

FIELD EMISSION MEASUREMENTS OF SOLID FUEL STOVES IN YUNNAN, CHINA

BY

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THESIS

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ABSTRACT

Emission factors of carbon monoxide (CO), particulate matter (PM_{2.5}), organic carbon (OC), and elemental carbon (EC), as well as combustion efficiency and particle optical properties were measured during 37 uncontrolled cooking tests of residential stoves in Yunnan Province, China. Fuel mixtures included coal, woody biomass, and agricultural waste. Compared to previously published emission measurements of similar stoves, these measurements have higher CO and PM_{2.5} emission factors. Real-time data show two distinct burn phases: a devolatilization phase after fuel addition with high PM_{2.5} emissions, and a solid-fuel combustion phase with low PM_{2.5} emissions. The average emission factors depend on the relative contributions of these phases, which are affected by the services provided by the stoves. Differences in stove and fuel characteristics that are not represented in emission inventories affect the variability of emission factors much more than do the type of solid fuel or stove. In developing inventories with highly variable sources such as residential solid-fuel combustion, we suggest that (1) all fuels should be accounted for, not just the primary fuel; (2) the household service provided should be emphasized rather than specific combinations of solid fuels and devices; and (3) the devolatilization phase should be explicitly measured and represented.

This work also advances the development of portable sampling instruments for climate-relevant emission characterization with regard to usability, reliability, and accuracy.

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CHAPTER 1: INTRODUCTION*

1.1 BACKGROUND

Approximately half the world's population, including roughly half of China, uses solid fuels, including coal, wood, charcoal, agricultural residue, and dung, for household cooking and heating¹. Solid fuels are burned in a wide range of household stoves which often have simple combustion chambers. The resulting low combustion efficiency and uncontrolled exhaust emits pollutants such as carbon monoxide (CO), particulate matter (PM), volatile organic compounds (VOC), nitrogen oxides (NO_x), and sulfur dioxide (SO₂). Health effects from exposure to stove emissions include strokes, heart attacks, chronic obstructive pulmonary disease, respiratory infections, and lung cancer. The Institute for Health Metrics and Evaluation attributes over 2.5 million deaths per year to household air pollution exposure from solid fuel stoves.¹ In China, the dominant source of indoor air pollution is from solid fuels,² despite many years of improved stove programs.^{3,4} In Northeastern Yunnan Province, the region of this study, lung cancer rates are among the highest in China, and decades of health studies have shown a strong link between lung cancer and household air pollution from coal stoves.^{5,6} Despite understanding the linkages of residential solid fuel combustion with health impacts, air pollution from residential stoves persists as a major global health burden.

In addition to health effects, emissions of residential solid fuel stoves contribute to global and regional climate change by altering chemical and physical processes in the atmosphere that affect clouds, weather patterns, and the global energy balance. Estimates of globally-averaged climate forcing by aerosols from residential solid fuel stoves range from 0.4 Wm⁻² to -0.3 Wm⁻².⁷ Solid fuel stoves are estimated to be responsible for approximately one quarter of global primary organic aerosol (POA) and black carbon (BC) emissions.^{7,8} POA is a major climate-cooling species⁷, although some POA, known as brown carbon, has been attributed to climate warming.⁹ BC is the second or third largest anthropogenic climate-warming species. China is considered the largest contributor of global BC emissions.¹⁰ Residential solid fuel stoves are estimated to be responsible for approximately half of Asian BC emissions.¹¹ BC emitted in some parts of Asia is of particular importance due to the proximity of the Himalayan glaciers, where BC deposition on

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snowpack amplifies local radiative forcing of BC by an order of magnitude and contributes to the melting of glaciers.¹² The Himalayan region, known as the Third Pole, is the third largest ice mass in the world after the Arctic and Antarctic. It provides fresh water for 20% of the world's population. Melting of Himalayan glaciers is causing changes in water availability, including increased flooding in some regions, and loss of water in other regions.^{13,14} This work is part of a larger study to better understand BC emissions in regions adjacent to the Himalayas.

Climate forcing estimates are obtained from simulations of atmospheric chemistry and transport that require emission rates and particle composition as inputs; those models also assume particle optical properties to estimate changes in radiative balance. Measurements are limited of emission quantity, particulate composition -- such as mass fractions of black carbon and organic carbon, and optical properties. Because emissions from solid-fuel stoves are strongly dependent on operating conditions, emission characteristics under real, rather than prescribed, operation are important in providing realistic model inputs. Cooking events simulated in laboratory testing may not be representative of field performance and may underestimate emissions.¹⁵⁻²⁵ Real-time measurements of emissions play an important role in identifying factors that lead to high emissions.

This work advances the development of portable sampling instruments for climate-relevant emission characterization with regard to usability, reliability, and accuracy, and applies those instruments to a field campaign in northeast Yunnan Province, China. The Yunnan field campaign served the dual purpose of producing emission measurements of coal stoves and of field testing emission equipment. The equipment development described here has informed further development of emissions equipment for residential stoves, heating stoves, brick kilns, and diesel engines.

1.2 PREVIOUS EMISSION MEASUREMENT EQUIPMENT

Field measurements of solid fuel cookstove emissions have been scarce because the stoves are usually located in rural areas that are difficult to access with conventional emission measurement equipment. The development of portable sampling instruments in recent years has made it practical to sample at homes in remote locations, which has led to an increase in the number of field measurements to fill data gaps. Emission sampling requires two physical

components: a method of capturing air from the exhaust stream and instrumentation to measure the captured emissions.

Some previous studies have used a portable or semi-portable collection hood attached to a duct and blower to capture stove emissions. A small sample was drawn continuously from the duct to instrumentation, and emission ratios were used to determine emission metrics.^{16,17,19} Johnson et al.¹⁶ collected gaseous emissions in Tedlar bags for subsequent laboratory analysis by gas chromatography. Johnson et al.¹⁷ and Coffey et al.¹⁹ measured CO and CO₂ concentrations directly in the sample train with gas sensors. Several studies have used portable and semi-portable sampling systems to sample chimney stove emissions.^{16,17,21,22,26-31} In some of these studies, emissions were sampled through a probe located at or near (within 1 meter beyond) the chimney outlet.^{21,22,29-31} Du et al.²² mixed forced dilution with the emission sample, while the others did not add forced dilution, although some dilution occurred naturally in the plume before the sample entered the sample probe. In other studies, the emissions were sampled directly from the chimney through a port in the chimney wall.^{16,17,26-28} In these studies, forced dilution was mixed with the emission sample. Sun et al.²⁶ calculated emission factors using the chimney flow rate, while all others calculated emission factors from emission ratios using the carbon balance method.

Extensive development was done on sampling equipment (described in Chapter 2: Emission Equipment Development and Appendix A: Book of Fumitron). This equipment was based on the principles of the ARACHNE portable partial capture sampling system used in Roden et al.³² and Roden et al.¹⁵ which was a collection of laboratory equipment mounted to a cart. Since then, subsequent iterations of the equipment described in Weyant et al.³³ and Weyant et al.³⁴ have become smaller, and more portable, and led to the design of equipment used in this work. Other field campaigns have used similar portable partial capture sampling systems.^{18,23,25,29}

1.3 PREVIOUS MEASUREMENTS OF EMISSIONS

Several studies have reported Chinese stove emissions for a large variety of coal and biomass fuel types. Most previous studies were controlled tests with prescribed burn sequences intended to simulate natural stove operating practices.^{26-28,35-44} Some emissions have been measured using prescribed cooking tasks or burn sequences of water boiling tests.^{27,28,36,43} Some prescribed burn sequences are representative of space heating.^{26,37-42} Shen et al.⁴⁴ measured emissions of

prescribed burn sequences representative of cooking. Zhang et al.³⁵ did not report the burn sequence.

Several studies were uncontrolled field tests that measured emissions during cooking.^{21,25,29,30} Chen et al.³¹ measured during cooking and space heating. Most of these studies used individual fuel types, and not the fuel mixtures that are common in many locations.

This work complements previous studies by providing emission measurements for complete burn sequences with user-chosen fuel mixtures of coal and biomass during uncontrolled cooking events in homes in northeastern Yunnan Province, China. In addition, the work uses real-time data to identify the causes of high-emitting events.

CHAPTER 2: METHODS*

2.1 SELECTION OF HOMES, STOVES, FUELS, AND COOKING EVENTS

Stove emissions were measured in nine rural mountain villages (approximately 2000 m elevation) in Fuyuan and Xuanwei counties of the Qujing prefecture in northeastern Yunnan Province, China (see map Appendix A: Figure 11) during July and August 2013. The region was chosen for its predominant use of coal fuel, which sets it apart from many other regions surrounding the Himalayas that predominantly use biomass fuel. The sample of households was selected in conjunction with in-country partners (Qujing Center for Disease Control and Prevention and local medical clinics). Households in each village were selected to have stove and fuel types common to that village, and further refined to households with willing participants and accessible chimney outlets for the homes using chimney stoves. Most homes in the study region used either chimney or portable solid-fuel stoves. Most of the homes selected were connected to a centralized electricity grid, and usually also had an electric hot plate, induction heater, or electric rice cooker.

We use the term “cooking event” to describe the preparation of a meal on a single stove, from the start to cessation of cooking. Measurements were taken during 37 cooking events in 22 households, including two cooking events measured on the same stove with similar fuel mixtures in 10 of the households. The cooking events ranged from 35 to 212 minutes in length, with an average length of 112 minutes, and a total sampling time of 70 hours. Thirty-two cooking events had complete burn sequences that include stove ignition. Five cooking events did not include stove ignition because the stove remained burning from a previous cooking task. The cooking events were uncontrolled, meaning that household residents performed their normal daily cooking tasks by choosing their own foods, cooking procedures, and fuels with no direct influence from research technicians. A variety of cooking tasks were observed including boiling, simmering, frying, and reheating. The most common task was heating water. Food was cooked in pots, pans, woks, and roasted directly on the coals. Pig fodder was stewed in large woks. In some cooking events, the stove was filled with fuel, then ignited, and not tended again the entire

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cooking event. In other cooking events, fuel was added in stages. Sometimes the fuel bed was prodded and tamped, ashes were removed, and the fuel bed was fanned by waving a hand-held fan or using an electric blower.

Fourteen chimney stoves were sampled that resembled the improved stove delivered by the Chinese National Stove Program in the 1980s (Figure 1a).³ These stoves had similar design features between households, but varying dimensions. They were built into kitchen counters with masonry materials (bricks, blocks, ceramic tiles, and mortar) and iron cooktops. Fuel was batch-loaded from the cooktop and intake air was supplied from a passage below the combustion chamber. One free-standing cast iron chimney stove was also sampled. Chimneys one to three stories (3 – 9 meters) tall were built into the wall of each home.

Seven portable stoves were sampled that resembled metal buckets with grates to suspend the fuel and intake air holes at the bottom (Figure 1b). Two of these portable stoves were designed specifically for burning coal honeycomb briquettes and had a layer of ceramic insulation on the inside wall (Figure 1c). The portable stoves were usually ignited outside and then brought inside the home after the smoke had subsided, but sometimes they remained outside or inside the home for the entire cooking event.



Figure 1: Stove types. a) Chimney stove with the chimney built into the wall of the house (n = 25). b) Portable bucket stove (n = 9). c) Portable stove for coal honeycomb briquettes (n=3).

Household residents were requested to operate their stoves and choose fuels according to their normal practices in that season. Coal was used in the form of raw chunks and honeycomb briquettes. Industrially manufactured honeycomb briquettes were made of pulverized coal with a clay binder pressed into a cylindrical shape with holes. The holes in the briquette provide an air

flow path during combustion and give the briquette a honeycomb appearance that leads to the name.

Lit with lighters and matches, biomass fuels were used to ignite the stoves. Biomass and coal were premixed in the combustion chamber before lighting or added in series over a period of several minutes after lighting. The biomass fuels were categorized into corncobs and woody fuels, for the purpose of estimating the fuel composition and energy content, and for attributing differences in emissions to fuels. Corn was a predominant crop in the study region, and dried corncobs without kernels were an abundant fuel. Woody fuel refers to a large variety of tree biomass, including milled lumber, split wood scraps, branches, twigs, leaves, pine needles, and cones.

Fuel type was recorded for all cooking events, and fuel mass consumed was measured for 21 cooking events by weighing the fuels loaded into the stove and the remaining char. Fuel analysis, including heating value and composition, is presented in Appendix B: Fuel Analysis.

2.2 EMISSION SAMPLING

Stove emissions were sampled using the partial capture method, and emission factors were calculated using the carbon balance method.^{36,45,46} This method involves capturing a small, representative sample of the plume (partial capture method) from which the ratio of each pollutant to total carbon is calculated, after subtraction of background concentrations. The fuel mass that contributed to the air concentration of carbon is inferred using the fuel carbon fraction (carbon balance method, equations in Appendix C: Emission Factor Calculations).

The portable sampling system is shown below in Figure 2 and Figure 3, and is documented in detail in Appendix I: Book of Fumitron. Since its first description,³² the partial capture sampling system has been used in multiple field campaigns,^{15,48} and has undergone several improvements. The entire system fits in two hand-held cases and consists of a sensor box (10 kg), batteries, dilution sample probes for chimney and non-chimney stoves, and accessories for handling filters and performing flow checks (20 kg). The sensor box has sample pumps that draws a gas sample through the filters and sensors (Figure 3). It records real-time sensor readings and also collects integrated particulate samples. Improvements in the latest version of the sensor box (described in Chapter 4: Emissions Equipment Development) include continuous measurement of background CO₂ and CO.

