DISSERTATION

RESIDENTIAL COOKSTOVE EMISSIONS: MEASUREMENT AND MODELING FROM THE LAB AND FIELD

Submitted by

Kelsey Bilsback

Department of Mechanical Engineering

In partial fulfillment of the requirements

For the Degree of Doctor of Philosophy

Colorado State University

Fort Collins, Colorado

Fall 2018

Doctoral Committee:

Advisor: Dr. John Volckens

Elizabeth Barnes
Shantanu Jathar
Anthony Marchese
Jeffrey Pierce

ABSTRACT

RESIDENTIAL COOKSTOVE EMISSIONS: MEASUREMENT AND MODELING FROM THE LAB AND FIELD

Emissions from solid-fuel cookstoves, which result from poorly controlled combustion, have been linked to indoor and outdoor air pollution, climate forcing, and human disease. The adverse effects of cookstoves have motivated commitment of substantial time and resources towards development of "improved" cookstoves that operate more efficiently and reduce emissions of harmful air pollutants. However, once disseminated to cookstove users, improved cookstoves often do not ameliorate air quality to a level that substantially reduces health risks or negative environmental impacts. Several critical knowledge gaps related to the emissions and performance of "improved" cookstoves exist; attempting to address these gaps is the subject of this dissertation.

Widely-used laboratory testing protocols overestimate the ability of improved stoves to lower emissions. In this work, we develop and validated a novel laboratory test protocol entitled the Firepower Sweep Test. We find that the Firepower Sweep Test reproduces the range of $PM_{2.5}$ and CO emissions observed in the field, including high emissions events not typically observed under current laboratory protocols. We also find that firepower is modestly correlated with emissions, although this relationship depends on stove-fuel combination. Our results justify incorporating multiple-firepower testing into laboratory-based protocols, but demonstrate that firepower alone cannot explain the observed variability in cookstove emissions.

Cookstoves emit many pollutants; however, most studies only measure fine particulate matter ($PM_{2.5}$) and carbon monoxide (CO). In this work, we present an extensive inventory of air pollutants emitted from wood, charcoal, kerosene, and liquefied petroleum gas (LPG) cookstoves. One-hundred and twenty pollutants, including $PM_{2.5}$, CO, organic matter, elemental carbon, inorganic ions, carbohydrates, ultrafine particles, volatile organic compounds, carbonyls, and polycyclic aromatic hydrocarbons, are included in this inventory. Our results demonstrate that, while most improved stoves tend to reduce $PM_{2.5}$ and/or CO emissions, reductions $PM_{2.5}$ and/or CO emissions do not always correspond to reductions of other harmful pollutants. These findings highlight the need to characterize the full emissions profile of "improved" cookstove designs before they are disseminated to users.

Accurate emissions data are critical inputs for models that aim to quantify the impacts of cookstoves on climate and health. Currently, model inputs are primarily derived from laboratory experiments that do not represent in-home use. In this work, we present a relatively inexpensive technique that uses a temperature measurement made at the combustion chamber outlet to estimate firepower. These firepower estimations have the potential to provide valuable information about the range of firepowers over which cookstoves are operated at during real-world use. We also demonstrate that in-use firepower measurements from "improved" cookstoves can be combined with laboratory emissions data from the Firepower Sweep Test to estimate in-use emissions using linear regression models. We find that the model predictions are accurate enough to determine which International Standards Organization emissions tier a given "improved" stove is likely to fall under.

ACKNOWLEDGEMENTS

First and foremost, thank you to my advisor, John Volckens. Thank you for your patience as I wrote (and rewrote) my first manuscript; your personal and professional advice; your unwavering support during times of struggle; and your willingness to let me travel across the globe to collect cookstove emissions data. Thank you for your incredible mentorship in technical research and communication over the last five years. I have cherished the opportunity to work for you. Thank you to my dissertation committee—Elizabeth Barnes, Shantanu Jathar, Anthony Marchese, and Jeffrey Pierce—for your time, insightful feedback, and expertise.

Thank you to the amazing EPA project team, including Christian L'Orange, Jack Kodros, Jeff Pierce, Michael Johnson, Rose Eilenberg, Eric Lipsky, Allen Robinson, and Subu. Thank you for your comments on the many, many drafts of my firepower sweep paper. Thank you to Jessica Tryner, James Tillotson, Kevin Dischino, Kelly Hixson, Brooke Reynolds, James Liacos, Nelson Salvatore, the Mizia Research Group, and the CSU Control Room Team for providing additional technical support and assistance during the Cookstove Shootout.

Thank you to Christian L'Orange and Michael Johnson for your leadership and assistance during the inevitable stress-inducing instrument failures of field work. Thank you to the incredible field personnel, including Jack Kodros, Rose Eilenberg, Eric Lipsky, Brooke Reynolds, Megan Benka-Coker, and Kristen Fedak. Thank you to all of the local hosts and field teams at the Center for Research in Energy and Energy Conversion, Kampala University, Tsinghua University, and in Honduras (i.e., Maggie Clark's research group). Thank you to all of the individuals who opened your homes to our team during the field campaign; I will be forever grateful for your hospitality.

Thank you to Ethan Walker, Lizette van Zyl, and Jordan Dahlke who did the majority of the grunt work for the NIH emissions testing project. Thank you Christian L'Orange for your technical support and Kristen Fedak for project management. Thank you Nicholas Good for spearheading the data analysis and illustrating what "tidy" R code should look like. Thank you Ander Wilson and Lauren Heck for your technical expertise and answering my endless statistics-related questions. Thank you John Mehaffy, Amy Sullivan, Arsineh Hecobian, Pierre Herckes and Yong Zhou for conducting the emissions sample analysis; Ander Wilson for answering endless questions about statistics. Thank you Jennifer Peel for your leadership and guidance.

Thank you to the rest of the Volckens research group including Eric Wendt, Casey Quinn, Rebecca Foos. Thank you Christian L'Orange for answering my endless questions throughout the last five years. Thank you Jessica Tryner for reading many, many draft of my writing and your motivational words.

Thank you to the wonderful community at the Powerhouse Energy Campus and all the incredible people I have met working there, especially Jonas Adler, Torben Grumstrump, Colin Gould, and Ali Akherti. Thank you to my unofficial mentors, especially Ellison Carter and Sheryl Magzamen.

The work presented here was enabled through funding by the U.S. Environmental Protection Agency (RD83543801), National Institutes of Health (ES023688), the Department of Mechanical Engineering Graduate Teaching Assistantship, and the College of Engineering Graduate Teaching Fellowship (GTF). Thank you to Sam Bechara and Tammy Donahue for being amazing mentors during my GTF and Kimberly Catton for organizing the GTF experience.

Finally, thank you to my mother, father, Evan, Jenna, and Kevin. You have been my unwavering support team. I could not have accomplished any of this work without you.

TABLE OF CONTENTS

CHAPTER 1: INTRODUCTION AND BACKGROUND 1

1.1 HEALTH AND ENVIRONMENTAL EFFECTS OF HOUSEHOLD COOKSTOVES 1
1.2 "IMPROVED" COOKSTOVES 2
1.3 LABORATORY TESTING PROTOCOLS FOR HOUSEHOLD COOKSTOVES 2
1.4 EMISSIONS CHARACTERIZATION OF HOUSEHOLD COOKSTOVES 3
1.5 EMISSIONS ESTIMATES FOR HOUSEHOLD COOKSTOVES 4
1.6 OVERVIEW AND OBJECTIVES OF THIS WORK 4

CHAPTER 2: THE FIREPOWER SWEEP TEST: A NOVEL APPROACH TO COOKSTOVE LABORATORY TESTING 6

2.1 INTRODUCTION 6
2.2 METHODS 9
2.2.1 Testing protocol 9
2.2.2 Stove-Fuel Technologies 12
2.2.3 Instrumentation 12
2.2.4 Data cleaning and performance metrics 13
2.2.5 Correlation analysis 15
2.2.6 Linear models 16
2.3 RESULTS AND DISCUSSION 17
2.3.1 FST and WBT operating conditions 17
2.3.2 FST and WBT emissions 21
2.3.3 FST, WBT, and field emissions 24
2.3.4 Firepower and emissions 26
2.3.5 Indoor Air Quality Ramifications 29
2.3.6 The ISO multiple-firepower test 30
2.3.7 Limitations and Future Work 31
2.3.4 CONCLUSIONS 33

CHAPTER 3: A COMPREHENSIVE LABORATORY ASSESSMENT OF AIR POLLUTANT EMISSIONS FROM RESIDENTIAL COOKSTOVES 35

3.1 INTRODUCTION 35
3.2 MATERIALS AND METHODS 36
3.2.1 Test Matrix 36
3.2.2 Test Protocol 37
3.2.3 Emissions Measurements 38
3.2.4 Data Processing and Emissions Metrics 40
3.2.5 Regression Analysis 41
3.3 RESULTS AND DISCUSSION 42
3.3.1 Overview 42
3.3.2 $PM_{2.5}$ composition 43
3.3.3 Carbon monoxide 45
3.3.4 Ultrafine particles 46
3.3.5 Polycyclic aromatic hydrocarbons 47
3.3.6 Volatile organic compounds 48
3.3.7 Carbonyl compounds 50
3.3.8 Carcinogenic compounds 51
3.3.9 $PM_{2.5}$ and CO as predictors for co-emitted pollutants 52
3.3.10 Synthesis and implications for cookstove research 53

CHAPTER 4: COUPLING LABORATORY AND FIELD MEASUREMENTS TO ESTIMATE AIR POLLUTANT EMISSIONS FROM COOKSTOVES....56

4.1 INTRODUCTION 56
4.2 MATERIALS AND METHODS....58
4.2.1 Overview of field and laboratory campaigns....58
4.2.2 Firepower measurements....59
4.2.3 Emissions measurements 61
4.2.4 Modified combustion efficiency 61
4.2.5 Linear model development....62
4.2.6 Laboratory-to-field emissions estimation 62
4.3 RESULTS AND DISCUSSION 63
4.3.1 Temperature versus firepower in the laboratory....63
4.3.2 Operating conditions in the laboratory and field....67
4.3.3 The relationship between firepower and emissions in the laboratory and field....69
4.3.4 Predicting emissions during an in-field cooking event....71
4.3.5 Limitations and Future Work....74
4.5 CONCLUSIONS 74

CHAPTER 5: CONCLUSIONS AND PRACTICAL IMPLICATIONS....76

REFERENCES 78

APPENDIX A....85

APPENDIX B 106

APPENDIX C 110

CHAPTER 1: INTRODUCTION AND BACKGROUND

1.1 Health and Environmental Effects of Household Cookstoves

Forty percent of the global population, or about three billion people, rely on solid fuels (e.g., wood, charcoal, agricultural residue, and animal waste) as their primary energy source.[1] Solid fuels are burned in cooking technologies (i.e., cookstoves) that operate under poor combustion conditions and in poorly ventilated homes; these conditions result in high emissions of harmful air pollutants.

Previous research has linked cookstoves with fine particulate matter concentrations that are several times higher than World Health Organization's guideline for indoor air quality.[2,3] Household air pollution is one of the top-ten risk factors for death and disease on the planet.[4] Health outcomes that have been associated with solid fuel use include: upper and lower respiratory tract infections, chronic obstructive pulmonary disease, asthma, low birth weights, and cataracts.[5] The health effects of air pollution emitted from cookstoves are complicated by the fact that cookstove users may have poor nutrition, limited access to health care, and other risk factors associated with low-income countries.[6]

Particle- and gas-phase emissions from cookstoves also affect climate and local air quality; this is especially true in regions of widespread cookstove use such as sub-Saharan African, Latin America, and Southeast Asia.[7–9] For example, cookstoves emit greenhouse gases—including carbon dioxide and methane—that may have a net climate warming effect if the fuels being burned are not harvested sustainably. Aerosols also have effects on the atmosphere: scattering or absorbing radiation (i.e., the direct effect), changing cloud properties (i.e., the aerosol indirect and cloud-albedo effects), changing the absorption of snow (i.e., the snow-albedo effect), or changing the temperature profile of the atmosphere (i.e., the semi-direct effect).[10] Other species that are emitted, such as volatile organic compounds, may lead to atmospheric chemical reactions that affect the quantity and composition of aerosols (i.e., secondary organic aerosols) or greenhouse gases[11] (e.g., ozone) in the atmosphere.

1.2 "Improved" Cookstoves

Substantial time and resources have been invested to develop "improved" cookstoves that are engineered to operate more efficiently and to reduce emissions of air pollutants. Examples of engineering improvements include: insulating the combustion chamber to prevent heat loss and adding an electric fan to improve fuel-air mixing. Improved cookstove technologies have shown promise in laboratory settings. However, once disseminated they often do not result in emissions reductions that are below the World Health Organization's recommendations clean indoor air.[3,12] Several critical knowledge gaps related to cookstove emissions and performance still exist; addressing these matters is the subject of this work.

1.3 Laboratory Testing Protocols for Household Cookstoves

Cookstove emissions testing is primarily conducted in controlled laboratory settings due to the costs and logistical challenges of conducting tests in a cookstove user's home. The Water Boiling Test[13] is the most established and commonly-used laboratory testing protocol. During the test, emissions, fuel use, and thermal efficiency are measured while a pot of water is brought to a boil twice and then simmered. The Water Boiling Test was designed to reduce test-to-test variability so that engineers could isolate which cookstove design component might reduce emissions; however, several studies have shown that the Water Boiling Test greatly underpredicts both the magnitude and variability of emissions seen during in-home testing.

The primary shortcoming of the Water Boiling Test is that it only simulates emissions during two types of cooking tasks that do not represent the diversity of cooking behaviors seen throughout the world. The protocol is also ambiguous, which has led to stoves that are optimized to perform well only under the Water Boiling Test conditions and to discrepancies in how the protocol is applied, making it difficult to compare performance between different stove types. More robust laboratory protocols are needed that address the gaps between cookstove performance during laboratory testing and in-home use, if laboratory-derived data are to be used to reliably provide information about in-home cookstove performance.

1.4 Emissions Characterization of Household Cookstoves

Cookstoves often operate under poor combustion conditions involving low flame temperatures, poor mixing dynamics, and localized fuel-rich or fuel-lean regions in the combustion chamber. The resulting emissions are composed of thousands of oxidized and partially-oxidized organic and inorganic constituents that depend on stove type, fuel type, and operating conditions.

Many of the constituents of cookstove smoke have been associated with adverse effects on human health[14,15] and the environment;[7,11] however, the majority of cookstove emissions studies conducted to date have only quantified fine particulate matter and carbon monoxide emissions.[16–18] Historically, emissions studies have primarily focused on these two constituents of cookstove smoke, because they are the only two pollutants with established guidelines for cookstoves.[19–21] Although fine particulate matter and carbon monoxide are important, when considering the health and environmental impacts of cookstoves, they do not give the full picture of the effects of cookstove smoke. More comprehensive datasets of cookstove emissions are needed to improve our understanding of how cookstoves might affect public health and the environment.

1.5 Emissions Estimates for Household Cookstoves

Accurate emissions data are critical to quantifying the impact of cookstoves on climate, air quality, and health.[7,8,22] Despite the fact that current laboratory emissions inventories do not represent in-home cookstove performance, these data are used to benchmark cookstove performance,[21] estimate indoor and outdoor air pollution concentrations,[7,22] choose treatment options for epidemiological studies,[23] and select stoves for large-scale control trials.[24]

Compared to laboratory testing, field campaigns provide realistic emissions data but are logistically intensive and often cost prohibitive. The expenses associated with field campaigns often limit both the number of measurements taken within a household and the number of households than can be monitored; these data likely provide a limited picture of cookstove performance given that cooking practices may vary within a single household or between households in the same region. Statistical tools that link laboratory and field emissions data have the potential to provide better emissions estimates and insights into why laboratory and field emissions data do not always agree.

1.6 Overview and Objectives of this Work

This work presents three novel contributions for reducing the uncertainties surrounding cookstove emissions. Chapter 2 introduces and provides validation for a novel laboratory emissions testing procedure, called the Firepower Sweep Test, which was designed to provide a more robust estimate of cookstove emissions performance in the laboratory than current standardized procedures. Chapter 3 presents one of the largest cookstove emissions inventories to date including one-hundred and twenty cookstove smoke constituents measured from twenty-six stove-fuel combinations. Chapter 3 also presents a linear regression analysis to explore whether $PM_{2.5}$ and CO are correlated with of other co-emitted pollutants. In Chapter 4, we present statistical tools that combine laboratory emissions data and stove-usage patterns from in-home cooking practices that can be used to better estimate field emissions data.

CHAPTER 2: THE FIREPOWER SWEEP TEST: A NOVEL APPROACH TO COOKSTOVE LABORATORY TESTING[1]

2.1 Introduction

An estimated forty-one percent of the world's population relies on energy from solid-fuel cookstoves to prepare their daily meals.[1] Household air pollution emitted by these stoves leads to poor indoor air quality and is ultimately an environmental risk factor for global morbidity and mortality. Additionally, particulate matter and greenhouse gas emissions from cookstoves have been linked to anthropogenic climate forcing and reduced outdoor air quality.[25,26]

Over the past several decades, "improved" cookstoves have been engineered to increase heat transfer efficiency and lower emissions. These improved stoves often show promise when tested in a laboratory setting; however, once disseminated, the same improved stoves may not perform substantially better than traditional stove designs.[3] Failure of improved cookstoves to reduce emissions, as well as other issues such as the continued use of traditional stoves alongside improved stoves (i.e., stove stacking), lead to persistent poor indoor air quality in regions where solid-fuel use is prevalent.

The Water Boiling Test (WBT)[27] is the most established and commonly-used laboratory protocol for evaluating cookstove performance. WBT data have been used to simulate indoor and outdoor air pollution levels,[28] choose treatment options for epidemiological studies, select stoves for large-scale randomized control trials,[24] and to rate stoves relative to published performance guidelines,[21] even though the scientific community widely acknowledges that this test overpredicts the ability of improved stoves to lower emissions and ameliorate indoor air

---

[1] This chapter was published as a manuscript: Bilsback, K. R.; Eilenberg, S. R.; Good, N.; Heck, L.; Johnson, M.; Kodros, J. K.; Lipsky, E. M.; L'Orange, C.; Pierce, J. R.; Robinson, A. L.; Subramanian, R; Tryner, J; Wilson, A; Volckens, J. The Firepower Sweep Test: A Novel Approach to Cookstove Laboratory Testing. *Indoor Air* **2018**.

quality.[27,29–32] The WBT protocol was initially developed to reduce test-to-test variability, so that engineers could better observe how small variations in stove design might lead to changes in emissions, thermal efficiency, or fuel consumption rates.[13] Several protocols that resemble the WBT exist[13,33,34], all with the objective of measuring thermal efficiency, fuel usage, and the emissions produced while a pot of water is brought to a boil twice and subsequently simmered.

There are several reasons why emissions measured during the WBT often do not represent emissions from real-world cooking. Primarily, the WBT only simulates the emissions during two tasks (boiling and simmering a pot of water), which do not represent the diversity of cooking practices used throughout the world. The WBT protocol also gives little direction as to how a stove should be run during testing. This ambiguity, which provides an opening for testers to tweak the protocol, has led to: (a) stoves optimized to perform well only under the WBT protocol and (b) adjustments to the protocol that cause a specific stove design to perform well. Additionally, the WBT does not provide guidelines for isolating and characterizing operating conditions such as start-up, shut-down, or refueling. Instead, emissions are averaged across a single cooking task, and the influence of specific operating conditions on the overall emissions can be difficult to discern. A more ideal test protocol would be (1) generalizable, meaning the results would be informative regardless of local cooking practices, (2) unambiguous, making it difficult to "cheat the system", and (3) transparent, allowing stakeholders to easily identify operating conditions that lead to high-emissions events.

Several research groups have proposed measuring emissions at different operating firepowers rather than measuring emissions during a specific cooking task. Prasad et al.[35] published a protocol designed to vary stove firepower by feeding a cookstove several batches of fuel, at different intervals, while measuring the thermal energy transferred to a pot full of water.

Later, Johnson et al.[36] proposed a 'burn cycle' approach, which involved measuring the combustion efficiency and carbon emission rate (a proxy for firepower) from stoves in the field, and then reproducing the distribution of field data in the laboratory using an approach similar to Prasad et al.[35]. The Heterogeneous Testing Protocol (HTP) was designed to assess stove performance across three discrete firepowers – high, medium, and low – using two different pot sizes.[37] Another protocol, the Indonesia Clean Stoves Initiative WBT, entails identifying burn cycles used in a community of interest, by measuring the heat flux to a pot of water during a cooking task in the field, and then developing a laboratory test that uses the identified burn cycles to heat a pot of water.[38] Most recently, a laboratory-based protocol was published through the International Standards Organization (ISO 19867-1:2018).[21] This protocol requires that emissions be quantified at three distinct firepower outputs. Each test-phase includes start-up, shut-down, and steady-state operation at a unique firepower.

None of the aforementioned protocols have been validated with field data, and thus there is no evidence that cookstove performance during these tests matches performance during real-world use. Furthermore, some of these protocols are only designed to characterize the cooking cycles of a specific user-audience but likely have limited generalizability. Another key limitation is that a multiple-firepower protocol assumes a relationship between firepower and emissions. There is literature precedence for this relationship;[39–47] however, the studies that have assessed emissions and firepower have only looked at binary trends (i.e., low vs. high firepower) or a single stove-fuel combination. Thus, this relationship has not been rigorously evaluated.

Laboratory test protocols should be able to simulate the range of emissions observed during in-home cooking, and therefore provide data that are reasonably linked with indoor air quality. We investigated a multiple-firepower test protocol to assess whether this approach could improve laboratory performance testing. In this work, we (1) developed a generalized multiple-firepower testing approach, called the Firepower Sweep Test [FST], (2) compared FST results to WBT and field measurements to assess whether a firepower-based method provides a more realistic representation of the range of potential in-home emissions and performance data, and (3) assessed the relationships between firepower and emissions, using correlation analysis and linear model selection.

2.2 Methods

2.2.1 Testing protocol

The FST was designed to examine performance by systematically operating a cookstove at multiple power outputs. Firepower is the rate of heat release from combustion, typically measured in kilowatts. Cookstove users can achieve differing firepowers by varying the fuel-feed rate, the mass of fuel in the combustion chamber, fuel spacing, and the air flow rate (in forced-air stoves). Adjusting firepower in a solid-fuel stove is analogous to turning the control knob on a modern electric or gas stove. High firepowers are used for tasks like boiling water or searing meat, while low firepowers are used for tasks like simmering rice or legumes. The following relationship can be used to model cookstove firepower:

$$FP = LHV_{fuel} \cdot \dot{m}_{fuel} \tag{2.1}$$

where FP is the operational firepower of the stove, LHV_{fuel} is the lower heating value of the fuel, and $\dot{m}_{fuel}$ is the burn rate of the fuel.

Variants of the FST were developed for continuous-feed wood stoves (Appendix A1), batch-fed charcoal stoves (Appendix A2), and batch-fed wood stoves (i.e., gasifiers; Appendix A3). The FST for continuous-feed wood stoves is described hereafter, while the protocols for the

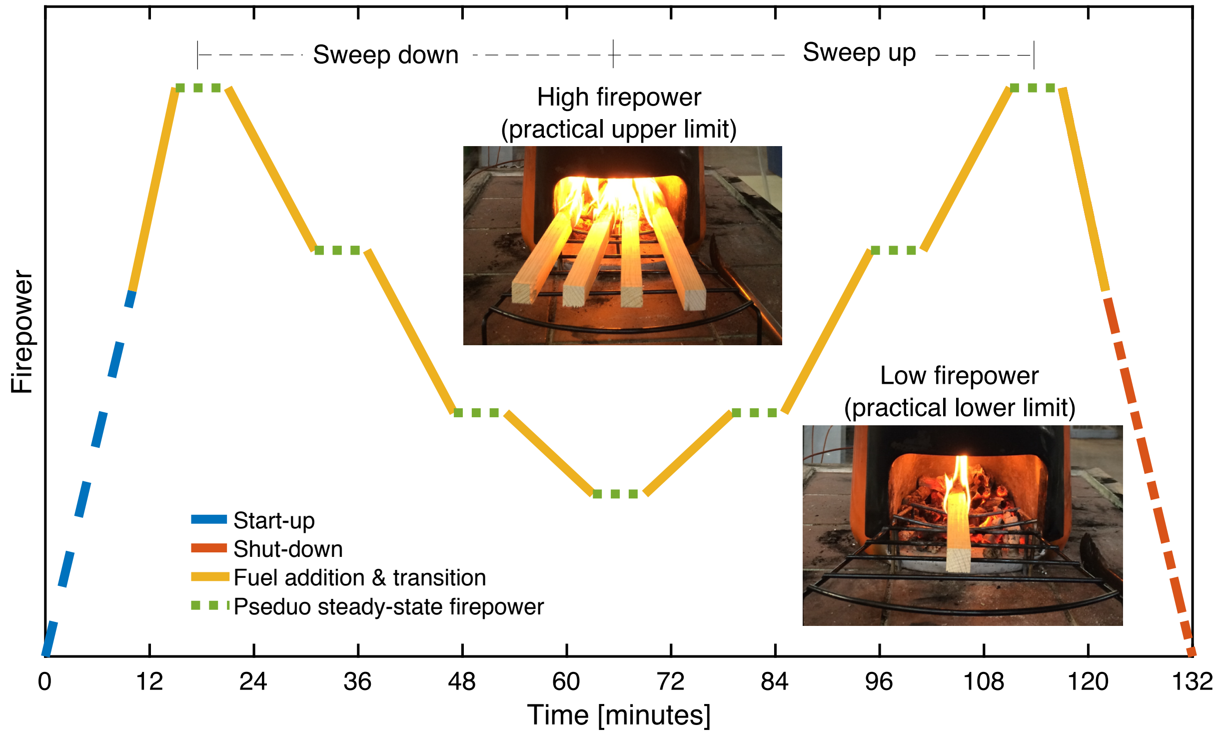


Figure 2.1: *A conceptual overview of the Firepower Sweep Test (FST) for wood stoves. Images of high- and low-firepower points that designate the practical upper limit and practical lower limit, respectively, for this rocket-elbow stove design are included.*

batch-fed stoves that are described in the Appendix. The FST protocol consisted of seventeen timed segments and lasted a total of 132 minutes. As depicted in Figure 2.1, the test phases included start-up, sweep-down (decreasing firepower), sweep-up (increasing firepower), and shut-down. During the sweep-down and sweep-up phases, the stove was operated through several target firepowers by controlling the fuel feed rate and the number of sticks fed into the combustion chamber. Temperature at the combustion chamber outlet (Appendix A5) was used as a proxy for firepower, because this and other studies[42,43] have demonstrated a linear relationship between combustion chamber outlet temperature and firepower (Appendix A4). Although the sensitivity of this measurement to ambient air temperature and thermocouple placement are not well-understood, a temperature measurement is likely to be more useful than exhaust CO_2

concentration for estimating firepower in a field setting, where total capture hoods are not available. In this study, the continuous-feed wood stoves were tested at four target firepowers; however, the FST could be adapted to include more steps if a stove-fuel combination has a large range of working firepowers (Appendix A6). Suggested time intervals, fuel batch sizes, and target combustion chamber outlet temperatures for continuous-feed wood, batch-fed charcoal, and batch-fed wood stoves are listed in Appendix A1, Appendix A2, and Appendix A3, respectively.

The FST began with a start-up phase in which start-up fuel was lit with a match and consumed. Next, the operator alternated between refueling and steady-state firepower events at designated time intervals. During steady-state events, the operator attempted to keep the combustion chamber outlet temperature as close to the target as possible. This was accomplished by controlling the amount of fuel in the combustion chamber and the fuel feed rate. Time-averaged measurements, such as filter-based fine particulate matter ($PM_{2.5}$) and BC emissions (as well as thermal efficiency, which is not reported here), were taken during start-up, shut-down, and the steady-state events. Continuous-reading instruments were operated throughout the test to quantify 'event-based' emissions profiles. No produced charcoal was removed during the test. During the final segment of the sweep, the tester shut-down the stove by putting any burning fuel in closed ash pot to cut off the air supply to the flame and extinguish the fuel. Any remaining charcoal was left to smolder in the stove for the duration of the shut-down event.

Ideally, the maximum and minimum firepowers achieved during the FST should represent the full range of firepowers that could be achieved for a given stove design. Agenbroad et al.[42] theorized that a stove's maximum firepower occurs at an excess air ratio slightly above 0%, although this firepower is difficult to achieve in practice. The lowest firepower point is constrained by the fact that combustion will cease below a certain temperature threshold. In this study, fuel charge sizes were kept roughly the same among all continuous-feed wood stoves. One wood stick and four sticks were used for the low- and high-firepower events, respectively. The sticks were approximately 1.5 cm x 1.5 cm x 12 cm. In future studies that involve testing over a range of firepowers, we recommend adjusting fuel batch sizes based on more rigorous pilot testing of individual stoves (Appendix A6).

2.2.2 *Stove-Fuel Technologies*

A total of 21 stove-fuel combinations were tested, in triplicate, using the FST (Appendix A7). Stove technologies included: a three-stone fire, an artisan clay stove, two rocket elbow stoves, three large chimney stoves, four charcoal stoves, and two gasifier stoves. Three wood fuels (Douglas fir, eucalyptus, and red oak), two charcoal fuels (lump hardwood and coconut charcoal), and one pellet fuel (eucalyptus pellets) were used. Single replicate tests were conducted with milled red oak in a forced-draft gasifier red oak and coconut briquettes in an improved, metal charcoal stove. Fuel properties are presented in (Appendix A8).

2.2.3 *Instrumentation*

Descriptions of the emissions sampling methodologies can be found in the literature. [42,43,48–50] Briefly, emissions were captured and diluted in a total-capture hood using a constant-volume displacement pump. Integrated particle emissions were sampled isokinetically from the main exhaust stream, drawn through a cyclone (URG, 16.7 Lpm) to provide an aerodynamic size cut of 2.5 μm, and then collected on a filter substrate.

Mass of $PM_{2.5}$ was measured gravimetrically using Teflon-coated glass fiber filters (Pallflex Fiberfilm T60A20, 47mm). To measure BC, the change in light attenuation through each filter was measured using a Magee Sootscan Transmissometer (Model OT21) and then converted to a BC surface density using a calibration curve (Appendix A9); the conversion was only developed for continuous-feed wood stoves. Filter samples were treated and handled according to standardized procedures. Daily laboratory blanks and weekly background samples were taken.

Gas-phase pollutants were characterized using a Testo 350 flue-gas analyzer and a Siemens ULTRAMAT gas analyzer. The gas analyzers were operated according to the manufacturers' recommendations and calibrated daily. Some filter (<1%) and gas (24%) measurements were excluded due to experimental issues (e.g., instrument failure, sample contamination, insufficient quality).

