capstudy.org) conducted by the Liverpool School of Tropical Medicine (LSTM), two FDCS: Philips and ACE-1, the latter being an updated variant of the former with modifications for durability and usability (cost ~90 USD) were distributed among communities to assess the impact of stove technology on the incidence and outcomes of pneumonia in children. In contrast, in Balaka, the non-governmental organization (NGO) Concern Universal (CU; www.concern-universal.org) distributed *Chitetezo Mbaula* (CM) stoves, a low-cost (~ 1-2 USD), locally produced, natural draft clay stove, with the main objective of reducing fuel use by users and with the program funded via sale of carbon credits (www.goldstandard.org/projects/malawi-biomass-energy-conservationproject).

Measurements in rural households were taken during routine cooking activities in September-October 2015. In-home stove use was limited to cooking of traditional foods such as *Nsima* (a corn-flour porridge, the staple food of Malawi) or rice, preparing vegetable or meat dishes and heating water for bathing purposes. Stoves tested included the traditional three stone fire (TSF), *Chitetezo Mbaula*, Philips and ACE – 1 (Figure A2, Appendix). In addition to in-home testing, emissions from Controlled Cooking Tests (CCT) (R. Bailis, 2007) on larger, wood-burning institutional stoves were measured at the Jacaranda School for Orphans in Blantyre city. Institutional stoves are used where a large number of people are fed; these stoves are distinguished from in-home stoves by their large size to accommodate a dedicated, large cooking pot (80-100 L) which fit snugly into the stove body, typically enabling more efficient heat transfer. The institutional stoves tested for this study were the traditional TSF, Aleva (AL), Mayankho (MA) and the JumboZama (JZ). The JumboZama is a scaled up version of the rocket gasifier stove Zama Zama (Rocket Works, Durban, South Africa) built inside a masonry housing. Section 3.2 discusses the performance of institutional stoves. Figure A3 shows the institutional stoves tested in this campaign. Table A1 summarizes tests conducted during the campaign.

3.2 Sampling Methodology

In-home emission measurements were performed using the portable Stove Emissions Measurement System (STEMS). Figure A3 shows a schematic and photograph of the STEMS. The STEMS is powered by a 12 V battery and measures real-time (2 second) concentrations of carbon dioxide (CO2), carbon monoxide (CO), temperature, relative humidity (RH) and particle light scattering (Bsp; also used as a proxy for real-time PM2.5 mass concentration) with a laser photometer (635 nm wavelength). Real-time STEMS data were logged via a laptop. Integrated filter samples were collected on two 47mm diameter filter trains with equal flows for gravimetric and thermo-optical OC/EC Analysis. One of the filter trains contained a quartz filter and the other train contained a Teflon filter followed by a backup quartz filter downstream to correct for gas phase absorption artifacts (Subramanian, Khlystov, Cabada, & Robinson, 2004). A six-armed stainless steel probe with radial sampling ports centered in equal areas was used to capture a representative sample of emissions (C.A. Roden, Bond, Conway, & Pinel, 2006). Emissions were naturally diluted before being sampled by the probe located approximately 1 - 1.5 m above the cookstove. The emission factors were derived based on the carbon balance approach (Described in Section 2.3) which does not require all emissions to be captured. Dilute emissions passed through conductive sampling tubing to the STEMS where a cyclone (BGI SCC 2.654) provided a 2.5 µm size cut before measurement. For each cooking session, background measurements were taken for 5-10 minutes either before or after the cooking session. Wood fuel was set aside before the start of cooking and wood moisture and weight recorded as per the standard Kitchen Performance Testing protocol. Wood moisture content was measured by a moisture meter (Lignomat mini-Ligno S/DC) by taking the average of three random pieces of wood at three locations. The difference in the weight of the wood before and after the cooking session was used to determine the wood consumed. The fire was started up by lighting matches or by blowing on charcoal left from the previous cooking session. Households often used char from the previous cooking session to start the fire. It was not feasible to weigh leftover char during the study which results in a small overestimation of the wood consumed. Measurements from Smith et al. (2000) indicate that 1 kg of wood can result in up to 161 grams of char being formed. This non-accountability of char negatively biases the wood consumed, hence leading to a positive bias in emission rates calculated in this study (Section 2.3). We employ a conservative uncertainty of 20% in the wood weight measured due to non-accountability of char weight in our wood weight measurements. A brief survey was conducted after testing to get user feedback on performance and perception of alternative stoves.

Teflon filters were weighed before and after the campaign using a microbalance (Mettler Toledo UMX-2) to derive gravimetric PM2.5 concentrations for each test. Teflon filters were equilibrated in the weighing chamber with controlled temperature (22 ± 2 °C) and RH (35 ± 2.5 %) for 24 hours before weighing. Quartz fiber filters were pre-baked in a laboratory oven at 550 Carbon (OC/EC) analysis of quartz fiber filters with a Sunset OC/EC Analyzer used a modified NIOSH protocol with longer step durations to ensure complete removal of OC on heavily loaded filters. Table A2 in the Supporting Information gives the details of the protocol. Gas sensor calibrations were performed before and after the field campaign using custom calibration gas cylinders.

Flows were regularly checked and calibrated in the field with a bubble flow meter. The light scattering sensor in the STEMS was calibrated with emissions from wood burning in the laboratory against a Photoacoustic Extinctiometer (PAX) at 870 nm (Figure A5) with a R^2=0.87. (Weyant et al., 2014) estimated an uncertainty of 40 % in a similar scattering cell of 660 nm wavelength against a nephelometer at 530 nm. Field blank filters (N=4) were used to correct gravimetric PM2.5 concentrations. Real-time PM light absorption at an optical wavelength of 880 nm was measured using an AE-51 MicroAeth (AethLabs) operated in conjunction with the STEMS. To avoid excessive loading on the filter, frequent filter ticket changes in the field and have accurate flow control through the instrument, the internal pump of the instrument was bypassed and an external flow meter (Honeywell AWM3150V) and vacuum line added. The flow rate during operation varied from 10-25 cm^3 min^{-1}.

An acknowledged effect of filter loading on MicroAeth absorption measurements were corrected via the algorithm described by Park et al. (2010a). Additional details on absorption calculations and loading correction is described in Supporting Information Section A1.

3.3 Emission Factor and Emission Rate calculation

Fuel based emissions factors (EF; gram pollutant per kg fuel consumed) were calculated using the carbon balance method, assuming that carbon comprises of 50% dry wood by weight and all gaseous carbon in the wood is emitted as CO and CO2. Since the sum of background-corrected CO and CO2 concentrations serves as a tracer for the amount of dry fuel consumed the carbon balance does not require all emissions to be captured. Other carbonaceous species (e.g. gaseous hydrocarbons) contribute a small fraction of carbon in emissions and are neglected in this calculation (M. Johnson et al., 2008; C.A. Roden et al., 2006; Shen et al., 2013; Zhang et al., 1999). The average emission rate for a cooking session was determined by equation (1) where EF is determined by carbon balance and dry wood consumed is the weight of the wood with estimated water weight subtracted.

$$Emission\ Rate\ (mg\ min^{-1}) = EF\ (g\ kg^{-1})\ \ \frac{Dry\ wood\ consumed\ (kg)}{Cooking\ duration\ (\mathrm{min})}\ \ 1000\,\frac{mg}{g} \qquad (1)$$

Since the institutional stove tests followed the CCT protocol for specific food preparation activities, fuel- and food-based EFs (in grams of pollutant emitted per kg of fuel and food input, respectively) were determined. The cooking task was fixed and ingredients to make the porridge included water (3 - 4 buckets of 20 L capacity), soy flour (usually 20 kg sacks) and salt (0.3 - 0.4 kg). Each ingredient's weight was recorded before mixing in the cooking pot, total ingredient weight was used in food-based EF calculation.

CHAPTER 4: RESULTS AND DISCUSSION

4.1 Pollutant Emission Factors and Emission Rates

Figure 1 shows real-time (2 s and 1 minute average) concentrations of CO, CO_2, particle absorption and scattering coefficients (B_{ap}; B_{sp}) for representative FDCS and traditional household stove tests. Cooking session durations ranged from 19 to 233 minutes (median = 49 minutes). Gravimetric $PM_{2.5}$ concentrations derived from filters weights for in-home tests correlated well (R^2=0.87) with the averaged particle scattering coefficients, suggesting that real-time scattering data can be used a proxy for real-time $PM_{2.5}$ mass concentrations (Figure A6). Figure 1 also shows single scattering albedo (SSA; fraction of scattering to total extinction; (B_{sp} / (B_{ap} + B_{sp})) and modified combustion efficiency (MCE; ratio of ΔCO_2 to the sum of ΔCO and ΔCO_2, where Δ indicates background-corrected concentrations in ppm) are shown on the right axes. A lower SSA signifies a greater contribution from absorption to total aerosol extinction and tends to be associated with positive radiative forcing, while higher MCEs indicate more efficient combustion due to a greater extent of fuel carbon oxidation. All cooking events were characterized by a scattering spike at startup, as observed by (C.A. Roden et al., 2006) (evident in the Figure 1 as SSA exceeding 0.5 during start of test). Observations of cooking activity showed that addition and adjustment of wood predominantly resulted in spikes in absorption for FDCS and spikes in scattering for traditional cookstoves. While FDCS tests typically had a large scattering peak only at the startup and overall particle extinction was dominated by absorption, extinction from traditional stove tests were dominated by scattering during testing. As a result, a comparatively higher SSA and lower MCE for traditional stoves is observed as compared to

FDCS. Test-average SSA at an optical wavelength of 880 nm was highest for Traditional stoves (0.36), followed by CM (0.28) and FDCS (0.25) indicating that particles from all 'alternative' stoves are more absorbing than those from traditional stoves and thus greater specific warming.

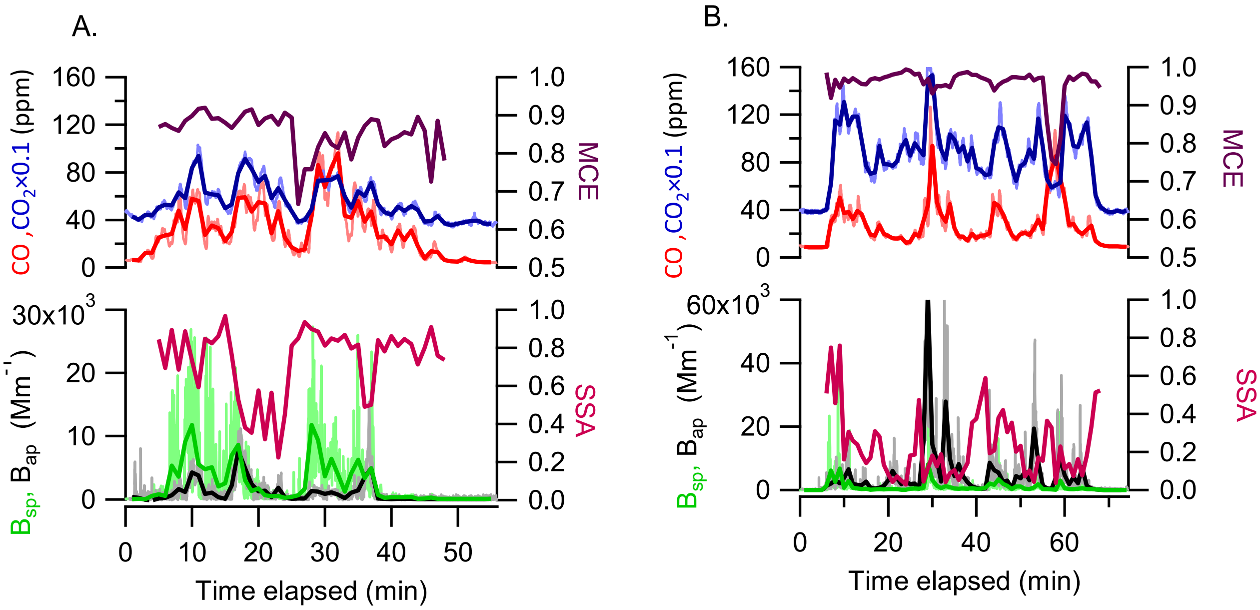

Figure 1. Time series of CO, CO_2, particle light scattering (B_{sp}) and absorption (B_{ap}) measured during in-home cookstove testing of: A. Traditional; B. Philips. Single scattering albedo (SSA) and modified combustion efficiency (MCE) are shown against right axes for emission periods only. CO_2 is divided by a factor 10. Note that the axis for optical properties is different for each graph. Lighter traces indicate real-time (0.5 Hz) data and darker traces indicate 1 minute averages.