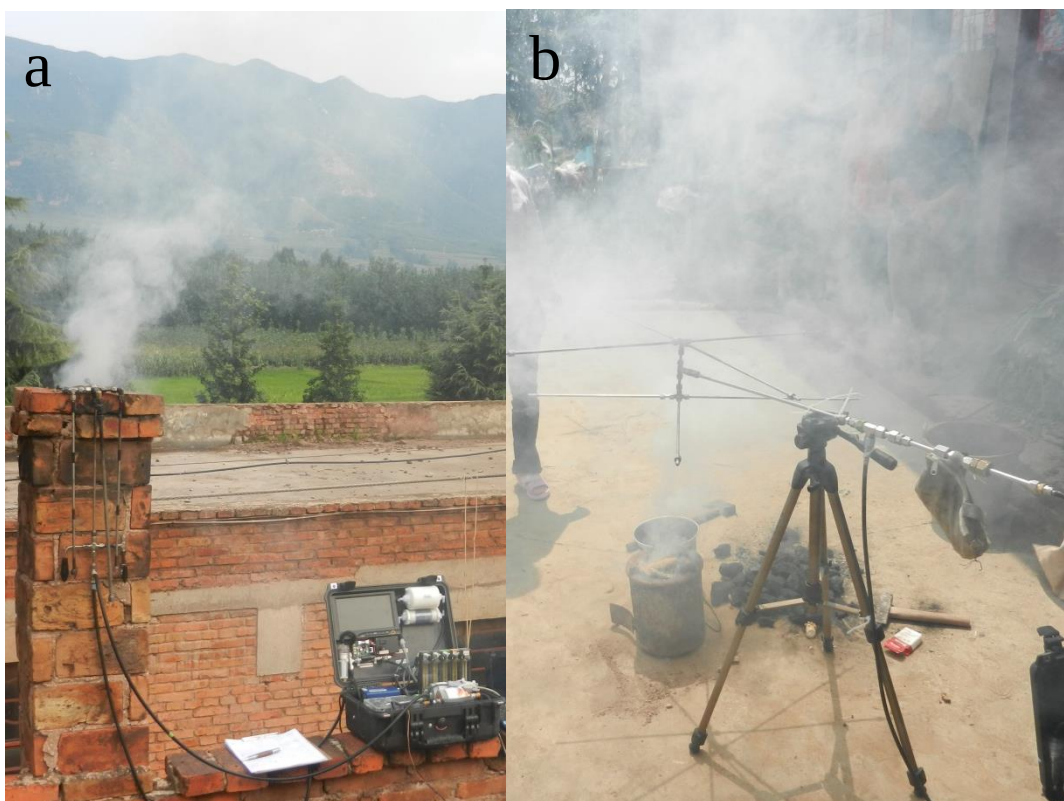


Figure 2: Portable sampling system. a) Sensor box sampling a chimney stove. b) 8-arm sampling probe for non-chimney stoves.

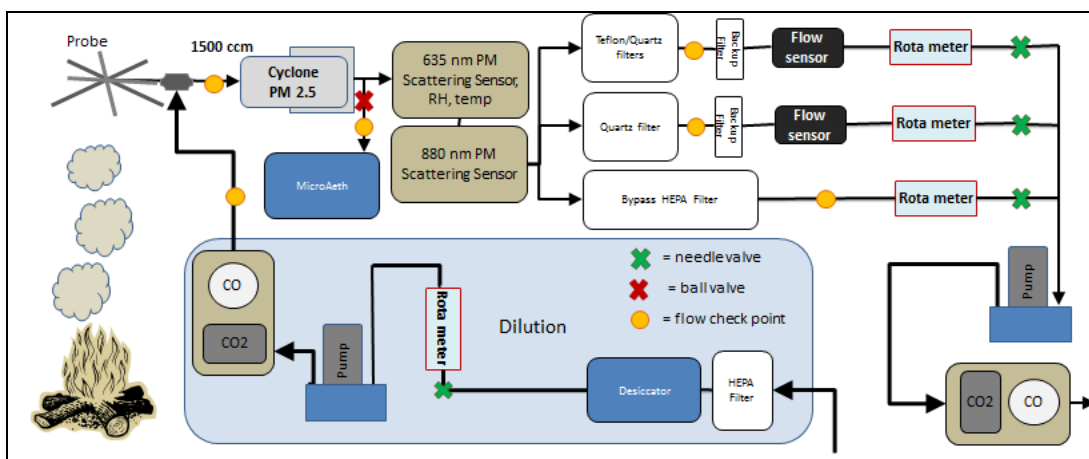


Figure 3: Sampling system flow schematic.

The chimney probe is a stainless steel tube, similar to an industrial stack sampling probe, located at the top outlet of the chimney. For portable stoves without a chimney, an 8-armed stainless steel sample probe was used (described in Roden et al.)³² with 24 sample inlet points designed to capture a representative sample through a cross-section of the plume without installing hoods that can interfere with cooking practice. For non-chimney stoves, emissions

from frying food (not from the stove) were also collected, but these emissions were relatively low compared to the stove emissions. PM emissions from frying food were observed during three cooking events, accounting for about 5% of the total PM emissions of a burn cycle.

Immediately downstream of the sample probe, the sample is mixed with dry dilution air to cool the sample to ambient temperatures, reduce the humidity below 50%, and reduce the sample concentrations to an appropriate range for the sensors. At the entrance into the sensor box, the sample passes through a cyclone with a 2.5 μm cut point to remove large particles of dust and ash that should not be counted as combustion emissions.

Primary and background CO was measured with Alphasense CO-AX electrochemical cells on Waltech potentiostat boards. Primary and background CO₂ was measured with CozIR SprintIR Wide Range nondispersive infrared sensors. Gas sensors received a two-point calibration with zero and span gas before and after the field campaign. The measured calibration drift and sensor specifications were used to estimate measurement uncertainty.

PM light absorption was measured at 880 nm with an MicroAeth model AE51. To minimize the number of filter changes, the flow was reduced to 20 ccm. The filter cartridge was changed between one and ten times per cooking event. PM light scattering was measured at two wavelengths, red (635 nm) and infrared (880 nm), by two optical PM sensors. The PM sensors were calibrated in the lab before and after the field campaign. The red wavelength sensor was calibrated against a TSI model 3563 3-wavelength nephelometer, and the infrared sensor was calibrated against a Droplet Measurement Technologies PAX Photoacoustic (870 nm). Background PM was determined with the scattering sensor by sampling background air for 10 minutes before and after each cooking event. The average background scattering coefficient was converted to PM mass concentration using the event-specific MSC.

Real-time scattering coefficients were often an order of magnitude higher than the calibrated range of the sensors ($b_{\text{sp}} > 100,000 \text{ Mm}^{-1}$) and beyond the range of a highly credible scattering measurement. Real-time absorption coefficients were also often higher than the AE51 could measure, resulting in real-time data gaps for events when large quantities of dark PM are emitted. Furthermore, the filter-based absorption measurement method utilized in the AE51 is subject to several known artifacts.⁴⁷ Therefore, the scattering and absorption coefficient data and dependent calculated metrics reported in this work (optical emission factors, single scattering albedo or SSA, and optical cross-sections) are considered approximate and used for relative

comparisons. Despite the uncertainty, the real-time optical scattering and absorption data are still valuable to show when PM was emitted during the cooking events.

The sampling system has a single stage of dilution, meaning the PM and gas samples have the same dilution ratio. The dilution ratio was set between 3:1 and 15:1 (dilution air : stove exhaust). However, the stove exhaust was diluted naturally before entering the probe, to result in an overall dilution ratio ranging between 100:1 and 1000:1.

PM_{2.5} samples were collected on 47 mm diameter polytetrafluoroethylene (PTFE) and quartz filters in parallel as described in Roden et al.³³ and Kirchstetter et al.⁴⁸ The PTFE filter was used for gravimetric analysis to determine the PM_{2.5} mass concentration. The quartz filter was used for thermal-optical analysis with a Sunset Laboratory OC/EC analyzer to determine organic carbon (OC) and elemental carbon (EC) mass concentrations. OC is the carbon component of organic matter (OM). The Sunset Analyzer PAR file is Appendix B: Sunset Analyzer PAR File. A second backup quartz filter located directly downstream of the PTFE filter collected semi-condensable organic carbon that passed through the PTFE filter. The carbon collected on the backup quartz filter was used as an estimate of adsorbed organic carbon on the primary quartz filter, and was subtracted from that value. The flow through each filter was set with needle valves, measured before and after each sampling event with a bubble meter, and monitored in real-time with digital flow sensors. Gravimetric and OCEC filters were stored and transported in sealed petri dishes below 0°C to minimize volatilization losses. Gravimetric filters were weighed with a Cahn C-31 microbalance at University of Illinois. Filters were conditioned at 25 °C and 50 % RH for 24 hours before weighing. Field blanks were collected and subtracted from the sample filter mass.

Real-time data were recorded every two seconds for CO concentration, CO₂ concentration, light scattering by particles at 635 and 880 nm wavelength (b_{sp} , m⁻¹), light absorption by particles at 880 nm wavelength (b_{ap} , m⁻¹), sample humidity, sample temperature, probe temperature, and filter flow rates. Concentrations of CO and CO₂ attributable to combustion exhaust were determined by subtracting real-time background concentrations from real-time sample concentrations. Background concentrations were measured at 1-2 meters horizontal distance from the plume to capture the air parcel that mixed with the plume. The background concentrations depend on ventilation and were between 0 and 50 ppm for CO and

400-2000 ppm for CO₂. Real-time emission data are reported with averages over one-minute time intervals.

2.3 DATA PROCESSING

Emission factors (EF) provide a normalized comparison of emissions for different technologies or activities and are used to estimate total emissions for use in climate and pollution models. Fuel mass-based emission factors (EF_m) of CO, PM_{2.5}, OC, and EC are reported as grams of pollutant per kilogram fuel consumed (g/kg), and energy-based emission factors (EF_e) are reported as grams of pollutant per MJ of heat content of the fuel consumed (g/MJ) (see Appendix B: Emission Factor Calculations). Energy-based emission factors are common for use in emission inventories, but differ from the metric used in standards, grams of pollutant per MJ of heat delivered. Emission rates (ER, g/hr) are also reported by multiplying EF by the fuel consumption rate.

Modified combustion efficiency (MCE) is an indicator of combustion completeness and is a proxy for nominal combustion efficiency (Equation 1). An MCE of unity indicates complete combustion. For solid-fuel stoves, MCE generally ranges between 0.8 and 1.

$$\text{MCE} = \frac{\text{CO}_2(\text{ppm})}{\text{CO}(\text{ppm}) + \text{CO}_2(\text{ppm})} \quad (1)$$

Total carbon (TC) refers to the carbon component of particulate matter and is the sum of OC and EC, while PM_{2.5} includes the oxygen and hydrogen bound to that carbon as well as other non-carbonaceous components. The EC/TC ratio is a dimensionless parameter that indicates the fraction of aerosol carbon in the form of elemental carbon. The OC/TC ratio indicates the fraction of PM carbon in the form of organic carbon.

To assist in estimating the climate impact of particles, we also report some properties that affect radiative transfer in the atmosphere, especially light absorption. EC is reported as a proxy for BC. The single scattering albedo (SSA) is a dimensionless parameter that characterizes absorptivity (Equation 2).

$$\text{SSA} = \frac{b_{\text{sp}}}{b_{\text{sp}} + b_{\text{ap}}} \quad (2)$$

An SSA of zero indicates pure light absorption and a value of unity indicates pure light scattering. Lower SSA values indicate darker PM, usually because of a higher BC fraction.

Integrated filter samples do not provide information about real-time PM emission. Therefore, we use the emission factor metrics EF_{scat} and EF_{abs} (with units of m^2/MJ) as proxies, assuming that the scattering and absorption coefficients are approximately proportional to the real-time concentrations of PM and EC concentration, respectively. The PM scattering coefficient b_{sp} is related to PM concentration through the mass scattering cross-section (MSC), which has units of m^2 (area of light scattered) per gram PM (m^2/g). Likewise, the absorption coefficient b_{ap} is related to EC concentration through the mass absorption cross-section of EC (MAC_{EC} , m^2/g). Although MSC and MAC_{EC} values vary with particle size and composition, this variation is small relative to the variation in PM concentration throughout a cooking event.

Measurement uncertainty was estimated for each input parameter as 68% confidence intervals and then propagated to the final output metrics using first order error propagation. Input uncertainties are tabulated in Appendix G: Measurement Uncertainty Estimates.

CHAPTER 3: FIELD EMISSION MEASUREMENT RESULTS*

3.1 FUEL CONSUMPTION

Figure 4 shows the quantity of fuel used in each cooking event. Overall, the dominant fuel type was coal (66% by mass, 72% by energy), followed by corncobs (22% by mass, 18% by energy), and woody fuels (12% by mass, 10% by energy). Although all events relied on coal for most of the energy needs, coal was always mixed with some type of biomass.