2.2.4 Data cleaning and performance metrics

Data clean-up was conducted in MATLAB 2017b. All emissions data were corrected for background gas and particle concentrations. Data from the gas analyzers were collected at one-second resolution and then averaged over each firepower interval. Filter measurements below the limit of detection (LOD) or limit of quantification (LOQ) (less than 15% of data reported here) were replaced by the $LOD/\sqrt{2}$ or the $LOQ/\sqrt{2}$, respectively.

Firepower was estimated using the following formula:

$$FP = \frac{LHV_{fuel} \cdot M_C \cdot Q_t}{Y_{C,fuel}} \left(\frac{\rho_{CO_2} \cdot \Delta CO_2}{M_{CO_2}} + \frac{\rho_{CO} \cdot \Delta CO}{M_{CO}} \right) \tag{2.2}$$

where FP is firepower, LHV_{fuel} is the lower heating value of the fuel, M denotes the molecular weight of carbon (M_C) or specific exhaust components (M_{CO}, M_{CO_2}), $Y_{C,fuel}$ is the mass fraction of carbon in the fuel, ρ_{CO_2} and ρ_{CO} represent the density of CO_2 and carbon monoxide (CO) in the exhaust, respectively, Q_t is the total volumetric flow rate of air through the hood, and ΔCO_2 and ΔCO are the background-corrected mixing ratios of CO_2 and CO. This formulation utilizes the carbon-balance method to estimate the total mass of fuel burned, with the assumption that emissions of gas-phase hydrocarbons and carbonaceous particulate matter are negligible relative to emissions of CO_2 and CO. Emissions factors for CO, $PM_{2.5}$, and BC (in units of mass of pollutant per mass of dry fuel burned) were calculated using a similar carbon-balance approach:

$$EF_i = \frac{m_i}{m_{fuel}} = \frac{m_i \cdot Y_{C,fuel}}{\Delta CO_2 + \Delta CO} \tag{2.3}$$

Where EF_i is the emissions factor of pollutant i, m_i is the background-corrected mass of pollutant i, and m_{fuel} is the mass of fuel burned.[51] Emissions factors based on the mass of fuel burned are presented in the main text because, often, they are the only metric reported in field studies where total capture hoods are not available. Emissions on a per-useful-energy-delivered basis and per-time basis will be provided in a Colorado State University online data repository.

Modified combustion efficiency (MCE) was calculated using the method published by Ward and Hao [52]:

$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \tag{2.4}$$

Given that CO is the most abundant product of incomplete combustion, MCE is a good proxy for the extent to which combustion is complete. Higher MCE values indicate more complete combustion (with the maximum being 1), while lower MCE values indicate less complete combustion. MCE is widely-used to indicate cookstove performance in both lab and field applications and is considered to be a reasonable proxy for actual combustion efficiency.[52] MCE has been used in cookstove and biomass burning applications to indicate whether combustion conditions are primarily flaming or smoldering,[40] which has important implications for emissions.

Data were analyzed using R (version 3.4.1); the code is hosted at: https://github.com/kbilsback/stoves_epa_star. We present operating conditions (firepower and

MCE) and emissions metrics ($PM_{2.5}$, BC, and CO) as a function of stove type, fuel type, and test phase. For the analysis, the stoves were broken into three categories based on the FST protocol design: continuous-feed wood stoves, batch-fed charcoal stoves, and batch-fed wood stoves. Within these categories, stove designs with similar combustion chamber geometry and materials were analyzed together. Details, including which specific stove designs fell into each category, are given in Appendix A7.

The FST data were compared to WBT data (Jetter et al.[53] and Medina et al.[54]) and field data (Roden et al.;[55] Wathore et al.;[56] and data collected by our group in India, Honduras, or Uganda[57]). Where possible, we compared data between test protocols (e.g., FST, WBT, field) using the same cookstove technology and fuel species; however, given the data available, comparing the exact same stove-fuel combination was not always possible. WBT data from tests conducted with 'wet' fuels were excluded from our analysis, because the fuels used in the FST were all air dried (moisture content ~6-8%). Since moisture content is often not reported in field studies, we did not exclude any field data based on reported moisture content.

2.2.5 Correlation analysis

The correlation between firepower and BC, firepower and CO, and firepower and $PM_{2.5}$ was quantified using Spearman's rho.[58] This metric was chosen because the calculation does not require any assumptions about (1) the distribution of the underlying data (i.e., the data do not need to be normally distributed) or (2) the relationship between the independent and dependent variables (e.g., linear, exponential, power relationship, etc.). Spearman's rho is a measure of the strength and directionality of a monotonic relationship in data. Rho values fall between negative one and one, where the sign designates the directionally (e.g., positive or negative) of the relationship and the value indicates the strength of the relationship (positive or negative one being a perfect monotonic relationship).

2.2.6 Linear models

The relationship between firepower and emissions was also evaluated using model selection. Ten candidate linear models were chosen for comparison (Appendix 12). Predictors included firepower and MCE as continuous variables and stove-fuel combination as a categorical variable. Outcomes included $PM_{2.5}$, CO, and BC emissions factors. The candidate models included every combination of the three predictors as additive terms. In addition we considered models with an interaction between stove-fuel combination and either MCE or firepower. MCE was not used as a predictor for CO emissions, however, as MCE is a proxy for CO emissions. We found that measurements taken within the same sweep were statistically independent.

Small sample-size-corrected Akaike information criterion (AICc) was used for model selection.[54] Unlike conventional model selection metrics, such as the coefficient of determination (R^2), AICc does not require models being compared to be nested and accounts for both the goodness-of-fit and the simplicity of the model (i.e., AICc favors models with fewer predictors). Although we did not use R^2 for model selection, the amount of variance explained (i.e., R^2) by each of the top models is reported in the results. We also present the Akaike weight, which can be interpreted as the probability that a specific model is the most optimal according to AICc, relative to the other candidate models.[55] Given that the Akaike weights of all candidate models should sum to 1.0, an Akaike weight close to 1.0 would indicate that the highest-performing model is very likely to be the best model for the data, while a lower Akaike weight indicates that there were other models that performed almost as well as the top model.

Separate models were developed for continuous-feed wood stoves and batch-fed charcoal stoves. Models were not developed for batch-fed wood stoves because there were limited stoves tested in this category (compared to the other stove categories). Estimates and the 95% confidence intervals of for each of the top models are given in Appendix A13. Significance levels of the interactions and diagnostic plots of the top models are given in Appendix 14 and Appendix 15, respectively.

2.3 Results and Discussion

2.3.1 FST and WBT operating conditions

Operating conditions observed during the FST and WBT are shown in Figure 2.2. For continuous-feed wood stoves, the FST captured several low-firepower events that the WBT missed entirely; these included start-up events (low firepower and high MCE), shut-down events (low firepower and low MCE), and several steady-state events.

In the WBT, stove start-up (i.e., fuel ignition) is not captured separately from steady-state operation. As a result, differences in firepower and MCE between start-up and steady state conditions cannot be identified. Lower than average firepowers may be seen during start-up because the stove and fuel are cool relative to other phases of the test. Additionally, the average firepower may be low during the start-up phase if the tester fails to add enough heat to the cookstove-fuel system using the match flame. If this occurs, then combustion will not be self-sustaining (i.e., the flame will extinguish) and several attempts may be required to light the stove. Ignition is especially difficult in wood combustion, where the fuel must be heated to release combustible gases. In poorly-insulated cookstoves, such as a three-stone fire, high heat release rates are needed to create a self-sustaining reaction.

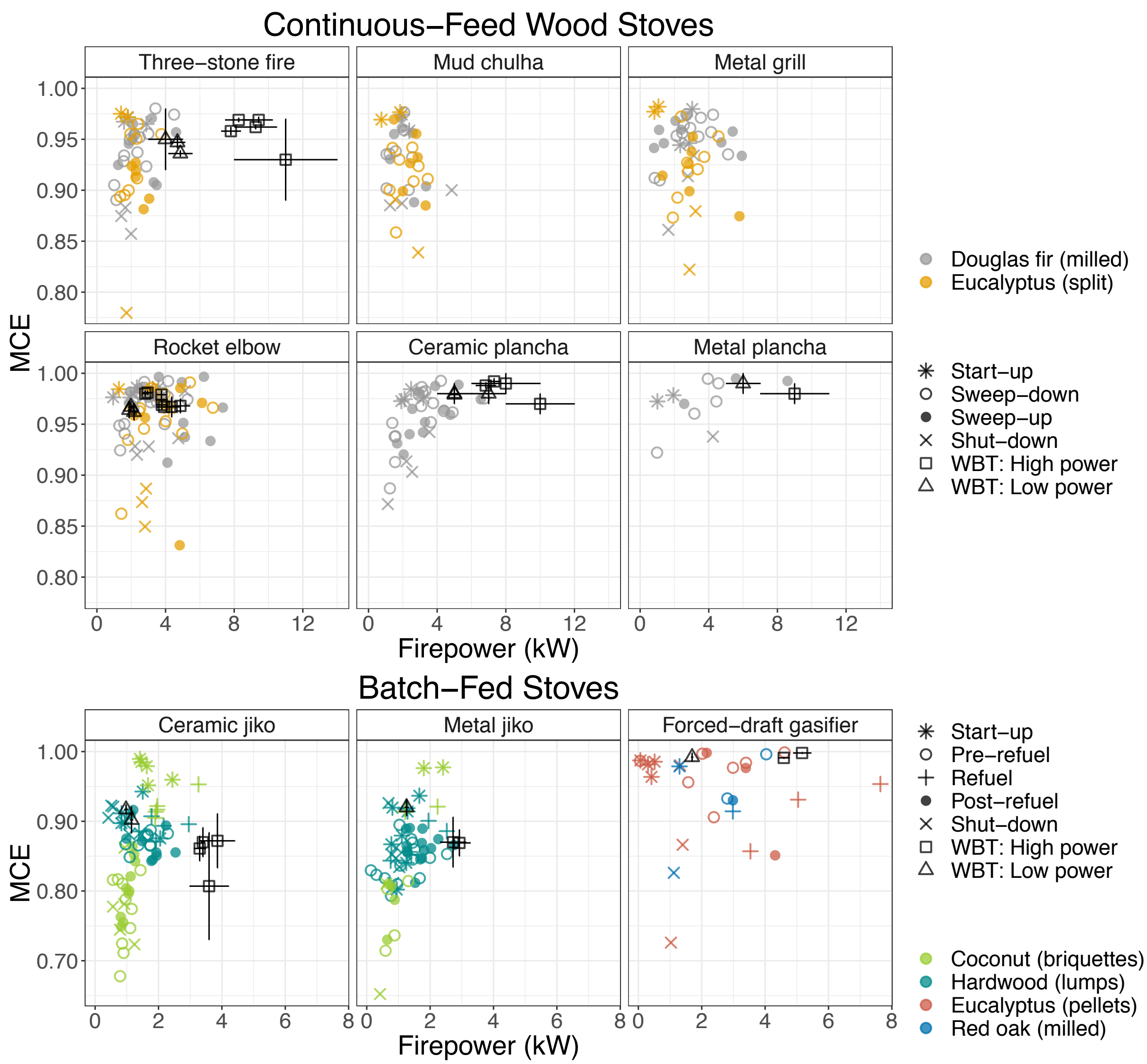


Figure 2.2: *Firepower (FP) versus modified combustion efficiency (MCE) measurements made during the Firepower Sweep Test (FST) and Water Boiling Test (WBT). High power (hot-start and cold-start) and low-power (simmer) data from the WBT are shown in black and are presented as replicate-averages with error bars that designate one standard deviation.*

Shut-down events in continuous-feed wood stoves resulted in lower firepowers and lower MCEs than the WBT. Three minutes into the shut-down phase of the FST, any flaming fuel was removed from the stove and extinguished by putting the fuel inside a covered pot. Then, any leftover charcoal was left to smolder in the stove for the remainder of the test. The smoldering combustion (i.e., with no flame) captured during shut-down of continuous-feed stoves was likely not captured in the WBT, because the WBT explicitly instructs the user to stop the test as soon as the pot of water reaches a boil (i.e., before the flaming fuel is extinguished).[13]

The WBT is also not likely to capture the steady-state, low-firepower operating conditions seen during the FST because the WBT procedure instructs the user to “bring the [pot] rapidly to a boil without being excessively wasteful of fuel.”[13] This instruction implies that the stove should be run at a high firepower, which likely does not represent all in-home cooking practices. High-power operation requires close monitoring, which may not occur if users are completing other task (i.e., child care, cleaning, etc.) simultaneously.[55]

In two cases, the WBT procedure included higher firepowers than the FST: the three-stone fire and ceramic plancha. To ensure that the FST was not biased by stove type, the fuel feed rates were not changed between competing technologies in this study. Because traditional stoves suffer from greater heat loss compared to insulated stoves, higher fuel feed rates are required to operate a traditional stove at a given useful power output. As a result, traditional stoves are often run at higher fuel consumption rates during the WBT than improved stoves. In this study, holding fuel feed rates constant resulted in the traditional stoves being operated at lower firepower levels compared to the WBT. The WBT protocol does not provide guidelines on fuel batch sizes.

For the two charcoal stoves, the FST produced a substantially wider range of MCEs than the WBT, with the WBT tending to favor higher MCEs. The WBT also did not capture the high-firepower events during refueling. When charcoal stoves were refueled during the FST, a fresh batch of room-temperature fuel was added on top of the hot charcoal fuel bed. Then, a small amount of additional fuel was soaked in kerosene and lit with a match. The high-firepower, high-MCE operation seen during refueling was likely not seen during the WBT because the WBT procedure does not explicitly require refueling to be characterized. Failure to capture refueling events is problematic because they occur frequently in the field, especially during longer-duration cooking events.

As with other stove types, the WBT procedure for batch-fed wood stoves missed several important operating regimes, including refueling and shut-down. During refueling, room-temperature fuel was added on top of the hot fuel bed, resulting in disruption of the secondary flame and a rapid release of unburned combustible gases.[49,50] During shut-down, firepowers and MCEs were lower than during the WBT because the secondary flame was extinguished.[13]

Operating conditions were more dependent on fuel type for batch-fed stoves than for continuous-feed wood stoves. For charcoal stoves, a larger range of MCEs was observed after switching from hardwood lumps (MCE ranged from 0.87 to 0.90) to coconut charcoal (MCE ranged from 0.77 to 0.91). Similarly, a wider range of MCEs were measured when operating the batch-fed wood stoves with red oak (from 0.92 to 0.97) instead of eucalyptus pellets (from 0.98 to 1.00). Fuel selection may be more important in determining the operating conditions of a batch-fed stove because the user has less control over the combustion process when fuel is fed in batches (rather than continuously). Additionally, batch-fed stoves often use processed fuels, which tend to have more variable properties (e.g., volatile content for charcoal and bulk density for pelletized fuels) than raw wood fuels, since the properties of processed-fuels depend on how the fuels were refined.

The FST intentionally targets and captures several operating conditions that the WBT misses, such as those seen during low-firepower events, refueling, and shut-down. For example, if a batch-fed stove was used in the field, the duration of an in-home cooking event could be shorter or longer than boiling a five-liter pot of water, thus decreasing or increasing the number of refuels needed to carry out the task. Characterizing refueling events separately, as is done in the FST, would allow the average firepower or MCE to be weighted by the typical number of refuels seen in a particular region. Capturing each operating phase separately also makes the FST potentially more robust and generalizable than the WBT. In theory, during the WBT, a fuel batch size could be chosen so that fewer or no refuelings are needed to boil the pot of water during the WBT. By requiring that each operating phase be captured separately, and by pre-defining a fuel feed rate and target temperature, there is less incentive to modify the protocol to better fit a particular stove design.

2.3.2 FST and WBT emissions

Scatterplots of $PM_{2.5}$ versus CO emissions from the FST and WBT are shown in Figure 2.3. For continuous-feed wood stoves, the FST captured a much wider range of $PM_{2.5}$ and CO emissions than the WBT. For example, the WBT missed the high-$PM_{2.5}$ emissions seen during some start-up events ($PM_{2.5}$: from 1.08 to 4.52 g/kg-fuel; CO: from 24 to 36 g/kg-fuel). Although there was a lot of variability during the FST start-up, BC emissions were often high compared to other phases of the test. The flaming combustion of volatiles that occured during start-up likely facilitated soot formation, resulting in higher BC and total $PM_{2.5}$ mass emissions compared to other phases of the test. Sullivan et al.[59] also observed high $PM_{2.5}$ emissions during start-up events.

The WBT often did not capture the low-$PM_{2.5}$ emissions (from 0.51 to 1.37 g/kg-fuel) and high-CO emissions (from 54 to 117 g/kg-fuel) seen during the shut-down segment of the FST. When the stove was extinguished, the volatile content of the fuel left in the stove was lower and there was minimal flaming combustion compared to other phases of the test. As a result, there was likely less soot production (leading to lower $PM_{2.5}$ emissions) and slower CO to CO_2 conversion rates (leading to higher CO emissions).

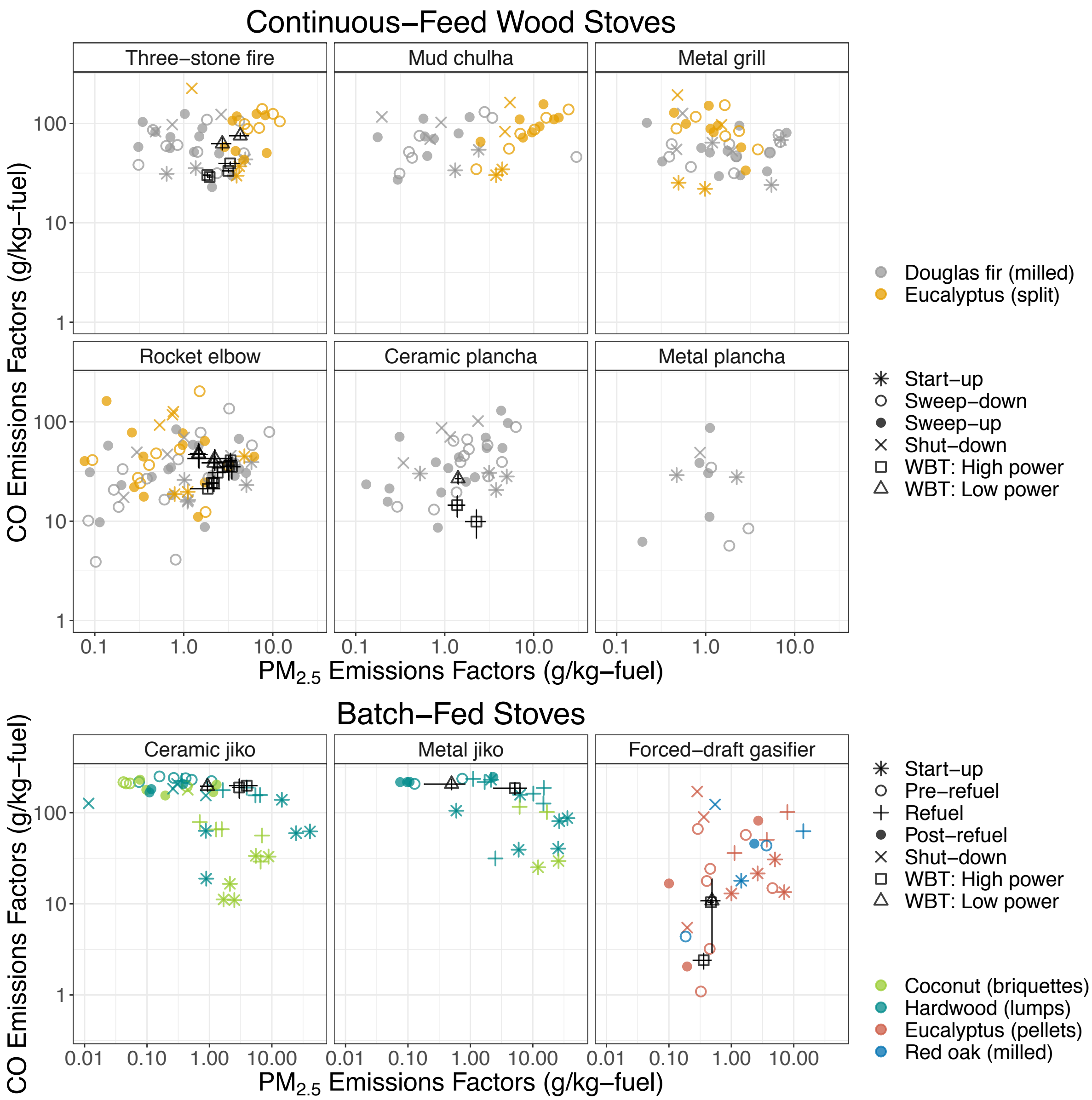


Figure 2.3: *Fine particulate matter ($PM_{2.5}$) and carbon monoxide (CO) emissions factors from the Firepower Sweep Test (FST). High power (hot-start and cold-start) and low-power (simmer) data from the WBT are shown in black and are presented as replicate-averages with error bars that designate one standard deviation.*

For continuous-feed wood stoves, the FST captured higher $PM_{2.5}$ emissions than the WBT. This result was unexpected given that the three-stone fire reached higher firepowers when tested under the WBT. This discrepancy could be caused by differences in fuel type, stove design, or more careful fire tending practices during the WBT compared to the FST. Although there was a lot of variability in emissions, the highest $PM_{2.5}$ emissions, excluding those associated with start-up, were generally associated with the highest firepower test point. For rocket-elbow and other improved stoves, capturing high-firepower operating conditions and the associated high-$PM_{2.5}$ emissions is important because users often prefer cooking with higher firepowers in real-world settings.[46]

The FST also captured much lower emissions factors than the WBT. Low emissions were typically observed during the lowest-firepower test point, or shutdown (for $PM_{2.5}$). The WBT encourages higher fuel feed rates and misses the lowest-firepower observed during the FST for continuous-feed stoves. Even the simmer phase, the lowest firepower phase of the WBT, requires operation at higher firepowers than the FST.

For batch-fed charcoal stoves, $PM_{2.5}$ emissions were dominated by start-up and refueling events. An accelerant, kerosene, was used during both of these phases, which led to flaming combustion of volatile fuel (kerosene), rather than surface oxidation of char, likely increasing soot production (and total $PM_{2.5}$ mass). The CO emissions were systematically lower during start-up (from 27 to 84 g/kg-fuel) and refueling (from 72 to 188 g/kg-fuel) of charcoal stoves compared to other phases of the test, which may have been due to higher heat-release rates and different chemistry during flaming combustion of volatile fuel components compared to reaction of oxygen with the solid charcoal surface. The high-$PM_{2.5}$ and low-CO emissions profile seen during start-up and refuel was not captured during the WBT.

Similarly, for the batch-fed wood stoves, the WBT missed the high-emissions events seen during the start-up, refueling, and shut-down phases of the FST. Emissions during steady-state operation were also often higher during the FST than the WBT, although this difference did not appear to be driven by firepower.

The FST captures a larger range of emissions than the WBT, making the FST potentially more generalizable to cooking practices in different regions. The WBT often misses the unique emissions profiles seen during start-up, shut-down, refueling, low firepowers, and very high fuel loading. By isolating and characterizing these operating conditions, the FST provides more transparency than the WBT. Many events missed entirely by the WBT are often associated with higher $PM_{2.5}$ and/or higher CO emissions in the FST. If events such as shut-down, "overfueling" (for continuous-feed stoves), and refueling (for batch-fed stoves) are not included in an emissions estimate or are included at the wrong frequency, then test-aggregated emissions measurements may be biased low compared to in-home cooking practice.

2.3.3 FST, WBT, and field emissions

$PM_{2.5}$ and CO emissions factors from the FST, the WBT, and uncontrolled, in-home cooking events are plotted in Figure 2.4. The FST resulted in a larger range of $PM_{2.5}$ and CO emissions factors than the WBT or the field data for all stoves except the ceramic planchas. Furthermore, the range of emissions factors captured by the FST encompasses the range of emissions captured by both the WBT and the field data for all stoves except for the ceramic planchas (CO and $PM_{2.5}$ emissions) and the three stone fire ($PM_{2.5}$ emissions only).

The fact that the FST is able to span the range of $PM_{2.5}$ and CO emissions factors observed in the field, for most stove designs, is impactful because it demonstrates that the FST is generalizable to in-home cooking practices. The fact that the FST spans the range of emissions seen in this particular set of field data is also notable given the range of stoves, fuel, and cooking practices used across the four countries used for comparison (Honduras, India, Malawi, Uganda).

Furthermore, the high emissions factors observed during the FST (but not the WBT) primarily come from events that are not characterized independently with the WBT: start-up, shut-down, refueling (for batch-fed stoves), and “overfueling” (for continuous-feed stoves).

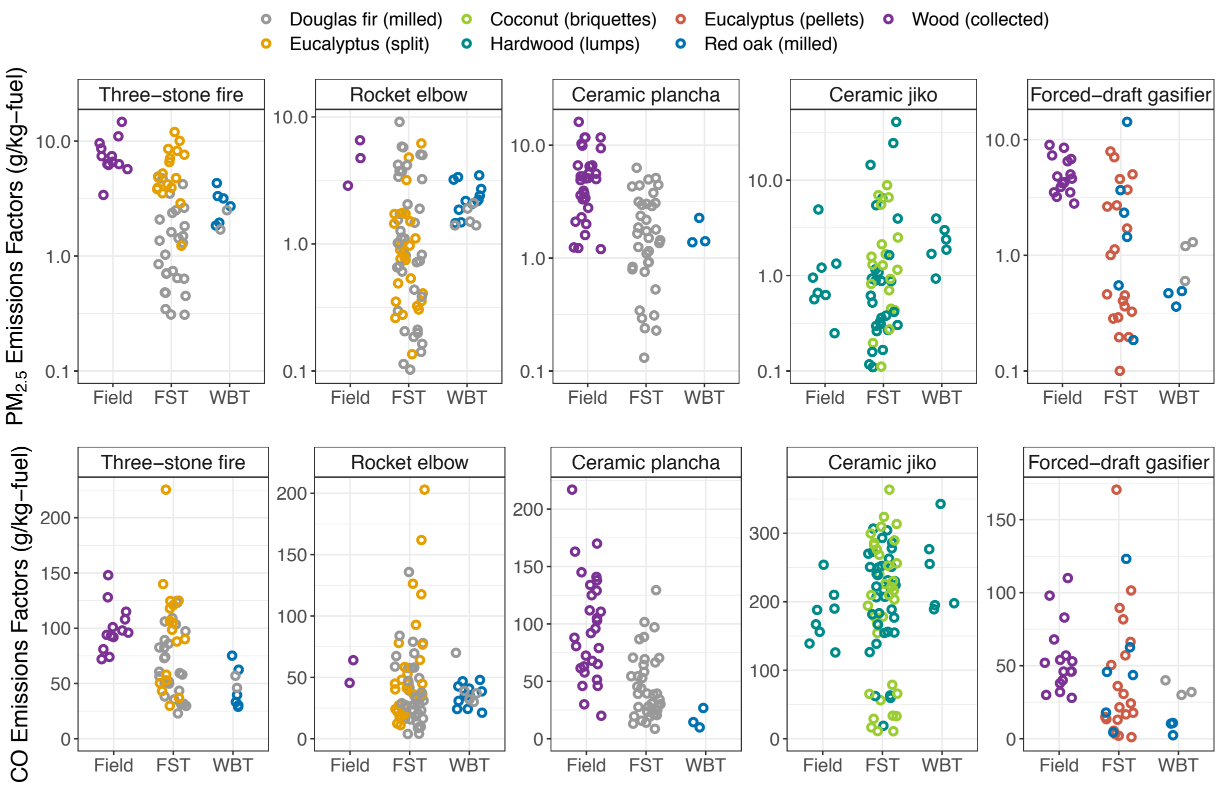


Figure 2.4: *Fine particulate matter ($PM_{2.5}$) and carbon monoxide (CO) emissions factors from the Firepower Sweep Test (FST), the Water Boiling Test (WBT), and in-home cooking (Field). The field data are from tests conducted in Honduras, India, Malawi, or Uganda. The fuel and the way it was processed are indicated by the color of the marker.*

The primary advantage of the FST is that the test reproduces the range of emissions seen in field studies. However, an important distinction to note is that mean emissions factors aggregated across the entire FST are not necessarily comparable to mean emissions from a specific cooking event in the field (because the FST was designed to span stove operating ranges, not to mimic typical cooking tasks). Thus, this multiple-firepower approach creates a protocol that is more generalizable than a procedure based on a specific task (e.g., boiling a pot of water).

The results shown in Figure 2.4 demonstrate that a multiple-firepower approach may be able to span the range emissions observed during in-home cooking (for most stove types), while the WBT systematically underpredicts the emissions observed during uncontrolled field tests. The median emissions under the FST will not mimic median emissions reported in the field, since absolute emissions depend on specific stove use pattern. The FST is simply designed to characterize various cookstove operating conditions independently to identify the *range* of emissions factors likely to be observed during real-world use.

2.3.4 Firepower and emissions

Correlation plots of firepower versus $PM_{2.5}$, firepower versus BC, and firepower versus CO are shown in Figure 2.5, Appendix 10, and Appendix 11, respectively. Our results indicate that firepower was correlated with $PM_{2.5}$, BC, and CO emissions factors; however, the correlation varied in strength and directionality across stove-fuel combinations (e.g., for $PM_{2.5}$, Spearman's rho ranged from -0.15 to 0.92). For a given stove, the directionality and strength of the correlation between firepower and $PM_{2.5}$ emissions was also not conserved across fuel types. For example, for the three-stone fire, the correlation was stronger for Douglas fir (0.63) than for eucalyptus (0.02). Similarly, for batch-fed charcoal stoves, the coefficients were much higher for coconut charcoal fuel (ceramic jiko: 0.74; metal jiko: 0.92) than lumped hardwood (ceramic jiko: 0.16; metal jiko: 0.05). These results indicate that fuel selection likely has a substantial impact on the relationship between firepower and emissions for wood- and charcoal-fueled stoves. Similar results were seen for BC and CO emissions.