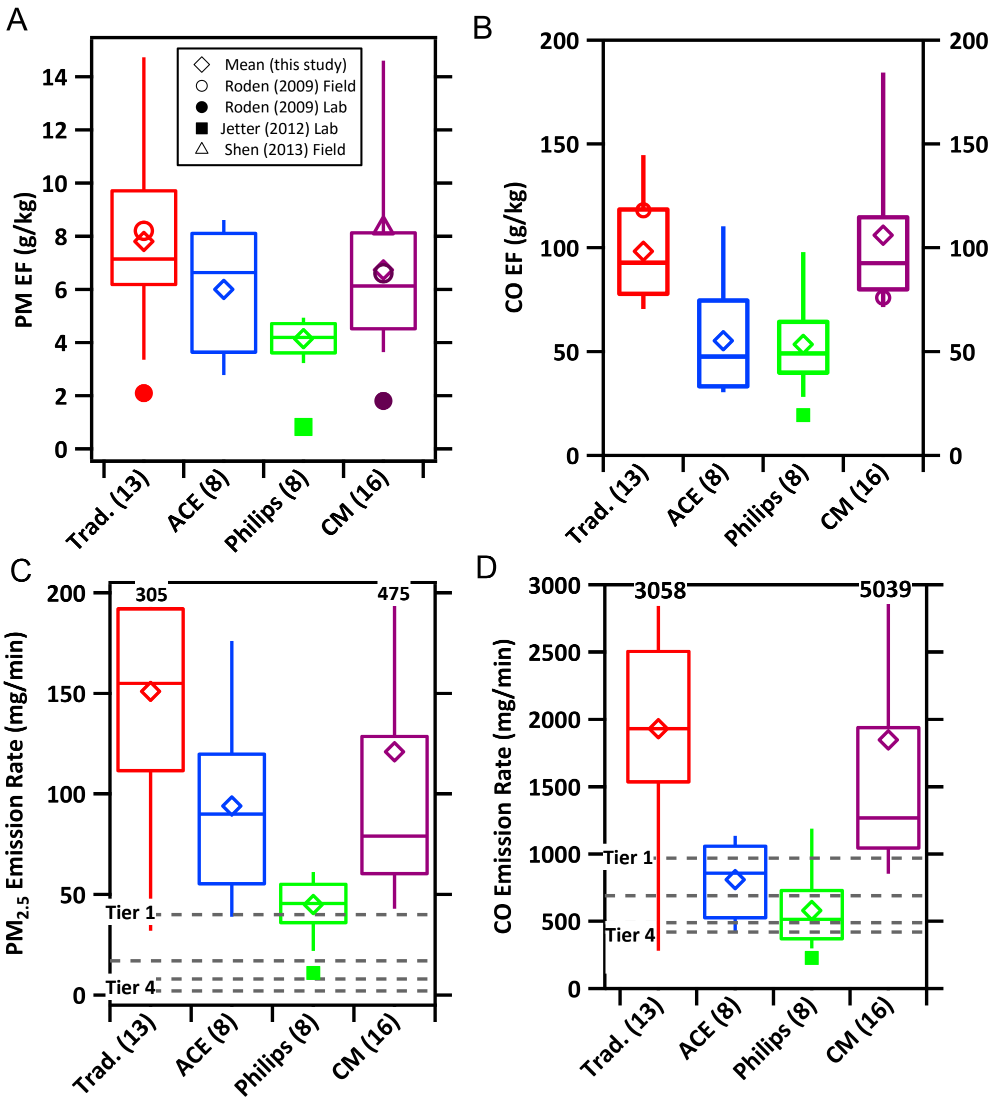

Figure 2. Box plots of pollutant emission factors and emission rates. The box represents the interquartile range of 25th and 75th percentile. The whiskers indicate the 5th and 95th percentile. The horizontal line in the box is the median (50th percentile). 95th percentile whiskers for some of the stoves were out of scale on the y axis and are indicated by numbers on the top axis. Panel A: $PM_{2.5}$ EF; Panel B: CO EF; Panel C: PM ER, Panel D: CO ER. Tier values for panel C and D taken from (Johnson and Chiang 2015). The legend for all panels is in panel A. Data from (Shen et al. 2013) is for a movable metal stove used in China. Data from (Roden et al. 2006) is for an improved Patsari Stove.

Comparing to mean $PM_{2.5}$ (4233 μg/m^3) and CO (48 ppm) concentrations measured in plumes above traditional cookstoves, FDCS plumes showed 36 and 44 percent reductions respectively, whereas CM showed only 4 % reductions in $PM_{2.5}$ and an increase of 15% in CO concentrations respectively.

Traditional cookstoves had the highest $PM_{2.5}$ EF (Figure 2A) of 7.8 ± 2.9 g kg^{-1} (average ± 1 SD) which is similar to field observations in Honduras (8.5 ± 1.6 g kg^{-1}; N=12) (Roden et al. 2006). In a field study in China, EF's of $PM_{2.5}$ ranged from 8.1-8.5 g kg^{-1} for a simple wood burning metal stove (Shen et al. 2013). The Philips emitted the least PM (4.1 ± 0.6 g kg^{-1}), with 47% lower mean emissions than the traditional stoves; a statistically significant reduction ($p < 0.005$ from paired t-test) Emissions from CM and ACE were lower than those from traditional stoves on average, but the difference was not statistically significant ($p = 0.761$ and 0.127, respectively).

CO EFs (Figure 2B) ranged from 28-198 g kg^{-1}, with traditional cookstoves the highest (98 ± 26 g kg^{-1}) and both FDCS showing the highest, statistically significant reductions of 45%. ($p < 0.005$). Mean CO emission factor for CM was 106 g kg^{-1}, which is slightly higher than traditional stoves. High emissions and variability for CO emission factor for this stove could be attributed to some of the CM cookstoves being broken, hence leading to inefficient combustion. The average CO emission factor for traditional stoves was comparable to field measurements from traditional stoves in Honduras (116 ± 55 g kg^{-1}) (Roden et al. 2006).

Although $PM_{2.5}$ and CO EF's obtained in this study are similar in range to other field studies, they are considerably higher than those observed in laboratory studies. Our mean

$PM_{2.5}$ EF for traditional stoves is roughly 3.7 times higher than laboratory tests by Roden et al. (2009). For the Philips, laboratory measurements by the US EPA found 80 % (0.83 g kg^{-1}) and 65% (19 g kg^{-1}) lower mean $PM_{2.5}$ and CO EFs respectively when compared to our field results.

Fuel based emission factors as shown in Figure 2A and 2B are normalized by fuel consumed and fail to take into account effect of two important parameters: actual cooking time and the amount of fuel consumed, and hence total emissions for each cooking session cannot be estimated. These parameters depend on the efficiency of the cookstove and is not reflected in fuel based EF's. ER's on the other hand include the effect of these two parameters and can be calculated using Equation 1.

Figure A7 in the Supporting Information shows the box plot of wet-basis (as measured) wood consumption rate in kg hr^{-1}. Traditional stoves consumed the most fuel with a mean consumption rate of 1.44 kg hr^{-1} with mean reductions from the CM, ACE and Philips being 26%, 27% and 51 % respectively. Panels C and D in Figure 2 show the $PM_{2.5}$ and CO ERs calculated using these values and Eq. 1; also shown are tier boundaries for indoor emission rates (www.iso.org/iso/catalogue_detail?csnumber=61975).

Figures 2C shows that trend in $PM_{2.5}$ ER (in terms of dry wood consumed) is similar to $PM_{2.5}$ EFs. For example, use of Philips reduced the ERs of both $PM_{2.5}$ and CO by 70% compared to traditional stoves. PM and CO ERs reported by Jetter et al. (2012) based on laboratory testing of Philips stove (for wet wood) show a *further* reduction of 76 % for $PM_{2.5}$ ER (11 mg min^{-1}; Tier 2) and 61 % reduction in CO ER (228 mg min^{-1}; Tier 4) compared our field values; further indication that laboratory tests can severely underestimate real-world emissions of even the most advanced biomass burning stoves.

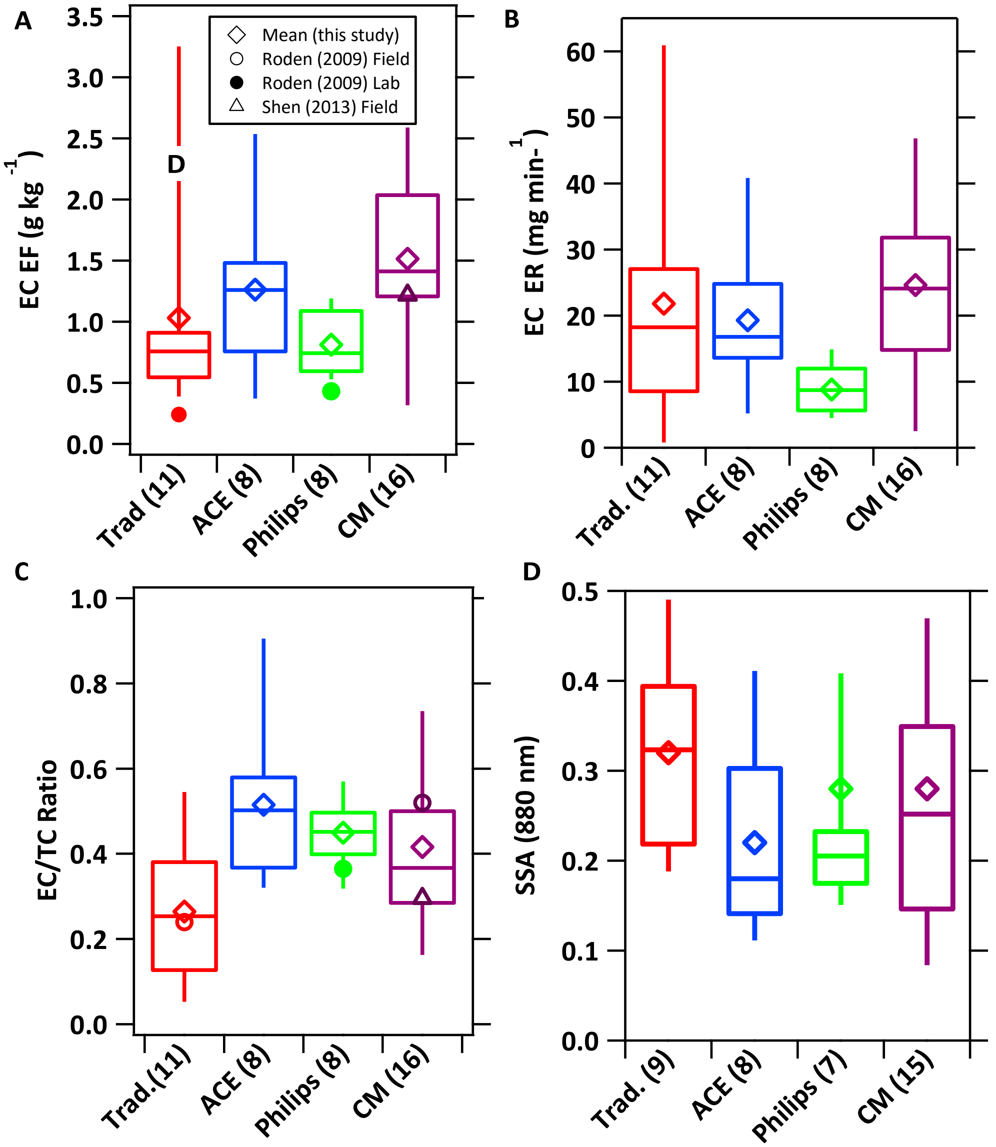

Figure 3. Box and whisker plots of A: EC EF; B: EC ER; C: EC/TC ratio; D: Single scattering albedo at 880 nm for different stove technologies tested. Data from (Shen et al. 2013) is for a simple metal stove.