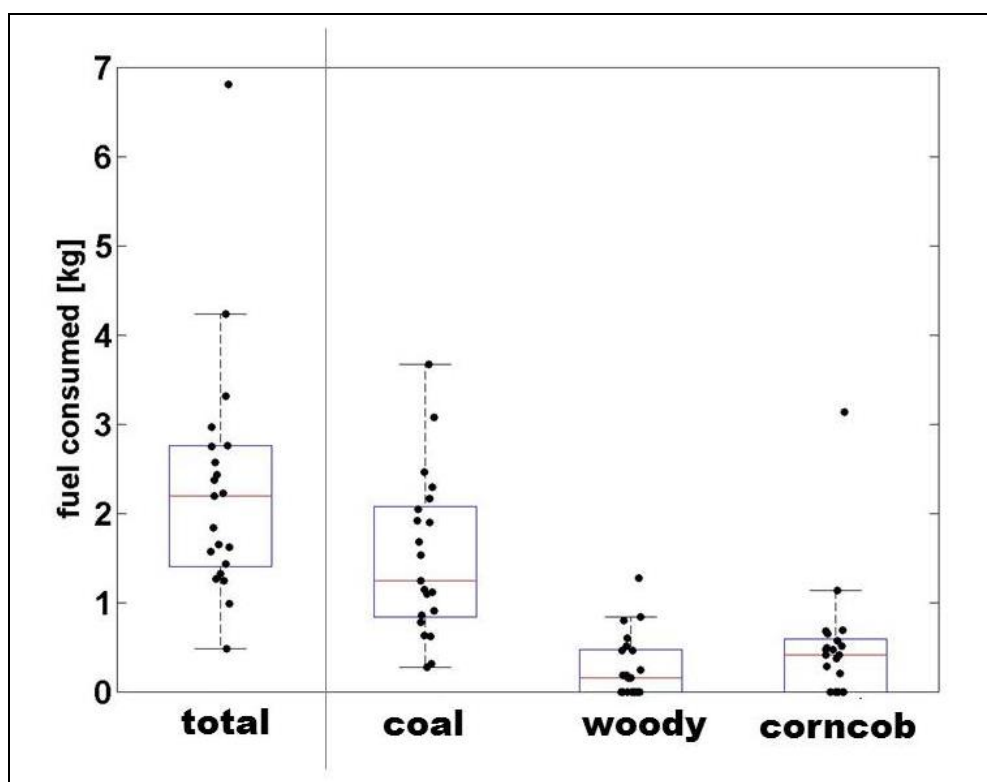


Figure 4: Mass of fuel consumed in each of the 21 cooking events where it was recorded. “Woody” biomass included wood, twigs, leaves, and pine cones.

3.2 MAGNITUDE AND VARIABILITY OF EMISSIONS

Tabulated results for each cooking event are in Appendix F: Summary of Cooking Events. Energy- and mass-based emission factors of CO, PM_{2.5}, OC, and EC are presented in Table 1 for cooking events that had complete burn sequences. The average EF_{e,CO} was 5.8±2.0

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g/MJ (mean $\pm$ 68% confidence interval), and $EF_{e,PM}$ was 0.63 ± 0.55 g/MJ. The confidence interval includes measurement uncertainty and inter-test variability but was dominated by inter-test variability. The $EF_{e,CO}$ range (2.4-10.5 g/MJ) and $EF_{e,PM}$ range (0.07-1.7 g/MJ) both span an order of magnitude. The distribution of CO and $PM_{2.5}$ emission factors for each cooking event is plotted in Figure 5. The average $EF_{m,CO}$ was 126 ± 44 g/kg and the average $EF_{m,PM}$ was 14 ± 9 g/kg. Correlation between PM emission and MCE was low ($R=0.34$). Compared to International Standards Organization cookstove performance targets,⁴⁹ nearly all cooking events with complete burn cycles score Tier 0, the worst tier, for both CO and $PM_{2.5}$ (assuming a thermal efficiency of 0.2 ± 0.1 (13,27,30-32)).

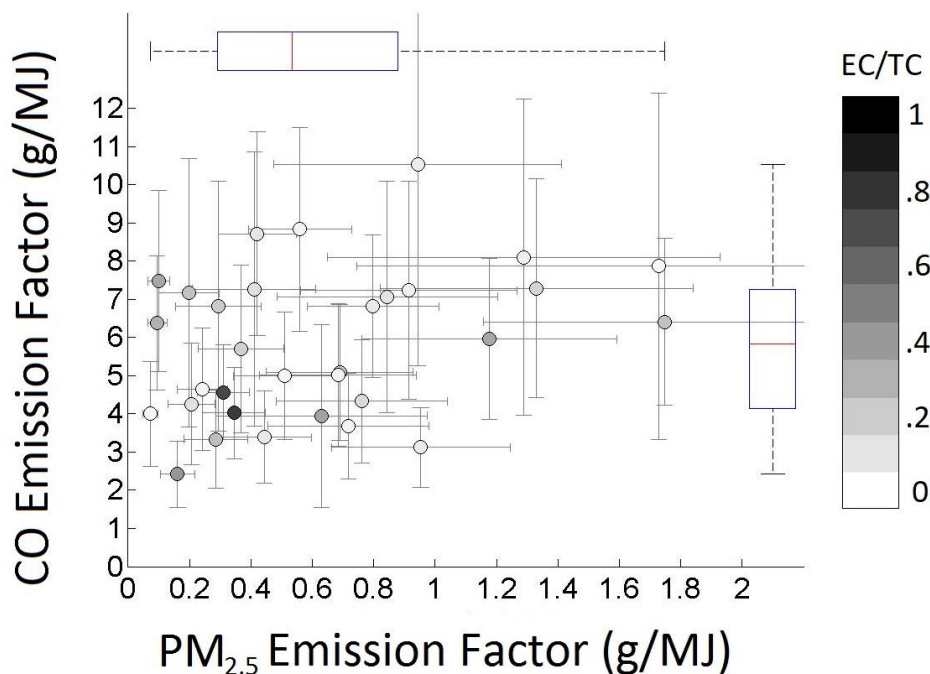


Figure 5: Energy based CO and PM emission factors for each cooking event that has a complete burn sequence. The boxes show quartiles for the overall distribution. The EC fraction is indicated by the shading of each point. Error bars show the measurement uncertainty.

Table 1: Average measured emission metrics for all events that had complete burn sequences (n=32).

Metric	abbreviation	mean	1 σ^a
Fuel Mass Based Emission Factor (g/kg _{fuel})	EF _{m,CO}	126	44
	EF _{m,PM2.5}	14	9.0
	EF _{m,OC}	9.7	7.5
	EF _{m,EC}	2.0	2.5
Energy Based Emission Factor (g/MJ)	EF _{e,CO}	5.8	2.0
	EF _{e,PM2.5}	0.63	0.45
	EF _{e,OC}	0.45	0.35
	EF _{e,EC}	0.09	0.1
Emission Rate (g/hr)	ER _{CO}	161	100
	ER _{PM2.5}	20	17
Modified Combustion Efficiency (-)	MCE	0.90	0.03
Single Scattering Albedo (-)	SSA ₈₈₀	0.68	0.17
Mass Scattering Cross-Section (m ² /g)	MSC ₆₃₅	4.2	1.8
	MSC ₈₈₀	3.1	1.4
Mass Absorption Cross-Section (m ² /g)	MAC _{EC,880}	12.6	5.0

^aThe 1 σ confidence interval is 68% of the Student's T-distribution.

TC accounted for 50 – 100% of the total PM mass, with a mean value of 79%. There is no correlation between composition (EC/TC) and PM_{2.5} emission factors ($R^2 = 0.07$). The mean EC/TC ratio was 0.18 ± 0.18 , indicating that TC was mostly OC. The range of EC/TC was 0.02 –

0.76. The distribution of EC/TC ratios is shown in Figure 6, and the EC/TC ratio is illustrated for each event in Figure 5.

The mean SSA was 0.68 ± 0.17 , with a minimum of 0.31 and a maximum of 0.96, which results in direct radiative forcing in the atmosphere at the time of emission that is approximately neutral (no cooling or warming).⁵⁰ The mean MSC was $4.2 \pm 1.8 \text{ m}^2/\text{g}$ at a wavelength of 635 nanometers and $3.1 \pm 1.4 \text{ m}^2/\text{g}$ at a wavelength of 880 nanometers. These MSC values were higher than for Honduran wood stoves ($2.2 \pm 0.6 \text{ m}^2/\text{g}$ at 530 nanometers).³² The average MAC_{EC} was $12.6 \pm 5.0 \text{ m}^2/\text{g}$.

PM composition other than carbonaceous material was not measured, but the PM mass can be explained by carbonaceous material (OM+EC). If all PM mass were carbonaceous material (OM+EC), then an OM/OC ratio of 1.2 produces the best fit between OM+EC and $\text{PM}_{2.5}$ ($R^2 = 0.94$). This ratio of 1.2 is at the low end of the range previously measured for urban aerosols in USA,⁵¹ and lower than the non-urban value suggested by Turpin et al.,⁵¹ of 2.1 ± 0.2 . The OM/OC ratio of 1.6 ± 0.2 for urban and 2.1 ± 0.2 for non-urban aerosol, suggested by Turpin et al.,⁵¹ would over-predict $\text{PM}_{2.5}$ for these households.

Figure 6 compares published test results with the measurements presented here. We are not aware of any other published emission data for the stove types and fuel mixtures measured in this study. However, several measurements have been published for laboratory tests and a few field tests burning individual fuel types in similar Chinese stoves, although some lack detailed stove descriptions. The published test results include coal fuel emissions (163 tests, 33 stoves)^{25,26,29-31,35-42,44} and biomass fuel emissions (56 tests, 13 stoves).^{21,25,27-31,36,43,44} Tests of wood and corn residue combustion were tabulated for the biomass reports. Conversions of the various emission metrics and statistical representations reported in these studies required for comparison are discussed in Appendix E: Emission Factors from Published Data.

Biomass fuels can be highly variable, making biomass categories imprecise and difficult to translate between studies. For example, “corn residue” may include other corn crop residues in addition to corncobs, such as corn stalks or corn chaff. “Woody fuels” in this study and other studies may refer to woody plants and crop residues, such as tree trunk, brush, twigs, or leaves, with tremendous variation in properties that affect emissions such as surface-to-volume ratio, chemical composition, and moisture content.

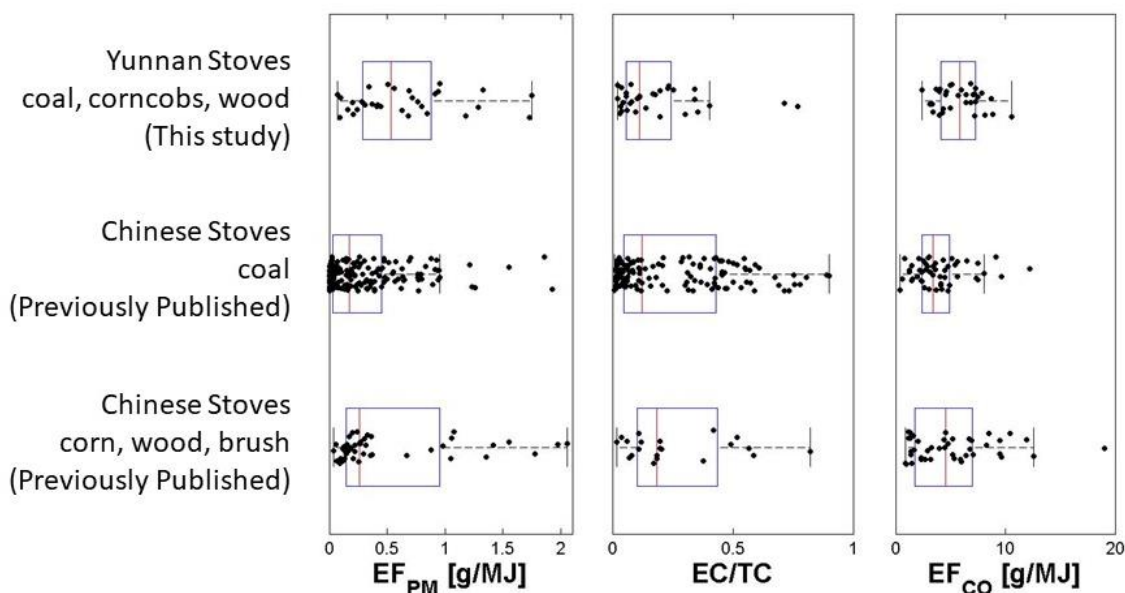


Figure 6: $EF_{e,PM}$, EC/TC, and $EF_{e,CO}$ measurements from this study and from previously published measurements of Chinese stoves. Boxplots summarize the distributions with boxes that show the 25%, 50% (median), and 75% percentile, and whiskers that show the range excluding outliers. Dots show individual test results. Four points are beyond the $EF_{e,PM}$ axis range, one for Chinese Stoves burning coal (6.2 g/MJ), and three for Chinese stoves burning biomass (3.2, 3.3, and 4.1 g/MJ).

The mean energy-based $PM_{2.5}$ emission factor measured in this study $EF_{e,PM}$ (0.63 g/MJ) was similar to the mean $EF_{e,PM}$ of published data for Chinese stoves burning biomass fuels (0.66 g/MJ), and about two times higher than published data for Chinese stoves burning coal fuels (0.33 g/MJ). The range (measured as boxplot whiskers) was similar to published results for coal fuel tests and biomass fuel test, except for one coal high emitter from Eilenberg et al.²⁵ (6.2 g/MJ) and three biomass high emitters from Du et al.²¹ (3.2, 3.3, 4.1 g/MJ). The mean EC/TC ratio (0.18) was lower than published results of coal fuel tests (0.25) and biomass fuel tests (0.26), but not significantly lower ($p > 0.1$). The range of EC/TC ratios measured was similar to published results for both coal and biomass fuel tests. The mean $EF_{e,CO}$ measured in this study (5.8 g/MJ) was similar to the mean $EF_{e,CO}$ for published biomass fuel tests (5.6 g/MJ), and higher than published data of coal fuel tests (4.0 g/MJ). $PM_{2.5}$ and CO emission factors measured in this study were significantly higher than published results of Chinese stoves burning coal fuels

($p < 0.01$), but not biomass fuels ($p > 0.79$). Reasons for differences between these field measurements and previously published data are explored below.