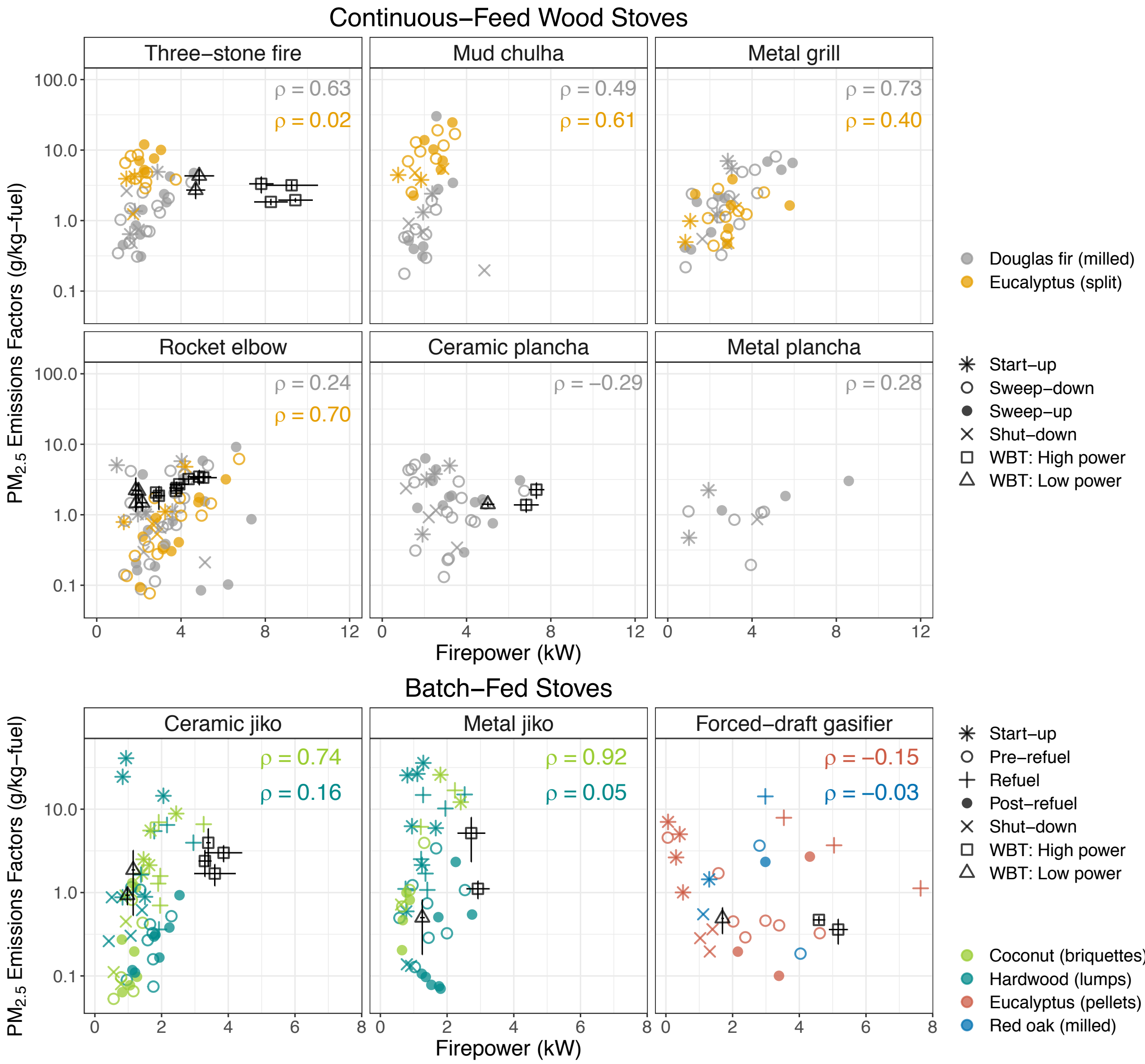


Figure 2.5: *Firepower versus particulate matter emissions factor data from the Firepower Sweep Test. Colors are used to identify the fuel type and shapes are used to identify the test phase. Spearman's rank correlation coefficient (ρ), which denotes the degree of correlation between firepower and particulate matter, was calculated for each stove-fuel combination (color-coded). High power (hot-start and cold-start) and low-power (simmer) data from the WBT are shown in black and are presented as replicate-averages with error bars that designate one standard deviation.*

The relationship between stove operating conditions (firepower and MCE) and emissions ($PM_{2.5}$, BC, and CO) was also quantified using linear models. The best-performing models based on AICc are listed in Table 2.1. All of the top models included both firepower and the stove-fuel type. Hence all the models had a unique intercept for each stove-fuel type and some models had an interaction term between stove-fuel type and either MCE (e.g., $PM_{2.5}$ from continuous-feed wood stoves) or firepower (e.g., BC emissions from continuous-feed wood stoves and CO on batch-fed wood stoves) resulting in a unique slope for each stove-fuel type. The presence of these variables further supports the result that, although firepower is correlated with emissions, the relationship between firepower and emissions is dependent on stove type and fuel type.

The modest R^2 values (ranging from 0.21 to 0.62) also demonstrate that firepower only explains some of the variance in emissions, indicating that there are factors beyond firepower that have not been accounted for in our models. This result is also supported by the fact that MCE only appears in the top model for $PM_{2.5}$ emissions from continuous-feed wood stoves. The association with MCE and $PM_{2.5}$ emissions has also been reported in the literature in cookstove[46,60,61] and open biomass-burning applications.

Both the correlation plots in Figure 2.5 and the top-models in Table 2.1 suggest a modest relationship between firepower and emissions, which likely varies from one stove-fuel combination to the next. This result supports the need for multiple-firepower testing that is calibrated to specific stove designs, as instructed in the FST protocol. Both the modest Spearman's rho and R^2 values also indicate that firepower alone cannot account for the observed variability in cookstove emissions. This result highlights the need to utilize field observations to pinpoint operating conditions, such as fanning the stove or quenching the flame using water, which may not follow the same firepower versus emissions trend seen during steady-state operation.

Table 2.1: *Highest-performing linear models for particulate matter ($PM_{2.5}$), black carbon (BC), and carbon monoxide (CO) emissions factors (EF) selected by small sample-size corrected Akaike information criterion (AICc), where FP is firepower, MCE is modified combustion efficiency, α_j are the coefficients for stove-fuel specific intercepts, β_j are the coefficients for stove-fuel specific slopes, (continuous-feed wood: j = 1,...,10; batch-fed charcoal: j = 1,...,4), γ is a constant coefficient across all stove-fuel combinations, epsilon (ϵ) is random error, R^2 is the coefficient of determination, and the Akaike weight represents the probability that, of the candidate models we selected, the top model is supported by the data.*

Continuous-feed wood stoves	R^2	Akaike weight
$ln(PM_{2.5}\ EF) = \alpha_j + \beta_j \cdot MCE + \gamma \cdot FP + \epsilon$	0.62	0.997
$ln(BC\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$	0.56	0.513
$ln(CO\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$	0.40	0.977
Batch-fed charcoal stoves	R^2	Akaike weight
$ln(PM_{2.5}\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$	0.21	0.390
$ln(CO\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$	0.58	0.814

2.3.5 Indoor Air Quality Ramifications

Simplified box-models of an indoor environment have demonstrated that, for episodic sources, the emissions rate and the amount of air pollution a person breathes in (i.e., intake) are independent.[62,63] This means that the total mass emitted per cooking event is the primary factor affecting intake and, therefore, the most important metric related to human health. In this manuscript, we report emissions on a per-fuel-mass basis and a per-useful-energy basis, rather than as total mass emitted, because the FST was designed to reproduce emissions during various operating conditions, rather than total emissions for a specific cooking event. The protocol was designed this way because task-based protocols, such as the WBT, have historically not been able to reproduce the total emissions of in-home cooking practices. We hypothesize that firepower-specific emissions data generated from protocols like the FST could be used in conjunction with information about local cooking practices (i.e., stove type, fuel type, and time-series measurements of firepower during cooking) to estimate in-home total mass emissions. This methodology could provide health researchers or policy makers with better information about the types of technologies that will provide real-world indoor air quality benefits. Validating the firepower-weighted emissions factor approach and understanding how FST data may be used to estimate total in-home emissions and air pollution concentrations is the subject of ongoing work.

In the absence of more comprehensive validation studies, we recommend using disaggregated FST emissions data as a means to examine whether a particular stove design is robust (i.e., whether a stove can produce low emissions under a variety of operating conditions). For example, a policy maker could compare the emissions of each FST phase when deciding which improved cookstove will be disseminated into local communities. By combining FST data with knowledge of local cooking practices, policy makers could make more informed decisions about which design might best improve local indoor air quality based on a specific community's cooking practices.

2.3.6 The ISO multiple-firepower test

The results of this work highlight the importance of a multiple-firepower test protocol, such as the one recently developed through the International Standards Organization (ISO 19867-1:2018). The ISO protocol provides guidance on testing stoves at three firepowers ("low," "medium," and "high"), as opposed to the seven proposed in the FST. The ISO protocol design is simpler (and less time consuming) than the FST, which is advantageous for resource-limited laboratories. Further cost-benefit analyses are needed to evaluate the ideal number of firepower test points. This will be the subject of ongoing work through the ISO 19867-1:2018 protocol revisions.

One concern regarding the ISO 19867-1:2018 protocol is that there is little guidance regarding the firepowers at which a given stove should be tested. In this work we find substantial overlap between the emissions seen at low and medium firepower test points. These results indicate the importance of testing at high firepower, and even "overfueled" testing conditions, to reproduce the high emissions events seen in the field. We recommend that users of the ISO 19867-1:2018 protocol test improved stoves at fuel-loading conditions much higher than those commonly used in the WBT.

Finally, the results of this study demonstrate the need to characterize emissions during non-steady state events—such as start-up, shut-down, and refueling—that sometimes lead to high emissions. The current ISO 19867-1:2018 protocol requires that start-up and shut-down emissions be captured, but does not require that they be captured separately from steady-state emissions.

2.3.7 Limitations and Future Work

A subset of the data are presented here; further description of the results, including tables of disaggregated data, aerosol properties, and real-time emissions profiles will be provided in a companion manuscripts.

Many factors beyond firepower cause deviations between laboratory and field emissions factors, including differences in fuel moisture content, the fuel source, degradation of the stove construction materials over time, cooking vessel geometry, and differences in regional cooking patterns. We acknowledge that addressing all of these aspects in a generalizable laboratory protocol would be impractical. However, the results of this work have demonstrated the value of a firepower-based protocol as a means to describe several important determinants of real-world stove emissions.

We attempted to compare FST data to WBT and field data using the same exact stove-fuel combinations. Unfortunately, finding studies that evaluated the same metrics of interest to our study with the same stove/fuel combinations was challenging; instead, we grouped stoves with with similar geometry and construction materials and fuels that were processed in similar ways. We expect that the differences in protocols result in greater differences in emissions than differences in fuel.

Some of the stoves with higher thermal masses (e.g., three-stone fire, ceramic plancha) were run at lower firepowers in the FST than the WBT, likely because the fuel consumption remained roughly the same across different stove designs. In future tests, the protocol for traditional stoves and chimney stoves could be modified to use more fuel, and possibly include more test points, so that these stoves are operated at higher firepowers during the FST, like they often are in the field.[46] These recommendations were incorporated into the pilot testing discussion in Appendix 6.

The primary focus of our study was investigating continuous-feed wood stoves and batch-fed charcoal stoves using the FST. Further work is needed to investigate whether the FST is a feasible alternative for additional stove-fuel combinations including liquid- and gas-fueled stoves (i.e., kerosene and liquefied petroleum gas).

Dilution ratios in the emissions hood can overestimate particle mass relative to more dilute ambient conditions due to differences in gas-particle partitioning of semivolatile organics;[64] however, the lower dilution ratios tested here likely represent the particle-vapor partition in an indoor environment before emissions are diluted to ambient concentrations downstream of the source. Future emissions measurements conducted in the field will be used to validate laboratory emissions factors. Additionally, secondary organic aerosol (SOA) formation was not accounted for, which means the results presented here will likely underestimate the aerosol-forming potential of cookstove emissions.[65]

2.3.4 Conclusions

In this work, we investigated the utility of a multiple-firepower cookstove testing procedure, called the FST, as an alternative to task-based laboratory testing. Most notably, we found that this multi-firepower test was able to reproduce the range of emissions observed during field studies, including the high emissions events typically not typically observed during controlled laboratory tests. When compared to data taken in Honduras, India, Malawi, and Uganda emissions measured during the the FST often spanned the range of emissions seen under in-home cooking practices. The FST also characterized non-steady state operating events such as start-up, shut-down, and refueling (for batch-fed stoves) independently, making high-emissions events easier to identify. Furthermore, our analysis indicates that, although firepower and emissions are correlated, the relationship is modest, which suggests the need for strategic field observations on in-home cooking practices since firepower alone cannot explain the observed variance in cookstove emissions, and depends on the stove-fuel combination, which demonstrates the need for multiple-firepower tests that are calibrated to specific stove designs.

These analyses suggest that multiple-firepower performance testing could constitute a substantial advancement towards more fully characterizing cookstove emissions in a laboratory setting and provide a compelling case for a multiple-firepower testing approach such as the one recently developed through the International Standards Organization (ISO 19867-1:2018). Such improved laboratory tests would benefit cookstove research by better predicting which technologies have low enough emissions to result in substantially improved indoor air quality, providing data that could lead to more strategic dissemination of improved stove technologies based on a specific community's cooking practices, and enabling more judicious deployment of field testing.

Our results suggest that cookstoves will likely perform worse under in-home cooking practices than under laboratory tests. The expanded range of emissions factors seen under the FST, especially those which represent higher emissions may be closer to actual operating conditions in the field. We recommend that designers prioritize improving cookstove performance under the highest emissions events (i.e., poorest performance), rather than mid-range emissions events, because these high-emission events likely contribute disportionately to cookstoves net emissions and exposures during in-home cooking practices.

CHAPTER 3: A COMPREHENSIVE LABORATORY ASSESSMENT OF AIR POLLUTANT EMISSIONS FROM RESIDENTIAL COOKSTOVES

3.1 Introduction

Household air pollution from solid fuels (i.e., wood, charcoal, coal, etc.) is a leading cause of death and disease worldwide.[4,22] Many constituents of cookstove smoke have known health and/or atmospheric effects. For example, fine particulate matter ($PM_{2.5}$) has been linked to long-term adverse health outcomes (e.g., respiratory tract infections and chronic obstructive pulmonary disease); carbon monoxide (CO) exposures have been linked to both acute and long-term health effects (e.g., low birth weight);[66–68] and individual compounds, such as benzene, formaldehyde, acetaldehyde, and certain polycyclic aromatic hydrocarbons (PAHs), have been classified as carcinogenic.[69,70] Additionally, cookstoves emit greenhouse gases, which absorb solar radiation; aerosols, which can affect the atmosphere by scattering or absorbing radiation and changing cloud properties;[71] and volatile organic compounds (VOCs), which are precursors to secondary organic aerosols[72,73] and tropospheric ozone.[74]

$PM_{2.5}$ and CO are the most commonly measured cookstove emissions; they are also the only pollutants for which there are published emissions guidelines. For example, the International Standards Organization recently published voluntary performance targets for $PM_{2.5}$ and CO (ISO 19867-3:2018). However, there is not clear evidence that reduced emissions of $PM_{2.5}$ and CO are indicative of reduced emissions of other health- or environment-relevant cookstove constituents.

Emissions data for other cookstove smoke constituents, beyond $PM_{2.5}$ and CO, are lacking. Some studies have reported information about $PM_{2.5}$ composition including elemental carbon, organic carbon,[75,76] and particle size[75–85] but few have quantified emissions at the compound level (e.g., VOCs,[86–89] carbonyl compounds,[89,90] and PAHs[91–94]). The studies that have measured emissions at the compound level have focused primarily on stove technologies used in China and Southeast Asia (e.g., built-in coal heating stoves). A few studies report speciated emissions from stoves used in other regions (e.g., Central America, Africa, and South-Central Asia), but only characterize a limited number of compounds (e.g., only formaldehyde or benzo[a]pyrene) emitted by a limited number of stove-fuel combinations, include limited replicate tests, and/or report limited metrics.[86,88,90,95–97]

To provide a more comprehensive dataset on cookstove emissions, we measured 120 particle- and gas-phase smoke constituents, including organic matter, elemental carbon, inorganic ions, carbohydrates, ultrafine particles, VOCs, carbonyls, and PAHs. We tested a total of 26 stove-fuel combinations that represent a range of technologies including traditional cookstoves (i.e., open fires), improved biomass cookstoves (i.e., stove which have been modified to lower emissions by adding insulation or a fan), and fossil-fuel cookstoves (i.e., kerosene and liquified petroleum gas [LPG] stoves). We also developed linear regression models to assess whether $PM_{2.5}$ and CO could be used to predict levels of other co-emitted pollutants.

3.2 Materials and Methods

3.2.1 Test Matrix

The stove-fuel test matrix is provided in Figure 3.1. A total of 87 tests were conducted, including three replicates for each stove-fuel combination and an additional nine make-up tests.

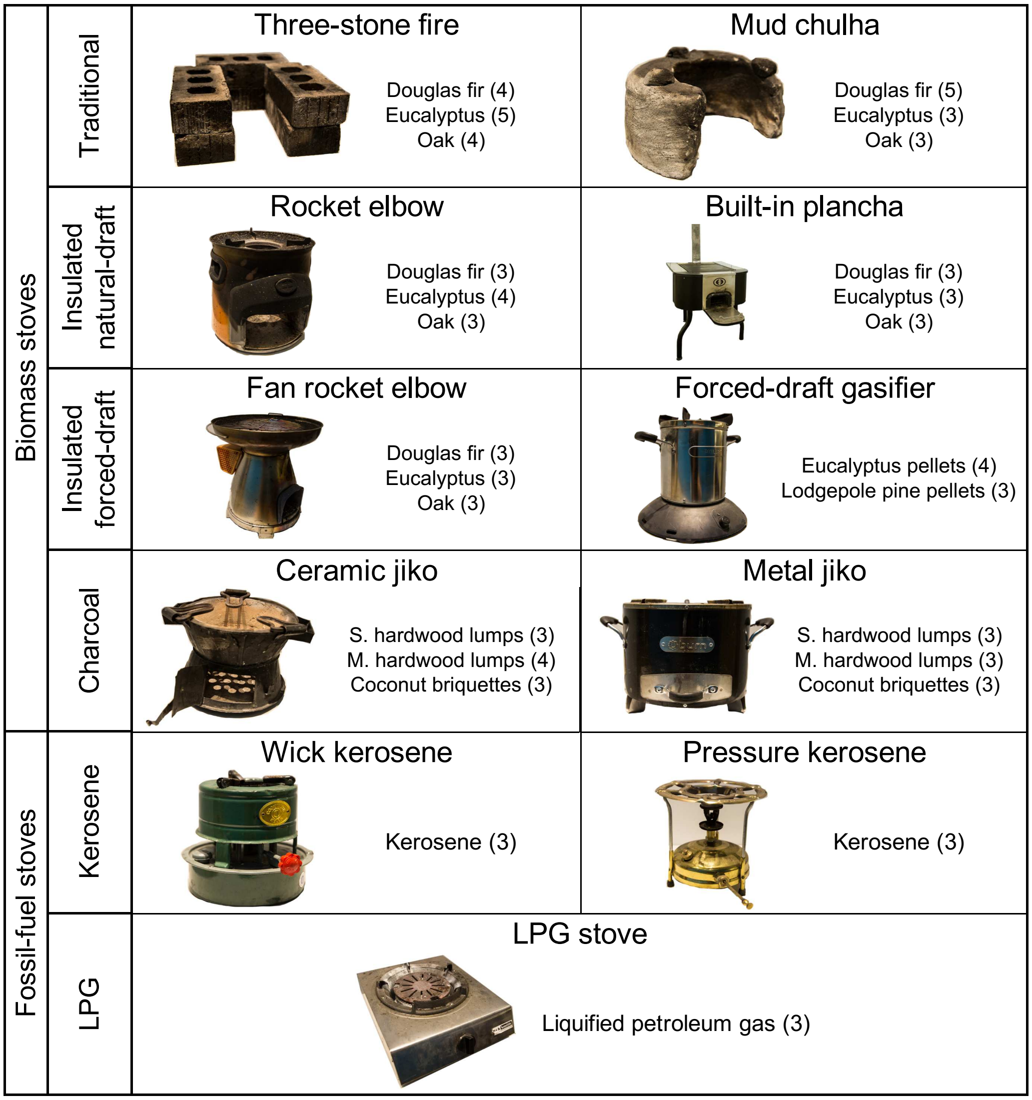


Figure 3.1: *Stove-fuel test matrix. The values in parentheses indicate the number of replicates conducted with each stove-fuel combination. Eighty-seven tests were conducted in total, which included three replicates for each stove-fuel combination and an additional nine make-up tests conducted on select stove-fuel combinations (to counterbalance replicates with substantial measurement issues). Makes and models of each stove design are presented in Appendix B1.*

3.2.2 Test Protocol

The Firepower Sweep Test was used for emissions testing; details of the protocol are provided elsewhere.[98] The protocol directs the user to operate the cookstove across a range of firepowers, in contrast to commonly-used laboratory protocols, which are primarily task based (i.e., boiling and simmering a pot of water).[98] Past work suggests that multiple-firepower testing captures a more realistic range of performance, relative to in-field use, than task-based testing.[98] Testing methodologies differed between continuous-feed biomass, batch-fed biomass, and fossil-fueled stoves due to differences in the combustion process among different stove-fuel combinations.[98] One emissions measurement was captured over the entire sweep (which did not include the stove's start-up and shut-down).

3.2.3 Emissions Measurements

A schematic of the instrumentation used in this study and approximate flow rates is given in Appendix B2. A custom-designed, total-capture hood was used for emissions testing. The hood flow rate was calibrated and monitored throughout the study using a hot-wire anemometer (Velometer AVM430-A; TSI Alnor; Huntington Beach, CA). Sample line flows were measured in triplicate before and after each test (mini-Buck Calibrator M-30; A.P. Buck, Inc; Orlando, FL).

Size-selective inlets (16.7 liters per minute, 2.5 µm cyclones; URG; Chapel Hill, NC) and isokinetic-flow rates were used for all particle-phase measurements. Teflon filters (46.2 mm, 2 µm PTFE $PM_{2.5}$ Air Monitoring Membrane Filter; Tisch International; North Bend, OH) were analyzed gravimetrically for $PM_{2.5}$ mass (MX5 Microbalance; Mettler Toledo; Columbus, OH). Quartz filters (Tissuquartz #2500QAT-UP; Pall Corporation; Port Washington, NY) were used to collect organic carbon and elemental carbon (OCEC Carbon Aerosol Analyzer 5L; Sunset Laboratory, Inc; Tigard, OR), organic carbon absorption artifacts (this filter was placed in-line behind the Telfon filter),[99] inorganic anions (DX-500 Ion Chromatography System; Dionex; Waltham, MA) and cations (ICS-3000 Ion Chromatography System; Dionex; Waltham, MA), particle-phase PAHs (Agilent 6890/5973 Inert, Agilent Technologies, Santa Clara, CA), and carbohydrates (1260 Infinity; Agilent Technologies; Santa Clara, CA).[100,101] Two polyurethane foam plugs (PUF Replacement P226-92; SKC-West, Inc; Fullerton, CA) were placed in series behind one of the quartz filters to measure gas-phase PAHs. To minimize contamination, the quartz filters were baked at 800°C and the polyurethane foam filters were sonicated in acetone and then in dichloromethane/methanol/hexane mixture (and then air dried) before testing. Filter housings and cartridges were cleaned first with dish soap and deionized water and then rinsed with a dichloromethane/methanol/hexane mixture before use.

VOCs were captured in a vacuum canister fitted with a critical orifice (CS1200ES Sampler; ENTECH Instruments, Inc.; Simi Valley, CA) and speciated using gas chromatography-mass spectrometry (GC-17A; Shimadzu; Kyoto, Japan).[102] Gas-phase aldehydes and ketones were collected using dinitrophenylhydrazine (DNPH) cartridges (Sep-Pak DNPH-Silica Cartridge; Waters Corp; Milford, MA) that were placed in-line behind an ozone scrubber (Sep-Pak Ozone Scrubber Potassium Iodide Cartridge; Waters Corp; Milford, MA) and analyzed using high-performance liquid chromatography (HPLC 1050 series; Agilent Technologies; Santa Clara, CA).

Online instrumentation included a particle sizer (Scanning Mobility Particle Sizer Spectrometer 3938, TSI Incorporated, Shoreview, Minnesota) for ultrafine particles, gas analyzers for CO, carbon dioxide (CO_2), and methane (CH_4) measurements (ULTRAMAT/OXYMAT 6 and ULTRAMAT 23; Siemens; Munich, Germany), and a thermocouple that measured the temperature of the water in the cooking pot (K-type, OMEGA Engineering Inc., Norwalk, CT). The scanning mobility particle sizer was set up after a secondary dilution system composed of a Venturi pump (Open Flow Venturi Single Stage

Vacuum Generator; Gast; Benton Harbor, MI) and a series of CO_2 sensors used to estimate secondary dilution rates (LI-820 CO_2 Analyzer; LI-COR; Lincoln, NE).

Laboratory blanks were collected daily. For time-integrated instrumentation, two-hour background measurements were conducted on a weekly basis (18 tests in total). For online instrumentation, five-minute background measurements were conducted at the beginning and end of each test.

3.2.4 Data Processing and Emissions Metrics

Data processing and analyses were conducted in R (version 3.4.1); the code is published on Github: https://github.com/nickgood/stoves_nih_2016_git. Emissions measurements were blank and background corrected and based on study-averaged measurements. Samples that were below a given analytic method limit of detection (LOD) were replaced with $LOD/\sqrt{2}$, and background-corrected values that were less than or equal to zero were replaced with zeros. Particle size distribution data were corrected for secondary dilution on a test-by-test basis (secondary dilution ratios ranged from 2.7 to 100). Particle losses in the venturi pump ahead of the SMPS were not corrected for in the post-analysis.

Fine particulate organic carbon measurements were converted to organic matter (i.e., primary organic aerosol). A conversion factor of 1.5 for both wood and pellet fuels and 1.2 for charcoal and fossil fuels was chosen. The conversion factor for wood was lower than those used in other studies (e.g., 1.9),[57,103] because the high ion emissions observed in this study would have led to an overestimation of $PM_{2.5}$ mass with a higher conversion factor. See Appendix B3 for a plot of the mass balance. The conversions factors chosen here are within the range of previous measurements.[73]

Ultrafine particle concentrations were calculated by summing the number of particles between the bins with median sizes closest to 10 and 100 nanometers (measured as electric mobility diameter).

We refer to compounds as carcinogenic if they have been classified as a "known" or "reasonably anticipated" human carcinogens by the National Toxicology Program[69] and/or classified as a Group 1- (carcinogenic to humans) or Group 2A- (probably carcinogenic to humans) compound by the International Agency for Research on Cancer.[70] Relevant compounds include benzene, formaldehyde, styrene, acetaldehyde, isoprene, benz[a]athracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, cyclopenta[cd]pyrene, dibenzo[a,h]anthracene, and indeno[1,2,3-cd]pyrene (see Appendix B4 for classifications). The IARC also classifies "indoor emissions from household combustion of biomass fuel," referring the entire smoke mixture, as Group 2A. However, since the following analyses are focused on the constituents of cookstove smoke we only included compounds that have been classified on a compound level in the analysis.

3.2.5 Regression Analysis

Linear regression models were used to evaluate whether $PM_{2.5}$ and CO (independent variables) can be used as predictors for other co-emitted species (dependent variables). Both individual pollutants (e.g., formaldehyde, isoprene, etc.) and pollutant groups (e.g., three-ring PAHs, ketones, etc.) were evaluated as outcomes (dependent variables). Seven models, using all permutations of two continuous variables ($PM_{2.5}$ and CO emissions) and one categorical variable (stove type), were compared to determine which models were the best predictors of emissions. Model coefficients of determination (R-squared values) were used to compare models. Because the R-squared metric favors models with more independent variables, model performance was also compared using the Bayesian information criterion. R-squared values are presented hereafter because the conclusions of the analysis were the same with both R-squared and the Bayesian information criterion, but the interpretation of R-squared is more intuitive.

For each pollutant (outcome), only observations that had both $PM_{2.5}$ and CO data were retained to ensure that R-squared values were comparable between models (i.e., nested). All pollutant variables were log-transformed to satisfy linear regression assumptions. Stove types with less than two data points for a particular pollutant or pollutant category (due to experimental issues or measurements that were below background) were excluded to prevent overfitting. Due to fewer replicates and more emissions that were below background, fossil fuel stoves were more likely to be excluded from the analysis than other stove types.

3.3 Results and Discussion

3.3.1 Overview

Emissions on a per-energy-delivered basis (mg/MJ_d) are presented here. In the following analyses, emissions from improved biomass stoves and fossil-fuel stoves are described relative to the three-stone fire, because the three-stone fire is the most commonly-used traditional cookstoves.[104] Several smoke constituents are presented by groups rather than as individual constituents including inorganic ions, which are grouped together, and VOCs, carbonyls, and PAHs, which are grouped by chemical structure (e.g., VOCs are grouped by bond complexity and PAHs are group by number of rings).

Total emissions and emissions composition on a per-energy-delivered basis varied widely among the stove types tested. There was also substantial variability in emissions measurements across replicates on each stove type. Compared to biomass stoves, fossil-fuel stoves reflected less variability, because they were limited to one fuel type and the operating conditions were more easily controlled. Wide variability in emissions is typical of in-home cooking practices; however, the variability seen in this study may still under represent the variability seen during in-home cooking factors that differ between the laboratory and field (e.g., fuel moisture content and fire tending practices).

3.3.2 $PM_{2.5}$ composition

Among the stoves tested, $PM_{2.5}$ emissions were highest from traditional biomass stoves followed by improved biomass stoves and fossil-fuel stoves (Figure 3.2). Relative to a three-stone fire, improved biomass stoves and fossil-fuel stoves reduced average $PM_{2.5}$ emissions by 44-70% and 99-100%, respectively. The decreased $PM_{2.5}$ emissions of fossil-fuel stoves, relative to biomass stoves, are likely attributable to the higher volatility of kerosene and LPG, which combined with the specific designs of these stoves, promotes more complete fuel-air mixing and, thus, more complete combustion. The pressurized kerosene and LPG stoves, in particular, employ the venturi effect to premix vaporized fuel with air.

Of the wood stoves tested, the insulated natural-draft stoves (rocket elbow: mean = 376 [range = 43-887] mg/MJ_d; built-in plancha: 361 [146-523] mg/MJ_d) and insulated forced-draft stoves (fan rocket-elbow: 124 [24-242] mg/MJ_d; gasifier: 266 [74-632] mg/MJ_d) had substantially lower average $PM_{2.5}$ emissions than the traditional stoves (three-stone fire: 670 [293-1609] mg/MJ_d; mud chulha: 998 [200-2117] mg/MJ_d) [Figure 3.2]. The lower $PM_{2.5}$ emissions of the insulated wood stoves, compared to traditional wood stoves, is likely attributable to better fuel-air mixing and reduced thermal losses, which help maintain the high temperatures needed to oxidize particulate matter.[105] Charcoal stoves also resulted in substantial reductions in total $PM_{2.5}$ emissions relative to the traditional stoves tested (ceramic jiko: 168 [72-330] mg/MJ_d; metal jiko: 202 [106-278] mg/MJ_d).