EC emission data are shown in Figure 3, which shows box plots of EC EFs, ERs, EC/TC ratios and SSA for cookstove technologies tested. All quantities show high variability, most likely due to differing configurations, uncontrolled nature of combustion and usage of the stoves coupled with the varying skills of the cook. EC EFs and EC/TC ratios for intervention stoves generally show an increasing trend relative to the TSF, though this is moderated for ERs due to the reduced fuel use, especially for the Philips stove. EC EFs for CM were similar to observations for other alternative cookstoves in other field studies (Roden et al. 2009b; Shen et al. 2013). Gasifiers had a highest EC/TC ratios (0.48) followed by CM (0.42) and Traditional cookstoves (0.28). EC/TC ratio for open biomass burning averages 0.1 (Reid et al. 2005b), which is significantly lower than primary cookstove emissions observed in this study.

Figure A8 in the Appendix displays the relationship between MCE with SSA and EC/TC ratio with SSA for in-home cookstove testing. We observe a general trend of decreasing MCE and decreasing EC/TC ratio with increasing SSA. Hence, larger EC/TC fraction for FDCS indicate more efficient burning (as indicated by higher MCE) and more specific global warming (as indicated by lower SSA). This is again consistent when comparing Figure 4C and 4D, with an increasing EC/TC ratio and decreasing SSA when moving across rudimentary to improved stove technologies. Figure 1 shows that decreasing values of SSA with increasing MCE is observed and can be explained by the fact that more BC is produced during the flaming part of the burning (hence a higher EC/TC ratio), when MCE is highest, and more OC and less BC is produced during the smoldering part of burning when MCE is lowest (lower EC/TC Ratio) (McMeeking et al. 2009; Pokhrel et al. 2016).

Table A5 in the SI gives a summary of all in-home stove tests performed during the campaign.

4.2 Institutional Stoves – Food and fuel based pollutant emission factors

Food and fuel based EF's were estimated for institutional stoves are listed in Table 1. EFs for PM and CO followed a consistent trend, with the Institutional TSF (I-TSF) the highest, followed by the AL, MA, and JZ CO and PM emissions were reduced by similar amounts, with Mayankho showing reduced emissions of 67-68% for CO and PM and JumboZama reducing CO and PM emissions by 73-75 %. Averaged wood consumption rate was highest for the Aleva (5.2 kg hr^{-1}), followed by I-TSF (4.18 kg hr^{-1}), Mayankho (3.85 kg hr^{-1}) and JumboZama (3.30 kg hr^{-1}). This wood consumption rate serves as a basis for energy demand for institutional stoves in Section 3.3. EC fractions in total carbon generally increased compared to I-TSF. Food based PM EF and CO EF follow the same trend as their fuel based counter parts, with JZ exhibiting the >80 % reductions for both. Lower EF values for JZ as compared to MA is also due to a smaller cooking pot size and reduced cooking time. Further extensive comparisons are difficult due to low number of tests for institutional stoves compared to in-home stoves and lack of emissions data for these stoves in literature. Table A5 in the SI gives a summary of all institutional stove tests performed during the campaign.

Table 1. Food and fuel based emission factors for institutional cookstoves. N indicated number of samples. Numbers are averages and numbers in brackets represent standard deviation.

	Fuel Based EF (g/kg fuel)				Food based EF (g/kg food)		
Stove	**CO EF**	**PM EF**	**EC EF**	**EC/TC**	**CO EF**	**PM EF**	**EC EF**
TSF (N=3)	105	7.1	0.58	0.18	7.9	0.53	0.042
	(9)	(1.3)	(0.21)	(0.07)	(1.4)	(1.11)	(0.010)
Aleva (N=1)	43	3.5	0.35	0.25	4.7	0.40	0.038
Mayankho (N=3)	33	2.4	0.73	0.61	2.21	0.16	0.049
	(15)	(0.6)	(0.23)	(0.23)	(0.98)	(0.04)	(0.017)
Jumbozama (N=3)	29	1.8	0.5	0.45	1.5	0.09	0.024
	(13)	(0.5)	(0.36)	(0.24)	(0.8)	(0.03)	(0.013)

4.3 Real-time optical properties

Reporting of test averages fails to give information on real-time emissions characteristics such as burn conditions, combustion efficiency, particle properties, and proxies for emission quantities describe dominant combustion phases and their relative contributions to total emissions (Chen et al. 2012). Patterns of Real-Time Emissions Data (PaRTED) analysis (Chen et al. 2012) was used to evaluate quantities and optical characteristics of emissions based on real-time data. In this analysis, MCE and SSA are calculated for each combustion event (1 minute); MCE is a proxy for combustion efficiency indicating the conversion of fuel carbon to CO_2. A bivariate histogram with MCE on the y-axis and SSA on the x-axis is weighted by instantaneous scattering emission factor ($IEF_{scat,}$ representing the amount of light scattering related to the particulate emissions normalized by mass of fuel consumed in that time) and normalized by the total emissions to show the percent contribution for each combustion event towards the total aerosol emissions. This weighting is reasonable, as it was for Chen et al., given that scattering shows a strong and consistent correlation ($R^2 = 0.87$) with average PM concentration from gravimetric analysis (Fig. S5). Additional information on this approach and calculations can be found in the Supporting Information of (Chen et al. 2012).

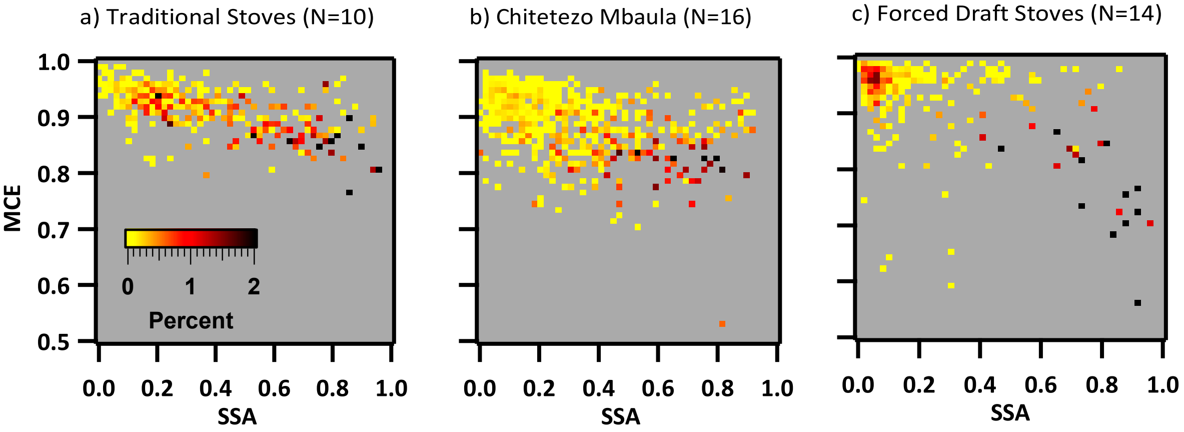

Figure 4. Bivariate histogram of MCE and SSA weighted by particle emissions (Parted Plots) for a. Traditional; b. Chititezo Mbaula and c. FDCS. Bottom axis is the single scattering albedo (SSA) at 880 nm. Left axis is the modified combustion efficiency (MCE). N indicates the number of tests included for this analyses. Note that 3 of the traditional stove tests were excluded from this analysis due to lack of either CO_2 (1 test) or absorption data (2 tests).

Figure 4 shows PaRTED plots for traditional, CM and FDCS (ACE-1 and Philips combined) emission tests. Each point on the graph represents the SSA and MCE at which a combustion event (1 minute) may occur and the color scale indicates the percent contribution of that event to the total scattering (~PM mass) emissions. Higher emission contributions (darker colored cells) tend to be clustered at relatively lower MCE's and higher SSA's, indicating that these events are emit more scattering particles/particle mass. For the FDCS, the majority of the combustion events occur at high MCE (>0.9) and low SSA (<0.2), indicating more efficient burning and emission of more absorbing particles for most part of their operation. High emission events (MCE < 0.9; SSA > 0.4) as indicated by the darker colors usually occurred at the startup of the cooking session. This extent of contribution to startup can be seen by stacking time series of the running IEF_{scat} against the normalized test time for each test in the respective stove type as seen in Figure A9. We observe that IEF_{scat} peaks during the startup phase. IEF_{scat} is normalized by the net gaseous carbon emitted (CO and CO_2). Although the test averaged IEF_{scat} is highest for Traditional and lowest for FDCS, we see a much higher peak for gasifiers. This is due to the fact that there is a relatively lower gaseous carbon emitted during these periods which results in these high peaks. These observation are consistent for FDCS stoves as shown in Figure 1B. This analysis suggests that stove type and cooking activity influenced the light scattering and light absorbing properties of the emitted particles.

While SSA for pure BC aerosol is reported to be in the range of 0.15-0.3 at an illumination wavelength of 530 nm (Roden et al. 2006), we observe several instances of

SSA's in the range of 0-0.2 for FDCS stoves at 880nm. Average SSA values were highest for traditional stoves (0.38 ± 0.16) and lowest for FDCS (0.22 ± 0.09). SSA decreases with increasing wavelength for biomass burning emissions and scattering is strongly dependent on particle size (Bergstrom et al. 2007).Further, (Just et al. 2013) observed in laboratory tests that a FDCS produced up to 3 times more particles at 30 nm than the traditional stoves. To further illustrate the effect of higher wavelength and lower particle size, Mie theory calculations were performed on pure BC aerosol with a refractive index of 1.85 + 0.71i (Bond and Bergstrom 2006). Figure A10 in the Appendix shows the dependence of optical properties (scattering, absorption and SSA) with particle diameter at two wavelengths of 530 and 880 nm. Considering the 30 nm particles, the SSA is reduced by a factor of 4.5 due to increase in wavelength from 530 to 880 nm. Comparing across particle sizes at 880 nm, SSA for a 100 nm particle is approximately 33 times higher than SSA for a 30 nm particle. Hence, lower SSA values in this study is attributed to reduced particle size and a higher optical wavelength of measurement.

The averaged mass scattering cross section (MSC; ratio of scattering to gravimetric $PM_{2.5}$ concentration) is 0.87 ± 0.31 m^2 gm^{-1} at an optical wavelength of 880 nm and is lower than MSC values reported by (Reid et al. 2005a) for dry biomass burning smoke, which is in the range of 3.6 to 4.3 m^2 gm^{-1} at an optical wavelength of 550 nm while (Roden et al. 2006) determined the average MSC of 2.2 ± 0.6 m^2 gm^{-1} at 530 nm for in-field use of cookstoves. Lower MSC values in this study is again attributed to wavelength dependence of scattering. Averaged mass absorption cross-section (MAC; ratio of B_{ap} EF to EC EF) for all tests was 13.2 ± 4.8 m^2 g^{-1} ,which is within the limits of the AE-51 MAC of 12.5 m^2 g^{-1} (Cheng 2013).