3.3 CAUSES OF EMISSION VARIABILITY

3.3.1 Burn Phases

Cooking events in these households exhibited two distinct “burn phases”, defined as periods of distinct combustion conditions. When fuel was added, a devolatilization phase occurred with high $\text{PM}_{2.5}$ emission, which lasted about 20 to 60 minutes. It was followed by a solid combustion phase with negligible $\text{PM}_{2.5}$ emission (Figure 7a). These burn phases are familiar to the millions of people who use residential coal stoves, have been acknowledged in other studies,^{25,37,38,31} and are confirmed in these real-time emission data. Cooking events were manually separated into the two burn phases by visual inspection of the real-time optical scattering data. The devolatilization phases were identified as periods of $\text{PM}_{2.5}$ emission that started after fuel addition and ended when the $\text{PM}_{2.5}$ emission faded. In all cases, emission eventually reduced to a low threshold near the detection limit of the optical scattering sensor. The solid combustion phases were marked as the periods of low $\text{PM}_{2.5}$ emission that followed the devolatilization phase, and continued until the cooking event ended, or fuel was added that triggered another devolatilization phase. Over all cooking events, the devolatilization phase occupied 40% of the time and accounted for 98% of $\text{PM}_{2.5}$ emissions (Figure 7b) and a similar fraction of BC emissions (not shown). 93% of $\text{PM}_{2.5}$ emissions occurred in 20% of the burning time. The highest $\text{PM}_{2.5}$ emissions usually occurred during stove ignition when the stove and fuel bed were cold. However, CO emission factors were not significantly different ($p = 0.25$) between the devolatilization phase ($\text{EF}_{\text{e,CO}} = 7.5 \pm 2.5 \text{ g/MJ}$) and solid combustion phase ($\text{EF}_{\text{e,CO}} = 5.7 \pm 3.6 \text{ g/MJ}$). In several instances, additional coal or biomass fuel was added to the stove, after the stove was hot and a glowing coal bed was established. Sometimes these fuel additions did not cause additional PM emissions, and sometimes these fuel additions resulted in smaller devolatilization periods that produced less $\text{PM}_{2.5}$ emissions than cold-start ignition.

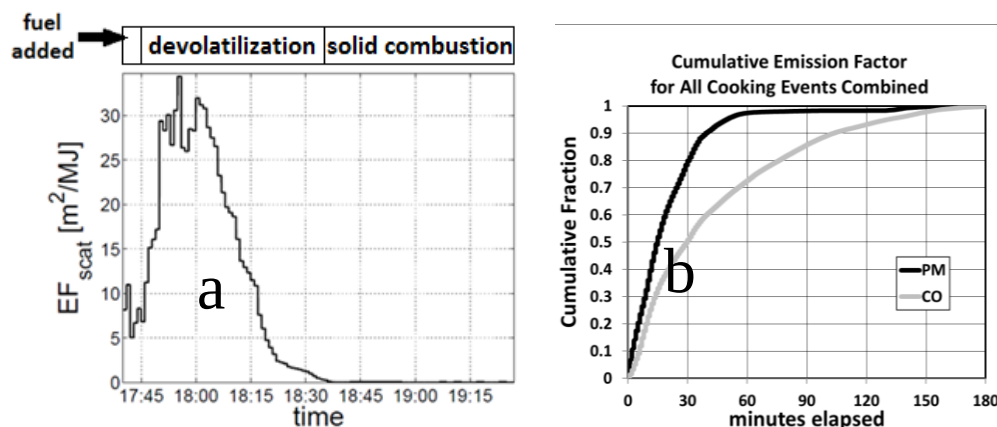


Figure 7: a) Instantaneous scattering emission factor of a typical cooking event illustrating the devolatilization and solid combustion phases. b) Cumulative optical scattering (PM) and CO emission factors vs minutes elapsed for all cooking events combined. The majority of PM is emitted at the start of the cooking events, while CO is emitted more evenly throughout the cooking events. Cumulative emissions for individual cooking events are plotted in Figure 8.

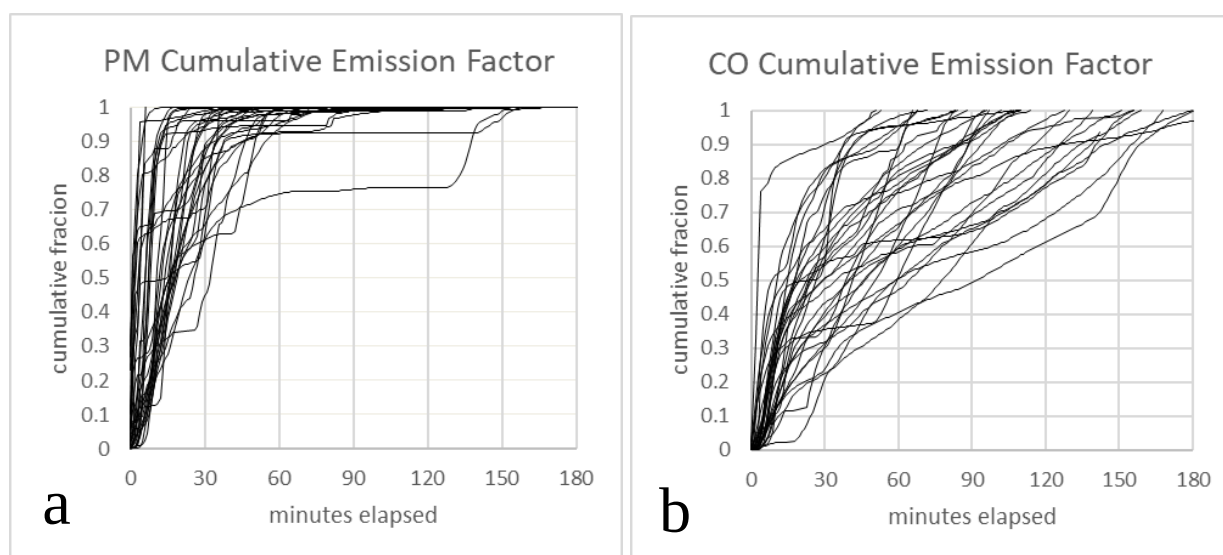


Figure 8: a) Cumulative scattering (PM) emission factors vs minutes elapsed for each cooking event with a full burn cycle. b) Cumulative CO emission factors vs minutes elapsed for each cooking event with a full burn cycle. The majority of PM is emitted at the start of the cooking events, while CO is emitted more evenly throughout the cooking events.

Previously published coal emission measurements in Figure 6 do not include the cold-start ignition emissions that account for most of the PM_{2.5} emissions during these cooking events. Most tests started the fire with a hot bed of charcoal,^{25,26,37-42} some with kerosene,³⁶ and for others the ignition method was not mentioned.^{29-31,35,40} One set of field tests was started with wood chips, but the emissions were not collected until after the coal ignited.⁴⁴ Bond et al.³⁷ and

Sun et al.²⁶ also excluded cold start emissions from reported emission factors with the reasoning that cold-start ignition is rare and unrepresentative. Our measurements show that cold-start ignition is common when stoves are used intermittently for cooking, and that realistic ignition is a major source of PM_{2.5} emissions.

The emissions reported for a cooking event are strongly dependent on when the cooking event occurred within the burn sequence. Five cooking events had incomplete burn sequences that excluded some or all of the devolatilization phase, and average PM_{2.5} emission factors for these events were lower than the lowest value of the 32 complete cooking events. After some cooking events, the coals were not extinguished and the solid combustion phase continued with little PM emission. These emissions were not measured, but they can be estimated. On average, about half the fuel loaded into the stove was consumed during the cooking event, and about half remained in the stove as hot coals. If the hot coals continued to burn until the fuel was completely consumed, then the total fuel consumption would double with no additional PM_{2.5} emissions, lowering the average PM_{2.5} emission factors by about half. On the other hand, if the remaining coals were extinguished and then included in a subsequent cooking event, the emission factors measured here would reflect net emissions. As practices varied from home to home, no general adjustment factors are possible. The inclusion of the cold-start ignition and exclusion of the burnout may explain why PM_{2.5} emission factors measured during this study are higher than previously published coal stove PM emission factors.

The variance in emission factors during the devolatilization phase accounted for 92% of the variance of the overall PM_{2.5} emission factor, 94% of the variance in overall BC emission factor, and 57% of the overall CO emission factor (Appendix H: Emission Factor Variance). Variability in the devolatilization phase dominated partly because of the large magnitude and variance, and partly because the devolatilization phase occupied a relatively large fraction of the fuel consumption (65±14%). If that phase, instead, occupied 10±5% of fuel consumption (about 20±10 minutes of a 3 hour burn sequence)—plausible operation for a heating stove—and the emission factors for each phase and their variability remained the same, then it would still explain 64% of the variance in PM_{2.5}. Uncertainty in the fraction of that phase would contribute another 29% of the variance.

3.3.2 Variability due to Fuel

Laboratory tests of Chinese coal stoves suggest that gaseous and particle emissions are dependent on coal composition. In general, coals containing more tar and volatile matter produce larger PM emission factors in cooking stoves.^{26,35,37,41,42} Coal honeycomb briquettes have lower PM emission factors and EC fraction compared to raw coal chunks.^{30,31,36-38,41} Indoor air pollution studies in this region indicate that different types of coal produce different amounts of PM.^{5,6} Emissions are also affected by fuel type for biomass-burning Chinese stoves.^{21,26-28,44}

To investigate whether variability in emissions was caused by biomass type, the 32 cooking events with coal and biomass fuel mixtures were classified into three groups based on the type of biomass mixed with the coal: (1) coal and woody fuel (n=7), (2) coal and corncob fuel (n=13), and (3) coal, corncob, and woody fuel (n=12). Emissions between the three groups were not significantly different using an independent t-test with one exception: mixtures of coal, corncobs, and woody fuels had lower EC fraction ($p=0.01$) and higher SSA ($p=.03$) than the other two fuel mixtures. The emissions of lignite (n=3) and honeycomb (n=1) coals lie within the range of emissions for bituminous coal for all emission metrics, although the use of coal other than bituminous was small, following practice in the region. Weak correlations were found between emission factors and the fraction of wood, corncobs, and total biomass in the fuel mixture ($r<0.37$).

It is likely that emissions are affected by fuel type and fuel characteristics, as suggested in previous studies.^{5,6,18,21,26-28, 31,35-38,41,42,44} However, those studies evaluated individual fuel types, rather than the mixtures of multiple fuel types observed in this study. It is not practical to apportion emissions to individual fuels in a mixture, because emissions may differ from the sum of emissions from isolated fuel types. In reality, every burn sequence had a unique fuel mixture that included both coal and biomass. The woody fuels included several unknown tree species and many plant forms. For example, pine included milled lumber, split branches, bark, small twigs, needles, and cones. Each form had a unique set of material properties that could affect combustion. The measured bulk composition of coal was similar to bituminous coal, but the heterogeneous nature of coal could create further variability across cooking events. Furthermore, any major grouping does not account for the fuel moisture content, shapes and sizes of fuel pieces, fraction of each fuel in the mixture, or the relative timing of fuel additions.

3.3.3 Variability due to Stove Type

Chimney stoves and portable stoves had similar magnitude and variability of emissions. $EF_{e,PM}$ for chimney and portable stoves was 0.63 ± 0.42 and 0.64 ± 0.53 g/MJ, respectively. $EF_{e,CO}$ for chimney and portable stoves was 5.69 ± 1.94 and 6.13 ± 2.07 g/MJ, respectively. Using an independent t-test, the only significant difference in emissions between the two types is that chimney stoves produced darker $PM_{2.5}$ than portable stoves ($SSA = 0.64 \pm 0.17$ vs. 0.78 ± 0.12 , $p = 0.01$) with a higher EC fraction ($EC/TC = 0.24 \pm 0.20$ vs. 0.07 ± 0.05 , $p = 0.001$).

Within each stove type, there was a large variation in design aspects that could affect emissions, such as geometry, dimensions, and materials. No two stoves were identical. In order to attribute emission variability to stove type, at least one of the stove types should have more similar design features than the stove types measured in this study, or a much larger sample size would be required. However, as with fuels, the stove designs in this population reflect real variability.

3.3.4 Inherent Variability

Stove operating practices of household residents determine what fuels are added, when they are added, and how the fuels are layered and mixed in the stove, which in turn affects emissions. In each of 10 households, two cooking events were measured on the same stove, and similar fuels were burned in each of the two events. The relative mean difference between devolatilization phase $PM_{2.5}$ emissions for these duplicate cooking events was 51%. Even when variables are isolated as much as possible by considering the same stove, similar fuels, and only the devolatilization phase, there remains an inherent variability due to the dynamic nature of combustion combined with variability in fuel characteristics and operating practices. Furthermore, the individual variables evaluated in the preceding analysis are not separable, but rather correlated in ways that are difficult to observe. This finding suggests a revised approach to representing emissions that is discussed next.

3.4 IMPLICATIONS FOR REPRESENTATIVE EMISSION INVENTORIES

Emissions have often been quantified by combining information about mass or energy of fuels used, devices in which they are used, and emission factors for fuel-device pairs. It is natural to suggest that measurements of fuel-device combinations would improve atmospheric emission

inventories or estimates of exposure, yet the observations reported here suggest that other factors dominate the emission variability.

(1) Laboratory testing, although useful for stove evaluation and for improving combustion technologies, may not include major factors that affect emissions, such as fuel mixtures or operating procedures. In such a case, laboratory findings cannot be extrapolated to infer in-use performance and emissions.

(2) Instantaneous emissions vary greatly with time and in response to household services. Therefore, field measurements must capture entire burn sequences under realistic operating conditions in order to provide accurate emission factors. The measurements reported here were for intermittent burn sequences typical of cooking, yet in the winter, the same stoves are also operated continuously to provide heat, and the contribution of the ignition phase would differ.

(3) Factors other than fuel-device combinations, such as variability in fuel and stove quality and operating procedure, affect stove performance and emissions. This variability means that the sample size required to distinguish the performance of specific fuel-device combinations or the influence of specific fuel characteristics may be impractically large.

(4) Fuel mixtures are common in real settings, and their emission factors may not be the same as the weighted emission factors from multiple separate fuels. Mixing of fuels cannot be separated from the household services, because fuels may be selected for different functions required to perform a task, such as ignition, low-power cooking, or all-night heating.