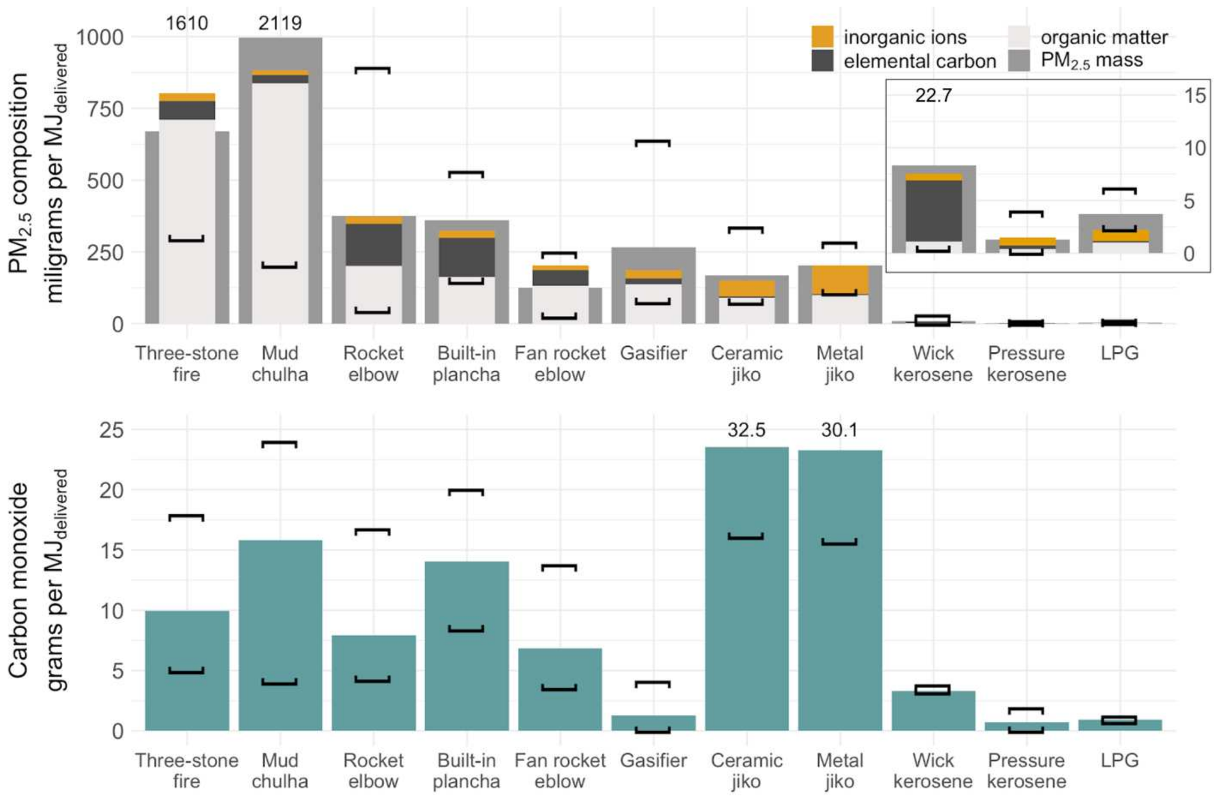


Figure 3.2: *Emissions of fine particulate matter ($PM_{2.5}$), elemental carbon, organic matter, inorganic ions, and carbon monoxide. The height of each colored bar represents replicate-averaged emissions for each stove type (including replicates across all fuel types). Square brackets indicate the range of emissions measured from each stove type (including all fuel types). On the top panel, the square brackets represent the range of $PM_{2.5}$ emissions, rather than the range of the summed constituents. For scaling purposes, numeric values are sometimes provided at the top of the plot rather than an upper bracket.*

Of the $PM_{2.5}$ constituents measured, primary organic matter constituted the largest fraction of $PM_{2.5}$ emitted for all stoves, except for the wick kerosene stove, which tended to emit more elemental carbon (Figure 3.2). Traditional biomass stoves had the highest organic matter emissions with improved biomass stoves representing a 72-87% and fossil-fuel stoves all representing a 99.9% decrease in average organic matter emissions relative to the three-stone fire. The traditional biomass stoves tested in this study were uninsulated and thus had greater heat loss to the environment, compared to improved biomass stoves. Greater heat loss likely leads to regions of lower localized combustion temperatures, which can promote organic aerosol formation.

The two insulated natural-draft stoves had the highest elemental carbon emissions of all the stoves tested. Relative to the three-stone fire, the rocket-elbow stove had a 124% average increase and the built-in plancha stove had an 105% average increase (other improved biomass stoves and fossil-fuel stoves had a 18-94% and 91-100% average decrease in elemental carbon emissions, respectively) (Figure 3.2). The increased elemental carbon emissions observed in insulated natural-draft cookstoves could be explained by the presence of the insulated combustion chambers of the improved stoves that were tested, which are likely to lead to higher temperatures that promote soot-particle formation and growth.

On average, the two charcoal stoves had the highest emissions of inorganic ions for all the stoves tested. The high average ion emissions from charcoal stoves were driven by the coconut briquette fuel, which emitted more inorganic ions (233 [70-345] mg/MJ_d) than particulate organic matter (47 [34-57] mg/MJ_d) and elemental carbon (1.4 [0.22-4.5] mg/MJ_d) combined (Figure 3.2); the high ion emissions were likely due to the high ash content of the coconut charcoal fuel. Relative to the three-stone fire, the ceramic jiko, metal jiko, and gasifier stoves had a 120%, 289%, and 10% increase in average ion emissions, respectively. Other improved biomass stoves (i.e., the rocket elbow, built-in plancha, fan rocket elbow stoves) 1-37% decrease, while fossil-fuel stoves had a 96-98% decrease in average ion emissions, relative to a three-stone fire.

3.3.3 Carbon monoxide

Charcoal cookstoves had the highest average CO emissions (ceramic jiko: 24 [16-32] g/MJ_d; metal jiko: 23 [16-30] g/MJ_d) [Figure 3.2]. Carbon monoxide emissions factors for the ceramic jiko and metal jiko stoves were 137% and 135% higher than the three-stone fire, respectively. The high CO emissions from charcoal-fueled stoves were likely attributable to the primary oxidation process of charcoal fuels. When charcoal is burned, oxygen reacts directly with the fuel surface to produce CO, often under conditions that yield lower heat-release rates, than wood-based fuels or fossil-fuels.[105] Lower heat release rates likely result in lower temperatures in the combustion zone, which can inhibit oxidation of CO to CO_2.

All improved wood stoves had decreased average CO emissions relative to the three-stone fire (rocket elbow: 20%; fan rocket elbow: 31%; gasifier: 87%) with the exception of the built-in plancha stove, which led to a 42% increase in average CO emissions (Figure 3.2). Advances in wood cookstove technology (e.g., improved insulation or the use of electric fans to promote mixing) resulted in lower average CO emissions compared to the three-stone fire. All fossil-fuel stoves emitted substantially less CO emissions that the three-stone fire, resulting in a 67%, 93%, and 91% decrease in average CO emissions of the wick kerosene, pressure kerosene, and LPG stoves, respectively.

3.3.4 Ultrafine particles

The fan rocket elbow had the highest ultrafine particle emissions on average (4.7e16 [2.1e15-7.8e16] particles/MJ_d) [Figure 3.3 - 13% higher than the three-stone fire. Other improved biomass stoves emitted from 24-68% fewer ultrafine particles than the three-stone fire. The smallest reductions in number of particles were from the gasifier (24%) and built-in plancha (29%), while the metal jiko (61%), rocket elbow (63%), and ceramic jiko (68%) resulted in more substantial reductions in ultrafine particle emissions. Fossil-fuel stoves had substantially larger reductions in ultrafine particles, relative to the three-stone fire, resulting in 97%, 98% and 89% reductions for the wick kerosene, pressure kerosene, and LPG stoves, respectively.

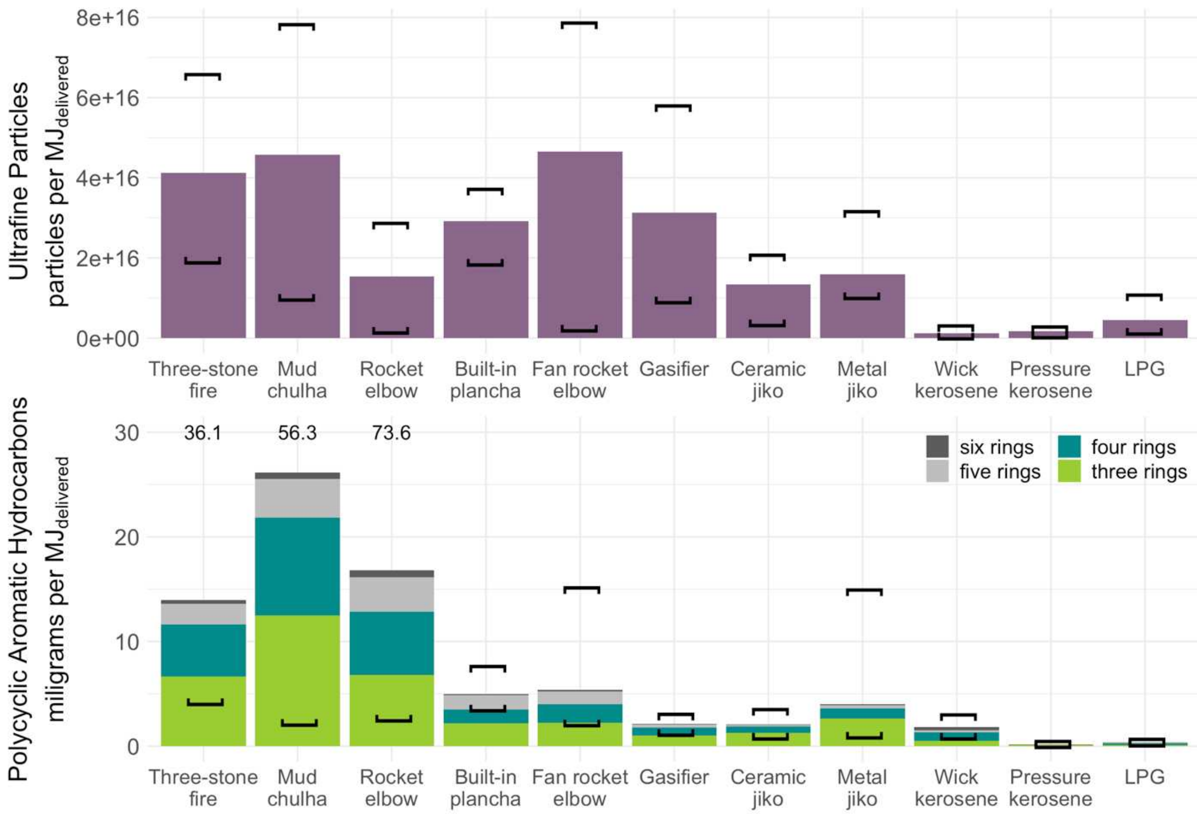


Figure 3.3: *Emissions of ultrafine particles (approx. 10-100 nm) and polycyclic aromatic hydrocarbons (PAHs). The height of each colored bar represents replicate-averaged emissions for each stove type (including replicates across all fuel types). Square brackets indicate the range of emissions measured from each stove type (including replicates across all fuel types). For scaling purposes, numeric values are sometimes provided at the top of the plot rather than an upper bracket.*

Ultrafine particles form via nucleation and condensation of organic exhaust gases or from incomplete oxidation of soot particles. Particles that originate as organic gas are likely to form in regions with lower combustion temperatures. Forced-draft biomass stoves (e.g., fan rocket elbow and gasifier) may have more low-temperature regions because internal fans push relatively cool ambient air into the combustion chamber to facilitate fuel-air mixing.[85]

3.3.5 Polycyclic aromatic hydrocarbons

The mud chulha stove had the highest average PAHs emissions (26 [2.2-55] mg/MJ_d) followed by the rocket elbow stove (17 [2.5-73] mg/MJ_d) and three-stone fire (14 [4.1-35] mg/MJ$_d$) [Figure 3.3]. Relative to the three-stone fire, the rocket elbow stove emitted 20% higher levels of PAHs, on average, while other biomass stoves reduced average PAH emissions by 61-85%. Stoves with higher PAH emissions often also had higher elemental carbon emissions, likely because PAHs are precursors for soot (i.e., elemental carbon) and thus forms under similar conditions (e.g., high-temperature, fuel-rich regions of the combustion chamber, which promote PAH formation and subsequent soot particle condensation and growth. Fossil-fuel stoves consistently resulted in substantial decreases in PAH emissions (wick kerosene: 87%; pressure kerosene: 99; LPG: 97%), relative to the three-stone fire.

On a mass basis, gas-phase PAH emissions were higher than particle-phase emissions (Figure 3.3). For example, three-ring PAHs, which are primarily found in the gas phase, made up 41-48% of total PAH emissions for wood stoves; 61% and 67% for the ceramic jiko and metal jiko respectively; and 26%, 54%, and 30% for the wick kerosene, pressure kerosene, and LPG stoves, respectively. Contrastingly, six-ring PAHs, which are primarily found in the particle phase, made up 1-4% of total PAHs for wood stoves; 3% for both charcoal stoves; and 17%, 35%, and 14% for the wick kerosene, pressure kerosene, and LPG stoves, respectively.

3.3.6 Volatile organic compounds

The total VOC emissions were highest for traditional biomass stoves including the three-stone fire (1157 [155-2251] mg/MJ$_d$) and mud chulha (2010 [1185-3254] mg/MJ$_d$) [Figure 3.4]. Relative to the three-stone fire, improved biomass stoves had a 69-92% decrease in total average VOC emissions. VOCs can be emitted if biomass fuel that has been volatilized escapes the combustion zone without being completely oxidized. Lower combustion temperatures in traditional stoves (due to poor thermal insulation and high excess-air ratios) could contribute to higher emissions of unburned hydrocarbons.

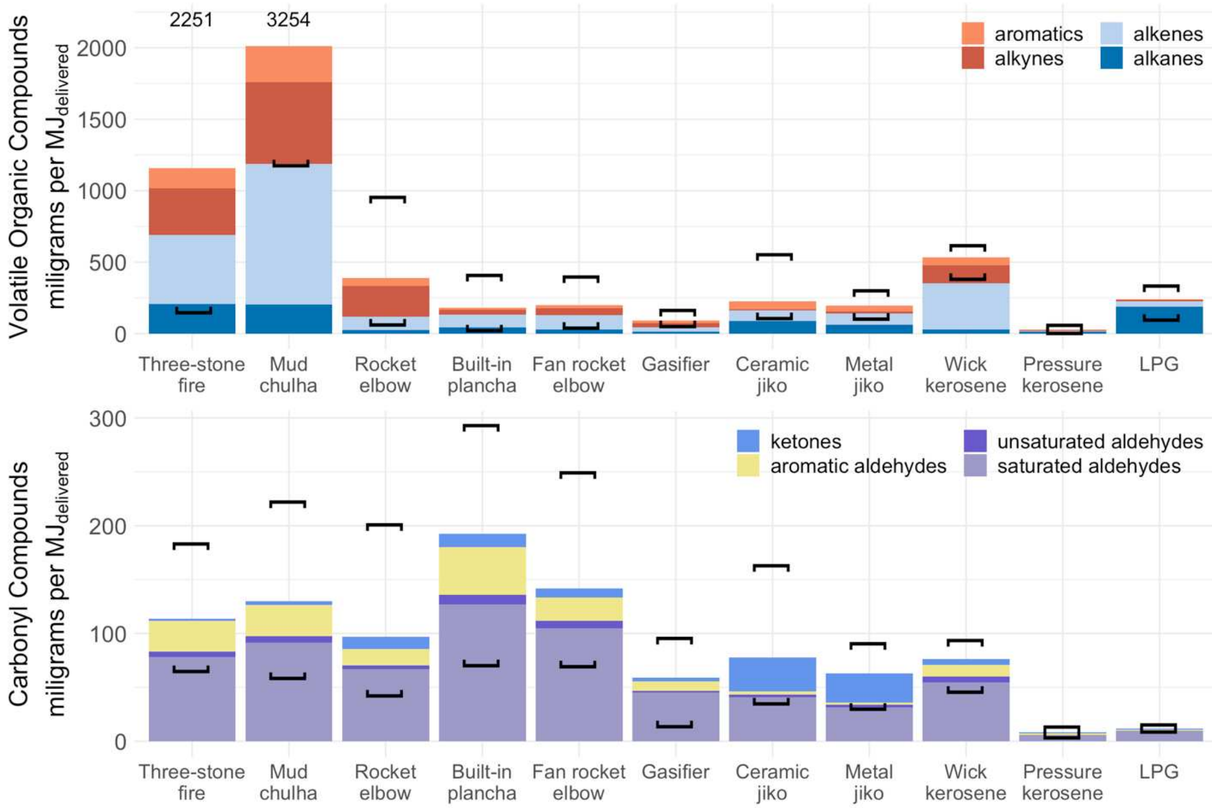


Figure 3.4: *Emissions of volatile organic compounds (VOCs) and carbonyls. The height of each colored bar represents replicate-averaged emissions for each stove type (including replicates across all fuel types). Square brackets indicate the range of emissions measured from each stove type (including replicates across all fuel types). For scaling purposes, numeric values are sometimes provided at the top of the plot rather than an upper bracket.*

The wick kerosene, pressure kerosene, and LPG stoves resulted in a 54%, 98%, and 79% decrease in emissions relative to the three-stone fire (Figure 3.4). In contrast to other pollutants, the wick kerosene stove (534 [388-608] mg/MJ_d) had higher VOCs emissions than any of the improved biomass stoves stoves and the LPG stove (239 [103-328] mg/MJ_d) emitted higher levels of VOCs than all improved biomass stoves except the rocket elbow. Ethene, an intermediary alkene in the kerosene oxidation pathway, was the most abundant compound emitted from the wick kerosene stove (50%). Propane, an alkane and a major constituent of LPG, was the most abundant VOC emitted from the LPG stove on average (75%), indicating that much of the VOC emissions from the LPG stove were from unburnt fuel.

VOC composition varied across stove type (Figure 3.4). Traditional biomass stoves and most improved biomass stoves emitted alkenes in the largest fraction (three-stone fire: 42%; mud chulha: 49%; built-in plancha: 50%; fan rocket elbow: 49%; gasifier: 34%; metal jiko: 40%), while the rocket elbow emitted the largest fraction of alkynes (59%) and the ceramic jiko emitted the most alkanes (39%). Of the fossil-fuel stoves tested, the pressurized stoves (pressure kerosene: 56%; LPG: 79%) emitted primarily alkanes, while the wick kerosene stove emitted primarily alkenes.

3.3.7 Carbonyl compounds

The built-in plancha (193 [71-299] mg/MJ_d) and the fan rocket elbow (142 [71-248] mg/MJ_d) emitted the most total carbonyls (Figure 3.4) - 70% and 25% more than the three-stone fire, respectively. Increased aldehyde emissions from these stoves might have occurred because both stoves have designs (i.e., a chimney or a fan) that lead to higher excess air ratios, which may lead to low-temperature regions where aldehydes are not completely oxidized.[105] The other improved biomass stoves that were tested, led to a 15-48% decrease in total carbonyl emissions relative to the three-stone fire. The wick kerosene, pressure kerosene, and LPG stoves led to a 33%, 93%, and 89% reduction in total carbonyl emissions relative to the three-stone fire. The wick kerosene reductions in total carbonyls were modest compared to other fossil-fuel stoves. As with the biomass stoves, the lower heat release rates associated with the wick kerosene stove could have resulted in lower temperatures and thus more carbonyl formation compared to the pressurized stoves.

Formaldehyde was the most abundant carbonyl compound emitted across all cookstoves, making up 39-44% of total carbonyl emissions, on average, for stoves wood-fuel stoves; 25% and 20% for the ceramic jiko and metal jiko, respectively; and 42%, 53%, and 60% for the wick kerosene, pressure kerosene, and LPG stoves, respectively. Acetaldehyde was also emitted in abundance from the ceramic jiko (25%) and metal jiko (28%) stoves.

3.3.8 Carcinogenic compounds

The mud chulha (193 [71-299] mg/MJ$_d$) and rocket elbow (193 [71-299] mg/MJ$_d$) emitted the highest levels of particle-phase carcinogenic compounds, representing a 56% and 38% increase relative to the three-stone fire, respectively (Figure 3.5). Other improved biomass stoves emitted 41-95% fewer particle-phase carcinogens (by mass), while the wick kerosene, pressure kerosene, and LPG stoves emitted 87%, 99%, and 97% fewer particle-phase carcinogens relative to the three-stone fire. All of the particle-phase carcinogens were also PAHs, thus total PAHs generally followed the same trends as the particle-phase PAHs.

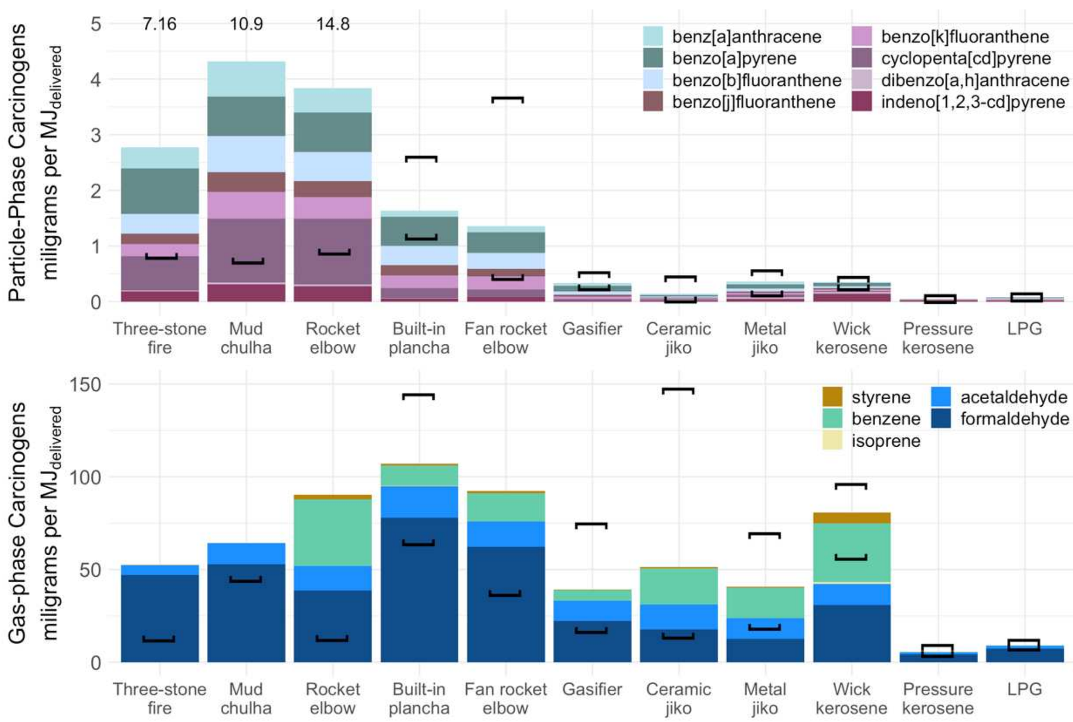


Figure 3.5: *Particle- and gas-phase emissions that are classified as "known" or "reasonably anticipated" human carcinogens. The height of each colored bar represents replicate-averaged emissions for each stove type (including replicates across all fuel types). Square brackets indicate the range of emissions measured from each stove type (including replicates across all fuel types). For scaling purposes, numeric values are sometimes provided at the top of the plot rather than an upper bracket.*

The mud chulha (237 [44-363] mg/MJ_d) and the three-stone fire (152 [12-239] mg/MJ_d) had the highest gas-phase carcinogen emissions. Improved biomass stoves had a 30-74% decrease in gas-phase carcinogens, while the wick kerosene, pressure kerosene, and LPG stoves resulted in 47%, 96%, and 94% decreases in gas-phase carcinogens, respectively. Benzene was the most abundant gas-phase carcinogen emitted from traditional wood stoves (three-stone fire: 66%; mud chulha: 63%), while the gas-phase carcinogens of other stoves were dominated by the carcinogenic carbonyls (i.e., formaldehyde and acetaldehyde).

3.3.9 $PM_{2.5}$ and CO as predictors for co-emitted pollutants

The results of the linear regression analysis are plotted in Figure 3.6. The R-squared values of the models for carcinogenic compounds (n = 13) and pollutant groups (n = 14) are plotted in Figure 3.6. Stove type (i.e., stove make/model) was the most important predictor of co-pollutant emissions. Stove type alone explained between 23-86% of the variability in emissions, while including one or more emissions variables in addition to stove type only resulted in slightly higher R-squared values ($PM_{2.5}$ and stove type: 24-87%; CO and stove type: 26-86%; $PM_{2.5}$, CO, and stove type: 26%-87%). This result indicates that adding information about $PM_{2.5}$ and CO does not add any additional information beyond knowing the stove type. Therefore, $PM_{2.5}$ and CO are likely not good proxies for co-emitted pollutants. Thus, reductions in $PM_{2.5}$ and CO might not necessarily result in proportional reductions of all harmful pollutants. This finding highlights an important issue because, currently, cookstove emissions standards only target $PM_{2.5}$ and CO. As a result, emissions from the majority of stove designs are not characterized beyond $PM_{2.5}$ and CO before the stoves are disseminated to users.

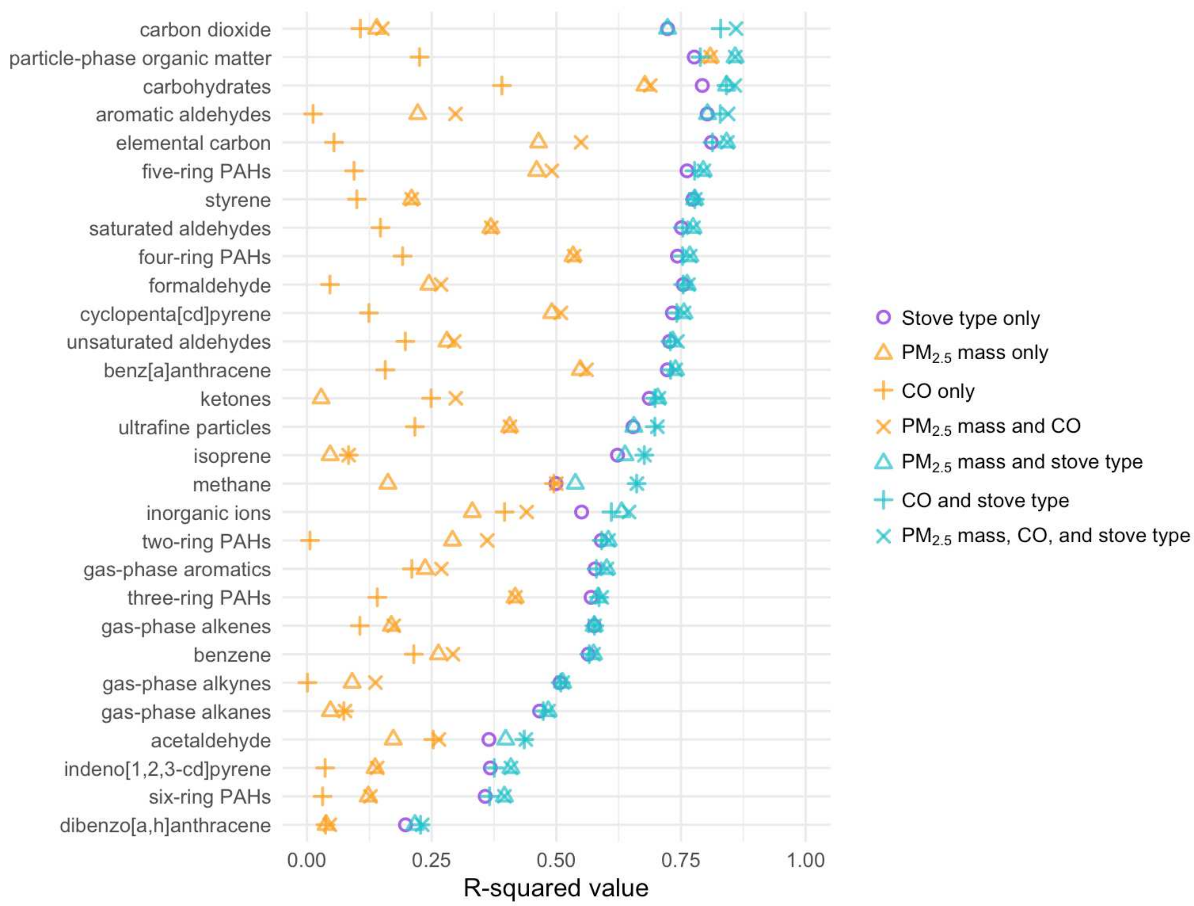


Figure 3.6: *Coefficients of determination (R-squared) values for linear regression models that use particulate matter ($PM_{2.5}$) mass, carbon monoxide (CO), and stove type (n = 11), as predictors of co-emitted pollutants.*

3.3.10 Synthesis and implications for cookstove research

We found above-background levels for 119 out of 120 cookstove smoke constituents observed in this study, evidencing the diversity of cookstove smoke constituents. Further, we find that total emissions and composition on a per-energy-delivered basis varied widely among the stove types tested and across test replicates (Figures 3.2-3.5). The latter, resulted in substantial overlap in the range of mass of emissions measured on a per-energy-delivered basis (or numbers, in the case of ultrafine particles). The variability is likely due to differences in stove operation across replicates and the different fuel types tested (in the case of biomass stoves). The variability demonstrated here is larger than other laboratory studies, but may still underpredict real-world variability.

Compared to the traditional biomass stoves tested, most improved biomass stoves emitted less air pollution per unit energy delivered (Figures 3.2-3.5). However, there were several exceptions. On average, relative to the three-stone fire, the rocket elbow had a 125% increase in elemental carbon, a 20% increase in PAHs, and a 38% increase in particle-phase carcinogens; the built-in plancha had an 105% increase in elemental carbon and a 70% increase in carbonyls; the fan-rocket elbow had a 13% increase in number of ultrafine particles and a 25% increase in carbonyl; and the gasifier had a 10% increase in ions. These results indicate that while the design features of improved wood stoves (e.g., insulated combustion chambers and enhanced fuel-air mixing) decreased emissions of many pollutants, they may also lead to increased levels of some smoke constituents due to the complex of biomass combustion. On average, relative to the three-stone fire, the ceramic jiko had a 120% increase in ions and a 137% increase in CO and the metal jiko had an 289% increase in ions and a 135% increase in CO. The increased ions and carbon monoxide were likely due to the differences in fuel composition (i.e., the higher ash content of the charcoal fuels tested) and the differences in charcoal combustion (primarily surface oxidation) compared to flaming combustion of wood fuels, respectively.

Of the smoke constituents measured, the LPG stove consistently had substantial emissions reductions relative to the three-stone fire (>79% decrease) [Figures 3.2-3.5]. The lower emissions are most likely attributable to the fact that the pressurized LPG stove design premixes vaporized fuel with air prior to combustion. Recently, stakeholders have begun promoting cleaner, liquid or gas fueled stoves (e.g., LPG, ethanol, etc.) over improved biomass stoves, arguing that improved biomass stoves do not reduce emissions substantially enough to provide substantial health or environmental benefits. We found that the emissions from the LPG stove were consistently lower than the improved biomass stoves across the pollutants observed in this work [Figures 3.2-3.5]; however, LPG stoves may not always be a feasible alternative to traditional biomass stoves in regions that lack access to LPG infrastructure.