3.3 Climate impacts of different stove options

To allow integrated comparisons of stove emission impacts, 20 and 100 year global warming commitment (GWC) in tons of equivalent CO_2 were estimated based on the measured pollutant emission factors in this study (CO_2, CO, OC and EC) following the method of (Grieshop et al. 2011). Where possible, assumptions were made to be consistent with those applied during carbon credit calculations. The Clean Development Mechanism suggests a default value of 0.81 for the fraction of non-renewable biomass (fNRB) for Malawi (www.cdm.unfccc.int/DNA/fNRB/index.html) which was used for this study although (Bailis et al. 2015) reported that this fraction may not be representative due to several factors including large spatial variations in fNRB, uncertainties in fuel use data and failure of models to respond to behavioral changes in response to scarcity of wood among the users. GACC recommended thermal efficiency of 10 % was assumed for baseline cookstove technologies (http://carbonfinanceforcookstoves.org/implementation/certification-process/baseline-guidelines) and annual fuel use were calculated based on fuel use reductions observed in this study (Figure A7). The global warming potential values were recommended by the Gold Standard and also published by the Intergovernmental Panel on Climate Change (IPCC) (IPCC 2014).

Other components commonly used for estimating GWC include CH_4, total non-methane hydrocarbons (TNMHC) and N_2O. GWP calculations by (Johnson et al. 2008) for field emissions in Mexico indicated that CH_4 was the largest non-CO_2 contributor to GWC (EC was not measured). Since we did not measure CH_4, we used ratios of CO to CH_4 EFs from laboratory results in literature and extrapolated to estimate CH_4 EFs. We incorporated

CH_4 to CO ratio of 0.05 for Philips and ACE based on laboratory results by Jetter et al. (2012) for the Philips FDCS. A CH_4 to CO ratio of 0.08 based on data compiled by (Grieshop et al. 2011) was used for traditional and *Chititezo Mbaula* stoves as the CO EF distributions did not show significant difference. TNMHC and N_2O has been reported to contribute a small fraction of GWC for biomass emissions (Johnson et al. 2008; Smith et al. 2000; Zhang et al. 2000) and their effects are not considered here. We compared climate impacts of in-home and institutional stoves and also included effects of modern LPG stoves. LPG, often being regarded as one of the cleanest technology across the cookstove spectrum was included in this analysis relatively quantify health and climate benefits.

Baseline (referred to as HH-Trad.) fuel consumption for in-home use is assumed to be 2.6 tons per annum based on a survey of fuel use (See SI Section S2). Assuming a thermal efficiency of 10 % for baseline technologies and wood heating value of 15 MJ kg^{-1}, we calculate the energy demand of each household to be approximately 10.6 MJ day^{-1}. This energy demand was set as a basis and we incorporated fuel use rate reductions observed in this study (Figure A7). The focus here is the influence of real-world emission factors on estimated impacts. The original analysis used values from laboratory studies, whereas data from this study is representative of real-world emissions (except for LPG, which was taken from (Grieshop et al. 2011)). Additional details on GWC calculations, including assumptions for institutional stoves are given in the Supporting Information.

Figure A11 and Figure 8 shows the GWC values estimated across a 20 and 100 year horizon respectively. As expected, traditional stoves show the highest contribution across both time scales, followed by the CM, ACE and Philips. For institutional stoves, the GWC was highest for the institutional TSF (I-TSF) followed by the Aleva, Mayankho and JumboZama. Across a 100 year horizon for household stoves; CM, ACE and Philips show overall reductions of 15%, 24 % and 54 % respectively from the HH-Trad stoves, with highest warming contributions coming from CO_2 (67-76 %) followed by EC (23 – 37 %). All the other species considered here contribute less than 10% to the net warming. In contrast, results from (Grieshop et al. 2011) indicate much higher reductions of 59 % and 93 % for improved and forced draft stoves respectively when compared to traditional stove technologies; further indication of discrepancy between lab and field studies (although this was done on a basis of 50% non-renewability of biomass and a daily energy demand of 20 MJ day^{-1}). For institutional stoves, we see reductions ranging from 30% for the Aleva to 58% for the JZ when compared to I-TSF. Due to the high expected combustion efficiency for a gaseous fuel, PICs contribute minimally to the GWC associated with LPG, thus the majority was contributed by CO_2 emissions from this non-renewable fuel. Warming impact of LPG was 83 % lower than the Philips stove. Across a 20 year horizon, EC, being a more effective short term enforcer to global warming, contributed the most to total positive GWC, ranging from 49 to 61 % across all wood burning stove types. Contribution by CO_2 was second highest for Philips (36 %) and ACE (28 %) whereas CH_4 contribution exceeded that of CO_2 for traditional and CM stoves. Across a 100 year horizon, CO_2 followed by EC had the largest contributions.

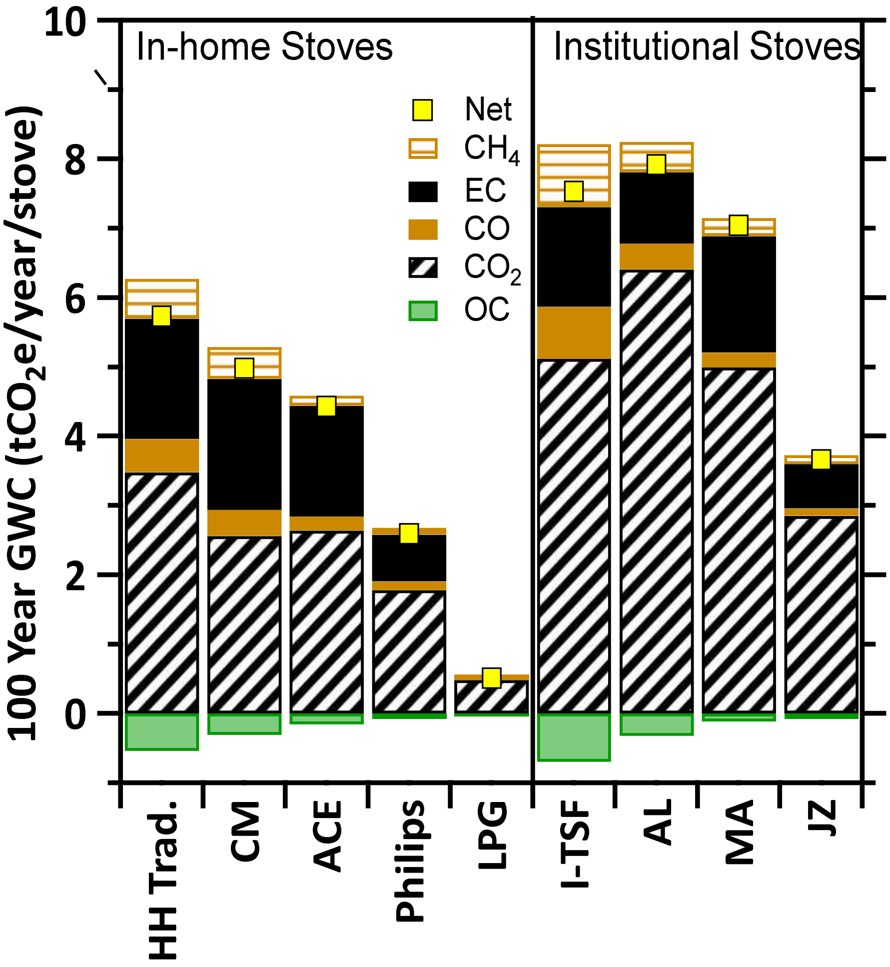

Figure 5. 100 year GWC values for one year of use of in-home stoves (L) and institutional stoves (R) from major short- and long-lived climate forcing species emitted by stoves. CH_4 component is estimated based on the ratio of CH_4 and CO emission factors from other sources. Note that the daily energy use is different for institutional stoves. In-home stoves are assumed to be used every day of the year. Institutional stoves are assumed to be used for 5 days a week for 40 weeks a year with an energy basis based on averaged fuel use data measured in this study.

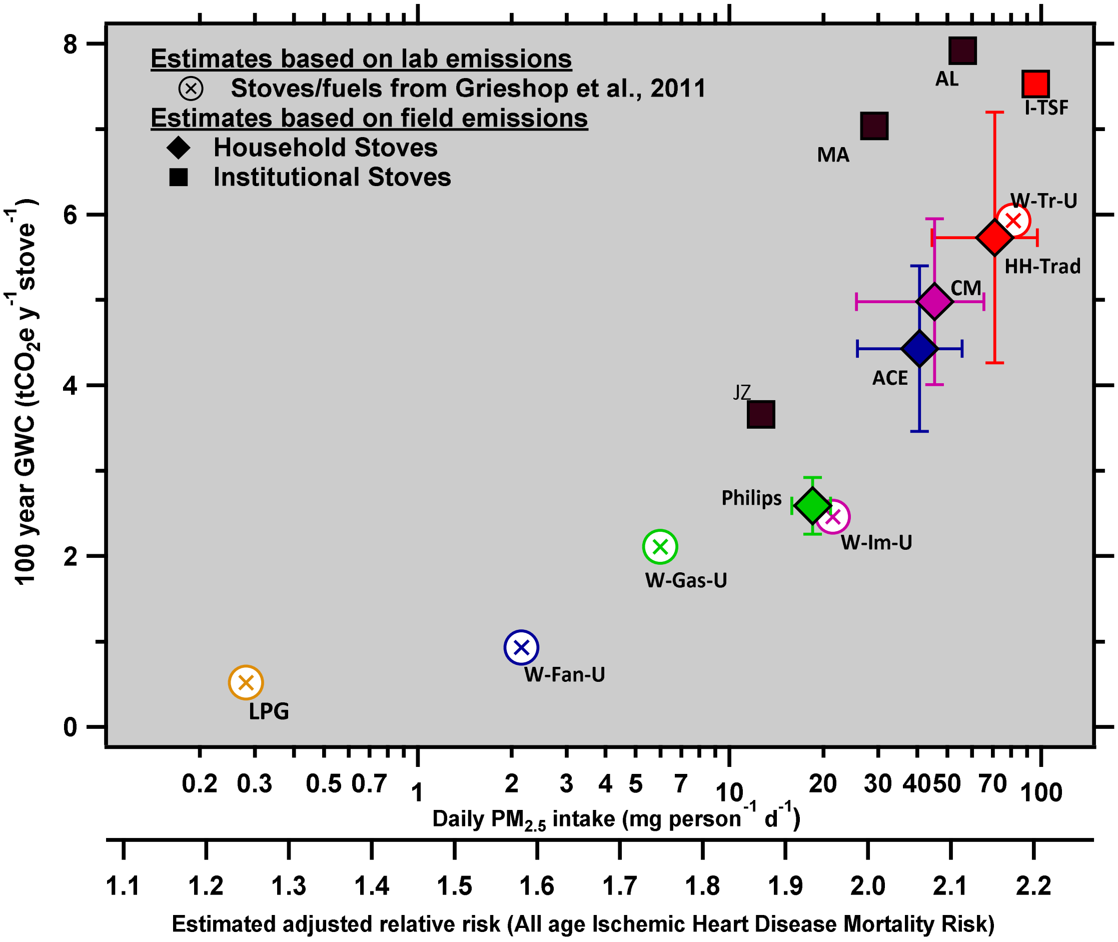

Figure 6. Health and climate impacts of various stove-fuel combinations based on laboratory emission test data (shown with circles and error bars; adapted from (Grieshop et al. 2011)) along with the central estimates from current study shown in marked with diamonds for in-home stoves and squares for institutional stoves. W-Tr-U : Traditional unvented wood burning stoves; W-Im-U: Improved unvented wood-burning stoves; W-Gas-U: Unvented wood burning gasifier stoves; W-Fan-U : Wood burning fan stove. HH-Trad.:Household Traditional; CM: Chitetezo Mbaula; I-TSF: Institutional three stone fire; AL: Aleva; MA: Mayankho; JZ: Jumbozama

Emissions are largely a concern due to their impacts on climate warming and human exposure. Figure 6, adapted from (Grieshop et al. 2011) estimates the PM intake/mortality risk (horizontal axis) and GWC (vertical axis) of several cookstove technologies evaluated in that study, with added estimates made based on data from in-home testing during this study. See the original paper for the methodology for making the PM intake estimates based on assumptions of energy demand (15 MJ household^{-1} day^{-1}) and biomass non-renewability (50%), which makes use of intake fraction to link emissions to human exposure and intake and the dose-response relationship from (Pope et al. 2009) to estimate adjusted relative risk of mortality due to cardiopulmonary and cardiovascular disease.