The observations reported here, in combination with other field measurements,³⁴ indicate that testing many fuel-device combinations is unlikely to reduce uncertainty in estimated emission rates. We offer three suggestions to improve emission inventories:

(1) Total fuel consumption should include secondary fuels required for ignition and supplementation, which are currently neither reported in fuel use statistics nor included in emission inventories. Emissions of these fuels are inseparable from those of the primary fuel, so that only a multiplier is needed to estimate their contribution. In this sample, the multiplier to obtain total fuel consumption from coal consumption (calculated as total fuel over coal fuel) would be 1.4 on a fuel energy basis, and 1.5 on a fuel mass basis. A similar multiplier was used by Chen et al.³⁷ to calculate household fuel consumption of multiple fuels as coal equivalent.

(2) Operator behavior and the demand for specific household services produces differences in operation that often dominate emission characteristics, causing large variability for

the same apparent fuel-stove combination. Therefore, instead of attributing emissions to a specific stove type, we suggest that household services should be identified along with the broad fuel-device category that provided those services, e.g. “traditional coal stoves used for cooking.” Separate devices should be distinguished only when differences in their in-field performance are statistically significant. In this study, the standard error of the average emission factor was determined with 15% with about 30 tests, which is sufficient for atmospheric modeling.

(3) The devolatilization phase that includes ignition contributes the largest proportion of PM emissions and their variability for these fuel mixtures. This phase should be explicitly measured and represented in inventories and policies to reduce emissions. The devolatilization phase may have a differing contribution when the same device is used for different uses, such as intermittent cooking and continuous heating.

The formal packaging of a device, its fuel, its ignition, and its operation as a system that meets a set of household needs may encourage more realism in laboratory tests and emission projection scenarios. Although the changes we propose account for some causes of variability, they do not assist in confirming improvements when performance is highly variable.⁵² Reducing the performance variability itself, rather than seeking to distinguish improvements in a highly variable population, could be a worthy focus for stove programs. Fuel-device combinations with low variability are most likely to achieve significant improvements in environment and human health.

CHAPTER 4: EMISSION EQUIPMENT DEVELOPMENT

This chapter summarizes our development of portable equipment to measure solid fuel combustion emissions. The equipment is specialized to characterize emissions for estimating climate impacts. It measures gases (CO and CO₂), particle light scattering and absorption, and collects filter samples for gravimetric and compositional analysis.

4.1 PREVIOUS EQUIPMENT

The first ARACHNE (Ambulatory Real-time Analyzer of Climate and Health-related Noxious Emissions) was constructed in 2005 by Chris Roden at University of Illinois (Figure 4a). It was a collection of laboratory equipment mounted to a cart. From 2005 - 2012 the instrument was developed by Justin Ellis and Cheryl Weyant to be smaller, lighter, and more portable. At least three iterations were constructed, which we refer to as ARACHNE 2, ARACHNE 3, and ARACHNE 4 (Figure 9b, 9c, and 9d, respectively).



Figure 9: Previous ARACHNE instruments developed at the University of Illinois. a) ARACHNE.³² b) ARACHNE 2. c) ARACHNE 3. d) ARACHNE 4. Photos b), c), and d) are from Cheryl Weyant.

4.2 ARACHNE 5

The ARACHNE 5 (Figure 2a) was constructed in 2013 for measuring emissions of Chinese coal stoves presented in Chapter 3: Field Emission Measurement Results.

4.2.1 Layout

The ARACHNE 5 sensor box shell is the same Pelican case used in the ARACHNE 4, but the layout of components was redesigned to improve accessibility to all components for easier troubleshooting, repair, and replacement of parts. The PM and gas sensors are mounted to circuit boards that plug into sensor cases from Aprovecho's PEMS. The cases have a sealed pin connector for electrical connections and a removable lid that allow access to the sensors, while maintaining a reliable seal. The sensors can be replaced quickly in the field by swapping out a sensor board.

4.2.2 Flow Control

Metering valves were added to control the flow through each branch of the sample train, including metering valves for each filter flow. Better flow control helped reduce uncertainty of the filter-based concentration measurements. Rotameters were also added to each branch of the sample train to provide a visual indicator of the approximate flow rates, which helps as a quality assurance check, and makes it easier to set the flows.

4.2.3 Leaks

The ARACHNE 5 sample train was designed to be easier to leak test and less prone to leaks. Flow metering valves are connected in the sample train directly to the pump, effectively minimizing the components and number of connections that are exposed to full pump vacuum. The sample train sensor components are located upstream of the metering valves, where the sample train is near ambient pressure.

4.2.4 Power supply

The ARACHNE 5 power supply incorporates a buck boost switching regulator to ensure that all pumps and linear regulators are supplied with a constant 12 volts for a battery input voltage range of 8 – 30 volts. The buck boost allows the instrument to be powered by a variety of

battery packs and DC power supplies, while keeping the pumps and sensors from drifting or failing from power fluctuations.

4.2.5 Batteries

Li-ion batteries were used to power the ARACHNE 5 because they had the best balance of energy density, longevity, and cost. During the field campaign in China, two large (300 Wh) Li-ion batteries were confiscated at an airport. In subsequent field campaigns, smaller Li-ion batteries (100 Wh) have been transported and used successfully.

4.2.6 CO sensor

Small, portable, and low-cost electrochemical CO sensors are available on the market from demands in the fire and safety industries. A CO cell with hydrogen compensation (Alphasense CO-AX) is used in the ARACHNE5 to avoid cross-sensitives when measuring coal emissions, which could have higher hydrogen emissions than biomass. The CO calibration procedure was updated to specify calibration gases that contain O₂, which is a required species in the electrochemical cell reaction. Previous calibration gases in the lab had N₂ as a balance gas, which would cause CO sensors to drift during calibration. The balance gas was changed from N₂ to zero air (80% N₂, 20% O₂).

4.2.7 CO₂ sensor

Several models of small, portable, low cost NDIR CO₂ sensors are available on the market due to demands in the building ventilation industry. The Telaire sensor, used in previous versions of the ARACHNE and PEMS sensor boxes, had a couple issues. The sensor had an auto-zero feature that could not be disabled. The auto-zero was triggered only in rare instances, but when it was triggered, it changed the sensor calibration, which was unfortunate during field campaigns without access to calibration gas. The sensor decided when to auto-zero based on expected building air patterns, which are different than stove emission patterns.

Another issue with the Telaire sensor was that it caused interference in other sensor signals. The infrared light source was a glowing filament that switched off and on at approximately 1 hz. It was essentially an inductor that created a voltage spike in the ground plane every cycle. The frequency of the voltage spike was slightly different than the sample

frequency of the DAQ. When the voltage spike aligned with the DAQ sample, other sensor signals would shift noticeably. The interference was minimized by using large capacitors, large conductors, and larger capacity voltage regulator, but the interference was never eliminated.

In search of a better sensor, four small, portable NDIR CO₂ sensor models (Telaire T6615-10KF, Cozir Ambient 10k, SprintIR Ambient 10k, and SprintIR Wide Range 5%) were tested for response time, detection limit, and accuracy against a Horiba AI-220 laboratory gas analyzer. The SprintIR Wide Range was chosen for the ARACHNE 5 because it had the best accuracy and fastest response time. Its LED light source had low power consumption (35 mW) that did not cause an inductive voltage spike like the Telaire glowing filament, and therefore required a much smaller and simpler power supply. The sensor also had a good detection limit and the widest range (0 – 50,000 ppm) appropriate for cookstoves and kilns.

Subsequent field campaigns showed the SprintIR sensor has a long-term (monthly) drift problem. The drift could be managed by weekly zero-point calibrations during a measurement campaign, or by zero and span calibrations before and after a campaign to determine calibration corrections for the CO₂ data.

Subsequent testing showed that the CO₂ response time of the sensor box is determined by the sample train residence time, and a sensor with a T_{90} response time of 1 minute or less is adequate. The sensor was abandoned in future equipment for a newer model of sensor with internal reference to correct for long-term drift (Vaisala GMM 111).

4.2.8 Optical PM scattering sensor

The ARACHNE 5 has two PEMS PM light scattering sensors in series, one 635 nm and one 880 nm. The 880 nm sensor was added to match the 880 nm light absorption measurement of the Microaeth in order to calculate single scattering albedo at 880 nm without using an angstrom exponent. Measuring particle light scattering at two wavelengths allows determination of the scattering angstrom exponent, which could provide information on the particle sizes and composition. The 880 nm laser is invisible to the human eye, which made it difficult to align and focus during construction. Furthermore, it was a Class IIIa laser that posed a much higher risk of damage the human eye, than the red laser. It was later determined that the added value of the 880 nm scattering data was not worth the difficulties and risks of working with the infrared laser, and it was not used again in future equipment.

The scattering sensors were characterized against the TSI nephelometer (700, 550, and 450 nm), Radiance Research nephelometer (530 nm), and the DMT Photoacoustic (870 nm). Smoke was generated in a combustor and drawn into a barrel. All instruments sampled from the barrel simultaneously. For a given batch of smoke, the ARACHNE5 PM sensor outputs were linear with the reference instruments ($r^2 > 0.9$). However, different batches of smoke resulted in different regression slopes (Figure 10). The reference instruments also had different regression slopes with respect to each other between batches of smoke. Over all batches of smoke, the ARACHNE5 sensors agreed with the reference instruments within 30% relative error.

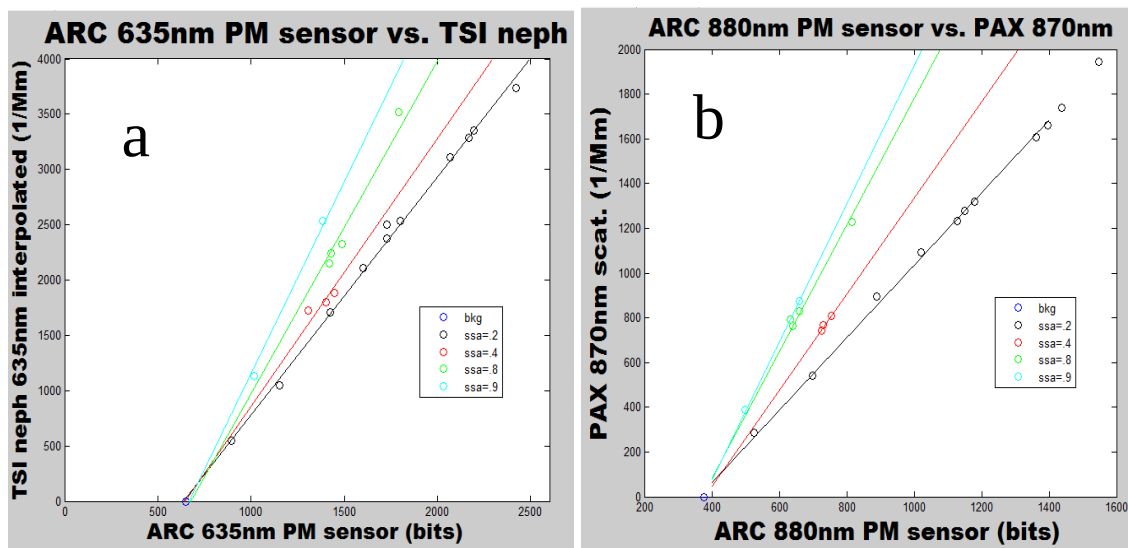


Figure 10: Inter-comparison between ARACHNE 5 scattering sensors and reference instruments. Four regression lines are shown, one for each batch of smoke, with single scattering albedo (SSA) of 0.2, 0.4, 0.8, and 0.9, respectively. For each regression r^2 is greater than 0.9. **a)** TSI nephelometer vs. 635 nm scattering sensor. **b)** DMT Pax vs 880 nm scattering sensor.

4.2.9 Optical PM absorption sensor

Particle light absorption is measured with a MicroAeth AE50. The MicroAeth was the smallest, most portable particle absorption sensor on the market. It is appropriate for measuring absorption coefficients below $10,000 \text{ Mm}^{-1}$, but the filter spot is too small to measure higher concentrations. The filter spot is quickly overloaded and the filter ticket must be replaced frequently, leaving data gaps and low-quality data. The cost of filter tickets can add up. The MicroAeth flow was reduced from 50 to 20 ccm to increase the time between filter changes, but the flow was not steady and filter changes were still too frequent.

4.2.10 Data Acquisition

The ARACHNE 5 has a Waltech data acquisition system (DAQ) which has lower signal noise than the NI-DAQ used in previous versions of the ARACHNE. High frequency signal noise is picked up by the successive approximation analog-to-digital converter (ADC) of the NI-DAQ, but this noise is filtered out by the delta-sigma ADC of the Waltech DAQ. The NI-DAQ requires proprietary LabVIEW software that requires an expensive license, gigabytes of drivers, and has a clunky GUI that is challenging to configure and change calibration constants. The Waltech DAQ data stream output is UART serial data, which is more reliable than the USB data stream output of the NI-DAQ. The UART data signal can be converted to RS-232, USB, wireless, or ethernet using many types of hardware adapters, and it can be read by many types of serial port software.

An attempt was made to incorporate a Raspberry Pi computer with a small monitor into the ARACHNE 5 in order to log data and view the real-time data on-board. However, the project was abandoned and never completed. As a fallback plan, the data was logged to an external computer using Teraterm serial port software and plotted in real-time using LiveGraph.

4.3. FUMITRON

Three Fumitrons were constructed in 2014 for measuring emissions of cooking and heating stoves in Nepal, China, and Alaska.

4.3.1 Layout

The Fumitron sensor box shell is the same Pelican case used in the ARACHNE5, but the layout of components was redesigned so the filter holders fit inside the case when the lid is closed. This allows the sensor box to be operated with the lid closed, which protects the sensor box from sun, rain, dust, and tampering by curious children. The filter holders can be installed before arriving at the measurement location, which can reduce setup time in households (installing filters, checking for leaks, setting flows), and reduce the chance of filter contamination.