Finally, we find that the ISO performance targets (i.e., absolute values of $PM_{2.5}$ and CO) can likely only be used to predict a subset of cookstove smoke constituents that are co-emitted with $PM_{2.5}$ and CO. Instead, developing stove-type-specific models or conducting comprehensive emissions assessment that target co-emitted pollutants of interest would likely be more informative.

CHAPTER 4: COUPLING LABORATORY AND FIELD MEASUREMENTS TO ESTIMATE AIR POLLUTANT EMISSIONS FROM COOKSTOVES

4.1 Introduction

Strong evidence suggests that air pollution emissions from solid-fuel cookstoves affect air quality and public health.[4,8,22] Accurate emissions data are critical inputs for models that aim to quantify cookstove impacts. Currently, emissions measurements are primarily derived from laboratory experiments where stoves are tested under controlled procedures. Laboratory studies have the potential to provide repeatable measurements at relatively low costs. Additionally, many emissions measurements that are crucial for understanding the impacts of cookstoves can only be feasibly measured in a laboratory (e.g., particle composition distributions and the atmospheric evolution of aerosols). However, most laboratory studies, to date, have systematically underestimated the magnitude and variability of emissions measured during in-home cooking practices.

Alternatively, field data collected during in-home cookstove use provide realistic emissions data.[57,106] However, field campaigns are logistically difficult. For example, the homes in many field testing sites do not have road access or access to electricity. Thus, low-cost battery-powered sensors must be used, the field, in place of high-quality stationary equipment that can be housed in laboratory testing facilities. Field campaigns are also cost prohibitive. The expenses associated with field campaigns limit both the number of measurements taken within a household and the number of households that can be monitored. Therefore, field data typically provide a limited view of cookstove emissions within a specific home or region, especially given the diversity of stove technologies, fuel types, and cooking practices that need to be characterized to understand the impacts of cookstoves on a global scale.

Descriptive analyses that compare cookstove laboratory and field emissions have been published;[98,107] however, analytical tools that link the two have not yet been established. For other combustion sources, such as biomass burning, models have been developed to provide some closure between laboratory- and field-emissions data. For example, several laboratory-based biomass-burning studies generated models that used modified combustion efficiency (MCE) to predict emissions factors; the models were then used to estimate emissions from wildfires using MCEs measured in the field.[108,109]

Previous studies on cookstoves have reported that stove firepower has modest predictive ability for some emissions.[42,43,98] However, models developed in the laboratory have not yet been combined with field-derived firepower data to estimate in-home cookstove emissions. To apply this technique to cookstoves, firepower measurements from the field are needed. The predominate technique for estimating firepower during an in-home cooking event is using a total-capture hood and the carbon-balance approach. This methodology is costly and intrusive, because it requires the construction of an emissions hood in a user's home and the operation of expensive (and cumbersome) gas analyzers. Another approach is using a real-time scale to track the fuel burn rate; however, this technique is not feasible for built-in stoves and can be complicated by the fact that the weight of the meal being prepared must be accounted for.[110]

To estimate in-home cookstove emissions using lab-derived models that use firepower to predict emissions, better techniques are needed to measure firepower when the use of total-capture hoods is not practical. Furthermore, given the recent publication of a standardized multiple-firepower testing approach, through the International Standards Organization (ISO

19867-1:2018); a low-cost technique for estimating firepower in the field could provide useful information for deciding which firepowers a stove should be tested at in the laboratory. In this work we (1) present a low-cost methodology for estimating stove firepower continuously in a field setting using temperature measured at the combustion chamber outlet and (2) estimate emissions from in-home cooking practices using laboratory-derived emissions models and firepower estimates from the field.

4.2 Materials and Methods

4.2.1 Overview of field and laboratory campaigns

Emissions measurements were made in ten homes in each of three field sites: Honduras, India, and Uganda. These locations were chosen to represent a variety of stove technologies and cooking practices across different cultures. Further details on the field sites are provided by Eilenberg et al..[57] In Honduras, built-in stoves with large metal griddles (i.e., planchas) and chimneys were tested. In India, traditional mud chulha stoves, which were built locally, and manufactured rocket-elbow stoves were tested. In Uganda, single pot ceramic charcoal stoves (i.e., ceramic jikos) were tested. Fuels were sourced locally by the cookstove user. The primary fuel in both Honduras and India was collected wood, while in Uganda charcoal was purchased at local vendors. At each home, measurements were conducted over the course of one individual cooking event (starting when the stove was lit and ending when the stove was shut down). All cooking events were uncontrolled, meaning the field team adopted all practical means possible to avoid influencing the user's practices.

In the laboratory, we tested stoves with similar (but not identical) designs to those that were used in the field. Side-by-side photos of the laboratory and field counterparts of each stove type are provided in Appendix C1. Two wood fuel types, Douglas fir and eucalyptus, were tested on continuous-feed wood stoves and two charcoal fuel types, hardwood lumps and coconut briquettes, were tested on the batch-fed charcoal stoves. Three replicate tests were conducted for each stove-fuel combination. Stoves were operated using the Firepower Sweep Test protocol during laboratory tests.[98] Briefly, this protocol instructs the user to operate the cookstove continuously across a range of targeted firepower levels, in order to quantify emissions across the full-range of a stove's working firepower output.[98]

4.2.2 Firepower measurements

The operational firepower of a cookstove is defined as the rate of heat release from combustion. Firepower can be modeled using the following equation:

$$FP = LHV_{fuel} \cdot \dot{m}_{fuel} \tag{4.1}$$

where FP is the operational firepower of the stove, LHV_{fuel} is the lower heating value of the fuel, and $\dot{m}_{fuel}$ is the burn rate of the fuel. Different firepowers can be achieved in a cookstove by varying the fuel-feed rate, the mass of fuel in the combustion chamber, or the fuel spacing. Changing firepower in a solid-fuel stove is analogous to turning the power knob on a modern electric or gas stove, where high firepowers are used for tasks like boiling water or searing meat, while low firepowers are used for tasks like simmering rice or legumes.

For tests conducted in both the laboratory and the field, fuel lower heating values were estimated by measuring the higher heating value of each fuel (C 200 Calorimeter System; IKA; Staufen, Germany) and subtracting the heat of vaporization of the water produced. Fuel burn-rate measurements, however, differed between the laboratory and the field. In the laboratory, the mixing ratios of carbon dioxide (CO_2) and carbon monoxide (CO) were measured downstream of the total-capture emissions hood. Then, the carbon balance approach was used to estimate fuel burn rate.[98] With this approach, firepower was tracked in real-time. In the field, firepower was measured using a time-integrated approach. The mass of fuel burned across an entire cooking event and the the duration of the cooking event were measured and used to calculate an average fuel burn rate. Tracking the fuel weight continuously throughout the cooking events was not feasible in this study, because the majority of stoves were built in (i.e., mud chulha and plancha stoves).

Previous work has demonstrated that temperature measured at the outlet of the combustion chamber of an improved cookstove can be used as a proxy for firepower.[42,43] To investigate this relationship, thermocouples were used to measure combustion exhaust temperatures in both the laboratory and the field (K-type thermocouples; OMEGA Engineering Inc.; Norwalk, CT). Photos illustrating thermocouple installation are provided in Appendix C2. In the laboratory, thermocouple data were recorded throughout the Firepower Sweep Test. In the field, thermocouple data were recorded for approximately two weeks using data loggers (OM-CP-TC101A; OMEGA Engineering Inc.; Norwalk, CT) wrapped in a custom-made layer of protective silicone. In the field, lengthy precautions were taken to ensure that the thermocouple placement was not moved relative to the flame across thermocouple deployments. Photos of securement mechanisms are also provided in Appendix C2.

In the lab and the field, thermocouple data were logged at 1.0 hertz and 0.5 hertz, respectively. In both locations, the ambient temperature, measured on the day of the test, was subtracted from all combustion chamber outlet temperature measurements. Therefore, the data presented in this work represent an increase in temperature (above ambient) due to combustion within the stove.

4.2.3 *Emissions measurements*

Laboratory emissions were characterized using a custom-designed, total-capture hood, as described by Eilenberg et al.[98] Emissions measurements included fine CO, CO_2, particulate matter ($PM_{2.5}$), and black carbon (BC). Separate emissions measurements were conducted during each of the Firepower Sweep Test protocol's targeted firepower points, as well as during start-up, shut-down, and refueling (for batch-fed stoves).

Field-based emissions were characterized using, a custom-designed portable emissions sampler. A six-arm probe, with several inlets along the length, was installed approximately one meter above non-chimney cookstoves or at the chimney exit. Emissions measurements included CO, CO_2, $PM_{2.5}$, and BC. Details of the data collection and processing are available elsewhere.[57]

4.2.4 *Modified combustion efficiency*

MCE was measured using the same approach in both the laboratory and the field, using the following relation:

$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} \tag{4.2}$$

where ΔCO_2 and ΔCO were the background-corrected mixing ratios of CO_2 and CO, respectively.[57] MCE is a proxy for the extent to which combustion is complete, because CO is the most abundant product of incomplete combustion. Values close to 1 indicate more complete combustion, while lower values indicate less complete combustion. MCE considered to be a reasonable proxy for actual combustion efficiency and is widely-used to indicator of cookstove performance. MCE is also used to indicate whether the combustion conditions are primarily flaming or smoldering, with higher MCEs indicating flaming combustion and lower MCEs indicating smoldering combustion.

4.2.5 *Linear model development*

Data collected in the laboratory was used to develop linear regression models to predict cookstove emissions as a function of stove firepower (continuous; fixed effect), MCE (continuous; fixed effect for predicting $PM_{2.5}$ emissions only), and stove type (categorical; fixed effect). Specific information about the model development is provided in Bilsback et al..[98] Briefly, outcomes included log-transformed values for $PM_{2.5}$,, CO, and BC emissions. Several models included an interaction term with stove type, which resulted in a independent model (i.e., unique slope and intercept) for each type of stove. The present work (unlike in Bilsback et al.)[98] did not consider fuel type as a fixed effect, for the sake of model simplification. In the field, fuels were sourced locally. Therefore, there was not always overlap between the exact fuel types tested in the laboratory and the field. To ensure that the models remained generalizable, only stove type (and not fuel type) was used as a categorical predictor. Additionally, the models from Bilsback et al.[98] were refit using emissions rates (i.e., emissions on a time basis) as outcomes, rather than emissions factors (i.e., emissions on a per-fuel-mass basis).

4.2.6 *Laboratory-to-field emissions estimation*

Based on the results of Bilsback et al.,[98] the following three models were used to predict field emissions:

$$ln(PM_{2.5}\ Rate) = \alpha_i \tag{4.3}$$

$$ln(PM_{2.5}\ Rate) = \alpha_i + \beta_i \cdot FP \tag{4.4}$$

$$ln(PM_{2.5}\ Rate) = \alpha_i + \beta_i \cdot FP + \gamma_i \cdot MCE \tag{4.5}$$

where FP is firepower; MCE is modified combustion efficiency; α_j are stove-specific intercepts; and β_j and γ_j are stove-specific slope coefficients. These three models represent increasing measurement complexity. Equation 4.3 is the baseline model, which represents only considering what stove type is being used (with no in-field measurements). Equations 4.4 and 4.5 represent adding a continuous firepower measurement and continuous MCE measurement in the field, respectively.

In the models where firepower was a predictor, real-time in-use firepower was estimated from the combustion chamber outlet temperatures. Household-specific conversion factors was derived to transform the temperature measured at the combustion chamber outlet to firepower for each household:

$$FP_i = exp\left(\frac{log(FP_{avg,H})}{\Delta T_{avg,H}} \cdot T_i\right) \tag{4.6}$$

where $FP_{avg,H}$ and $\Delta T_{avg,H}$ were the cooking-event averaged firepower and change in temperature (above ambient temperature), respectively and T_i and FP_i were individual temperature and firepower measurements made within the same household where the average measurements was taken. This conversion factor was used to estimate continuous firepower (at 0.5 hertz) over two-week thermocouple deployments.

One-minute averaged firepower and MCE estimates from each in-field cooking event were used to assess whether the laboratory emissions models had predictive ability for field data. Given the non-linear nature of the laboratory-derived models, continuous firepower and MCE measurements provided better estimates than cooking-event average measurements. The one-minute emissions estimates were averaged over the cooking event to compare model predictions to the test-averaged emissions measured in the field.

4.3 Results and Discussion

4.3.1 Temperature versus firepower in the laboratory

Scatter plots of temperature measured at the combustion chamber outlet versus log-transformed firepower are shown in Figure 4.1. Stoves with similar designs were grouped together. Each of the linear fits in Figure 4.1 represents a distinct test replicate (i.e., a unique Firepower Sweep Test) to demonstrate the reproducibility of the relationship between temperature and firepower for independent tests conducted for each stove type.

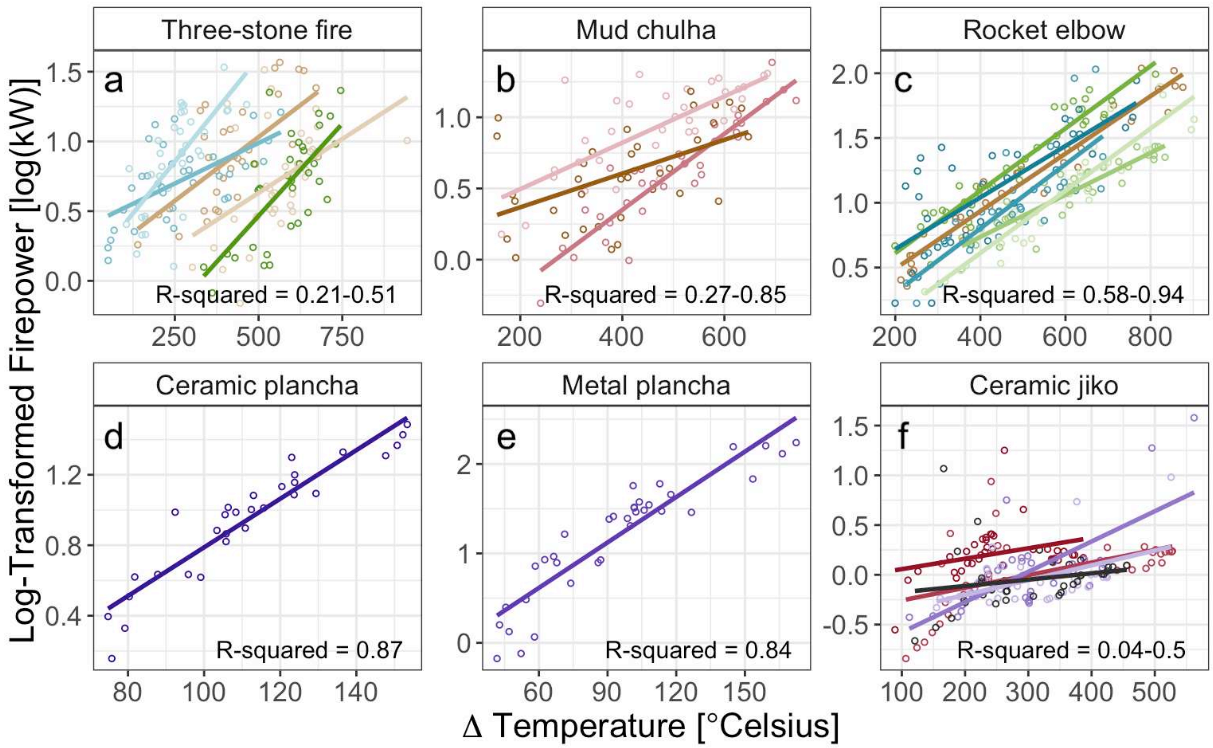


Figure 4.1: *Change in temperature (above ambient) at the combustion chamber outlet versus log-transformed firepower. Each linear fit represents a distinct test replicate (i.e., a unique Firepower Sweep Test). The range of model coefficients of determination (R-squared values) calculated across replicates are provided for each stove type.*

Overall, the data indicate that temperature measured at the combustion chamber outlet and log-transformed firepower are well-correlated. Although the strength of the correlation between temperature and log-transformed firepower varied by stove type, we find that the linear nature of the relationship, which has been documented elsewhere for a rocket-elbow stove,[42,43] is generalizable to other stove designs.

The relationship was weakest for stoves that did not have an enclosed combustion chamber (i.e., the three-stone fire [Figure 4.1a] and mud chulha [Figure 4.1b]). Due to the open combustion zones on these stoves, the thermocouple probe did not always sit a consistent distance away from the flaming fuel. For example, if the fuel bed shifted within the combustion zone, or new fuel was added, the fuel bed could move farther away from the probe, causing the temperature to decrease even if the stove's average firepower remained relatively stable. This circumstance is demonstrated by the wide range of firepower values measured at low temperatures in the mud chulha (Figure 4.1b). We found that the majority of the events where temperature fluctuations were not related to a change in firepower were short-lived. Thus, longer averaging periods generally led to stronger correlations between temperature and log-transformed firepower for stoves that had an open combustion chamber. Three-minute averages are depicted in Figure 4.1.

The open nature of the three-stone fire and mud chulha stove geometries also resulted in changes of the slope of the best-fit line between replicate Firepower Sweep Tests (Figure 4.1a and 4.1b). Between experiments, the stoves were taken down from the emissions testing platform, stored, and set up back in the emissions testing hood (often by different personnel, on a different day). This procedure resulted in variability in thermocouple placement at the outlet of the poorly defined "combustion chamber" and, thus, variability in the slope between temperature and log-transformed firepower between replicate tests.

Chimney stoves (i.e., the ceramic plancha and metal plancha) had the strongest correlations (Figure 4.1d and 4.1e). On chimney stoves, the thermocouple was installed at the base of the chimney, in the center of the pipe, by drilling a small hole into the side of the chimney. The thermocouple was situated much farther from the firebox than on stoves without chimneys and was held in place securely. The exhaust temperatures measured in chimney stoves were lower, but had higher correlations with firepower, likely because: (1) the exhaust gas temperature measured by the thermocouple was not affected by direct contact with the flame and (2) the placement of the thermocouple was more consistent from test to test.

Strong correlations were also observed on non-chimney stoves that had an enclosed combustion chamber (i.e., the rocket elbow), as shown in Figure 4.1c. In the absence of a chimney, the exhaust gas temperatures measured by the thermocouple may have been affected by direct contact between the thermocouple and the flame; however, the structure of the enclosed combustion chamber encouraged consistent thermocouple placement. This result supports the assertion that consistent thermocouple placement is important for maintaining the fidelity of the relationship between temperature and log-transformed firepower. Measurements taken using the rocket-elbow stove also resulted in little variation in the slope of the relationship between temperature and log-transformed firepower between replicate tests (Figure 4.1c). Thus, the relationship between temperature and log-transformed firepower was preserved between test replicates.

Overall, weaker correlations were observed on batch-fed charcoal stoves (Figure 4.1f). Given that the batch-fed ceramic jiko stove had a relatively open combustion chamber, compared to other stoves, the thermocouple location could have been affected by the refueling process. Furthermore, the large fluctuations in firepower that occurred during refueling did not always result in a consistent change in temperature at the combustion chamber outlet. However, strong correlations between temperature and log-transformed firepower were observed during steady-state operation of charcoal cookstoves.

Based on (1) the fact that the temperature was strongly correlated with firepower, for a specific stove design, and (2) the importance of consistent thermocouple placement to achieving a consistent relationship between temperature and firepower, a household-specific conversion factor was used to estimate firepower from field-based measurements of temperature at the combustion chamber outlet for a given stove (Equation 4.6).

To date, there is relatively little information available on in-use firepower for different cookstove designs. Multiple-firepower testing has recently been proposed as an alternative to task-based performance testing and has gained traction through the new laboratory protocol published by the International Standards Organization (ISO 19867-1:2018). Gathering information about the range of firepowers over which cookstoves are operated during in-use cooking could help inform the firepower levels that stoves should be tested at using the newly established protocol.

4.3.2 Operating conditions in the laboratory and field

Previous work suggests that a key component to establishing a relationship between the laboratory and field emissions data is ensuring that the range of operating conditions tested in the laboratory match (or overlap) with operating conditions observed in the field.[98] Thus, the range of firepower (calculated using a carbon balance in the laboratory and Equation 4.6 in the field) and MCE levels measured during the laboratory and field campaigns are plotted in Figure 4.2.

The degree of overlap between the range of firepower and MCE levels measured in the laboratory and field varied by stove type. For single-pot stoves with semi-enclosed combustion chambers (i.e., the rocket-elbow and ceramic-jiko stoves), there was good agreement between the range of firepowers and MCEs measured in the laboratory and the field. For the mud chulha stove, there was good agreement between the laboratory and field data at lower firepowers; however, the lab data missed the high-firepower points seen in the field. The discrepancy between the range of firepowers measured in the laboratory and field is likely due to a difference in the size of the mud chulha stoves tested in both settings. In the laboratory, the mud chulha stove was a single-pot semi-circle ring of cement, while in the field the mud chulha stoves were made locally and often were often large enough to hold two or even three pots (Appendix C1).

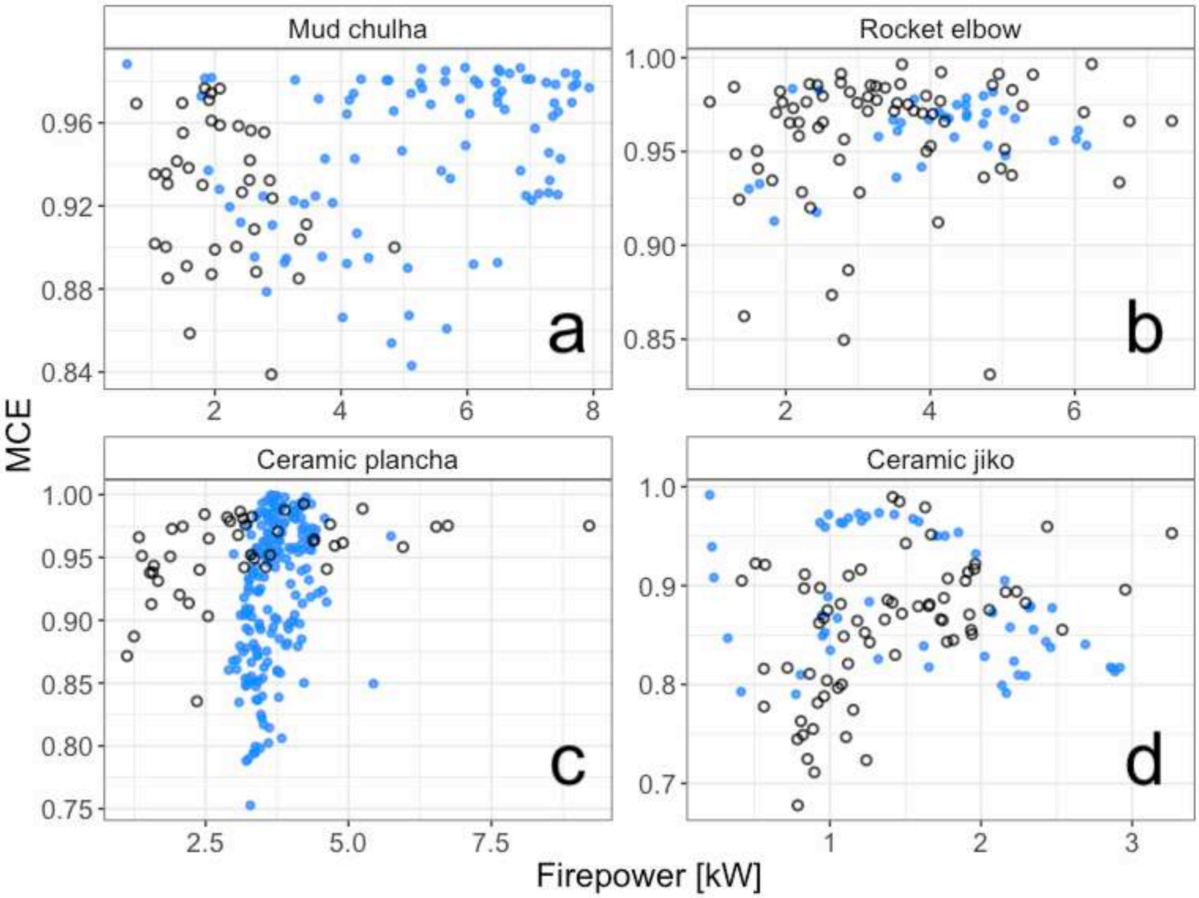


Figure 4.2: *Scatter plots demonstrating the range of firepower and modified combustion efficiency (MCE) levels measured in the lab (black) and the field (blue) for selected stove designs. Laboratory data was derived from the Firepower Sweep Test and field data were gathered from in-use cooking events in Honduras, India, and Uganda.*

There were also discrepancies in the range of operating conditions covered by the ceramic plancha stove in the laboratory and the field. In contrast to the mud chulha stove, however, the laboratory data covered the range of firepower seen in the field but not the range of MCEs measured in the field. In-use cooking patterns in Honduras demonstrated that the majority of built-in plancha stoves were operated continuously throughout the day. These cooking patterns led to more smoldering events (i.e., low MCEs at mid-range firepowers) than were captured in the laboratory setting. Only short-duration smoldering events were represented in the Firepower Sweep Test for continuous-feed wood stoves (approximately seven minutes during shutdown), while the smoldering events observed in Honduras were likely longer duration, possibly resulting in a different emissions profile as the stove bed and combustion chamber cooled.

4.3.3 The relationship between firepower and emissions in the laboratory and field

Before firepower was used to predict in-field emissions, we wanted to examine whether the relationship between firepower and measured emissions was conserved between the laboratory and field. Plots of firepower (calculated using a carbon balance in the laboratory and Equation 4.6 in the field) versus log-transformed measured $PM_{2.5}$ emissions, are shown in Figure 4.3 for both lab and field emissions. First-order linear regression model fits are overlaid on the correlation plots with and 95% confidence intervals. The linear fits were derived using the segment-averaged Firepower Sweep Test data and test-averaged field data are overlaid to validate the laboratory-based predictions.

Almost all of the field measurements, except for three measurements on the mud chulha stove (which captured higher firepowers than were tested in the laboratory), fell within the scatter of the relationship between firepower and log-transformed $PM_{2.5}$ laboratory data. This result is important, because it implies that the relationship between firepower and log-transformed $PM_{2.5}$ emissions is conserved between the laboratory and the field. The overall relationship between firepower and log-transformed $PM_{2.5}$ emissions is modest, likely because these linear regression plots do not contain additional variables such MCE and fuel type. However, the results shown in Figure 4.3 confirm that the relationship between firepower and emissions is conserved between the laboratory and the field; this result is an important advancement for connecting laboratory and field emissions and also serves to validate recent efforts by the community to encourage multiple-firepower testing in the laboratory setting (e.g., ISO 19867-1:2018).

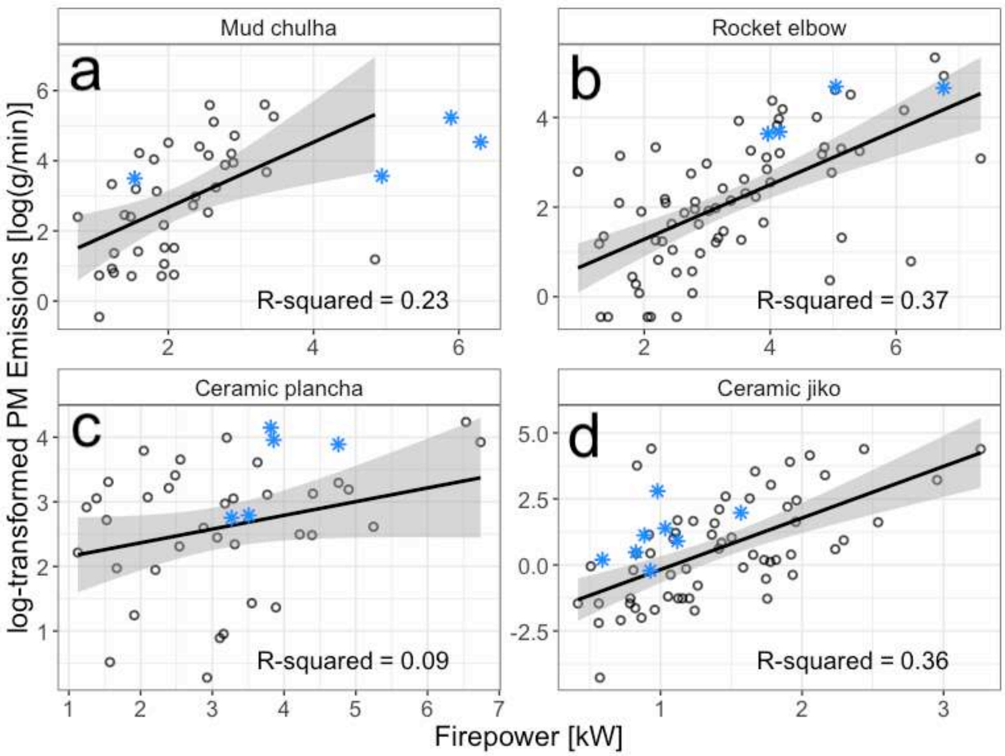


Figure 4.3: *Firepower versus measured emissions with lab data depicted by black markers and field data depicted by blue markers. The first-order linear regression line of best fit and 95% confidence interval is shown for each stove type. Only lab data were used to derive the line of best fit. The range of model coefficients of determination (R-squared values) calculated across replicates are provided for each stove type.*

The strength of the correlation between the laboratory measurements of firepower and emissions reported here is lower than in previous reports,[98] because fuel type was not included as a predictor in the models reported here (Figure 4.3). We chose not to include fuel type because a laboratory model that includes stove-fuel combination as a predictor (rather than just stove type) is less generalizable. Cookstove users typically collect their fuels in the area surrounding their homes or purchase fuel from local vendors. These fuel sourcing practices may result in a variety of fuel species being used simultaneously and thus more scatter in the relationship between firepower and emissions during in-home cookstove use. Developing a stove-fuel specific model would likely be more accurate than stove type alone. However, this approach would require substantially more data. We recommend that future work investigate the sensitivity of adding information about the fuels being used to the models.

The strength of the relationship between firepower and emissions in the laboratory varied by stove type. For example, the rocket-elbow stove had the strongest correlation between firepower and log-transformed $PM_{2.5}$ emissions (Figure 4.3b). The strength of this relationship is likely facilitated by the rocket-elbow cookstove design, which includes an insulated, enclosed combustion chamber. This geometry provides relatively consistent combustion conditions (e.g., combustion chamber geometry) at similar firepowers across the cooking cycle. Contrarily, emissions from traditional stoves that have larger, open combustion chambers are likely to be influenced by additional factors, beyond firepower, (e.g., stone placement on a three-stone fire).