Figure 9 is a dramatic demonstration of the implications of the performance decrement observed in our field measurements in terms of both health and climate impacts. In comparing equivalent stoves tested in different settings, we see that all wood burning stoves underperform in terms of emissions/exposure with their laboratory counterparts. For instance, climate impact of Philips is roughly 2.8 times higher and daily PM intake is 7.4 times higher for in-home field use compared to laboratory use. JZ and Philips show the least relative contribution to both exposure and climate warming among in-home and institutional stoves respectively, but are still associated with much greater GWC and PM intake values than for the 'benchmark' LPG stoves. For example, compared to the estimated impacts of LPG stove use, in-home use of a Philips stove results in 5 times higher GWC and around 57 times higher daily exposure. This corresponds to increase in adjusted relative risk (which follows a log-linear relationship with PM exposure) from 1.3 to 1.65although the Philips is still further off from LPG stoves, which has a relative risk of roughly 0.29, Impact estimates for HH Trad,, CM and three of the institutional stoves (I-TSF, MA and AL) are within the bounds of impact estimates based on laboratory tests of traditional stoves. . whereas best performing FDCS are reasonably within the bounds for laboratory performance of improved unvented wood-burning stoves (W-Im-U). Field observations of emissions from a range of stoves used in interventions give important insights into the potential for these technologies to meet their promised potential to mitigate the climate and health impacts associated with traditional technologies.

CHAPTER 5: CONCLUSIONS AND FUTURE WORK

Our results suggest that both simple ‘improved’ stoves and more advanced biomass stoves provide some benefits, but fall well short of addressing a range of important impacts. For example, the while the simple clay stove tested here reduces fuel use and PM emissions, we estimate it to provide relatively minimal reductions to mitigate health (risk reduction of ~35%) and climate (GWC reduction of 54%) impacts. Results from a recent study suggest that use of the CM reduces fuel consumption by approximately 34–43% (Malakini et al. 2014). In contrast, we see a slightly lower reduction of 26 % for the CM based on the fuel consumption rate (kg hr^{-1}). FDCS models, and especially the Philips stoves tested, reduced all emission rates relative to baseline cookstoves, through a combination of reduced fuel-based emission factors and fuel consumption rates, but are substantially higher than those measured in laboratory based measurements. This is likely in large part to these stoves not being used in accordance to manufacturer recommendations. For example, wood pieces sticking out of the gasifier stoves was a common observation in the field (Figure A1 in Appendix). This is in part due to the requirement for very small fuel pieces, which were not what was available/preferably harvested in this environment. Processing larger logs and branches to the size recommended for the FDCS models considered here (~1 x 5 cm) represents much extra work for the household, and was simply not usually completed. Such a burning practice does not lead to optimal combustion of the volatiles emitted from pyrolysis of wood in the combustion chamber. Results from lab testing could be viewed as an indication of the potential performance of the stove, but it is not realistic that such performance is achieved in field settings, where many factors may affect how the device is used. Such variables are not accounted for during lab tests, which focus on consistency and repeatability. The only way that this source of variability can be addressed is via a stove that is completely robust to changing fuel type/configuration or a situation in which a homogenized fuel source (e.g. pellets) is provided or readily available. The former is only likely possible in a more advanced combustion device (e.g. the enclosed heating stoves, often with catalytic after treatment of exhaust used in developed countries) that are beyond the budget of the target population, while the latter requires a close look at the broader system, beyond the stove.

Our observations thus highlight the need to expand the view beyond 'clean cookstoves' to clean and controlled cooking systems, which could provide significant health and climate benefits and ideally perform identically and reproducibly under both laboratory and field conditions. The approach favored by (Smith and Dutta 2011) is to focus efforts on a switch to modern appliances (e.g. electrical induction cookers and LPG) rather than promoting 'improved' biomass stoves. Concern Universal (CU), who has played a major role fostering local production groups and markets for improved cookstoves, suggest that the CM is perceived to produce less smoke in the kitchen, is safer, and reduces both cooking and fuelwood collection times and/or frequencies (Jagger and Jumbe 2016). This is consistent with the feedback survey for this stove, as all participants conveyed the same information on stove perception which was provided to them by CU during stove distribution. Another interesting point to note is that the communities in Balaka were not aware that advanced stove technologies such as the Philips even existed; thus altering the perception of Chitetezo Mbaula.

Lab testing, even if they do not replicate field performance is required for relative comparison of stove performance under similar conditions. However, current laboratory protocols fail to deliver real-world emissions thus prompting a need for a protocol to accurately represent emissions characteristics and quantities as observed in the field. This can be achieved by obtaining more field measurements at various locations across the world. One major drawback of current laboratory testing protocols is that they fail to replicate low-efficiency emissions such as smoldering, which is a common observation in the field. Hence, future lab testing protocols would do a better job at representing real-world emissions if a smoldering phase was included during testing. In depth characterization of field performance and emissions of both baseline and replacement stove technologies could be vital for analyzing health and climate benefits along with being used as inputs in model application.

One important component not included in this work is the effect of brown carbon (BrC), the absorbing component of OC across visible and ultra-violet wavelengths which is also responsible for global warming. Non-accountability for BrC could lead to substantial uncertainties in quantifying the effect of BrC, leading to misattribution of observed particulate matter absorption to BC, contributing to the discrepancy between models and observations (Saleh et al. 2014). However this was beyond the scope of this work and future research calls for accounting for this effect in models.

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APPENDIX

APPENDIX A

Section A1. BC loading correction

For filter based optical measurement of BC, the attenuation of light (ATN) is given by,

$$ATN = 100.\ln\left(\frac{I_0}{I}\right) \tag{1}$$

Where I_0 and I light intensities through a reference blank spot and spot of aerosol on the filter ticket respectively. The factor 100 is for convenience. Particle absorption (B_{ap}) is calculated by the following equation:

$$B_{ap} = BC.\sigma_{ATN} = \frac{10^6.A.\Delta ATN}{100.Q.\Delta t} \tag{2}$$

where BC is the black carbon concentration in $\mu g/m^3$; A is the area of the sample spot (7.1 x 10^{-6} m^2); Q is the volumetric flow rate in m^3/s; Δt is the sampling interval in s; ΔATN is the variation in the ATN during the period Δt, and σ_{ATN} is the apparent mass attenuation cross-section (MAC) for the black carbon that is collected on the filter in m^2/g (Cheng 2013).

Filter based optical measurement of BC is associated with loading effects. At low ATN values, the relationship between ΔATN is proportional to the BC concentration on the filter. As ATN increases, the measured BC concentration (or absorption) becomes underestimated (Bond et al. 1999; Drinovec et al. 2015; Park et al. 2010; Virkkula et al. 2005, 2007). Absorption from AE-51 was corrected for filter loading artifacts using the approach by Park et al. 2010. For this approach, the corrected absorption is given by:

$$B_{ap}(compensated) = (1 + k.ATN).B_{ap}(non \quad compensated) \tag{3}$$

In this approach, the average BC concentration in a one width ATN bin is plotted and the factor k is calculated based on the ratio of slope and intercept obtained from the linear fit of the plotted data. The basic idea behind this approach is that within a large data set, the probability of BC lying in an ATN bin same across all ATN bins i.e. the BC vs ATN slope should be close to 0 (Drinovec et al. 2015).

Section A2. Global warming commitment (GWC) calculation

Assumptions for calculations for GWC values. According to the Project Design Document for Gold Standard voluntary offset projects, baseline fuel consumption per household (n=252) in Balaka was on average 2.561 tonnes of fuelwood per annum. Fuel savings were determined from the average fuel consumption rate (in kg/hr) of the stoves studied in this work. The *Chitetezo Mbaula*, ACE and Philips reduced fuel usage by 26%, 27 % and 51 % respectively (Figure A5). Assuming a wood heating value of 15 MJ/kg, a daily energy use of 10.6 MJ per day per stove was calculated and set as a baseline for further calculations. The GWC is calculated by the equation:

$$GWC = \Sigma GWP_i \times GHG_i \tag{4}$$

where GWP_i is the 20 or 100 year global warming potential for each species (CO_2, CO, OC, BC/EC and CH_4) and GHG_i is the quantity of each species in terms of carbon equivalent. GWP values were taken from the IPCC 2013 report (IPCC, 2014) are listed in Table A3. LPG stoves were assumed to have a thermal efficiency of 53.6 % and a heating value of 45.8 MJ kg^{-1} (Grieshop et al. 2011)

For institutional stoves, it was assumed that the stove was used 2 times a day for 5 days a week for 40 weeks in a year. The amount of wood used for each cooking session was assumed to be the average weight based on this study. Figure A7 shows the 20 year GWC for both in-home and institutional stove tested during the study duration.

Section A3 PaRTED Analysis (Chen et al. 2012b)

The instantaneous scattering emission factor (IEF_{scat}) represents the amount of light scattering related to the particulate emissions emitted from the combustion of 1 kg of wood. The IEF_{scat} for each combustion event was estimated using the following equation:

$$IEF_{scat,i} = B_{sp,i}/C_{carbon,i}$$

Where:

$B_{sp,\,i}$ = scattering coefficient (Mm^{-1})

C_{carbon} = background corrected carbon concentration (ppm)

$IEF_{scat,i}$ = instantaneous scattering emission factor ($m^2\ kg^{-1}$wood)

Additional information on calculations can be found in the supporting information of (Chen et al. 2012b). The IEF_{scat} is a proxy for the relationship between light scattering and mass concentration during a combustion event based on the fuel usage. The measurement helps with the comparison of different light scattering and absorbing particle during different combustion events.

Table A1. Field Testing Summary

District	**Stove**	**No. of tests**	**Test Dates**	**Collaborating Partner**
Chikhwawa	Traditional	7	3 - 14 Sep 2015	CAPS Study: Liverpool School of Tropical Medicine
	ACE-1	8		
	Philips	8		
Balaka	Traditional	6	16 - 25 Sep 2015	Concern Universal
	*Chitetezo Mbaul*a	16		
Blantyre	Three Stone Fire	3	29 Sep – 3 Oct 2015	Clioma/Hestian/ Christa Roth
	Mayankho	3		
	JumboZama	3		
	Aleva	1		

Table A2. Temperature protocol for OC/EC analysis, Sunset Laboratory Analyzer

Mode	**Time (s)**	**Temperature (°C)**	**Power Constant**	**Time Constant (s)**	**Blower Mode**
Helium	10	1	0.001	100	0
Helium	-1	200	0.055	85	0
Helium	-1	310	0.055	85	0
Helium	-1	475	0.095	75	0
Helium	-1	615	0.15	45	0
Helium	-1	700	0.3	35	0
Helium	-1	550	0.001	100	16
Oxygen	90	550	0.18	65	0
Oxygen	90	625	0.18	42	0
Oxygen	90	700	0.2	36	0
Oxygen	90	775	0.27	32	0
Oxygen	90	850	0.25	25	0
Oxygen	-1	870	0.3	20	0
CalibrationOx	120	1	0.001	100	16
Offline	1	0	0.001	100	16

Note: Time of -1 indicates that the FID should return to baseline.

Table A3. 20 and 100 year Global Warming Potential Values used for this study

Species	**GWP 20 year**	**GWP 100 year**
CO_2	1	1
CO	5.9	1.9
BC/EC	2421.1	658.6
OC	-244.1	-66.4
CH_4	83.9	28.5

Table A4. Summary of Emission Factors and Optical properties for in-home emissions testing.