The Fumitron layout further improves usability and component accessibility for repairs. The layout provides space inside the case for additional batteries for 24 hours of runtime. Sample train flow check points (unions to connect the bubble meter inline) were moved to more

accessible locations. All sensors were moved to underneath the aluminum panel for protection for the sun. The gas sensors in the ARACHNE5 are located on the top side of the panel, and they drift in direct sunlight.

4.3.2 Flow Sensors

The Fumitron has four digital flow sensors, one for each branch of the sample train, to allow real-time measurement of the total system flow and dilution ratio. Measuring the real-time dilution ratio provides quality assurance, and allows corrections to be made to measured concentrations if the dilution ratio is changed during a sampling event.

Data Acquisition: Open-source firmware was written in C+ for the Waltech DAQ to make it configurable for the Fumitron sensor inputs. Open-source computer software was also written in Python to provide a simple, platform-independent interface for logging and plotting the output data stream.

A wireless transmitter was added that delivers the data stream to the computer without the use of a cord. An SD card logger was also added to log the data on-board the Fumitron. The addition of the wireless transmitter and SD card logger allow cookstove measurements to be less intrusive. After initial equipment setup, the equipment can be left unattended sampling in a kitchen for long periods of time, and all the data is reliably logged to the SD card. The data stream can be monitored anytime from outside the kitchen on a computer using the wireless transmitter. There is no need for the technician to be in the kitchen during cooking. By eliminating the computer cord connection, the equipment is more portable and easier to position in tight spaces. For brick kiln measurements, the data stream can be monitored from the ground while the equipment is sampling up on the platform.

4.3.3 Probe

A mounting bracket was made for the probe to simply and quickly attach it to any standard camera tripod. A full line of probe ornaments hit the market soon after, which quickly became popular in the international cookstove field measurement scene.

4.3.4 Documentation

The Fumitron is accompanied by thorough documentation. The Appendix I: Book of Fumitron includes operating instructions, diagrams, theory of operation, maintenance and repair, and parts lists. A fabrication guide was also written.

REFERENCES

- (1) Gakidou, E.; Afshin, A.; Abajobir, A.A.; Abate, K.H.; Abbafati, C.; Abbas, K.M.; Abd-Allah, F.; Abdulle, A.M.; Abera, S.M.; Aboyans, V. et al. Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 1990-2016: a systematic analysis for the Global Burden of Disease Study 2016. *Lancet*. **2017**, 390, 1345–1422; DOI:[10.1016/S0140-6736\(17\)32366-8](https://doi.org/10.1016/S0140-6736(17)32366-8).
- (2) Zhang, J.; Smith, K. R. Household Air Pollution from Coal and Biomass Fuels in China: Measurements, Health Impacts, and Interventions. *Environ. Health Perspect.* **2007**, 115 (6), 848–855; DOI: 10.1289/ehp.9479.
- (3) Edwards, R.D.; Liu, Y.; He, G.; Yin, Z.; Sinton, J.; Peabody, J.; Smith, K.R. Household CO and PM measured as part of a review of China's National Improved Stove Program. *Indoor Air*. **2007**, 17(3), 189-203.
- (4) Adria, O.; Bethge, J.P. Chinese National Improved Stove Program (CNISP). http://www.bigeen.net/en/policy/guide/appliances/policy_examples/12/.
- (5) He, X.; Chen, W.; Liu, Z.; Chapman, R. An Epidemiological Study of Lung Cancer in Xuan Wei County, China: Current Progress. Case-Control Study on Lung Cancer and Cooking Fuel. *Environ. Health Perspect.* **1991**, 94, 9-13.
- (6) Seow, W. J.; Hu, W.; Vermeulen, R.; Hosgood, H. D.; Downward, G. S.; Chapman, R. S.; ... Lan, Q. Household air pollution and lung cancer in China: a review of studies in Xuanwei. *Chin. J. Cancer*, **2014**, 33 (10), 471–475; DOI: 10.5732/cjc.014.10132.
- (7) Bond, T.C.; Streets, D.G.; Yarber, K.F.; Nelson, S.M.; Woo, J.H.; Klimont, Z.; A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res.* **2004**, 109, D14203; DOI:10.1029/2003JD003697.
- (8) Bond, T. C.; Doherty, S. J.; Fahey, D. W.; Forster, P. M.; Berntsen, T.; DeAngelo, B. J.; Flanner, M. G.; Ghan, S.; Karcher, B.; Koch, D.; et al. Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res.: Atmos.* **2013**, 118, 5380-5552; DOI: 10.1002/jgrd.50171.
- (9) Feng, Y.; Ramanathan, V.; Kotamarthi, V.R. Brown carbon: a significant atmospheric absorber of solar radiation? *Atmos. Chem. Phys.* **2013**, 13 (17), 8607-8621.
- (10) Lu, B.; Kong, S.; Han, B.; Wang, X.; Bai, Z. Inventory of atmospheric pollutants discharged from biomass burning in China continent in 2007. *China Environ. Sci.* **2011**, 31 (2), 186–194.
- (11) Lu, Z.; Zhang, Q.; Streets D. G. Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996-2010. *Atmos. Chem. Phys.* **2011**, 11, 9839-9864; DOI: 10.5194/acp-11-9839-2011.

- (12) Kopacz, M.; Mauzerall, D.; Wang, J.; Leibensperger, E.; Henze, D.; and Singh, K. Origin and radiative forcing of black carbon transported to the Himalayas and Tibetan Plateau. *Atmos. Chem. Phys.* **2011**, 11 (6), 2837-2852; DOI: 10.5194/acp-11-2837-2011.
- (13) Gao, J.; Yao, T.; Masson-Delmotte, V.; Steen-Larsen, H.C.; Wang, W. Collapsing glaciers threaten Asia's water supplies. *Nature*. **2019**, 565, 19-21.
- (14) Lutz, A. F.; Immerzeel, W. W.; Shrestha, A. B.; Bierkens, M. F. P. Consistent increase in High Asia's runoff due to increasing glacier melt and precipitation. *Nature Climate Change*. **2014**, 4, 587-592. DOI: 10.1038/nclimate2237
- (15) Roden, C. A.; Bond, T. C.; Conway, S.; Osorto Pinel, A. B.; 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; DOI: 10.1016/j.atmosenv.2008.05.041.
- (16) Johnson, M.; Edwards, R.; Frenk, C. A.; Masera, O. In-field greenhouse gas emissions from cookstoves in rural Mexican households. *Atmos. Environ.* **2008**, 42 (6), 1206-1222; DOI: 10.1016/j.atmosenv.2007.10.034.
- (17) Johnson, M.; Edwards, R.; Berrueta, V.; Masera, O. New Approaches to Performance Testing of Improved Cookstoves. *Environ. Sci. Technol.* **2010**, 44, 368-374; DOI: 10.1021/es9013294.
- (18) 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.
- (19) Coffey, E.; Muvandimwe, D.; Hagar, Y.; Wiedinmyer, C.; Kanyomse, E.; Piedrahita, R.; Dickinson, K.; Oduro, A.; Hannigan, M. 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; DOI: 10.1021/acs.est.7b02436.
- (20) Lombardi, F.; Riva, F.; Bonamini, G.; Barbieri, J.; Colombo, E. Laboratory protocols for testing of Improved Cooking Stoves (ICSs): A review of state-of-the-art and further developments. *Biomass Bioenergy*. **2017**, 98, 321-335.
- (21) Du, W.; Zhu, X.; Chen, Y.; Liu, W.; Wang, W.; Shen, G.; Tao, S.; Jetter, J.J. Field-based emission measurements of biomass burning in typical Chinese built-in-place stoves. *Environ. Pollut.* **2018**, 242, 1587-1597.
- (22) Deng, M.; Zhang, S.; Shan, M.; Li, J.; Baumgartner, J.; Carter, E.; Yang, X. The impact of cookstove operation on PM_{2.5} and CO emissions: A comparison of laboratory and field measurements. *Environ. Pollut.* **2018**, 243, 1087-1095; DOI: 10.1016/j.envpol.2018.09.064.

- (23) Garland, C.; Delapena, S.; Prasad, R.; L'Orange, C.; Alexander, D.; Johnson, M. Black carbon cookstove emissions: a field assessment of 19 stove/fuel combinations. *Atmos. Environ.* **2017**, 169, 140-149.
- (24) Medina, P.; Berrueta, V.; Martínez, M.; Ruiz, V.; Ruiz-Mercado, I.; Masera, O.R. Closing the gap between lab and field cookstove tests: Benefits of multi-pot and sequencing cooking tasks through controlled burning cycles. *Energy Sustainable Dev.* **2017**, 41, 106-111.
- (25) Eilenberg, S.R.; Bilsback, K.; Johnson, M.; Kodros, K.; Lipsky, E.; Naluwagga, A.; Fedak, K.; Benka-Coker, M.; Reynolds, B.; Peel, P.; Clark, M.; Shan, M.; Sambandam, S.; L'Orange, C.; Pierce, J.; Subramanian, R.; Volckens, J.; Robinson, A. Field measurements of solid-fuel cookstove emissions from uncontrolled cooking in China, Honduras, Uganda, and India. *Atmos. Environ.* **2018**, 190, 116-125; DOI: 10.1016/j.atmosenv.2018.06.041.
- (26) Sun, J.; Zhi, G.; Jin, W.; Chen, Y.; Shen, G.; Tian, C.; Zhang, Y.; Zong, Z.; Cheng, M.; Zhang, X.; Zhang, Y.; Liu, C.; Lu, J.; Wang, H.; Xiang, J.; Tong, L.; Zhang, X. Emission factors of organic carbon and elemental carbon for residential coal and biomass fuels in China- A new database for 39 fuel-stove combinations. *Atmos. Environ.* **2018**, 190, 241-248; DOI: 10.1016/j.atmosenv.2018.07.032.
- (27) Li, X.; Duan, L.; Wang, S.; Duan, J.; Guo, X.; Yi, H.; Hu, J.; Li, C.; Hao, J. Emission Characteristics of Particulate Matter from Rural Household Biofuel Combustion in China. *Energy Fuels.* **2007**, 21 (2), 845-851; DOI: 10.1021/ef060150g.
- (28) Li, X.; Wang, S.; Duan, L.; Hao, J.; Nie, Y. Carbonaceous Aerosol Emissions from Household Biofuel Combustion in China. *Environ. Sci. Technol.* **2009**, 43 (15), 6076-6081; DOI: 10.1021/es803330j.
- (29) Shen, G.; Tao, S.; Wei, S.; Chen, Y.; Zhang, Y.; Shen, H.; Huang, Y.; Zhu, D.; Yuan, C.; Wang, H.; Wang, Y.; Pei, L.; Liao, Y.; Duan, Y.; Wang, B.; Wang, R.; Lv, Y.; Li, W.; Wang, X.; Zheng, X. Field Measurement of Emission Factors of PM, EC, OC, Parent, Nitro-, and Oxy-Polycyclic Aromatic Hydrocarbons for Residential Briquette, Coal Cake, and Wood in Rural Shanxi, China. *Environ. Sci. Technol.* **2013**, 47 (6), 2998-3005; DOI: 10.1021/es304599g.
- (30) Shen, G.; Chen, Y.; Xue, C.; Lin, N.; Huang, Y.; Shen, H.; Wang, Y.; Li, T.; Zhang, Y.; Su, S.; Huangfu, Y.; Zhang, W.; Chen, X.; Liu, G.; Liu, W.; Wang, X.; Wong, M.H.; Tao, S. Pollutant Emissions from Improved Coal- and Wood-Fuelled Cookstoves in Rural Households. *Environ. Sci. Technol.* **2015**, 49 (11), 6590-6598; DOI: 10.1021/es506343z.
- (31) Chen, Y.; Shen, G.; Liu, W.; Du, W.; Su, S.; Duan, Y.; Lin, N.; Zhuo, S.; Wang, X.; Xing, B.; Tao, S. Field measurement and estimate of gaseous and particle pollutant emissions from cooking and space heating processes in rural households, northern China. *Atmos. Environ.* **2016**, 125, 265-271; DOI: 10.1016/j.atmosenv.2015.11.032.