4.3.4 Predicting emissions during an in-field cooking event

After evaluating (1) whether there was agreement between the operating conditions measured in the laboratory and the field and (2) whether the firepower and emissions followed the same trend in the laboratory and the field, three linear models were used to predict $PM_{2.5}$ emissions in the field: one that only used stove type as a predictor (Equation 4.3); one that used firepower and stove type as predictors (Equation 4.4); and one that used MCE, firepower, and stove type as predictors (Equation 4.5). The results of the model predictions are shown in Figure 4.4. The voluntary indoor emissions tiers published by the International Standards Organization (19867-3:2018) are also represented in Figure 4.4. Uncontrolled testing methodologies were used during field measurements. Thus, emissions factors on a per-energy-delivered basis were not measured. Instead, we compare the data collected to the indoor emissions tiers, which are presented as emissions rates. The chimney stoves tested during this study may technically be classified under the tiers presented in Figure 4.4 since the emissions measurements were made at the chimney exit. However, for model validation purposes we compared to the levels of the ISO emissions tiers. If the data falls into the shaded region used to represent one of the ISO tiers, this indicates that the stove has been classified correctly. If the data falls below the shaded region, this indicates that the model underestimated the emissions tier of that stove, while if the data point falls above the shaded region, the model overestimated the emissions tier of that stove.

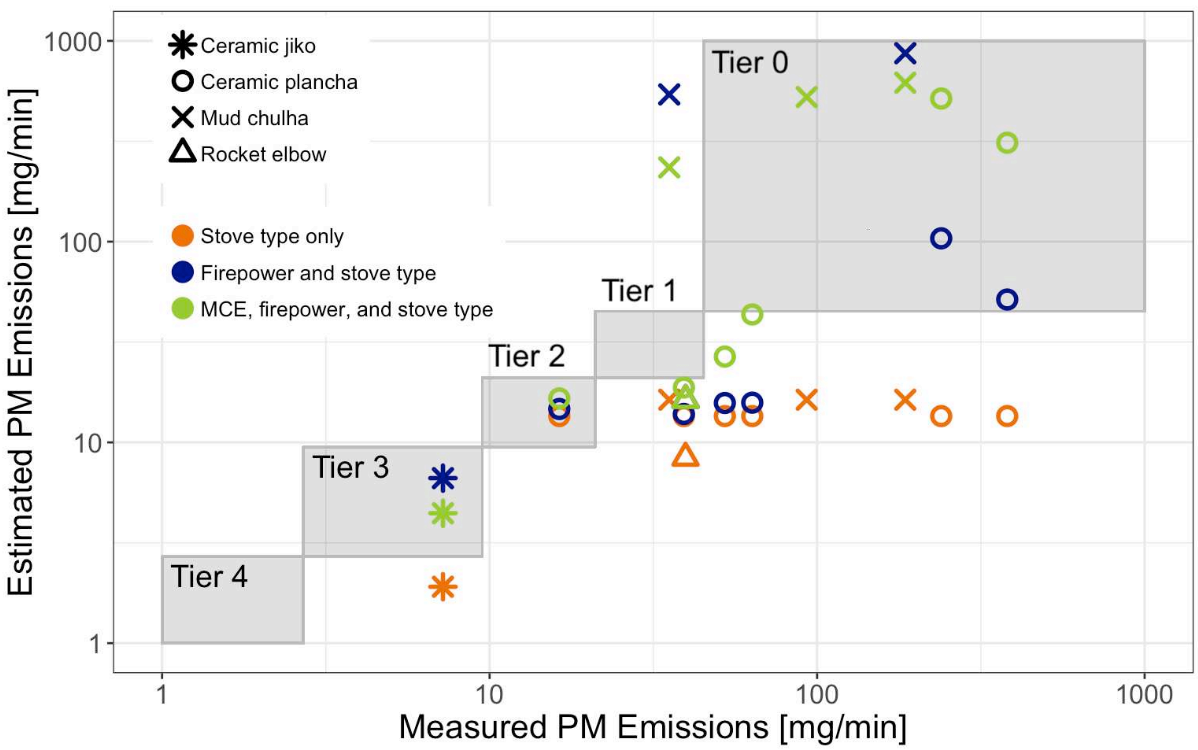


Figure 4.4: *Comparison of fine particulate matter ($PM_{2.5}$) emissions measured during in-home use and estimated using laboratory-derived models. Three models were used to predict in-use emissions: one that only used stove type as a predictor; one that used stove type and firepower as predictors; and one that used stove type, firepower, and modified combustion efficiency (MCE) as predictors. Each marker represents a single in-home test. The gray rectangles represent the voluntary indoor emissions tiers published by the International Standards Organization (19867-3:2018).*

When only stove type was a predictor (Equation 4.3), one out of eleven stoves were classified in the correct ISO emissions tier. This result demonstrates that the average $PM_{2.5}$ emissions rate measured during the Firepower Sweep Test will likely not represent the ISO emissions tier an in-use stove will fall under. The model that used both stove type and one-minute averaged firepower as predictors correctly classified the ISO emissions tier of five out of eleven stoves, constituting a 36% increase in the classification accuracy of the model represented by Equation 4.4 (compared to the baseline model). Thus, using information about the relationship between emissions and firepower derived from the lab, resulted in an increase in the predictive ability of the emissions model. The model that used stove type along with one-minute averaged firepower and one-minute averaged MCE as predictors correctly classified the ISO emissions tier of nine out of eleven stoves, constituting a 63% increase in the classification accuracy of the model represented by Equation 4.5 (compared to the baseline model). Thus, using information about the relationship between emissions and firepower and information about the relationship between emissions and MCE derived from the lab, resulted in an increase in the predictive ability of the emissions model (compared to both the baseline model and the model that used only information about firepower).

No notable increase in predictive ability of either the Equation 4.4 or Equation 4.5 models was observed, compare to the baseline model (Equation 4.3), when test-average firepower and test-averaged MCE measurements were used as predictors were used in place of continuous firepower and continuous MCE measurements (Appendix C3). This result was due to the non-linear relationship between firepower and emissions and MCE and emissions, which highlights the importance of using continuous measurement rather than test-average measurements for predictions.

4.3.5 Limitations and Future Work

We were not always able to test the exact same stove model between the laboratory and the field. Achieving this is notably difficult for traditional stoves, as many traditional stoves are handmade locally, thus making them difficult to obtain for laboratory testing. For example, the mud chulha stove tested in the laboratory was a single pot stove, while many of the mud chulha stoves tested in the field were made of different materials and were two- or three-pot stoves (Appendix C1). We recommend that future work include quantifying the sensitivity of the model estimates to using the exact stove design tested in the field, since the three-term models performed satisfactorily. Further we recommend the applicability of additional stove designs are validation before this approach is widely applied in practice.

In the field, our firepower calculations were based on the lower heating value of the primary fuel used each test. There were several instances, however, where other fuel sources such as small pieces of trash, corn cobs, and cow dung were added to the fire in addition to the wood or charcoal. These practices are representative of actual field operation and thus households where these types of events occured were not excluded from the analysis. The combustible materials added, in addition to wood and charcoal, likely had smaller lower heating values. Thus, the emissions calculations may be positively biased.

4.5 Conclusions

In this work, we present a technique for estimating firepower in the field. This technique, which uses a relatively inexpensive measurement of temperature at the combustion chamber outlet, has the potential to provide valuable information about the range of firepowers over which cookstoves are operated at during real-world use.

We also demonstrated that adding information about continuous-firepower and continuous-MCE can improve laboratory emissions estimates by 63%. We evaluated the accuracy of the estimates by identifying whether the models were able to predict the ISO emissions tier (19867-3:2018) a stove would fall under. These estimates are generally accurate enough to estimate the ISO emissions tier for a given stove. Although further validation of other stove types is needed, this result is an important contribution towards closing the gap between laboratory and field performance.

CHAPTER 5: CONCLUSIONS AND PRACTICAL IMPLICATIONS

In this work, I present several substantial contributions that reduce the uncertainties surrounding cookstove emissions. Historically, cookstove stakeholders (e.g., designers, health and social scientists, climate modelers, policymakers) have relied on tasked-based laboratory tests, such as the Water Boiling Test (WBT), to support cookstove design, indoor and outdoor air quality modeling, health studies, and policy efforts, even though laboratory data overpredict the ability of improved stoves to lower emissions during in-home cooking. In Chapter 2, I investigated the efficacy of multiple-firepower testing. The results demonstrate that the range of emissions seen under real-world conditions can be better replicated by varying firepower. Thus, multiple-firepower laboratory tests have the potential to better predict which technologies have low enough emissions to substantially improve indoor air quality.

In Chapter 4, I further demonstrate that, when combined with information about local cooking practices (e.g., in-field firepower and usage), data from the Firepower Sweep Test can be used to estimate which International Standards Organization performance tier a stove is likely to fall into. These results have the potential to enable more strategic dissemination of improved stove technologies based on a specific community's cooking practices. These modeling techniques, when combined with the proposed low-cost technique for estimating firepower, also have the potential to aid in the development and implementation of standardized testing procedures (e.g., ISO 19867-1:2018) at an international level.

One of the largest cookstove emissions inventories developed, to date, is presented in Chapter 3. This inventory will be publicly available through the Colorado State Library database services. These data, for the first time, demonstrate that reductions $PM_{2.5}$ and CO emissions do not necessarily correspond to significant reductions in all harmful pollutants emitted by solid-fuel cookstoves. The use of this data set will likely be far reaching. We present data that are of interest to air quality and climate modelers (e.g., volatile organic compounds, biomass burning tracers, particle size distributions, etc.) as well as health researchers (e.g., carcinogenic compounds, inorganic ions, etc.). We anticipate that these data will aid policy makers in selecting cookstoves to disseminate to communities, health researchers in selecting stoves for large scale randomized control trials or interpreting observed health effects after "improved" stoves are disseminated, to build data sets for source apportionment, and to compare simulated indoor air pollutant concentrations to health-based regulatory guidelines.

REFERENCES

(1) Bonjour, S.; Adair-Rohani, H.; Wolf, J.; Bruce, N. G.; Mehta, S.; Prüss-Ustün, A.; Lahiff, M.; Rehfuess, E. A.; Mishra, V.; Smith, K. R. Solid Fuel Use for Household Cooking: Country and Regional Estimates for 1980–2010. *Environ. Health Perspect.* **2013**, *121* (7), 784–790.

(2) World Health Organization. *WHO Guidelines for Indoor Air Quality: Selected Pollutants*; World Health Organization, 2010.

(3) Clark, M. L.; Peel, J. L.; Balakrishnan, K.; Breysse, P. N.; Chillrud, S. N.; Naeher, L. P.; Rodes, C. E.; Vette, A. F.; Balbus, J. M. Health and Household Air Pollution from Solid Fuel Use: The Need for Improved Exposure Assessment. *Environ. Health Perspect.* **2013**, *121* (10), 1120–1128.

(4) GBD 2013 Risk Factors Collaborators; Forouzanfar, M. H.; Alexander, L.; Anderson, H. R.; Bachman, V. F.; Biryukov, S.; Brauer, M.; Burnett, R.; Casey, D.; Coates, M. M.; et al. Global, Regional, and National Comparative Risk Assessment of 79 Behavioural, Environmental and Occupational, and Metabolic Risks or Clusters of Risks in 188 Countries, 1990-2013: A Systematic Analysis for the Global Burden of Disease Study 2013. *Lancet* **2015**, *386* (10010), 2287–2323.

(5) Kim, K.-H.; Jahan, S. A.; Kabir, E. A Review of Diseases Associated with Household Air Pollution due to the Use of Biomass Fuels. *J. Hazard. Mater.* **2011**, *192* (2), 425–431.

(6) Smith, K. R.; Bruce, N.; Balakrishnan, K.; Adair-Rohani, H.; Balmes, J.; Chafe, Z.; Dherani, M.; Hosgood, H. D.; Mehta, S.; Pope, D.; et al. Millions Dead: How Do We Know and What Does It Mean? Methods Used in the Comparative Risk Assessment of Household Air Pollution. *Annu. Rev. Public Health* **2014**, *35*, 185–206.

(7) Kodros, J. K.; Scott, C. E.; Farina, S. C.; Lee, Y. H.; L'Orange, C.; Volckens, J.; Pierce, J. R. Uncertainties in Global Aerosols and Climate Effects due to Biofuel Emissions. *Atmos. Chem. Phys.* **2015**, *15* (15), 8577–8596.

(8) Lacey, F. G.; Henze, D. K.; Lee, C. J.; van Donkelaar, A.; Martin, R. V. Transient Climate and Ambient Health Impacts due to National Solid Fuel Cookstove Emissions. *Proc. Natl. Acad. Sci. U. S. A.* **2017**, *114* (6), 1269–1274.

(9) Koch, D.; Bond, T. C.; Streets, D.; Unger, N.; van der Werf, G. R. Global Impacts of Aerosols from Particular Source Regions and Sectors. *J. Geophys. Res.* **2007**, *112* (D2).

(10) Intergovernmental Panel on Climate Change. *Climate Change 2013: The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press, 2014.

(11) Bond, T.; Venkataraman, C.; Masera, O. Global Atmospheric Impacts of Residential Fuels. *Energy for Sustainable Development* **2004**, *8* (3), 20–32.

(12) Thomas, E.; Wickramasinghe, K.; Mendis, S.; Roberts, N.; Foster, C. Improved Stove Interventions to Reduce Household Air Pollution in Low and Middle Income Countries: A Descriptive Systematic Review. *BMC Public Health* **2015**, *15*, 650.

(13) *The Water Boiling Test Version 4.2.3: Cookstove Emissions and Efficiency in a Controlled Laboratory Setting*; 2014.

(14) Torres-Duque, C.; Maldonado, D.; Perez-Padilla, R.; Ezzati, M.; Viegi, G. Biomass Fuels and Respiratory Diseases: A Review of the Evidence. *Proc. Am. Thorac. Soc.* **2008**, *5* (5), 577–590.

(15) R., P.-P.; A., S. Respiratory Health Effects of Indoor Air Pollution. *International Journal of Tuberculosis and Lung Disease* **2010**, *14* (9), 1079–1086.
(16) L'Orange, C.; Volckens, J.; Defoort, M. Influence of Stove Type and Cooking Pot Temperature on Particulate Matter Emissions from Biomass Cook Stoves. **2012**.
(17) Prapas, J.; Baumgardner, M. E.; Marchese, A. J.; Willson, B.; Defoort, M. Influence of Chimneys on Combustion Characteristics of Buoyantly Driven Biomass Stoves. *Energy for Sustainable Development* **2014**, *23*, 286–293.
(18) Arora, P.; Jain, S.; Sachdeva, K. Laboratory Based Assessment of Cookstove Performance Using Energy and Emission Parameters for North Indian Cooking Cycle. *Biomass Bioenergy* **2014**, *69*, 211–221.
(19) Bryden, K. M.; Bruce, N.; Peck, M. *WHO Indoor Air Quality Guidelines: Household Fuel Combustion*; World Health Organization: Geneva, 2015.
(20) *Guidelines for Evaluating Cookstove Performance*; IWA 11:2012; International Standards Organization, 2012.
(21) International Standards Organization. *Clean Cookstoves and Clean Cooking Solutions -- Harmonized Laboratory Test Protocols -- Part 1: Standard Test Sequence for Emissions and Performance, Safety and Durability*; 19867-1:2018; 2018.
(22) Kodros, J. K.; Carter, E.; Brauer, M.; Volckens, J.; Bilsback, K. R.; L'Orange, C.; Johnson, M.; Pierce, J. R. Quantifying the Contribution to Uncertainty in Mortality Attributed to Household, Ambient, and Joint Exposure to PM 2.5 From Residential Solid Fuel Use : Sensitivity in SFU Mortality Estimates. *GeoHealth* **2018**, *2* (1), 25–39.
(23) Peel, J. L.; Baumgartner, J.; Wellenius, G. A.; Clark, M. L.; Smith, K. R. Are Randomized Trials Necessary to Advance Epidemiologic Research on Household Air Pollution? *Current Epidemiology Reports* **2015**, *2* (4), 263–270.
(24) Mortimer, K.; Ndamala, C. B.; Naunje, A. W.; Malava, J.; Katundu, C.; Weston, W.; Havens, D.; Pope, D.; Bruce, N. G.; Nyirenda, M.; et al. A Cleaner Burning Biomass-Fuelled Cookstove Intervention to Prevent Pneumonia in Children under 5 Years Old in Rural Malawi (the Cooking and Pneumonia Study): A Cluster Randomised Controlled Trial. *Lancet* **2017**, *389* (10065), 167–175.
(25) Aunan, K.; Berntsen, T. K.; Myhre, G.; Rypdal, K.; Streets, D. G.; Woo, J.-H.; Smith, K. R. Radiative Forcing from Household Fuel Burning in Asia☆. *Atmos. Environ.* **2009**, *43* (35), 5674–5681.
(26) Venkataraman, C.; Habib, G.; Eiguren-Fernandez, A.; Miguel, A. H.; Friedlander, S. K. Residential Biofuels in South Asia: Carbonaceous Aerosol Emissions and Climate Impacts. *Science* **2005**, *307* (5714), 1454–1456.
(27) Berkeley Air Monitoring Group. *Stove Performance Inventory Report*; 2012.
(28) Grieshop, A. P.; Marshall, J. D.; Kandlikar, M. Health and Climate Benefits of Cookstove Replacement Options. *Energy Policy* **2011**, *39* (12), 7530–7542.
(29) Bailis, R.; Berrueta, V.; Chengappa, C.; Dutta, K.; Edwards, R.; Masera, O.; Still, D.; Smith, K. R. Performance Testing for Monitoring Improved Biomass Stove Interventions: Experiences of the Household Energy and Health Project. *Energy for Sustainable Development* **2007**, *11* (2), 57–70.
(30) Berrueta, V. M.; Edwards, R. D.; Masera, O. R. Energy Performance of Wood-Burning Cookstoves in Michoacan, Mexico. *Renewable Energy* **2008**, *33* (5), 859–870.
(31) Boy, E.; Bruce, N.; Smith, K. R.; Hernandez, R. Fuel Efficiency of an Improved Wood-Burning Stove in Rural Guatemala: Implications for Health, Environment and

Development. *Energy for Sustainable Development* **2000**, *4* (2), 23–31.
(32) Arora, P.; Jain, S. A Review of Chronological Development in Cookstove Assessment Methods: Challenges and Way Forward. *Renewable Sustainable Energy Rev.* **2016**, *55*, 203–220.
(33) *Indian Standard on Solid Biomass Chulha-Specification*; Bureau of Indian Standards, 1991.
(34) *General Specifications for Biomass Household Stoves*; Quality and Technical Supervision Bureau of Beijing Municipality, 2008.
(35) Prasad, K. K.; Krishna Prasad, K.; Sangen, E.; Visser, P. Woodburning Cookstoves. In *Advances in Heat Transfer*; 1985; pp 159–317.
(36) Johnson, M.; Edwards, R.; Berrueta, V.; Masera, O. New Approaches to Performance Testing of Improved Cookstoves. *Environ. Sci. Technol.* **2010**, *44* (1), 368–374.
(37) Makonese, T.; Pemberton-Pigott, C.; Robinson, J.; Kimemia, D.; Annegarn, H. Performance Evaluation and Emission Characterisation of Three Kerosene Stoves Using a Heterogeneous Stove Testing Protocol (HTP). *Energy for Sustainable Development* **2012**, *16* (3), 344–351.
(38) Sustainable Energy Technology and Research Center. *Indonesian Clean Stove Initiative Pilot Programme Water Boiling Test Methods and Product Evaluation Criteria*; 2014.
(39) Carter, E. M.; Shan, M.; Yang, X.; Li, J.; Baumgartner, J. Pollutant Emissions and Energy Efficiency of Chinese Gasifier Cooking Stoves and Implications for Future Intervention Studies. *Environ. Sci. Technol.* **2014**, *48* (11), 6461–6467.
(40) Chen, Y.; Roden, C. A.; Bond, T. C. Characterizing Biofuel Combustion with Patterns of Real-Time Emission Data (PaRTED). *Environ. Sci. Technol.* **2012**, *46* (11), 6110–6117.
(41) Yuntenwi, E. A. T.; MacCarty, N.; Still, D.; Ertel, J. Laboratory Study of the Effects of Moisture Content on Heat Transfer and Combustion Efficiency of Three Biomass Cook Stoves. *Energy for Sustainable Development* **2008**, *12* (2), 66–77.
(42) Agenbroad, J.; DeFoort, M.; Kirkpatrick, A.; Kreutzer, C. A Simplified Model for Understanding Natural Convection Driven Biomass Cooking stoves—Part 1: Setup and Baseline Validation. *Energy for Sustainable Development* **2011**, *15* (2), 160–168.
(43) Agenbroad, J.; DeFoort, M.; Kirkpatrick, A.; Kreutzer, C. A Simplified Model for Understanding Natural Convection Driven Biomass Cooking stoves—Part 2: With Cook Piece Operation and the Dimensionless Form. *Energy for Sustainable Development* **2011**, *15* (2), 169–175.
(44) Ballard-Tremeer, G.; Jawurek, H. H. Comparison of Five Rural, Wood-Burning Cooking Devices: Efficiencies and Emissions. *Biomass Bioenergy* **1996**, *11* (5), 419–430.
(45) Joshi, V.; Venkataraman, C.; Ahuja, D. R. Emissions from Burning Biofuels in Metal Cookstoves. *Environ. Manage.* **1989**, *13* (6), 763–772.
(46) Coffey, E. R.; Muvandimwe, D.; Hagar, Y.; Wiedinmyer, C.; Kanyomse, E.; Piedrahita, R.; Dickinson, K. L.; Oduro, A.; Hannigan, M. P. New Emission Factors and Efficiencies from in-Field Measurements of Traditional and Improved Cookstoves and Their Potential Implications. *Environ. Sci. Technol.* **2017**, *51* (21), 12508–12517.
(47) Dischino, K. Methods for Particulate Matter Emissions Reduction in Wood Burning Cookstoves, Colorado State University. Libraries, 2016.
(48) L'Orange, C.; DeFoort, M.; Willson, B. Influence of Testing Parameters on Biomass Stove Performance and Development of an Improved Testing Protocol. *Energy for Sustainable Development* **2012**, *16* (1), 3–12.
(49) Tryner, J.; Willson, B. D.; Marchese, A. J. The Effects of Fuel Type and Stove Design on Emissions and Efficiency of Natural-Draft Semi-Gasifier Biomass Cookstoves. *Energy for*

Sustainable Development **2014**, *23*, 99–109.
(50) Tryner, J.; Tillotson, J. W.; Baumgardner, M. E.; Mohr, J. T.; DeFoort, M. W.; Marchese, A. J. The Effects of Air Flow Rates, Secondary Air Inlet Geometry, Fuel Type, and Operating Mode on the Performance of Gasifier Cookstoves. *Environ. Sci. Technol.* **2016**, *50* (17), 9754–9763.
(51) Ferek, R. J.; Reid, J. S.; Hobbs, P. V.; Blake, D. R.; Liousse, C. Emission Factors of Hydrocarbons, Halocarbons, Trace Gases and Particles from Biomass Burning in Brazil. *J. Geophys. Res. D: Atmos.* **1998**, *103* (D24), 32107–32118.
(52) Darold E. Ward and Wei Min Hao. Projection of Emissions from Burning of Biomass for Use In Studies of Global Climate and Atmospheric Chemistry; Air & Waste Management Association.
(53) Jetter, J.; Zhao, Y.; Smith, K. R.; Khan, B.; Yelverton, T.; Decarlo, P.; Hays, M. D. Pollutant Emissions and Energy Efficiency under Controlled Conditions for Household Biomass Cookstoves and Implications for Metrics Useful in Setting International Test Standards. *Environ. Sci. Technol.* **2012**, *46* (19), 10827–10834.
(54) Medina, P.; Berrueta, V.; Martínez, M.; Ruiz, V.; Edwards, R. D.; Masera, O. Comparative Performance of Five Mexican Plancha-Type Cookstoves Using Water Boiling Tests. *Development Engineering* **2017**, *2*, 20–28.
(55) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O.; MacCarty, N.; Still, D. Laboratory and Field Investigations of Particulate and Carbon Monoxide Emissions from Traditional and Improved Cookstoves. *Atmos. Environ.* **2009**, *43* (6), 1170–1181.
(56) Wathore, R.; Mortimer, K.; Grieshop, A. P. In-Use Emissions and Estimated Impacts of Traditional, Natural- and Forced-Draft Cookstoves in Rural Malawi. *Environ. Sci. Technol.* **2017**, *51* (3), 1929–1938.
(57) Rose Eilenberg, S.; Bilsback, K. R.; Johnson, M.; Kodros, J. K.; Lipsky, E. M.; Naluwagga, A.; Fedak, K. M.; Benka-Coker, M.; Reynolds, B.; Peel, J.; et al. Field Measurements of Solid-Fuel Cookstove Emissions from Uncontrolled Cooking in China, Honduras, Uganda, and India. *Atmos. Environ.* **2018**, *190*, 116–125.
(58) Gibbons, J. D.; Chakraborti, S. Nonparametric Statistical Inference. In *International Encyclopedia of Statistical Science*; 2011; pp 977–979.
(59) Sullivan, B.; Allawatt, G.; Emery, A.; Means, P.; Kramlich, J.; Posner, J. Time-Resolved Particulate Emissions Monitoring of Cookstove Biomass Combustion Using a Tapered Element Oscillating Microbalance. *Combust. Sci. Technol.* **2016**, *189* (6), 923–936.
(60) L'Orange, C.; Volckens, J.; DeFoort, M. Influence of Stove Type and Cooking Pot Temperature on Particulate Matter Emissions from Biomass Cook Stoves. *Energy for Sustainable Development* **2012**, *16* (4), 448–455.
(61) Christian, T. J.; Yokelson, R. J.; Cárdenas, B.; Molina, L. T.; Engling, G.; Hsu, S.-C. Trace Gas and Particle Emissions from Domestic and Industrial Biofuel Use and Garbage Burning in Central Mexico. *Atmos. Chem. Phys. Disc.* **2009**, *9* (2), 10101–10152.
(62) Nazaroff, W. W. Inhalation Intake Fraction of Pollutants from Episodic Indoor Emissions. *Build. Environ.* **2008**, *43* (3), 269–277.
(63) Wilson, D. L. Clean Cookstoves Need Better Performance Guidelines. *Environ. Sci. Technol.* **2016**, *50* (18), 9805–9806.
(64) Lipsky, E. M.; Robinson, A. L. Effects of Dilution on Fine Particle Mass and Partitioning of Semivolatile Organics in Diesel Exhaust and Wood Smoke. *Environ. Sci. Technol.* **2006**, *40* (1), 155–162.