Test ID	Stove	PM EF	CO EF	EC EF	OC EF	EC/TC	MAC	MSC	B_{ap} EF
		g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	m^2 g^{-1}	m^2 g^{-1}	m^{2}kg^{-1}
1	Traditional Mud Stove	6.1 ± 0.4	70 ± 5	0.78 ± 0.10	1.26 ± 0.21	0.32 ± 0.05	n/a	0.63 ± 0.26	n/a
3	ACE	8.5 ± 0.9	110 ± 12	1.43 ± 0.23	1.06 ± 0.21	0.36 ± 0.07	n/a	0.80 ± 0.33	n/a
4	Philips	3.2 ± 0.4	52 ± 8	0.65 ± 0.11	0.81 ± 0.18	0.45 ± 0.10	n/a	0.48 ± 0.20	n/a
5	3SF	3.4 ± 0.4	72 ± 8	0.90 ± 0.14	1.64 ± 0.33	0.28 ± 0.06	5.2 ± 1.0	0.89 ± 0.37	4.6 ± 0.5
6	3SF	11.0 ± 2.3	148 ± 31	0.50 ± 0.12	4.72 ± 1.48	0.13 ± 0.04	9.9 ± 3.2	1.08 ± 0.49	4.9 ± 1.0
8	ACE	2.8 ± 0.3	30 ± 3	0.38 ± 0.06	0.39 ± 0.07	0.61 ± 0.11	17.0 ± 2.9	0.56 ± 0.23	6.4 ± 0.5
9	ACE	6.8 ± 0.8	57 ± 7	1.09 ± 0.17	1.88 ± 0.82	0.33 ± 0.15	13.7 ± 2.6	0.96 ± 0.40	14.9 ± 1.6
10	ACE	7.3 ± 0.8	40 ± 5	2.69 ± 0.41	0.28 ± 0.06	0.69 ± 0.14	5.6 ± 1.0	0.30 ± 0.13	15.0 ± 1.5
11	ACE	6.5 ± 0.6	32 ± 4	0.65 ± 0.10	1.11 ± 0.21	0.36 ± 0.07	13.3 ± 2.4	0.23 ± 0.10	8.6 ± 0.7
12	ACE	3.5 ±	53 ±	1.29 ±	0.93 ±	0.40 ±	11.4 ±	0.50 ±	14.7 ±

		0.4	6	0.19	0.17	0.08	2.0	0.21	1.3
13	ACE	9.0 ± 1.0	83 ± 10	1.55 ± 0.40	3.30 ± 0.67	0.16 ± 0.05	13.9 ± 3.9	1.68 ± 0.70	21.6 ± 2.3
14	Philips	5.0 ± 0.5	98 ± 9	1.23 ± 0.18	0.93 ± 0.18	0.41 ± 0.08	13.1 ± 2.3	0.55 ± 0.23	16.1 ± 1.4
15	Philips	4.2 ± 0.6	28 ± 5	0.53 ± 0.10	1.13 ± 0.25	0.37 ± 0.09	15.0 ± 3.4	1.06 ± 0.45	7.9 ± 1.0
16	Philips	4.2 ± 0.3	68 ± 5	0.83 ± 0.12	0.99 ± 0.17	0.38 ± 0.07	13.3 ± 2.1	0.72 ± 0.29	11.0 ± 0.8
17	Philips	3.5 ± 0.4	54 ± 5	0.59 ± 0.09	0.80 ± 0.16	0.45 ± 0.09	14.2 ± 2.5	0.57 ± 0.23	8.3 ± 0.7
18	3SF	14.7 ± 1.6	128 ± 14	0.39 ± 0.06	6.96 ± 1.37	0.08 ± 0.02	23.0 ± 4.5	1.12 ± 0.46	8.9 ± 0.9
19	Philips	3.9 ± 0.4	38 ± 5	0.95 ± 0.15	0.97 ± 0.19	0.40 ± 0.08	12.9 ± 2.4	0.75 ± 0.31	12.4 ± 1.2
20	3SF	9.6 ± 1.0	96 ± 11	0.59 ± 0.10	4.01 ± 0.82	0.14 ± 0.03	21.4 ± 4.2	1.25 ± 0.52	12.5 ± 1.3
21	Philips	4.8 ± 0.5	46 ± 6	1.09 ± 0.17	1.11 ± 0.22	0.37 ± 0.07	13.6 ± 2.5	0.94 ± 0.39	14.9 ± 1.5
22	Traditional Mud Stove	9.8 ± 0.9	136 ± 12	3.25 ± 0.47	2.71 ± 0.50	0.19 ± 0.04	7.2 ± 1.2	1.21 ± 0.50	23.3 ± 2.1
23	Traditional Mud Stove	7.1 ± 0.6	83 ± 8	2.17 ± 0.31	2.30 ± 0.43	0.22 ± 0.04	13.3 ± 2.2	0.93 ± 0.38	28.7 ± 2.4
25	Philips	4.3 ± 0.5	46 ± 6	0.62 ± 0.10	0.97 ± 0.21	0.41 ± 0.09	16.1 ± 3.1	0.48 ± 0.20	10.0 ± 1.1
26	ACE	4.6 ± 0.7	46 ± 7	1.29 ± 0.23	1.22 ± 0.28	0.35 ± 0.08	14.1 ± 3.0	1.65 ± 0.70	18.2 ± 2.3
27	CM	6.9 ± 0.6	108 ± 10	0.32 ± 0.05	0.78 ± 0.14	0.44 ± 0.08	16.2 ± 2.8	0.84 ± 0.35	5.1 ± 0.5
28	CM	3.7 ± 0.5	93 ± 13	1.43 ± 0.24	4.64 ± 1.03	0.12 ± 0.03	6.3 ± 1.3	1.17 ± 0.49	9.0 ± 1.1
29	CM	14.7 ± 1.0	156 ± 11	1.29 ± 0.17	6.67 ± 1.09	0.08 ± 0.01	0.0 ± 0.0	1.41 ± 0.57	26.5 ± 1.9
30	CM	8.0 ± 0.7	93 ± 9	2.06 ± 0.30	2.06 ± 0.38	0.23 ± 0.04	7.2 ± 1.2	1.08 ± 0.44	14.9 ± 1.3
31	CM	6.1 ± 0.7	117 ± 15	1.96 ± 0.32	2.07 ± 0.45	0.24 ± 0.05	30.6 ± 6.2	1.08 ± 0.45	59.9 ± 7.0
32	CM	6.7 ± 0.8	103 ± 13	2.40 ± 0.40	2.75 ± 0.60	0.19 ± 0.04	7.7 ± 1.6	0.94 ± 0.39	18.6 ± 2.2
33	CM	4.6 ± 0.5	89 ± 10	0.89 ± 0.14	1.62 ± 0.32	0.28 ± 0.06	12.1 ± 2.3	0.72 ± 0.30	10.8 ± 1.1
34	CM	8.1 ± 0.8	97 ± 10	2.78 ± 0.42	2.78 ± 1.13	0.25 ± 0.10	10.5 ± 1.9	1.04 ± 0.43	29.3 ± 2.9

35	CM	4.5 ± 0.5	78 ± 9	1.56 ± 0.25	0.86 ± 0.17	0.42 ± 0.09	11.8 ± 2.2	0.68 ± 0.28	18.4 ± 1.9
36	CM	3.6 ± 0.5	68 ± 9	1.38 ± 0.23	0.74 ± 0.16	0.46 ± 0.10	12.6 ± 2.6	0.63 ± 0.27	17.5 ± 2.1
37	CM	4.6 ± 0.5	73 ± 9	2.14 ± 0.34	0.77 ± 0.16	0.45 ± 0.09	9.9 ± 1.9	0.67 ± 0.28	21.3 ± 2.3
38	CM	5.7 ± 0.6	78 ± 8	1.23 ± 0.18	2.06 ± 0.40	0.23 ± 0.05	11.8 ± 2.1	0.79 ± 0.33	14.5 ± 1.4
39	CM	8.9 ± 0.8	177 ± 16	1.45 ± 0.21	3.66 ± 0.67	0.14 ± 0.03	12.6 ± 2.2	1.10 ± 0.45	18.2 ± 1.6
40	CM	6.1 ± 0.5	83 ± 7	1.20 ± 0.17	2.21 ± 0.39	0.22 ± 0.04	10.2 ± 1.7	1.08 ± 0.44	12.3 ± 1.0
41	CM	11.1 ± 0.9	199 ± 15	1.32 ± 0.18	3.77 ± 0.65	0.14 ± 0.02	12.7 ± 2.0	1.13 ± 0.46	16.8 ± 1.3
43	CM	4.5 ± 0.4	83 ± 8	0.81 ± 0.12	1.44 ± 0.27	0.29 ± 0.06	11.3 ± 2.0	0.82 ± 0.34	9.2 ± 0.8
44	3SF	6.3 ± 0.6	92 ± 9	0.53 ± 0.08	2.67 ± 0.50	0.19 ± 0.04	14.6 ± 2.6	0.96 ± 0.40	7.8 ± 0.7
45	3SF	6.3 ± 0.7	94 ± 11	0.74 ± 0.12	2.30 ± 0.47	0.22 ± 0.05	13.6 ± 2.7	0.83 ± 0.34	10.0 ± 1.1
46	3SF	6.3 ± 0.6	108 ± 10	0.91 ± 0.13	1.90 ± 0.35	0.25 ± 0.05	15.1 ± 2.6	0.65 ± 0.27	13.8 ± 1.2
48	3SF	5.7 ± 0.5	74 ± 7	0.74 ± 0.11	2.18 ± 0.39	0.22 ± 0.04	20.1 ± 3.4	0.66 ± 0.27	14.9 ± 1.2
49	3SF	7.4 ± 0.6	93 ± 8	0.86 ± 0.12	5.21 ± 0.91	0.10 ± 0.02	14.5 ± 2.4	0.79 ± 0.32	12.4 ± 1.0
50	3SF	7.4 ± 1.0	81 ± 12	n/a	n/a	n/a	n/a	0.77 ± 0.33	12.6 ± 1.7
51	Mayankho	2.3 ± 0.2	28 ± 3	0.94 ± 0.12	0.36 ± 0.06	0.64 ± 0.11	6.9 ± 1.0	0.81 ± 0.33	6.5 ± 0.5
52	TSF	6.6 ± 0.7	98 ± 12	0.82 ± 0.12	2.45 ± 0.51	0.21 ± 0.04	12.6 ± 2.3	0.73 ± 0.31	10.3 ± 1.2
53	Mayankho	1.8 ± 0.1	22 ± 2	0.77 ± 0.10	0.24 ± 0.04	0.72 ± 0.12	7.0 ± 1.0	0.34 ± 0.14	5.4 ± 0.4
54	TSF	6.2 ± 0.6	101 ± 10	0.48 ± 0.06	2.28 ± 0.43	0.22 ± 0.04	16.1 ± 2.6	0.50 ± 0.21	7.7 ± 0.7
55	TSF	8.6 ± 0.8	115 ± 11	0.44 ± 0.06	3.56 ± 0.66	0.15 ± 0.03	18.3 ± 3.0	0.83 ± 0.34	8.1 ± 0.7
56	JZ	2.1 ± 0.2	24 ± 4	0.22 ± 0.03	0.64 ± 0.12	0.50 ± 0.10	13.6 ± 2.4	0.18 ± 0.07	3.0 ± 0.3
57	Mayankho	3.0 ± 0.3	51 ± 5	0.49 ± 0.06	0.94 ± 0.17	0.40 ± 0.07	12.1 ± 1.8	0.70 ± 0.29	6.0 ± 0.5
58	Aleva	3.5 ±	43 ±	0.35 ±	1.07 ±	0.36 ±	14.2 ±	0.49 ±	4.9 ±