- (33) Weyant, C.; Athalye, V.; Ragavan, S.; Rajarathnam, U.; Lalchandani, D.; Maithel, S.; Baum, E.; Bond, T.C. Emissions from South Asian brick production. *Environ. Sci. Technol.* **2014**, 48 (11), 6477–6483, DOI: 10.1021/es500186g.
- (34) Weyant, C.L.; Chen, P.; Vaidya, A.; Chaoliu, L.; Qianggong, Z.; Thompson, R.; Chen, Y.; Edwards, R.; Shichang, K.; Shrestha, G.R.; Bond, T.C. Emission measurements from traditional biomass cookstoves in South Asia and Tibet. Submitted to *Environ. Sci. Technol.* **2019**
- (35) Zhang, Y.; Schauer, J.; Zhang, Y.; Zeng, L.; Wei, Y.; Liu, Y.; Shao, M. Characteristics of Particulate Carbon Emissions from Real-World Chinese Coal Combustion. *Environ. Sci. Technol.* **2008**, 42 (14), 5068-5073. DOI: 10.1021/es7022576.
- (36) Zhang, J.; Smith, K.R.; Ma, Y.; Ye, S.; Jiang, F.; Qi, W.; Liu, P.; Khalil, M.A.K.; Rasmussen, R.A.; Thorneloe S.A.; Greenhouse gases and other airborne pollutants from household stoves in China: a database for emission factors. *Atmos. Environ.* **2000**, 34, 4537-4549; DOI: 10.1016/S1352-2310(99)00450-1.
- (37) Bond, T.C.; Covert, D.S.; Kramlich, J.C.; Larson, T.V., Charlson, R.J. Primary particle emissions from residential coal burning: Optical properties and size distributions. *J. Geophys. Res.* **2002**, 107 (D21), 8347; DOI: 10.1029/2001JD000571.
- (38) Zhi, G.; Chen, Y.; Feng, Y.; Xiong, S.; Li, J.; Zhang, G.; Sheng, G.; Fu, J. Emission Characteristics of Carbonaceous Particles from Various Residential Coal-Stoves in China. *Environ. Sci. Technol.* **2008**, 42 (9), 3310-3315; DOI: 10.1021/es702247q.
- (39) Zhi, G.; Peng, C.; Chen, Y.; Liu, D.; Sheng, G.; Fu, J. Deployment of Coal Briquettes and Improved Stoves: Possibly an Option for both Environment and Climate. *Environ. Sci. Technol.* **2009**, 43 (15), 5586-5591; DOI: 10.1021/es802955d.
- (40) Ge, S.; Xu, X.; Chow, J.; Watson, J.; Sheng, Q.; Liu, W.; Bai, Z.; Zhu, T.; Zhang, J. Emissions of Air Pollutants from Household Stoves: Honeycomb Coal versus Coal Cake. *Environ. Sci. Technol.* **2004**, 38 (17), 4612-4618; DOI: 10.1021/es049942k.
- (41) Chen, Y.; Sheng, G.; Bi, X.; Feng, Y.; Mai, B.; Fu, J. Emission Factors for Carbonaceous Particles and Polycyclic Aromatic Hydrocarbons from Residential Coal Combustion in China. *Environ. Sci. Technol.* **2005**, 39 (6), 1861-1867; DOI: 10.1021/es0493650.
- (42) Chen, Y.; Zhi, G.; Feng, Y.; Fu, J.; Feng, J.; Sheng, G.; Simoneit, B. R. T. Measurements of emission factors for primary carbonaceous particles from residential raw-coal combustion in China. *Geophys. Res. Lett.* **2006**, 33 (20), L20815; DOI: 10.1029/2006GL026966.
- (43) Jetter, J.; Zhao, Y.; Smith, K.; Khan, B.; Yelverton, T.; DeCarlo, P.; and Hays, M. 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; DOI: 10.1021/es301693f.

- (44) Shen, G.; Yang, Y.; Wang, W.; Tao, S.; Zhu, C.; Min, Y.; Xue, M.; Ding, J.; Wang, B.; Wang, R.; Shen, H.; Li, W.; Wang, X.; Russell, A. Emission Factors of Particulate Matter and Elemental Carbon for Crop Residues and Coals Burned in Typical Household Stoves in China. *Environ. Sci. Technol.* **2010**, *44* (18), 7157-7162; DOI: 10.1021/es101313y.
- (45) Smith, K.R.; Khalil, M.A.K.; Rasmussen, R.A.; Thorneloe, S.A.; Manegdeg, F.; Apte, M. Greenhouse gases from biomass and fossil fuel stoves in developing countries: A Manila pilot study. *Chemosphere*. **1993**, *26*, 479-505.
- (46) 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; DOI: 10.1021/es3003348.
- (47) Bond, T.C.; Anderson, T.L.; Campbell, D. Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols. *Aerosol Science and Technology*. **2010**, *30* (6), 582-600; DOI: 10.1080/027868299304435.
- (48) Kirchstetter, T.W.; Corrigan, C.E.; Novakov, T. Laboratory and field investigation of the adsorption of gaseous organic compounds onto quartz filters. *Atmos. Environ.* **2001**, *35*, 1663-1671.
- (49) International Standards Organization ISO/TR 19867-3:2018 Clean cookstoves and clean cooking solutions -- Harmonized laboratory test protocols -- Part 3: Voluntary performance targets for cookstoves based on laboratory testing
- (50) Haywood, J.; Shine, K. The effect of anthropogenic sulfate and soot aerosol on the clear sky planetary radiation budget. *Geophys. Res. Lett.* **1995**, *22*, 603–606.
- (51) Turpin, B. J.; Lim, H. Species Contributions to PM_{2.5} Mass Concentrations: Revisiting Common Assumptions for Estimating Organic Mass. *Aerosol Sci. Technol.* **2001**, *35* (1), 602-610; DOI: [10.1080/02786820119445](https://doi.org/10.1080/02786820119445).
- (52) L'Orange, C.; Leith, D.; Volckens, J; DeFoort, M. A Quantitative Model of Cookstove Variability and Field Performance: Implications for Sample Size. *Biomass Bioenergy*. **2015**, *72*, 233-241.

APPENDIX A: STUDY AREA MAP

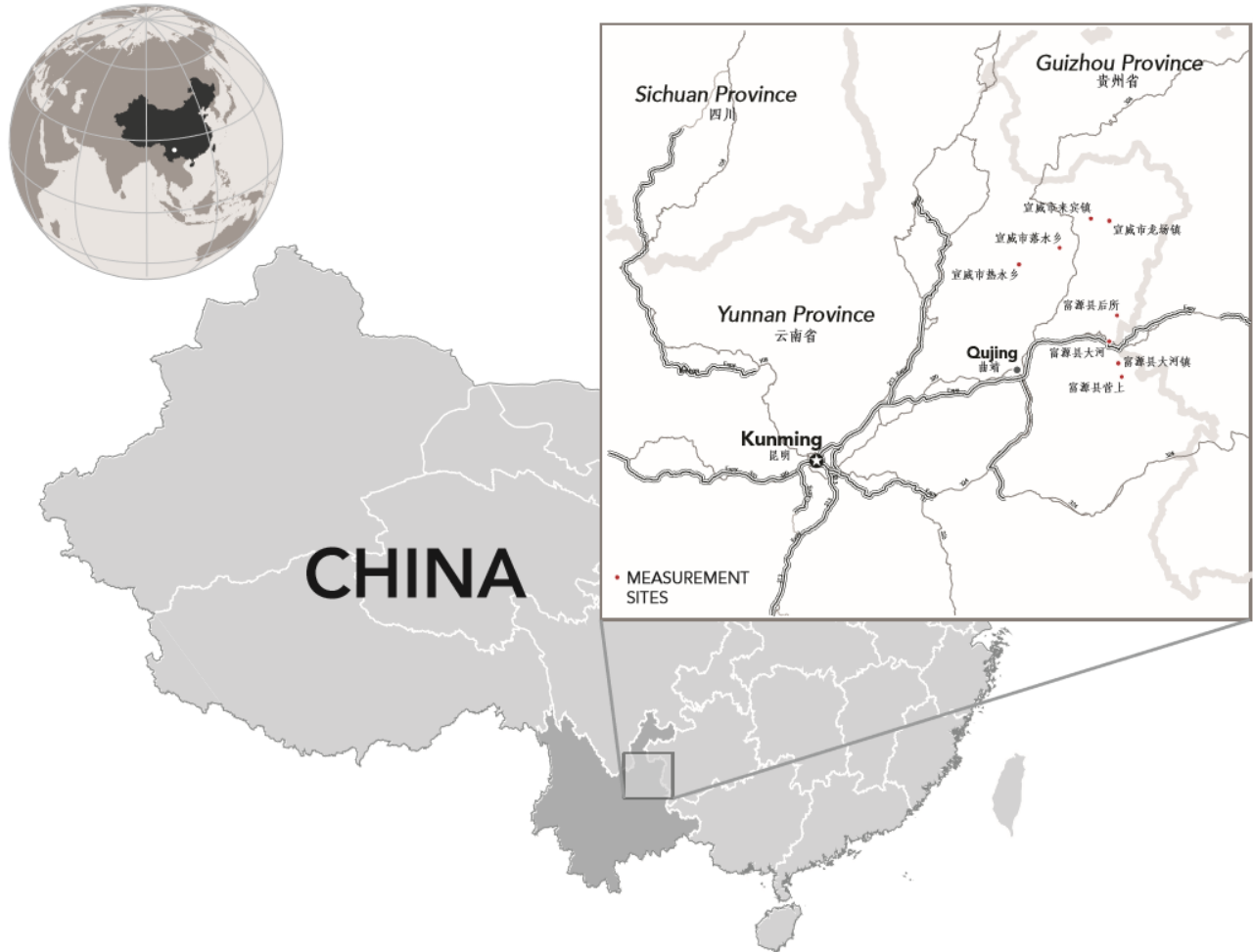


Figure 11: Study Area Map made by Andy Dang. Villages in the study area are red dots.

APPENDIX B: FUEL ANALYSIS

Small coal samples (5-50 grams) were taken from each household and sorted into three groups: honeycomb, lignite, and bituminous. Raw coal chunks include a large variety of particle sizes from powder up to 10 cm in diameter. We further divided raw coal chunks into two subgroups, lignite and bituminous, based on hardness, texture, and appearance. Lignite coal was softer, brownish, and friable, while bituminous coal was harder, shiny, and maintained integrity. The bituminous subgroup could also contain sub-bituminous and anthracite coal types. However, since these subgroups were mixed with biomass during burning and no significant differences were determined based on fuel mixture, we do not emphasize the distinction among coal fuels. Ultimate, proximate, and heating value analyses were performed on mixture-averaged samples of each coal group to determine mass fractions of carbon, hydrogen, nitrogen, oxygen, sulfur, fixed carbon, volatile matter, lower heating value, and higher heating value. Fuel analysis results are given below in Table 2.

Table 2: Proximate and Ultimate fuel analysis results for coals and corncobs. Woody fuel samples were not analyzed, and instead approximated from analysis of similar woody fuels published by the Biomass Energy Foundation.

	Lignite	Bituminous Sample 1	Bituminous Sample 2	Honeycomb Briquettes	Corncoobs	Woody
Fuel composition (% dry basis)						
Ash	37.0	20.7	28.0	72.2	1.4	
Volatile Matter	23.2	23.2	19.6	10.0	79.0	
Fixed Carbon	39.7	56.2	52.3	17.8	19.6	
Carbon	47.3	66.8	61.1	20.6	46.9	50.0
Hydrogen	3.0	4.4	3.8	0.8	6.2	
Nitrogen	1.0	1.3	1.1	0.4	0.5	
Oxygen	11.3	6.4	5.8	5.6	44.9	
Sulfur	0.3	0.3	0.2	0.3	0.1	
Fuel Heating Value (MJ/kg)						
LHV (MJ/kg)	17.5	26.6	24.5	7.0	17.4	18.5
HHV (MJ/kg)	18.3	27.6	25.3	7.2	19.0	

APPENDIX C: EMISSION FACTOR CALCULATIONS

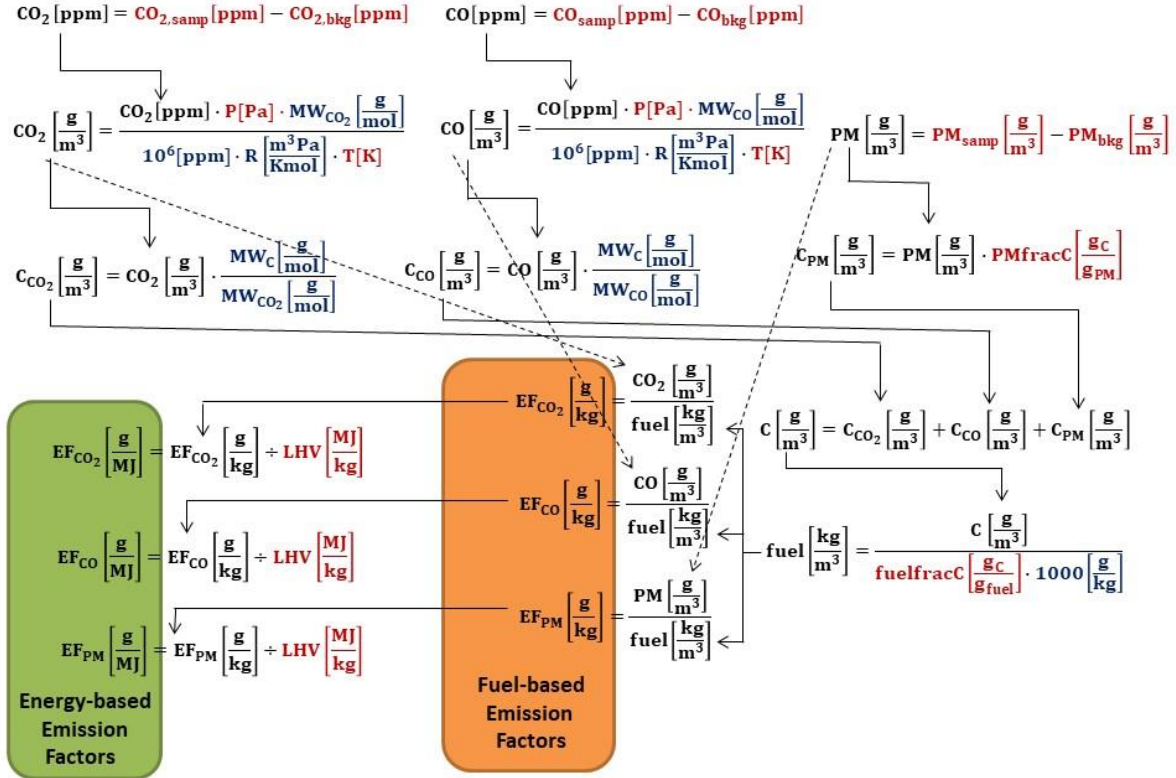


Figure 12: Partial Capture Carbon Balance Emission Factor Calculation Flow Chart. Red variables are measured inputs and blue variables are known constants.

The carbon fraction of PM **PMfracC** is determined from OCEC analysis:

$$PMfracC = (OC + EC) / PM \quad (3)$$

The fuel carbon fraction **fuelfracC** and lower heating value **LHV** were determined for each event by weighing each type of fuel in the mixture with a scale and obtaining fuel samples for laboratory analysis. Then mixture averaged fuel properties were calculated for each event by combining fuel laboratory analysis results and published fuel property data.