(65) Saliba, G.; Subramanian, R.; Saleh, R.; Ahern, A. T.; Lipsky, E. M.; Tasoglou, A.; Sullivan, R. C.; Bhandari, J.; Mazzoleni, C.; Robinson, A. L. Optical Properties of Black Carbon in Cookstove Emissions Coated with Secondary Organic Aerosols: Measurements and Modeling. *Aerosol Sci. Technol.* **2016**, *50* (11), 1264–1276.
(66) Pérez-Padilla, R.; Schilmann, A.; Riojas-Rodriguez, H. Respiratory Health Effects of Indoor Air Pollution. *Int. J. Tuberc. Lung Dis.* **2010**, *14* (9), 1079–1086.
(67) Torres-Duque, C.; Maldonado, D.; Pérez-Padilla, R.; Ezzati, M.; Viegi, G. Biomass Fuels and Respiratory Diseases. *Proc. Am. Thorac. Soc.* **2008**, *5* (5), 577–590.
(68) Naeher, L. P.; Brauer, M.; Lipsett, M.; Zelikoff, J. T.; Simpson, C. D.; Koenig, J. Q.; Smith, K. R. Woodsmoke Health Effects: A Review. *Inhal. Toxicol.* **2007**, *19* (1), 67–106.
(69) National Toxicology Program. NTP 12th Report on Carcinogens. *Rep. Carcinog.* **2011**, *12*, iii – 499.
(70) International Agency for Research on Cancer (IARC). Agents Classified by the IARC Monographs. *Agents Classified by the IARC Monographs, Volumes 1–121*, April, 18 2018.
(71) Intergovernmental Panel on Climate Change. *Climate Change 2013: The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press, 2014.
(72) Saliba, G.; Subramanian, R.; Saleh, R.; Ahern, A. T.; Lipsky, E. M.; Tasoglou, A.; Sullivan, R. C.; Bhandari, J.; Mazzoleni, C.; Robinson, A. L. Optical Properties of Black Carbon in Cookstove Emissions Coated with Secondary Organic Aerosols: Measurements and Modeling. *Aerosol Sci. Technol.* **2016**, *50* (11), 1264–1276.
(73) Reece, S. M.; Sinha, A.; Grieshop, A. P. Primary and Photochemically Aged Aerosol Emissions from Biomass Cookstoves: Chemical and Physical Characterization. *Environ. Sci. Technol.* **2017**, *51* (16), 9379–9390.
(74) Seinfeld, J. H. *Atmospheric Chemistry and Physics of Air Pollution*; Wiley-Interscience, 1986.
(75) Habib, G.; Venkataraman, C.; Bond, T. C.; Schauer, J. J. Chemical, Microphysical and Optical Properties of Primary Particles from the Combustion of Biomass Fuels. *Environ. Sci. Technol.* **2008**, *42* (23), 8829–8834.
(76) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O.; MacCarty, N.; Still, D. Laboratory and Field Investigations of Particulate and Carbon Monoxide Emissions from Traditional and Improved Cookstoves. *Atmos. Environ.* **2009**, *43* (6), 1170–1181.
(77) Johnson, M.; Edwards, R.; Alatorre Frenk, C.; Masera, O. In-Field Greenhouse Gas Emissions from Cookstoves in Rural Mexican Households. *Atmos. Environ.* **2008**, *42* (6), 1206–1222.
(78) Venkataraman, C.; Rao, G. U. Emission Factors of Carbon Monoxide and Size-Resolved Aerosols from Biofuel Combustion. *Environ. Sci. Technol.* **2001**, *35* (10), 2100–2107.
(79) Rapp, V. H.; Caubel, J. J.; Wilson, D. L.; Gadgil, A. J. Reducing Ultrafine Particle Emissions Using Air Injection in Wood-Burning Cookstoves. *Environ. Sci. Technol.* **2016**, *50* (15), 8368–8374.
(80) Just, B.; Rogak, S.; Kandlikar, M. Characterization of Ultrafine Particulate Matter from Traditional and Improved Biomass Cookstoves. *Environ. Sci. Technol.* **2013**, *47* (7), 3506–3512.
(81) Shen, G.; Gaddam, C. K.; Ebersviller, S. M.; Vander Wal, R. L.; Williams, C.; Faircloth, J. W.; Jetter, J. J.; Hays, M. D. A Laboratory Comparison of Emission Factors, Number Size Distributions, and Morphology of Ultrafine Particles from 11 Different Household

Cookstove-Fuel Systems. *Environ. Sci. Technol.* **2017**, *51* (11), 6522–6532.
(82) Tryner, J.; Volckens, J.; Marchese, A. J. Effects of Operational Mode on Particle Size and Number Emissions from a Biomass Gasifier Cookstove. *Aerosol Sci. Technol.* **2017**, *52* (1), 87–97.
(83) L’Orange, C.; Volckens, J.; DeFoort, M. Influence of Stove Type and Cooking Pot Temperature on Particulate Matter Emissions from Biomass Cook Stoves. *Energy for Sustainable Development* **2012**, *16* (4), 448–455.
(84) Patel, S.; Leavey, A.; He, S.; Fang, J.; O’Malley, K.; Biswas, P. Characterization of Gaseous and Particulate Pollutants from Gasification-Based Improved Cookstoves. *Energy for Sustainable Development* **2016**, *32*, 130–139.
(85) Caubel, J. J.; Rapp, V. H.; Chen, S. S.; Gadgil, A. J. Optimization of Secondary Air Injection in a Wood-Burning Cookstove: An Experimental Study. *Environ. Sci. Technol.* **2018**, *52* (7), 4449–4456.
(86) Tsai, S. M.; Zhang, J. J.; Smith, K. R.; Ma, Y.; Rasmussen, R. A.; Khalil, M. A. K. Characterization of Non-Methane Hydrocarbons Emitted from Various Cookstoves Used in China. *Environ. Sci. Technol.* **2003**, *37* (13), 2869–2877.
(87) Wang, Q.; Geng, C.; Lu, S.; Chen, W.; Shao, M. Emission Factors of Gaseous Carbonaceous Species from Residential Combustion of Coal and Crop Residue Briquettes. *Front. Environ. Sci. Eng. China* **2013**, *7* (1), 66–76.
(88) Stockwell, C. E.; Yokelson, R. J.; Kreidenweis, S. M.; Robinson, A. L.; DeMott, P. J.; Sullivan, R. C.; Reardon, J.; Ryan, K. C.; Griffith, D. W. T.; Stevens, L. Trace Gas Emissions from Combustion of Peat, Crop Residue, Biofuels, Grasses, and Other Fuels: Configuration and FTIR Component of the Fourth Fire Lab at Missoula Experiment (FLAME-4). *Atmos. Chem. Phys. Disc.* **2014**, *14* (7), 10061–10134.
(89) Klein, F.; Pieber, S. M.; Ni, H.; Stefenelli, G.; Bertrand, A.; Kilic, D.; Pospisilova, V.; Temime-Roussel, B.; Marchand, N.; El Haddad, I.; et al. Characterization of Gas-Phase Organics Using Proton Transfer Reaction Time-of-Flight Mass Spectrometry: Residential Coal Combustion. *Environ. Sci. Technol.* **2018**, *52* (5), 2612–2617.
(90) Zhang, J.; Smith, K. R. Emissions of Carbonyl Compounds from Various Cookstoves in China. *Environ. Sci. Technol.* **1999**, *33* (14), 2311–2320.
(91) Kim Oanh, N. T.; Oanh, N. T. K.; Dung, N. T. Emission of Polycyclic Aromatic Hydrocarbons and Particulate Matter from Domestic Combustion of Selected Fuels. *Environ. Sci. Technol.* **1999**, *33* (16), 2703–2709.
(92) Kim Oanh, N. T.; Oanh, N. T. K.; Albina, D. O.; Ping, L.; Wang, X. Emission of Particulate Matter and Polycyclic Aromatic Hydrocarbons from Select Cookstove–fuel Systems in Asia. *Biomass Bioenergy* **2005**, *28* (6), 579–590.
(93) Shen, G.; Wang, W.; Yang, Y.; Zhu, C.; Min, Y.; Xue, M.; Ding, J.; Li, W.; Wang, B.; Shen, H.; et al. Emission Factors and Particulate Matter Size Distribution of Polycyclic Aromatic Hydrocarbons from Residential Coal Combustions in Rural Northern China. *Atmos. Environ.* **2010**, *44* (39), 5237–5243.
(94) Shen, G.; Wang, W.; Yang, Y.; Ding, J.; Xue, M.; Min, Y.; Zhu, C.; Shen, H.; Li, W.; Wang, B.; et al. Emissions of PAHs from Indoor Crop Residue Burning in a Typical Rural Stove: Emission Factors, Size Distributions, and Gas−Particle Partitioning. *Environ. Sci. Technol.* **2011**, *45* (4), 1206–1212.
(95) Kandpal, J. B.; Maheshwari, R. C.; Kandpal, T. C. Comparison of CO, NO2 and HCHO Emissions from Biomass Combustion in Traditional and Improved Cookstoves. *Energy*

1994, *19* (11), 1151–1155.
(96) Gupta, S.; Saksena, S.; Shankar, V. R.; Joshi, V. Emission Factors and Thermal Efficiencies of Cooking Biofuels from Five Countries. *Biomass Bioenergy* **1998**, *14* (5-6), 547–559.
(97) Leavey, A.; Patel, S.; Martinez, R.; Mitroo, D.; Fortenberry, C.; Walker, M.; Williams, B.; Biswas, P. Organic and Inorganic Speciation of Particulate Matter Formed during Different Combustion Phases in an Improved Cookstove. *Environ. Res.* **2017**, *158*, 33–42.
(98) Bilsback, K. R.; Eilenberg, S. R.; Good, N.; Heck, L.; Johnson, M.; Kodros, J. K.; Lipsky, E. M.; L'Orange, C.; Pierce, J. R.; Robinson, A. L.; et al. The Firepower Sweep Test: A Novel Approach to Cookstove Laboratory Testing. *Indoor Air* **2018**.
(99) Subramanian, R.; Khlystov, A. Y.; Cabada, J. C.; Robinson, A. L. Positive and Negative Artifacts in Particulate Organic Carbon Measurements with Denuded and Undenuded Sampler Configurations Special Issue of Aerosol Science and Technology on Findings from the Fine Particulate Matter Supersites Program. *Aerosol Sci. Technol.* **2004**, *38* (S1), 27–48.
(100) Birch, M. E.; Cary, R. A. Elemental Carbon-Based Method for Monitoring Occupational Exposures to Particulate Diesel Exhaust. *Aerosol Sci. Technol.* **1996**, *25* (3), 221–241.
(101) Sullivan, A. P.; Holden, A. S.; Patterson, L. A.; McMeeking, G. R.; Kreidenweis, S. M.; Malm, W. C.; Hao, W. M.; Wold, C. E.; Collett, J. L. A Method for Smoke Marker Measurements and Its Potential Application for Determining the Contribution of Biomass Burning from Wildfires and Prescribed Fires to Ambient PM2.5organic Carbon. *J. Geophys. Res.* **2008**, *113* (D22).
(102) McGlenny, W. A.; Pleil, J. D.; Evans, G. F.; Oliver, K. D.; Holdren, M. W.; Winberry, W. T. Canister-Based Method for Monitoring Toxic VOCs in Ambient Air. *J. Air Waste Manage. Assoc.* **1991**, *41* (10), 1308–1318.
(103) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O. Emission Factors and Real-Time Optical Properties of Particles Emitted from Traditional Wood Burning Cookstoves. *Environ. Sci. Technol.* **2006**, *40* (21), 6750–6757.
(104) International Energy Agency. *Energy Access Outlook 2017*; 2017.
(105) Turns, S. *An Introduction to Combustion: Concepts and Applications*; McGraw-Hill Higher Education.
(106) Roden, C. A.; Bond, T. C.; Conway, S.; Benjamin, A.; Pinel, O. Emission Factors and Real-Time Optical Properties of Particles Emitted from Traditional Wood Burning Cookstoves. *Environ. Sci. Technol.* **2006**, *40* (21), 6750–6757.
(107) Roden, C. A.; Bond, T. C.; Conway, S.; Pinel, A. B. O.; MacCarty, N.; Still, D. Laboratory and Field Investigations of Particulate and Carbon Monoxide Emissions from Traditional and Improved Cookstoves. *Atmos. Environ.* **2009**, *43* (6), 1170–1181.
(108) Yokelson, R. J.; Burling, I. R.; Gilman, J. B.; Warneke, C.; Stockwell, C. E.; Gouw, J. de; Akagi, S. K.; Urbanski, S. P.; Veres, P.; Roberts, J. M.; et al. Coupling Field and Laboratory Measurements to Estimate the Emission Factors of Identified and Unidentified Trace Gases for Prescribed Fires. *Atmos. Chem. Phys.* **2013**, *13* (1), 89–116.
(109) Christian, T. J. Comprehensive Laboratory Measurements of Biomass-Burning Emissions: 1. Emissions from Indonesian, African, and Other Fuels. *J. Geophys. Res.* **2003**, *108* (D23), 955.
(110) Johnson, M.; Edwards, R.; Berrueta, V.; Masera, O. New Approaches to Performance Testing of Improved Cookstoves. *Environ. Sci. Technol.* **2010**, *44* (1), 368–374.

APPENDIX A

Appendix A1. Continuous-feed wood stove protocol summary

segment name		description	duration	fuel added	target temperature
start up		use a match to light the shims and start the stove, attempt to consume all of the shims during this sample	ten minutes	three shims	
		add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water (5 liters at room temperature) and preparing the emissions system	five minutes	four sticks (1.5 cm x 1.5 cm x 12 cm)	
	1	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	1200 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	three sticks	
	2	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	900 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	two sticks	
	3	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	750 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	one stick	
	4	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	650 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	two sticks	

	5	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	750 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	three sticks	
	6	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	900 K
		take the pot of water off the stove and post-weigh, add fuel, transition to next target combustion chamber outlet temperature, and prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	ten minutes	four sticks	
	7	put a new pot of water on the stove, maintain stove as close to the target temperature as possible	six minutes	none	1200 K
		prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none	
shut-down		after three minutes put any burning fuel in closed ash pot to cut off the air supply to the flame and extinguish the fuel, allow the charcoal bed to smolder for the remaining time	ten minutes	none	
total time: 132 minutes					

Appendix A2. Batch-fed charcoal stove protocol summary

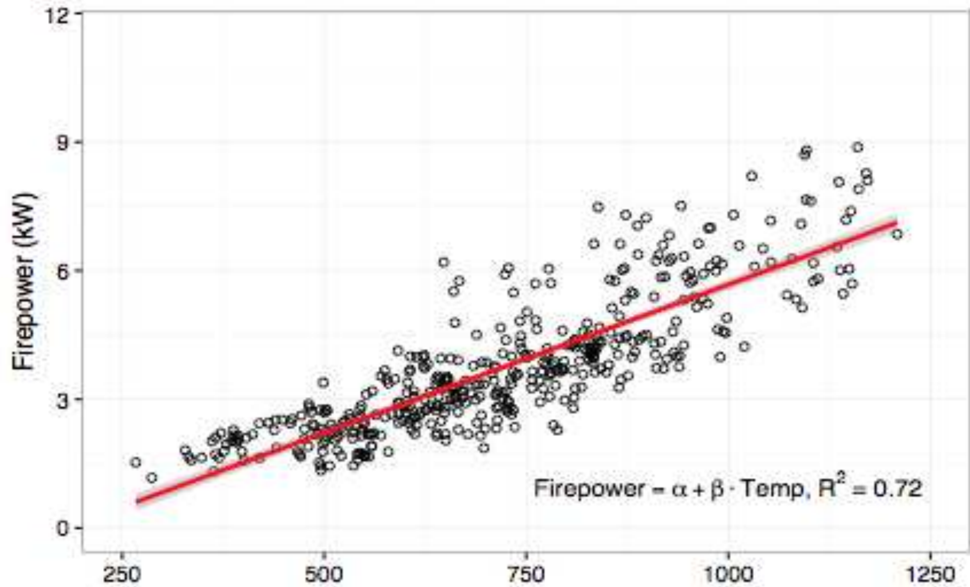
segment name		description	duration	fuel added
start-up		add the kerosene on top of the fuel bed and light the fuel, after three minutes put the water pot on the stove	ten minutes	fuel batch one (400 grams) and kerosene (approx. 15 grams)
pre-refuel		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water (5 liters at room temperature) and preparing the emissions system	five minutes	none
	1	put the pre-weighed pot of water on the stove	twenty minutes	none
		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none
	2	put the pre-weighed pot of water on the stove	twenty minutes	none
		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none
3		add the next batch of fuel and kerosene to the stove, light the fuel, after three minutes put the water pot on the stove	ten minutes	fuel batch two (400 grams) and kerosene (approx. 15 grams)
post-refuel		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none
	4	put the pre-weighed pot of water on the stove	twenty minutes	none
		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none
	5	put the pre-weighed pot of water on the stove	twenty minutes	none
		take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system	five minutes	none
shut-down		after three minutes pour remaining fuel into an ash pot, cut off the air supply and extinguish the fuel by placing a top on the pot, allow the charcoal bed to smolder for the remaining time	ten minutes	none
total time: 140 minutes				

Appendix A3. Batch-fed wood stove protocol summary

<table>
<tr><th colspan="2">segment name</th><th>description</th><th>duration</th><th>fuel added</th></tr>
<tr><td colspan="2">start-up</td><td>add the kerosene on top of the fuel bed and light the fuel, after three minutes put the water pot on the stove</td><td>ten minutes</td><td>fuel batch one (600 grams) and kerosene (approx. 15 grams)</td></tr>
<tr><td rowspan="5">pre-refuel</td><td></td><td>prepare for the next integrated sample by: weighing a new pot of water(5 liters at room temperature) and preparing the emissions system</td><td>five minutes</td><td>none</td></tr>
<tr><td>1</td><td>put the pre-weighed pot of water on the stove</td><td>twenty minutes</td><td>none</td></tr>
<tr><td></td><td>take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system</td><td>five minutes</td><td>none</td></tr>
<tr><td>2</td><td>put the pre-weighed pot of water on the stove</td><td>twenty minutes</td><td>none</td></tr>
<tr><td></td><td>take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system</td><td>five minutes</td><td>none</td></tr>
<tr><td colspan="2">3</td><td>add the next batch of fuel and kerosene to the stove, light the fuel, after three minutes put the water pot on the stove</td><td>ten minutes</td><td>fuel batch two (600 grams) and kerosene (approx. 15 grams)</td></tr>
<tr><td rowspan="3">post-refuel</td><td></td><td>prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system</td><td>five minutes</td><td>none</td></tr>
<tr><td>4</td><td>put the pre-weighed pot of water on the stove</td><td>twenty minutes</td><td>none</td></tr>
<tr><td></td><td>take the pot of water off the stove and post-weigh, prepare for the next integrated sample by: weighing a new pot of water and preparing the emissions system</td><td>five minutes</td><td>none</td></tr>
<tr><td colspan="2">shut-down</td><td>after three minutes pour remaining fuel into an ash pot, cut off the air supply and extinguish the fuel by placing a top on the pot, allow the charcoal bed to smolder for the remaining time</td><td>ten minutes</td><td>none</td></tr>
<tr><td colspan="5">total time: 115 minutes</td></tr>
</table>

Appendix A4. Correlation plot between temperature at the combustion chamber outlet and firepower

Data were collected during the Firepower Sweep Test for a single rocket elbow stove. The correlation includes data from tests with both Douglas fir and eucalyptus fuels.



Temperature (K)

Appendix A5. Photos of thermocouple placement at the combustion chamber outlet

Appendix A6. Guidelines for pilot testing

Continuous-feed wood stoves

Before testing a new stove/fuel combination using the Firepower Sweep Test (FST) for wood stoves, pilot testing is highly recommended to establish:

- A working range of firepower outputs,
- The duration of each firepower segment,
- The total amount of fuel needed to conduct the test, and
- The target combustion chamber outlet temperatures to be used for each integrated firepower sample.

The goal of pilot testing is to fill in (or extend) Appendix A1 for a specific stove/fuel combination. Pilot testing may not be needed as the user becomes more comfortable with the FST protocol, but the following instructions are recommended for new users or drastically different stove designs.

Obtain supplies and set up the workspace. Log combustion chamber outlet temperature measurements. Use a pot of water to keep the testing conditions as close to the field as possible. Start up the stove by igniting and consuming fire starters in the combustion chamber. After the stove is sufficiently hot and the starting fuel is nearly consumed, attempt to reach the highest combustion chamber outlet temperature possible by adjusting the fuel feed rate and number of sticks fed into the combustion chamber. Once the stove has reached its maximum practical firepower, attempt to keep the combustion chamber outlet temperature as consistent as possible for approximately six minutes. After the firepower of the stove is held steady for six minutes, note the average combustion chamber outlet temperature. This will be the target combustion chamber outlet temperature for the highest firepower point. Additionally, note the number of sticks required to maintain the stove at this firepower for six minutes. Ideally, no new sticks will be fed into the combustion chamber during an integrated firepower sample; the length of the fuel may need to be adjusted to fulfill this goal. After consuming the wood in the combustion chamber, try to achieve the lowest firepower possible. Typically, the lowest firepower is constrained by the fact that below a certain combustion chamber outlet temperature the flame cannot be maintained without extinguishing. To achieve the lowest firepower possible, reduce the fuel amount to a single stick and reduce the fuel feed rate until a consistently low temperature can be achieved for approximately six minutes. Note the lowest practically achievable combustion chamber outlet temperature. This will be used as the target combustion chamber outlet temperature for the lowest firepower point. Note the length of fuel required to maintain the stove at the lowest achievable firepower for six minutes.

Next, establish the target combustion chamber outlet temperatures for the medium-high and medium-low firepower samples. The stick quantities used for the medium-high and medium-low samples should be equally spaced between the high firepower and low firepower fuel quantities. For example, if four sticks are used at the highest firepower point and one stick is used at the lowest firepower point, then the medium-high firepower sample should use three sticks and the medium-low firepower sample should use two sticks. Attempt to maintain both the medium-high and medium-low firepower points for approximately six minutes. Note the combustion chamber outlet temperatures that can be consistently achieved for each of these firepower points. Finally, extinguish the stove. After the pilot test described above is successfully used to establish the appropriate firepower points and feed rates for the stove, the FST protocol may be run several times as described before completing a test with emissions measurements.

This will ensure that the tester has developed comfort with the protocol and that the timing of each test phase can be maintained. Finally, for low-emitting stoves, pilot gravimetric measurements should be made to ensure that the mass collected on the filters is above the limit of detection for gravimetric analysis.

Batch-fed charcoal and wood stoves

Unlike the FST for continuous-feed wood stoves, the charcoal test does not require establishing a working range of firepower outputs or target combustion chamber outlet temperatures. Instead, the pilot testing only involves running the stove as described in Appendix A2 or Appendix A3 before completing a test with emissions measurements. Pilot testing will ensure that the tester has developed comfort with the protocol and that the timing of each test phase can be maintained. Additionally, pilot test will establish if if segment durations or fuel batch sizes need to be adjusted needs to be adjusted for a specific stove/fuel combination.

Appendix A7. Stove-fuel test matrix and a list of the equivalent stove types from WBT and field testing

Continuous-feed wood stoves					
stove		fuel	make (model)	WBT equivalent	field equivalent
three-stone fire		Douglas fir (milled), eucalyptus (split)	artisan	three-stone fire[1,5]	three-stone fire[4]
mud chula		Douglas fir (milled), eucalyptus (split)	artisan	NA	NA
metal grill		Douglas fir (milled), eucalyptus (split)	Vikram	NA	NA
rocket elbow (model 1)		Douglas fir (milled), eucalyptus (split)	Envirofit (G3300)	Envirofit G3300[1], improved no chimney[2]	NA
rocket elbow (model 2)		Douglas fir (milled)	Chulika		NA

metal plancha	 (chimney not pictured)	Douglas fir (milled)	Envirofit (HM5000)	Ecostufa[5]	Eco H[2,3], Ecostufa[2,3]
ceramic plancha (model 1)		Douglas fir (milled)	artisan	Patsari[5], Onil[1,5]	Justa[2,3,6], Traditional[2,3]
ceramic plancha (model 2)	 (chimney not pictured)	Douglas fir (milled)	artisan		
Batch-fed charcoal stoves					
ceramic jiko (small)		hardwood (lumps), coconut (briquettes)	artisan	Jiko Ceramic[1], KCJ (Kenya Ceramic Jiko) Standard Stove[1]	all-ceramic jiko[6], metal-clad ceramic jiko (several designs)[6]
ceramic jiko (large)		hardwood (lumps), coconut (briquettes)	artisan		
improved metal jiko		hardwood (lumps) - one replicate only, coconut (briquettes)	BURN Design Labs	Jiko, Metal[1]	NA

metal jiko (small)		hardwood (lumps)	artisan		NA
metal jiko (large)		hardwood (lumps)	artisan		NA
Batch-fed wood stoves					
forced draft gasifier (model 1)		eucalyptus (pellets)	modular gasifier*	Philips Power Stove HD4012[1]	Philips HD4012LS[4], ACE-1[4]
forced draft gasifier (model 2)		eucalyptus (pellets), red oak (milled) - one replicate only	Philips (HD4012)		

*Tryner J, Tillotson JW, Baumgardner ME, Mohr JT, DeFoort MW, Marchese AJ. The Effects of Air Flow Rates, Secondary Air Inlet Geometry, Fuel Type, and Operating Mode on the Performance of Gasifier Cookstoves. *Environ Sci Technol*. 2016;50:9754–9763.
[1] Jetter J, Zhao Y, Smith KR, et al. Pollutant emissions and energy efficiency under controlled conditions for household biomass cookstoves and implications for metrics useful in setting international test standards. *Environ Sci Technol*. 2012;46:10827–10834.
[2] Roden CA, Bond TC, Conway S, Pinel ABO, MacCarty N, Still D. Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos Environ*. 2009;43:1170–1181.
[3] Roden CA, Bond TC, Conway S, Pinel ABO. Emission factors and real-time optical properties of particles emitted from traditional wood burning cookstoves. *Environ Sci Technol*. 2006;40:6750–6757.
[4] Wathore R, Mortimer K, Grieshop AP. In-Use Emissions and Estimated Impacts of Traditional, Natural- and Forced-Draft Cookstoves in Rural Malawi. *Environ Sci Technol*. 2017;51:1929–1938.
[5] Medina P, Berrueta V, Martínez M, Ruiz V, Edwards RD, Masera O. Comparative performance of five Mexican plancha-type cookstoves using water boiling tests. *Development Engineering*. 2017;2:20–28.
[6] Data collected in La Esperanza, Honduras as part of a partnership with Colorado State University Mechanical Engineering and Environmental Health
[7] Data collected in Kampala, Uganda as part of a partnership with Colorado State University and Makrere University

Appendix A8. Fuel properties

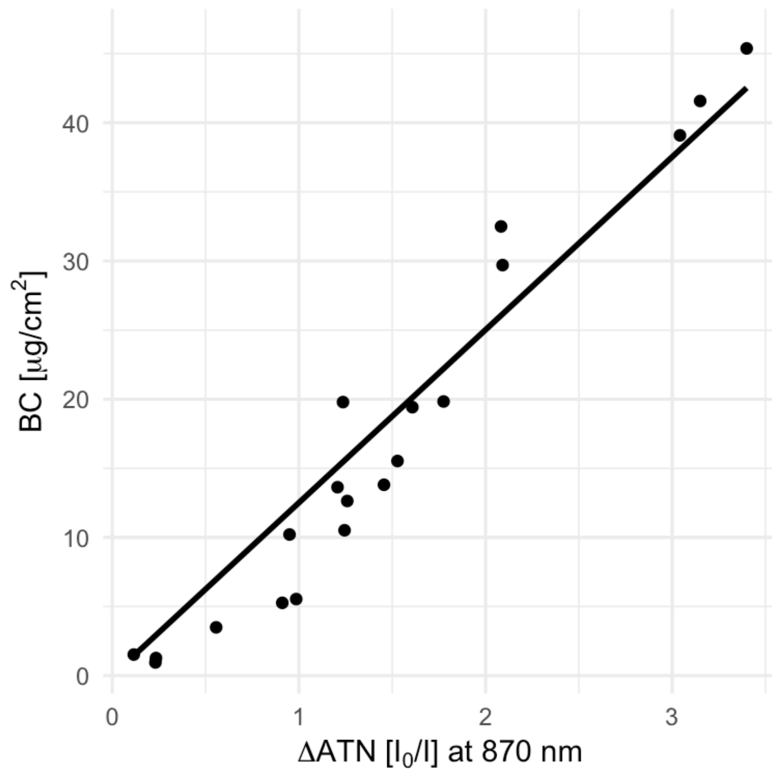
Reported properties are averages of at least three replicate analyses.

fuel		lower heating value*	carbon mass fraction
coconut (briquettes)		16100 kJ/kg	0.49
hardwood (lumps)		28100 kJ/kg	0.84
Douglas fir (milled)		17400 kJ/kg	0.50
eucalyptus (chopped)		18200 kJ/kg	0.49
red oak (milled)		17700 kJ/kg	0.49
eucalyptus (pellets)	Missing photo	16300 kJ/kg	0.50

* Higher heating values were measured directly using a bomb calorimeter (C 200 Calorimeter System; IKA; Staufen, Germany). These were converted to lower heating values assuming a 6% hydrogen content in wood-based fuels and a 2.5% hydrogen content in charcoal fuels. Values are reported on a dry basis.

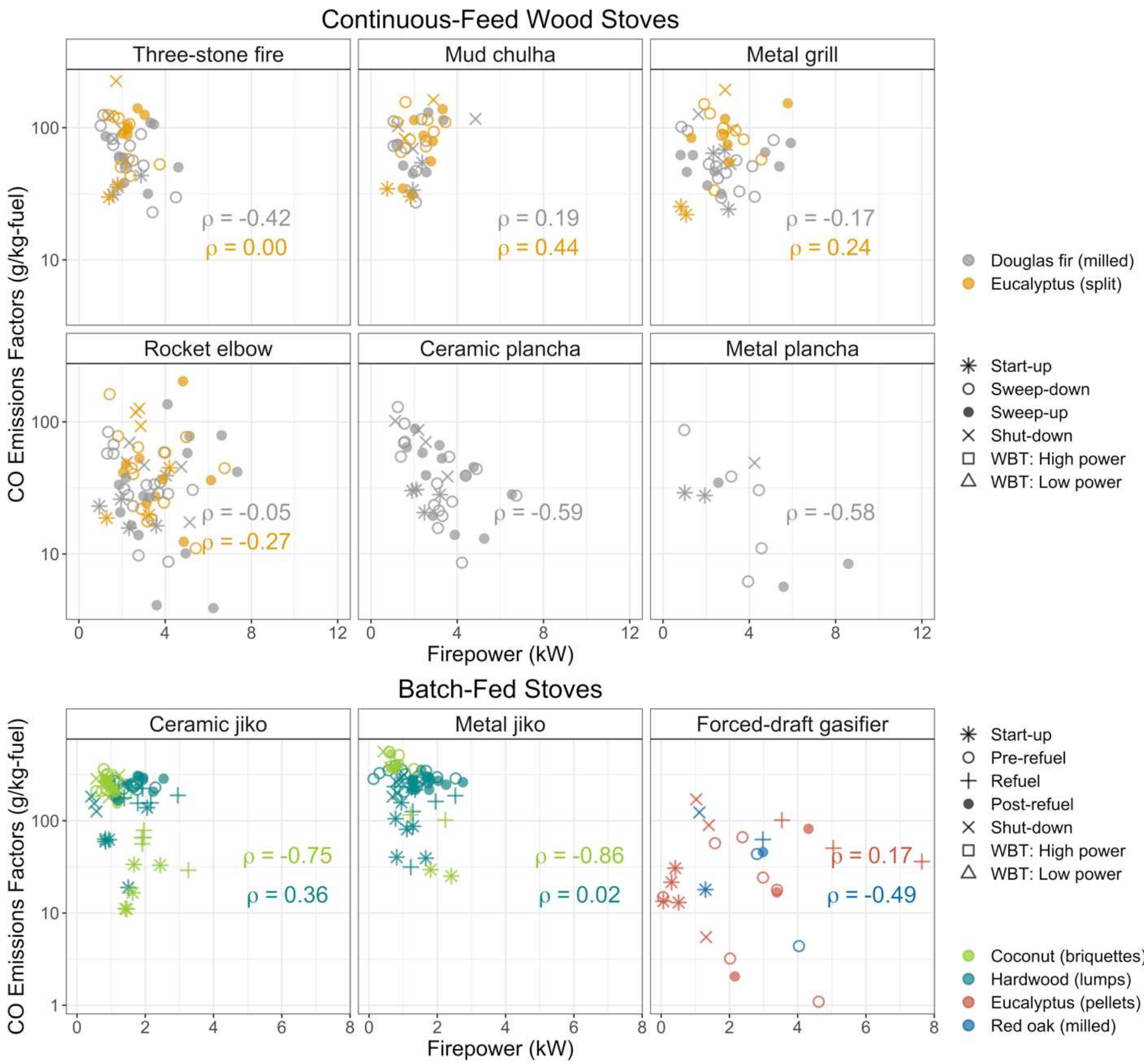
Appendix A9. Black carbon calibration curve (for continuous-feed wood stoves only)

The calibration curve to convert a change in attenuation (ΔATN) through a Pallflex Fiberfilm filter, to an equivalent black carbon surface concentration. The curve was developed using Pallflex Fiberflim and Tissuquartz filter samples collected in parallel. The ΔATN of the Fiberfilm filters was measured using the Magee Sootscan Transmissoemter (Model OT21) and the Tissuquartz filters were analyzed for equivalent black carbon (BC) using a Sunset Lab OC-EC Aerosol Analyzer (NIOSH 5040 Method). The particle samples were created using a rocket elbow stove design. Separate tests were conducted with two types of wood (eucalyptus and Douglas fir). No difference was seen between the two sources, so both source types were grouped together to develop the calibration curve. The analysis yielded an attenuation coefficient (σ_{ATN}) of 10.8 $cm^2/\mu g$ and a loading correction of 0.1418.