		0.3	4	0.04	0.19	0.06	2.1	0.20	0.4
59	JZ	2.1 ± 0.2	18 ± 2	0.90 ± 0.12	0.36 ± 0.07	0.63 ± 0.11	7.9 ± 1.2	0.24 ± 0.10	7.1 ± 0.6
60	JZ	1.2 ± 0.1	43 ± 4	0.37 ± 0.04	0.60 ± 0.10	0.51 ± 0.08	12.3 ± 1.8	0.61 ± 0.25	4.5 ± 0.3

Table A5. Summary of food and fuel emission factors and fuel use for Institutional Stoves

		Food Based EF		
Test ID	Stove	PM EF g kg^{-1}	CO EF g kg^{-1}	EC EF g kg^{-1}
51	Mayankho	0.16 ± 0.01	2.0 ± 0.2	0.066 ± 0.009
52	TSF	0.42 ± 0.02	6.3 ± 0.9	0.053 ± 0.008
53	Mayankho	0.11 ± 0.01	1.4 ± 0.2	0.049 ± 0.007
54	TSF	0.52 ± 0.03	8.5 ± 1.2	0.040 ± 0.006
55	TSF	0.65 ± 0.03	8.8 ± 1.3	0.033 ± 0.005
56	JZ	0.12 ± 0.01	1.3 ± 0.2	0.012 ± 0.002
57	Mayankho	0.19 ± 0.01	3.3 ± 0.3	0.032 ± 0.004
58	Aleva	0.38 ± 0.02	4.7 ± 0.6	0.038 ± 0.005
59	JZ	0.09 ± 0.01	0.8 ± 0.1	0.038 ± 0.006
60	JZ	0.06 ± 0.00	2.4 ± 0.2	0.020 ± 0.003

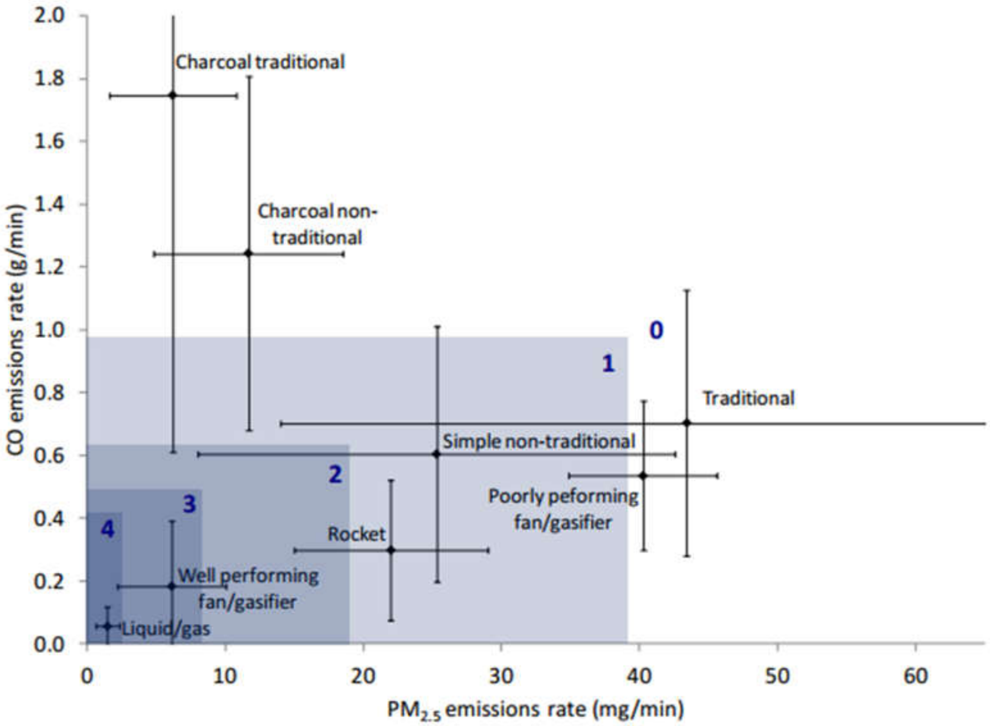

Figure A1. Taken from (http://cleancookstoves.org/resources_files/stove-performance-inventory-pdf.pdf) Indoor emissions performance for key stove/fuel classes across the IWA Tiers for the WBT. Error bars represent ± one standard deviation of the available tests sets. Stove/fuel classes with no error bars consist of two or less data points. Tiers are indicated by blue numbers.

Figure A2. Improved stove technologies tested in the Malawi field campaign. L to R: Philips, ACE-1 and *Chitetezo Mbaula*. Pictures by A. Grieshop and P. Jagger (*Chitetezo Mbaula*).

Figure A3. Institutional stoves. (L to R) Aleva, Mayankho, JumboZama

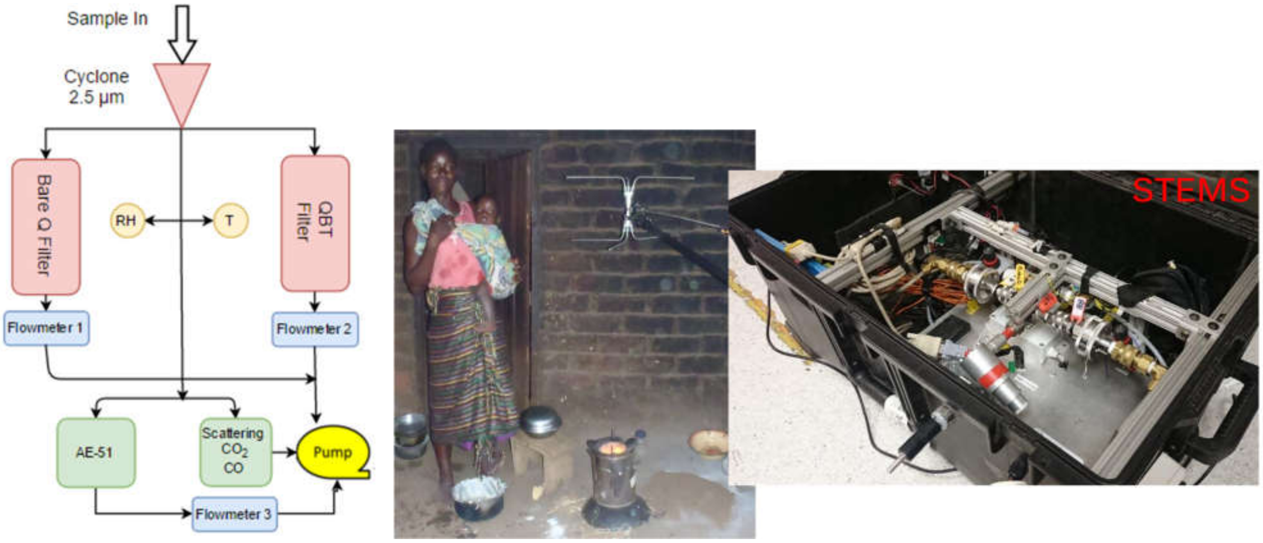

Figure A4. STEMS-1G Schematic and setup in the field. Each of the flowmeters have valves downstream for flow control (not shown).

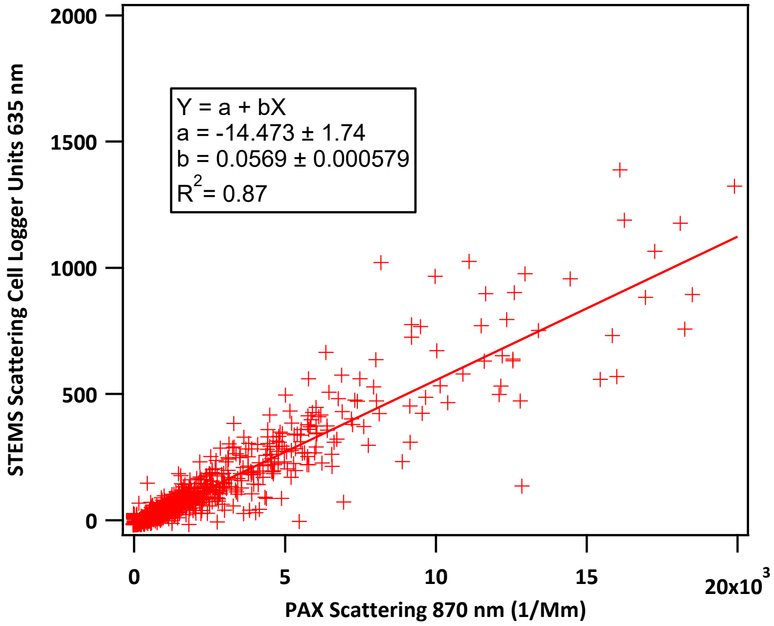

Figure A5. Calibration of the STEMS scattering cell against the PAX.

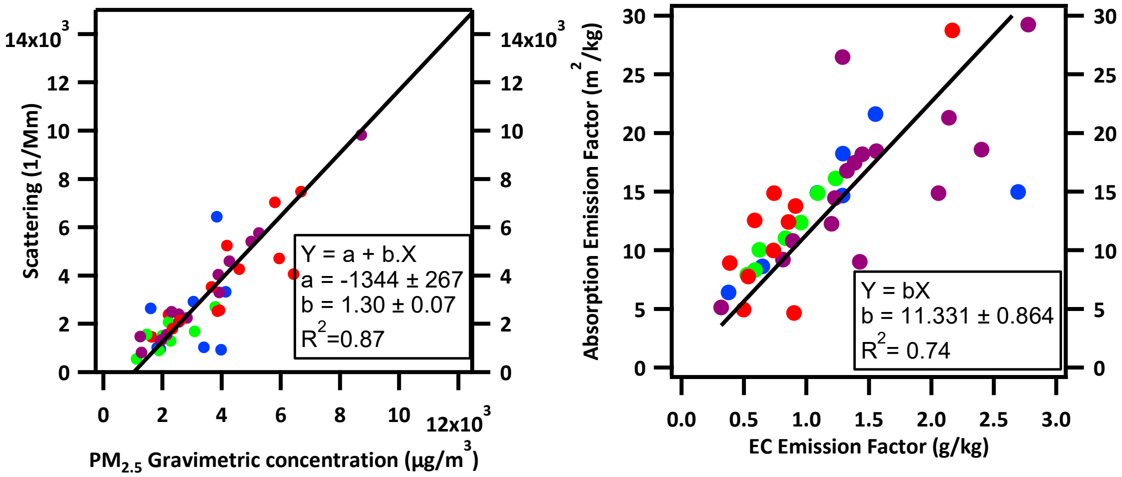

Figure A6. L: Relationship between averaged scattering and gravimetric PM concentrations. R: Relationship between absorption and EC emission factor. The slope is the averaged mass absorption cross section (MAC). The color of the data points are same as the colors used for the respective stoves in the box and whisker plots in previous figures.

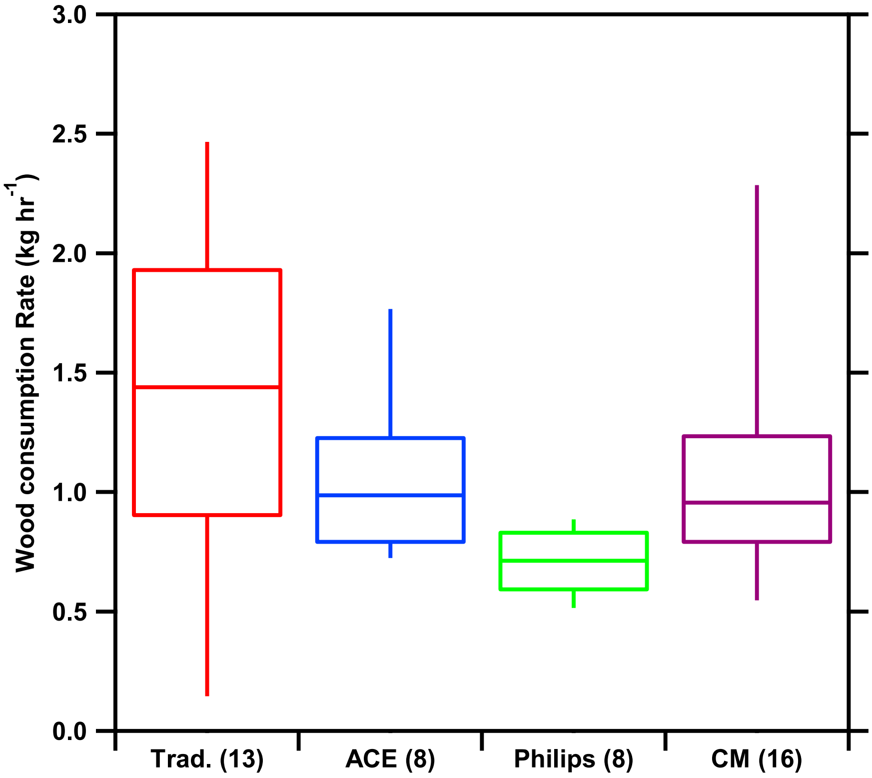

Figure A7. Box plot of fuel consumption in kg per hour for a single cooking session. Wood consumption rate is determined by dividing the wood used in a cooking task by the time required for cooking. Wood weight is not corrected for moisture. Background periods are not included in this calculation.

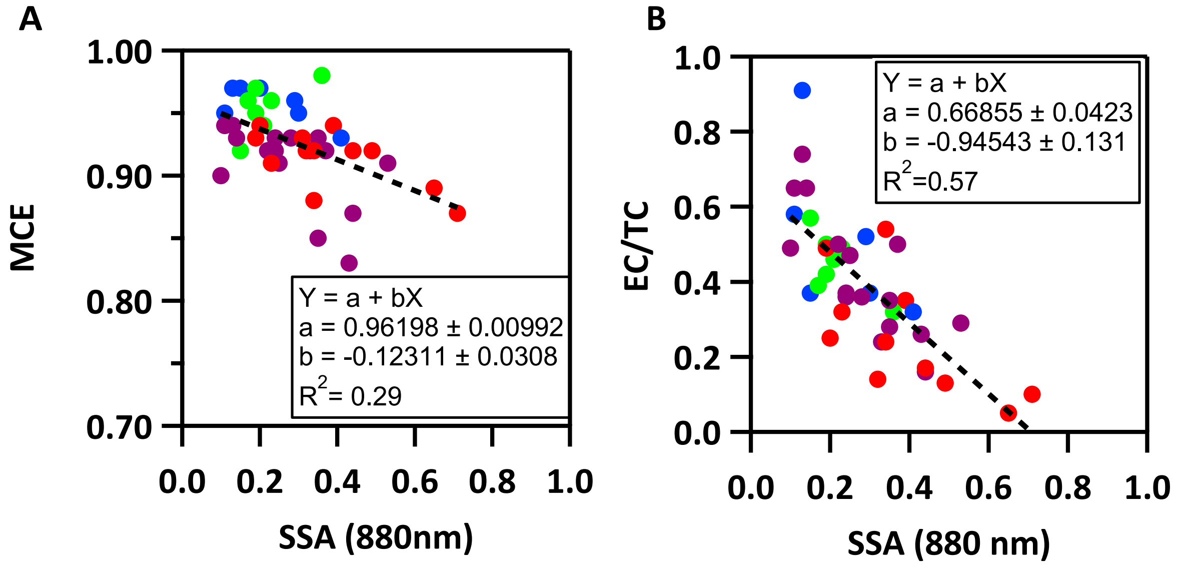

Figure A8. Relationship between A. SSA and MCE and B. SSA and EC for in-home cookstoves. The color of the data points are same as the colors used for the respective stoves in the box and whisker plots in previous figures.

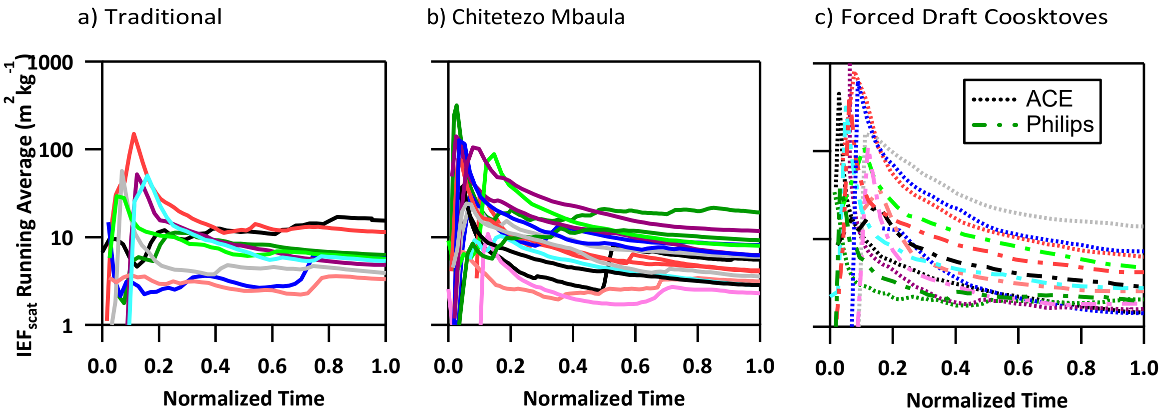

Figure A9 Running average of IEF_{scat} for different stove types normalized by the cooking time. Note that cooking time is different and shorter than the total sampling time; the latter includes background measurements. Each trace color indicates emissions from a single cooking session.

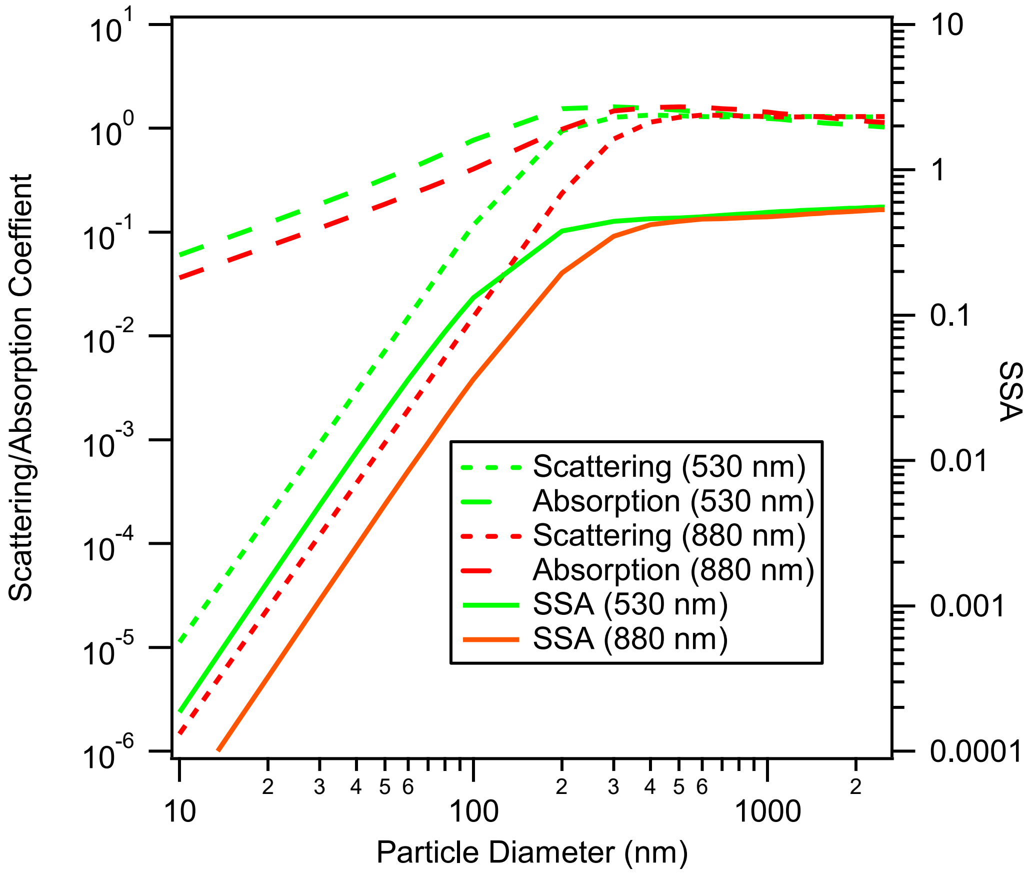

Figure A10. Dependence of optical properties: Scattering, Absorption (left axis) and SSA (right axis) with particle size and wavelength for pure BC aerosol on a logarithmic scale. Refractive index for BC used was 1.85 + 0.71i (Bond and Bergstrom 2006).

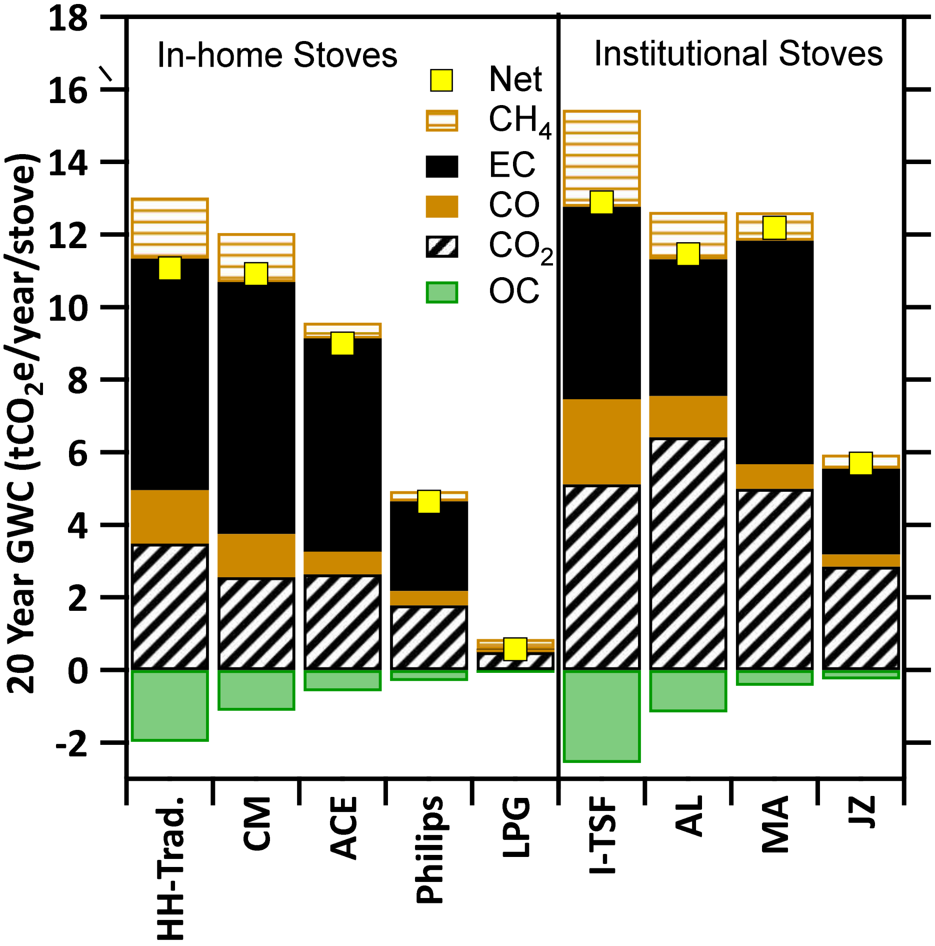

Figure A11 20 year GWC values for in-home and institutional stoves. 100 year GWC values for one year of use of in-home stoves (L) and institutional stoves (R) from major short- and long-lived climate forcing species emitted by stoves. CH_4 component is estimated based on the ratio of CH_4 and CO emission factors from other sources. Note that the daily energy use is different for institutional stoves. In-home stoves are assumed to be used every day of the year. Institutional stoves are assumed to be used for 5 days a week for 40 weeks a year.