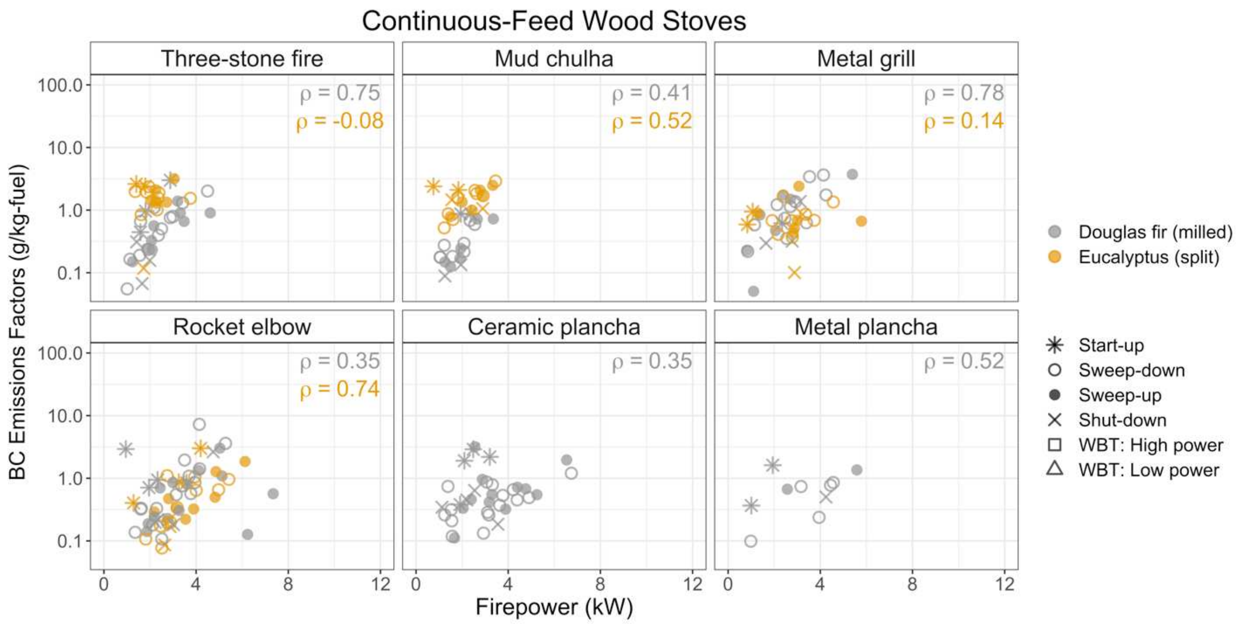
Appendix A10. Firepower versus carbon monoxide correlation plot

Firepower versus carbon monoxide (CO) emissions factors from the Firepower Sweep Test. Shapes are used to identify the test phase and colors are used to identify the fuel type.



Appendix A11. Firepower versus black carbon correlation plot

Firepower versus black carbon (BC) emissions factors from the Firepower Sweep Test. Shapes are used to identify the test phase and colors are used to identify the fuel type.



Appendix A12. Candidate linear models used in model selection

EF is the emissions factor of interest, FP is firepower, MCE is modified combustion efficiency, α_j are the coefficients for stove-fuel specific intercepts , β_j are the coefficients for stove-fuel specific slopes, (continuous-feed wood: j = 1,…,10; batch-fed charcoal: j = 1,...,4), γ and λ are constant slope coefficients across all stove-fuel combinations, ω is a constant slope coefficient across all stove-fuel combinations, and epsilon (ϵ) is random error. The asterisk (*) equations were used to predict carbon monoxide emissions, since MCE is nearly equivalent to CO emissions. FP and MCE were both scaled (i.e., normalized) before the models were fitted. (See SI14 for scaling factors.)

Model #	
1	$^*ln(EF) = \omega + \gamma \cdot FP + \epsilon$
2	$ln(EF) = \omega + \lambda \cdot MCE + \epsilon$
3	$ln(EF) = \omega + \lambda \cdot MCE + \gamma \cdot FP + \epsilon$
4	$^*ln(EF) = \alpha_j + \gamma \cdot FP + \epsilon$
5	$^*ln(EF) = \alpha_j + \beta_j \cdot FP + \epsilon$
6	$ln(EF) = \alpha_j + \lambda \cdot MCE + \epsilon$
7	$ln(EF) = \alpha_j + \beta_j \cdot MCE + \epsilon$
8	$ln(EF) = \alpha_j + \gamma \cdot FP + \lambda \cdot MCE + \epsilon$
9	$ln(EF) = \alpha_j + \beta_j \cdot FP + \lambda \cdot MCE + \epsilon$
10	$ln(EF) = \alpha_j + \beta_j \cdot MCE + \gamma \cdot MCE + \epsilon$

Appendix A13. Coefficients and 95% confidence intervals of the top regression models

EF is the emissions factor of interest, FP is firepower, MCE is modified combustion efficiency, α_j are the coefficients for stove-fuel specific intercepts , β_j are the coefficients for stove-fuel specific slopes, (continuous-feed wood: j = 1,…,10; batch-fed charcoal: j = 1,...,4), γ is a constant slope coefficients across all stove-fuel combinations, and epsilon (ϵ) is random error. FP and MCE were both scaled (i.e., normalized) before the models were fitted. The mean (μ) and standard deviation (σ) used to scale the FP and MCE variables are provided under each table.

Continuous-feed wood stoves

Fine particulate matter ($PM_{2.5}$) emissions factors:

$$ln(PM_{2.5}\ EF) = \alpha_j + \beta_j \cdot MCE + \gamma \cdot FP + \epsilon$$

	α_j	β_j	γ
mud chulha, Douglas fir (milled)	0.2 (-0.54, 0.93)	-0.41 (-0.91, 0.09)	0.61 (0.39, 0.84)
mud chulha, eucalyptus (split)	2 (1.05, 2.96)	-0.54 (-1.31, 0.22)	0.61 (0.39, 0.84)
metal grill, Douglas fir (milled)	0.59 (0.02, 1.17)	-0.11 (-1.1, 0.88)	0.61 (0.39, 0.84)
metal grill, eucalyptus (split)	0.57 (-0.37, 1.51)	0.39 (-0.37, 1.16)	0.61 (0.39, 0.84)
rocket elbow, Douglas fir (milled)	0.25 (-0.38, 0.88)	-1.02 (-1.69, -0.34)	0.61 (0.39, 0.84)
three-stone fire, Douglas fir (milled)	0.33 (-0.24, 0.91)	-0.07 (-0.79, 0.66)	0.61 (0.39, 0.84)
rocket elbow, eucalyptus (split)	-0.74 (-1.31, -0.17)	-0.03 (-0.48, 0.43)	0.61 (0.39, 0.84)
three-stone fire, eucalyptus (split)	1.82 (0.84, 2.81)	-0.28 (-1.1, 0.54)	0.61 (0.39, 0.84)
ceramic plancha, Douglas fir (milled)	0.6 (0.07, 1.12)	-1.23 (-1.86, -0.6)	0.61 (0.39, 0.84)
metal plancha, Douglas fir (milled)	0.33 (-1.05, 1.7)	-0.96 (-2.22, 0.3)	0.61 (0.39, 0.84)

Note: FP was transformed using μ = 3.00 and σ = 1.42. MCE was transformed using μ = 0.95 and σ = 0.03.

Black carbon (BC) emissions factors:

$$ln(BC\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$$

	α_j	β_j
mud chulha, Douglas fir (milled)	-0.43 (-1.58, 0.72)	1.09 (0.27, 1.91)
mud chulha, eucalyptus (split)	0.75 (-0.07, 1.56)	0.9 (-0.28, 2.08)
metal grill, Douglas fir (milled)	0.12 (-0.43, 0.67)	0.96 (0.36, 1.56)
metal grill, eucalyptus (split)	-0.28 (-0.87, 0.31)	0.07 (-0.7, 0.84)
rocket elbow, Douglas fir (milled)	-0.91 (-1.31, -0.5)	0.49 (0.1, 0.88)
three-stone fire, Douglas fir (milled)	-0.39 (-0.93, 0.15)	1.03 (0.34, 1.72)
rocket elbow, eucalyptus (split)	-1.42 (-1.94, -0.9)	0.99 (0.43, 1.54)
three-stone fire, eucalyptus (split)	0.55 (-0.38, 1.47)	0.13 (-1.32, 1.58)
ceramic plancha, Douglas fir (milled)	-0.84 (-1.27, -0.42)	0.39 (-0.02, 0.8)
metal plancha, Douglas fir (milled)	-0.93 (-1.84, -0.03)	0.66 (-0.18, 1.49)

Note: FP was transformed using μ = 3.00 and σ = 1.42. MCE was transformed using μ = 0.95 and σ = 0.03.

Carbon monoxide (CO) emissions factors:

$$ln(CO\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$$

	α_j	γ
mud chulha, Douglas fir (milled)	4.05 (3.6, 4.5)	-0.18 (-0.32, -0.04)
mud chulha, eucalyptus (split)	4.39 (3.93, 4.85)	-0.18 (-0.32, -0.04)
metal grill, Douglas fir (milled)	3.93 (3.56, 4.3)	-0.18 (-0.32, -0.04)
metal grill, eucalyptus (split)	4.47 (4.01, 4.92)	-0.18 (-0.32, -0.04)
rocket elbow, Douglas fir (milled)	3.42 (3.11, 3.72)	-0.18 (-0.32, -0.04)
three-stone fire, Douglas fir (milled)	4.05 (3.67, 4.42)	-0.18 (-0.32, -0.04)
rocket elbow, eucalyptus (split)	3.79 (3.41, 4.16)	-0.18 (-0.32, -0.04)
three-stone fire, eucalyptus (split)	4.39 (3.93, 4.86)	-0.18 (-0.32, -0.04)
ceramic plancha, Douglas fir (milled)	3.65 (3.33, 3.98)	-0.18 (-0.32, -0.04)
metal plancha, Douglas fir (milled)	3.05 (2.44, 3.66)	-0.18 (-0.32, -0.04)

Note: FP was transformed using μ = 3.00 and σ = 1.42. MCE was transformed using μ = 0.95 and σ = 0.03.

Batch-fed charcoal stoves

Fine particulate matter ($PM_{2.5}$) emissions factors:

$$ln(PM_{2.5}\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$$

	α_j	γ
ceramic jiko, hardwood (lump)	-1.52 (-2.36, -0.68)	0.39 (-0.15, 0.92)
ceramic jiko, coconut (briquettes)	-1.6 (-2.46, -0.74)	0.39 (-0.15, 0.92)
metal jiko, hardwood (lump)	-1.46 (-2.27, -0.65)	0.39 (-0.15, 0.92)
metal jiko, coconut (briquettes)	0.13 (-1.11, 1.37)	0.39 (-0.15, 0.92)

Note: FP was transformed using μ = 1.31 and σ = 0.53. MCE was transformed using μ = 0.83 and σ = 0.05.

Carbon monoxide (CO) emissions factors:

$$ln(CO\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$$

	α_j	β_j
ceramic jiko, hardwood (lump)	5.47 (5.33, 5.61)	0.05 (-0.04, 0.15)
ceramic jiko, coconut (briquettes)	5.29 (5.09, 5.5)	-0.28 (-0.56, 0)
metal jiko, hardwood (lump)	5.65 (5.56, 5.75)	-0.04 (-0.12, 0.04)
metal jiko, coconut (briquettes)	5.91 (5.47, 6.35)	-0.18 (-0.59, 0.24)

Note: FP was transformed using μ = 1.31 and σ = 0.53. MCE was transformed using μ = 0.83 and σ = 0.05.

Appendix A14. Table of p-values for top models coefficients

Significance of interaction terms (i.e., the stove-fuel specific slopes), where EF is the emissions factor of interest, FP is firepower, MCE is modified combustion efficiency, α_j are the coefficients for stove-fuel specific intercepts , β_j are the coefficients for stove-fuel specific slopes, (continuous-feed wood: j = 1,…,10; batch-fed charcoal: j = 1,...,4), γ is a constant slope coefficients across all stove-fuel combinations, and epsilon (ϵ) is random error. Significance levels were calculated by comparing the best model as selected by AICc with and without interaction terms via an analysis of variance (ANOVA) test. FP and MCE were both scaled (i.e., normalized) before the models were fitted. (See SI14 for scaling factors.)

Continuous-feed wood stoves	p-value
$ln(PM_{2.5}\ EF) = \alpha_j + \beta_j \cdot MCE + \gamma \cdot FP + \epsilon$	<0.0001*
$ln(BC\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$	0.0110
$ln(CO\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$	0.159
Batch-fed charcoal stoves	p-value
$ln(PM_{2.5}\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$	0.144
$ln(CO\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$	0.0239

* Note that for the $PM_{2.5}$ model for continuous-feed wood stoves an interaction between stove-fuel type and FP is significant when comparing a model with no interactions to a model with an interaction with FP ($p < 0.001$) and when comparing a model with both interaction terms (i.e., FP and MCE) to a model with only an interaction with MCE ($p = 0.0123$); however, the model with an interaction with MCE only was selected as the top-performing model with AICc.

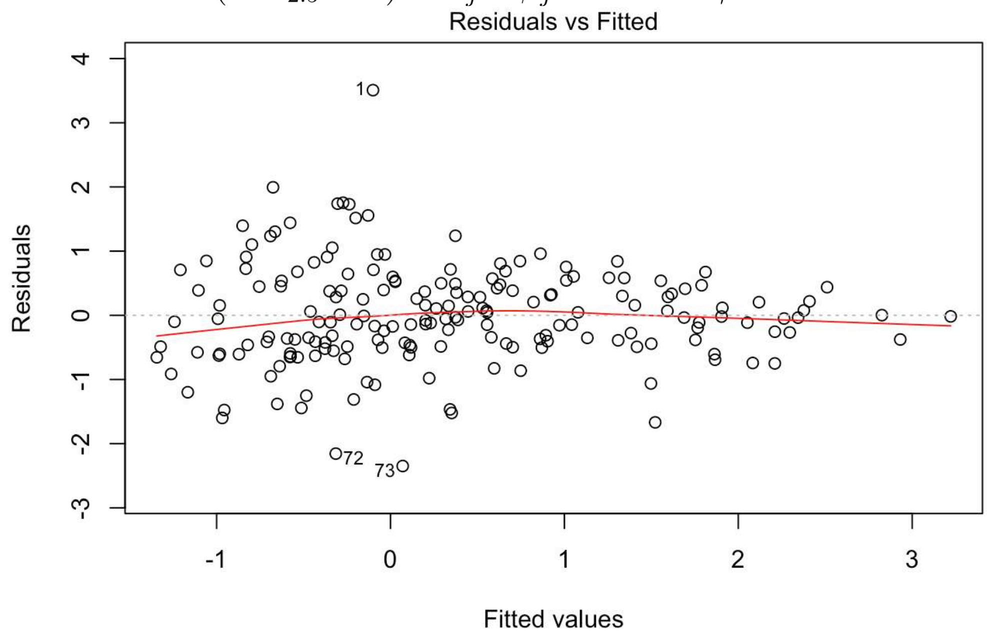
Appendix A15. Linear regression diagnostic plots (fitted values vs. residuals)

Data are the top linear regression models for each pollutant

Continuous-feed wood stoves

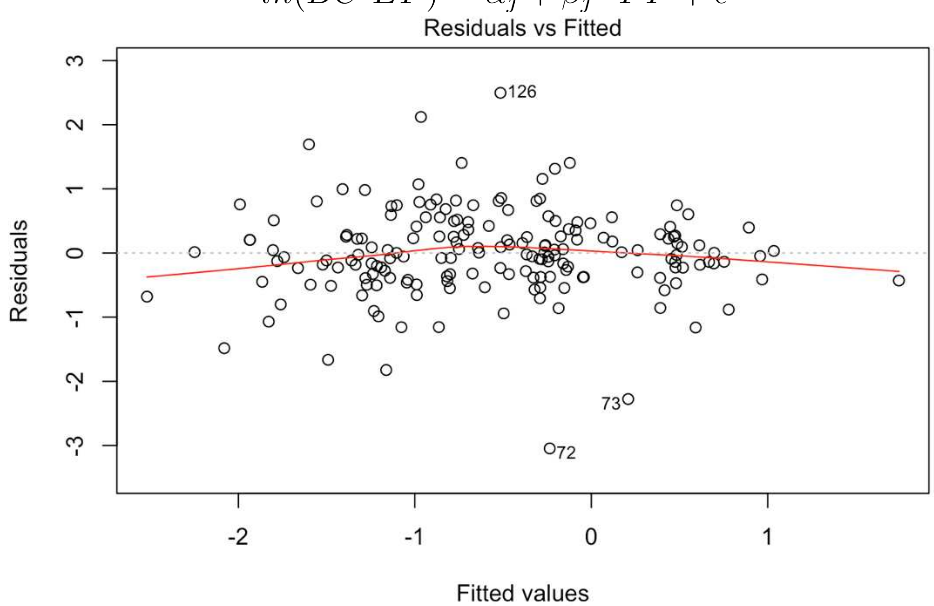
Fine particulate matter ($PM_{2.5}$) emissions factors:

$$ln(PM_{2.5}\ EF) = \alpha_j + \beta_j \cdot MCE + \gamma \cdot FP + \epsilon$$



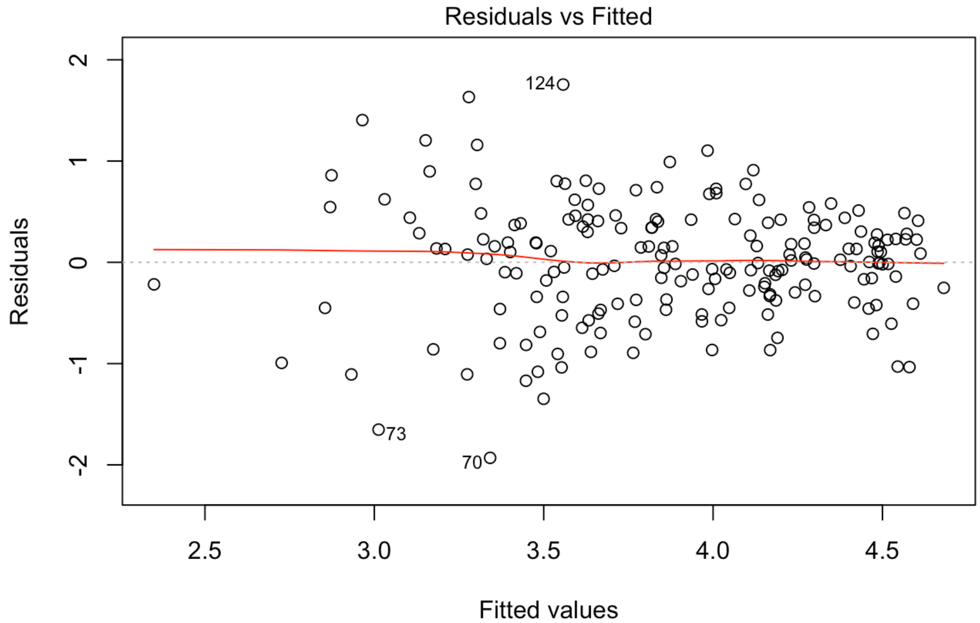
Black carbon (BC) emissions factors:

$$ln(BC\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$$



Carbon monoxide (CO) emissions factors:

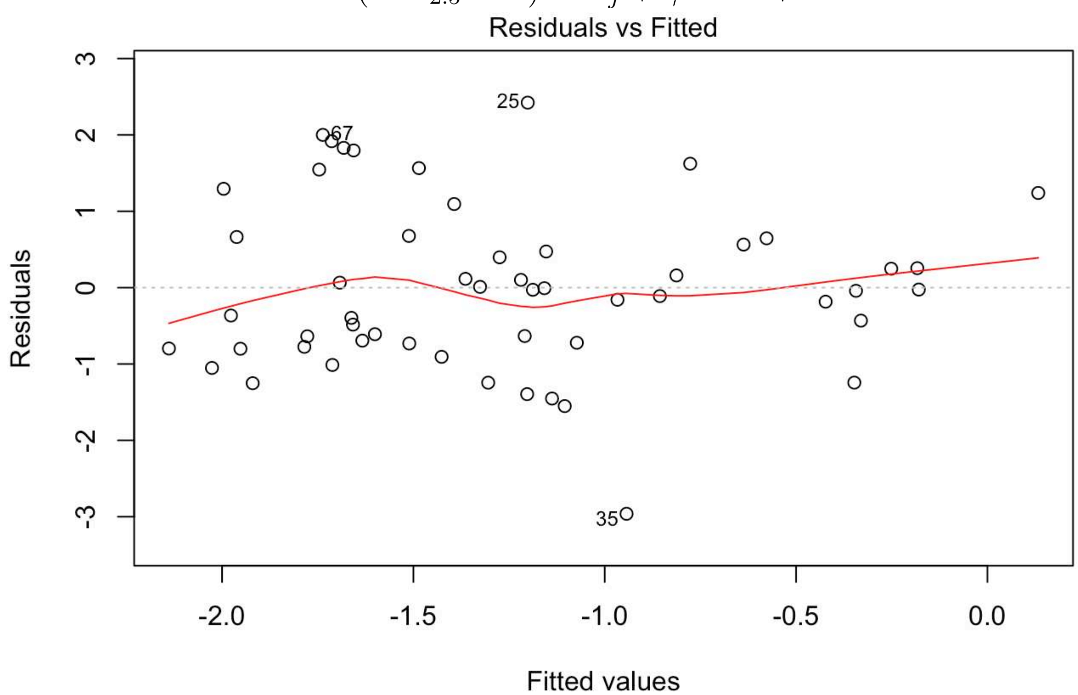
$$ln(CO\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$$



Batch-fed charcoal stoves

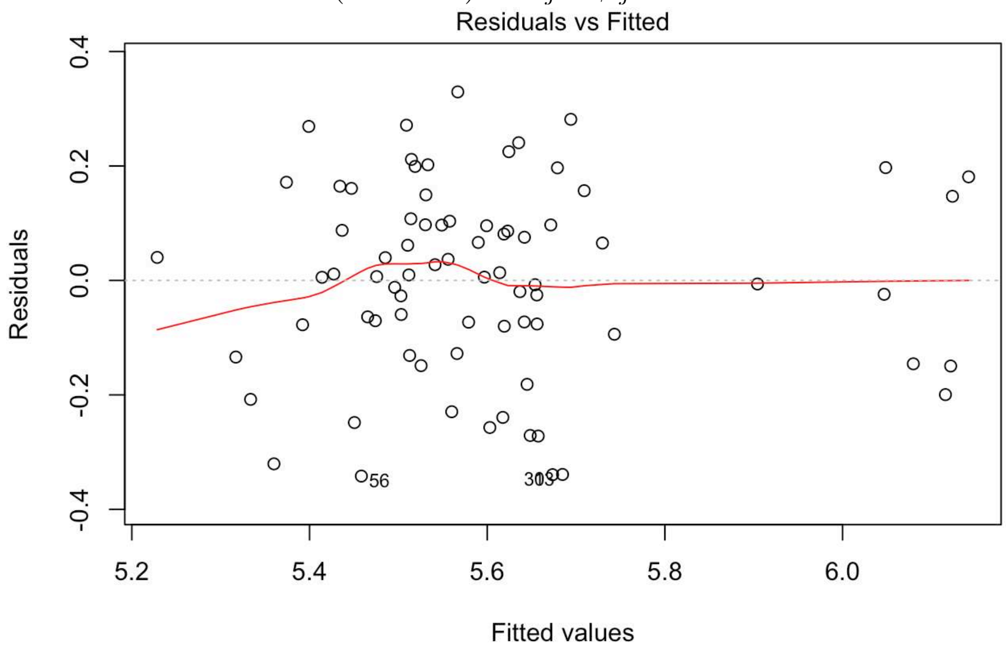
Fine particulate matter ($PM_{2.5}$) emissions factors:

$$ln(PM_{2.5}\ EF) = \alpha_j + \gamma \cdot FP + \epsilon$$



Carbon monoxide (CO) emissions factors:

$$ln(CO\ EF) = \alpha_j + \beta_j \cdot FP + \epsilon$$

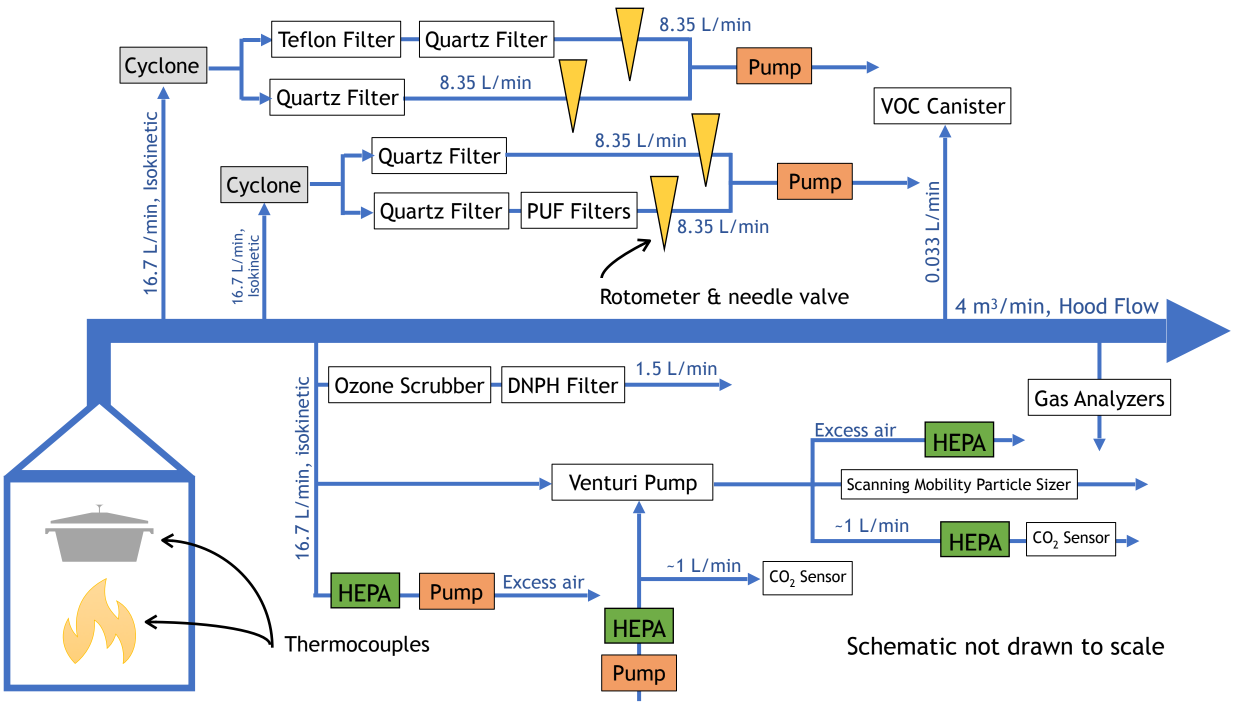


APPENDIX B

Appendix B1. Stove makes and models

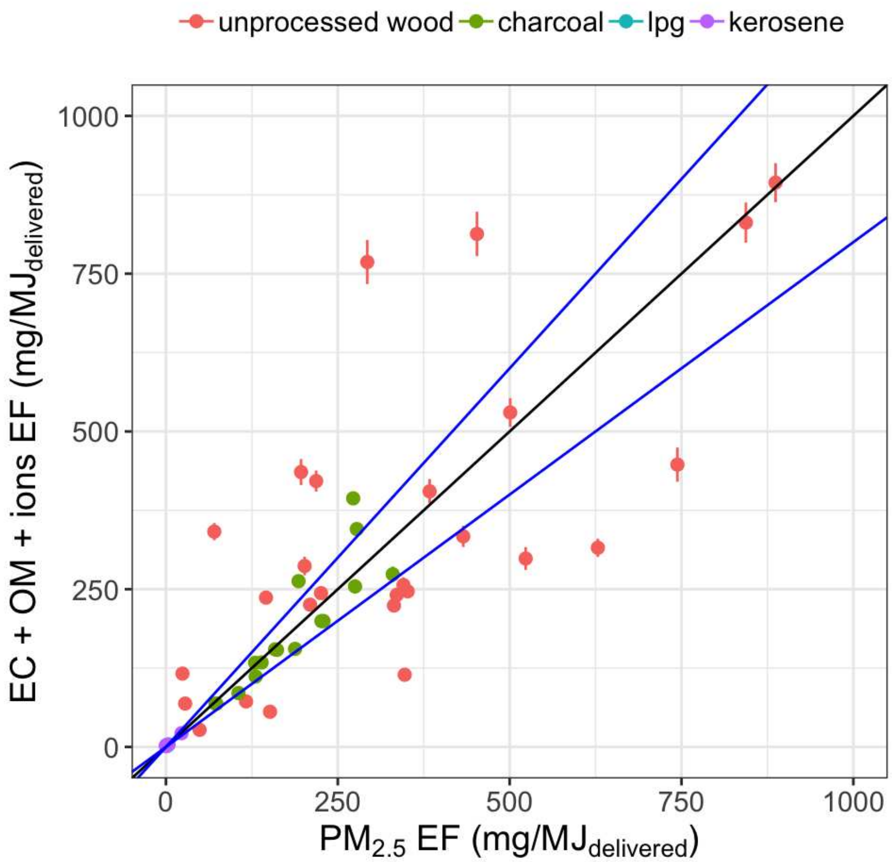
stove name	make	model
three-stone fire	artisan	artisan
chulha	artisan	artisan
rocket-elbow	Envirofit	G3300
built-in plancha (with chimney)	Envirofit	HM5000
fan rocket-elbow	Biolite	Homestove[1]
forced-draft gasifier	Philips	HD4012
ceramic charcoal	artisan	artisan
metal charcoal	BURN	jikokoa
wick kerosene	Cook 'n Lite	82
pressurized kerosene	Butterfly	2412
lpg	WokSmith	WS-C1-25K

Appendix B2. Instrumentation schematic for emissions testing



Appendix B3. Particle emissions mass balance

$PM_{2.5}$ mass versus the sum of inorganic ions, elemental carbon, and organic matter. The error bars represent the uncertainty from the Sunset OCEC analyzer measurement. The black line represent the line $y = x$ and the blue lines represent $\pm 20\%$ deviation from that line.



Appendix B4. List of carcinogenic compounds

pollutant	phase	NTP[1]	IARC[2]
benzene	gas	Known to be a human carcinogen	Group 1: Carcinogenic to humans
formaldehyde	gas	Known to be a human carcinogen	Group 1: Carcinogenic to humans
styrene	gas	Reasonably anticipated to be a human carcinogen	Group 2A: Probably carcinogenic to humans
acetaldehyde	gas	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
isoprene	gas	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
benz[a]anthracene	particle	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
benzo[a]pyrene	particle	Reasonably anticipated to be a human carcinogen	Group 1: Carcinogenic to humans
benzo[b]fluoranthene	particle	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
benzo[j]fluoranthene	particle	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
benzo[k]fluoranthene	particle	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans
cyclopenta[cd]pyrene	particle	NA	Group 2A: Probably carcinogenic to humans
dibenzo[a,h]anthracene	particle	Reasonably anticipated to be a human carcinogen	Group 2A: Probably carcinogenic to humans
indeno[1,2,3-cd]pyrene	particle	Reasonably anticipated to be a human carcinogen	Group 2B: Possibly carcinogenic to humans

[1] National Toxicology Program (NTP)
[2] International Agency for Research on Cancer (IARC)

APPENDIX C

Appendix C1. Side-by-side photos of the laboratory and field for each stove type

	Laboratory stoves	Field stoves
Ceramic jiko		
Ceramic plancha		
Mud chulha		
Rocket elbow		
Three-stone fire		

Appendix C2. Photos illustrating thermocouple installation and securement

Appendix C3. Model results with test-averaged firepower and MCE measurements