For calculations of emission factors for phases within a cooking event, the dilution ratio, fuel carbon fraction **fuelfracC**, and fuel energy content **LHV** were assumed to be constant throughout the cooking event.

Organic carbon, elemental carbon, and scattering emission factors were calculated by:

$$EF_{OC} \left[\frac{g}{kg} \right] = \frac{OC \left[\frac{g}{m^3} \right]}{fuel \left[\frac{kg}{m^3} \right]}, \quad (4)$$

$$EF_{EC} \left[\frac{g}{kg} \right] = \frac{EC \left[\frac{g}{m^3} \right]}{fuel \left[\frac{kg}{m^3} \right]}, \quad (5)$$

$$EF_{scat} \left[\frac{m^2}{kg} \right] = \frac{b_{sp} [m^{-1}]}{fuel \left[\frac{kg}{m^3} \right]}, \quad (6)$$

where the **fuel** variable is calculated using the above flow chart (Figure 12).

APPENDIX D: SUNSET ANALYZER PAR FILE

```
'modified NIOSH870.par
Minimum Step Time,45
Maximum Step Time,300
Helium, 10, 1, .001, 100, 0, -1
Helium, 120, 340, .025, 75, 0, 19
Helium, 120, 500, .08, 60, 0, 29
Helium, 120, 615, .12, 40, 0, 33
Helium, 180, 870, .30, 35, 0, 46
Helium, 45, 550, .001, 100, 16, 50
Oxygen, 90, 550, .18, 65, 0, 51
Oxygen, 90, 625, .18, 42, 0, 51
Oxygen, 90, 700, .20, 36, 0, 51
Oxygen, 90, 775, .27, 32, 0, 42
Oxygen, 90, 850, .25, 25, 0, 45
Oxygen, 120, 890, .30, 20, 0, 46
CalibrationOx, 120, 1, .001, 100, 16, 46
Offline, 1, 0, .001, 100, 16
*****
'format for above:
'Mode; time(sec); temperature(deg C); power constant; time constant; blower speed; Sample
Position Temp Offset
'power constant - .0001 to 1; think of it as a percentage
'typical power constats are .01 to .4; must be positive
'time constant (seconds) - 1 to 200 must be positive
'typical time constants - 10 to 120
'for low temperatures, use long time constant and low power
'for high temperatures, use higher power and shorter time constants
'blower speed - 0 to 16; 0 = off; 16 = full. Only use below 300 C, where 1 or 2 helps prevent
overshoots
```

APPENDIX E: EMISSION FACTORS FROM PUBLISHED DATA

Table 3: Summary of results taken from published emission studies of similar stoves burning coal or biomass fuels.^{21,25-31,35-44} For biomass fuel tests, only those burning wood, branches, and corn residue similar to biomass fuels in our study were considered for comparison. The results are reported in a variety of metrics that required conversion to the data points in Figure 6 for comparison with this study. Some results are reported as statistical representations (mean and standard deviation). These results are converted to individual data points assuming a normal distribution.

Publication (citation #)	Stoves, Fuels, and Lighting Method	Metrics ^a	Conversion to EF _e (g/MJ) ^a	# of data points converted
Zhang, 2000 (36)	11 stove/coal combinations. 5 stove/biomass combinations. 5 stoves, 5 coal types, 3 biomass types. Coal lit with kerosene	EF _{edel,TSP} , EF _{edel,CO} , η_{th} . Test replicates reported as mean and CoV. Single tests reported as single values.	EF _e = EF _{edel} × η_{th} . Mean and CoV converted to 3 data points: mean-SD, mean, mean+SD.	31 points for coal EF _{e,PM} , EF _{e,CO} . 16 points for biomass EF _{e,PM} , EF _{e,CO} .
Bond, 2002 (37)	3 stove/coal combinations. Bituminous and lignite burned in Stove 1 and honeycomb briquettes in Stove 2. Lit with hot bed of charcoal	EF _{m,TSP} , HV. Test replicates reported as mean and SD.	EF _e = EF _m / HV. Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	9 points for coal EF _{e,PM} .
Zhi, 2008 (38)	18 stove/coal combinations. 9 coal types burned as raw chunks in Stove 1 and honeycomb briquettes in Stove 2. Lit with hot bed of charcoal	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} (grams per kilogram dry ash-free fuel). Single tests reported as single values.	EF _e = EF _m / HV. Assuming HV = 30 MJ/kg dry ash-free fuel.	18 points for coal EF _{e,PM} , EC/TC.
Zhi, 2009 (39)	24 stove/coal combinations. 6 coal types burned as raw chunks and briquettes in each of 2 stoves. Lit with hot bed of charcoal	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} (grams per kilogram dry ash-free fuel). Single tests reported as single values.	EF _e = EF _m / HV. Assuming HV = 30 MJ/kg dry ash-free fuel.	24 points for coal EF _{e,PM} , EC/TC.
Ge, 2004 (40)	3 stove/coal combinations. 2 coal types burned in 1 stove. Lighting method not reported	EF _{edel,TSP} , η_{th} . Test replicates reported as mean and SD.	EF _e = EF _{edel} × η_{th} . Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	6 points for coal EF _{e,PM} .
Chen, 2005 (41)	5 stove/coal combinations. 5 coal types burned in 1 stove. Lit with hot bed of charcoal	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} (grams per kilogram dry ash-free fuel). Single tests reported as single values.	EF _e = EF _m / HV. Assuming HV = 30 MJ/kg dry ash-free fuel for bituminous and anthracite, and HV = 22 MJ/kg dry ash-free fuel for subbituminous.	5 points for coal EF _{e,PM} , EC/TC.
Chen, 2006 (42)	9 stove/coal combinations. 9 coal types burned in 1 stove. Lit with hot bed of charcoal	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} (grams per kilogram dry ash-free fuel). Test replicates reported as mean and SD.	EF _e = EF _m / HV. Assuming HV = 30 MJ/kg dry ash-free fuel. Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	27 points for coal EF _{e,PM} , EC/TC.

Table 3 continued (2/3)

Publication (main text citation #)	Stoves, Fuels, and Lighting Method	Metrics ^a	Conversion to EF _e (g/MJ) ^a	# of data points converted
Shen, 2010 (44)	5 stove/coal combinations. 1 stove/biomass combination. 5 coal types burned in 1 stove. Corn burned in another stove. Coal lit with wood chips but lighting emissions not included in reported emissions.	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} , HV. Test duplicates reported as mean and SD.	EF _e = EF _m /HV. Mean and SD converted to 2 data points: mean-SD, mean+SD.	10 points for coal EF _{e,PM} , EC/TC. 2 points for biomass EF _{e,PM} , EC/TC.
Shen, 2013 (29)	2 stove/coal combinations. 1 stove/biomass combination. Coal briquettes burned in Stove 1, coal cakes in Stove 2, and wood in Stove 3 Coal lighting method not reported	EF _{m,TSP} , EF _{m,EC} , EF _{m,OC} . Single tests reported as single values, two tests per stove.	EF _e = EF _m /HV. Assuming HV = 20 MJ/kg for coal briquettes, 7 MJ/kg for coal cakes, and 19 MJ/kg for wood.	4 points for coal EF _{e,PM} , EC/TC. 2 points for biomass EF _{e,PM} , EC/TC.
Li, 2007 (27)	1 stove/biomass combination. Maize residue burned in 1 stove.	EF _{edel,PM2.5} , EF _{edel,EC} , EF _{edel,OC} , EF _{edel,CO} , η_{th} . Test duplicates reported as mean and SD.	EF _e = EF _{edel} x η_{th} Mean and SD converted to 2 data points: mean-SD, mean+SD.	2 points for biomass EF _{e,PM} , EF _{e,CO} , EC/TC.
Li, 2009 (28)	2 stove/biomass combinations. Maize residue and woody fuel burned in 1 stove.	EF _{m,PM2.5} , EF _{m,EC} , EF _{m,OC} . Test replicates reported as mean and SD.	EF _e = EF _m /HV assuming HV measured in Li, 2007. Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	6 points for biomass EF _{e,PM} , EC/TC.
Jetter, 2012 (43)	1 stove/biomass combination. Corncobs burned in 1 stove.	EF _{edel,PM2.5} , EF _{edel,CO} , η_{th} . Test replicates reported as mean and SD.	EF _e = EF _{edel} x η_{th} Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	3 points for biomass EF _{e,PM} , EF _{e,CO} .
Du, 2018 (21)	7 stove/biomass combinations. 3 brick chimney stoves built in place. Stove 1: Sichuan: 10 corncob, 10 brushwood, 10 fuel wood Stove 2: Shanxi: 12 corncob, 10 fuel wood Stove 3: Guizhou, 6 corncob, 18 fuel wood	EF _{m,PM2.5} , EF _{m,CO} , EF _{m,OC} EF _{m,EC} . Test replicates reported as mean, SD, and range.	EF _e = EF _m /HV assuming HV measured in Li, 2007. Mean and range used as 3 data points: min, mean, max	21 points for biomass EF _{e,PM} , EF _{e,CO} . 7 points for biomass EC/TC
Chen, 2016 (31)	3 stove/coal combinations 1 stove/biomass combination Chimney stoves: Cooking with honeycomb briquettes and wood. Space heating with raw coal chunks and peat. Lighting method not reported	EF _{e,PM2.5} , EF _{e,CO} , EF _{e,OC} EF _{e,EC} . Reported as mean and SD.	Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	9 points for coal EF _{e,PM2.5} , EF _{e,CO} . 3 points for coal EC/TC 3 points for biomass EF _{e,PM2.5} , EF _{e,CO} . 1 point for biomass EC/TC

Table 3 continued (3/3)

Publication (main text citation #)	Stoves, Fuels, and Lighting Method	Metrics ^a	Conversion to EF _e (g/MJ) ^a	# of data points converted
Eilenberg, 2018 (25)	3 stove/coal combinations 1 stove/biomass combination Coal honeycomb briquettes burned in 1 portable stove and 4 chimney stoves. Raw coal burned in 3 chimney stoves. Wood burned in 1 chimney stove. Lighting method: hot coal bed	EF _{m,PM2.5} , EF _{m,CO} , EF _{m,OC} EF _{m,EC} . Reported for individual 9 events as mean and SD.	EF _e = EF _m /HV assuming HV = 30 MJ/kg for raw coal, 20 MJ/kg for coal briquettes, and 19 MJ/kg for wood.	8 points for coal EF _{e,PM2.5} , EF _{e,CO} , and EC/TC. 1 point for biomass EF _{e,PM2.5} , EF _{e,CO} , and EC/TC.
Shen, 2015 (30)	1 stove/coal combination 11 coal tests in 1 stove Lighting method not reported	EF _{m,TSP} , EF _{m,CO} , EF _{m,OC} EF _{m,EC} reported as mean and SD.	EF _e = EF _m /HV using HV = 20.9 MJ/kg from coal fuel analysis. Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	3 points for coal EF _{e,PM2.5} , EF _{e,CO} , and EC/TC. 1 point for coal EC/TC.
Sun, 2018 (26)	28 stove/coal combinations 2 stove/biomass combinations 7 coal types burned as honeycomb briquettes in 1 stove and raw chunks in 3 stoves. Corncobs and pine wood burned in 1 stove. Lighting method: hot coal bed (alcohol gel used for initial lighting, but lighting emissions not included in reported emissions).	EF _{m,OC} EF _{m,EC} .	EF _e = EF _m /HV assuming HV = 30 MJ/kg for dry ash free coal, and 19 MJ/kg for wood, and 17 MJ/kg for corncobs.	28 points for coal EC/TC 2 points for biomass EC/TC.
Zhang, 2008 (35)	3 stove/coal combinations 3 types of coal burned in 1 stove Lighting method not reported	EF _{m,PM2.5} , EF _{m,OC} EF _{m,EC} reported as mean and SD for each coal.	EF _e = EF _m /HV assuming HV = 30 MJ/kg for coal. Mean and SD converted to 3 data points: mean-SD, mean, mean+SD.	9 points for coal EF _{e,PM2.5} . 3 points for coal EC/TC.

^a Abbreviations:

EF_{edel} = emission factor, grams per megajoule delivered to the cooking vessel.

EF_e = emission factor, grams per megajoule fuel consumed.

EF_m = emission factor, grams per kilogram fuel consumed.

η_{th} = thermal efficiency (-).

HV = heating value, megajoules per kilogram fuel.

TSP = total suspended particulate matter.

SD = standard deviation.

CoV = coefficient of variation = SD/mean.

APPENDIX F: SUMMARY OF COOKING EVENTS

Table 4: General information and fuel data for each cooking event.

Test #	Date (yyyymmdd)	Stove Type	House ID	Village ID	Complete Burn Cycle?	Cooking Event Length (minutes)	Dry fuel mass consumed (kg) ^a										Carbon Fraction (-)		LHV (MJ/kg)	
							Bituminous Coal		Lignite Coal		Coal Briquettes		Woody		Corncobs					
							Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ		
3	20130722	chimney	h1	v1	Complete	55	0.85	0.21	0.00	0.00	0.00	0.00	0.80	0.12	0.00	0.00	0.59	0.10	22.7	4.0
4	20130723	chimney	h2	v1	Complete	160	2.29	0.38	0.00	0.00	0.00	0.00	0.46	0.08	0.00	0.00	0.63	0.11	23.9	4.1
5	20130723	chimney	h2	v1	Complete	53	1.11	0.28	0.00	0.00	0.00	0.00	0.46	0.08	0.00	0.00	0.61	0.13	23.1	4.9
6	20130724	chimney	h3	v1	Incomplete	98	?	N/A	0.00	0.00	0.00	0.00	?	N/A	0.00	0.00	0.58	0.12	23.0	4.6
7	20130724	chimney	h3	v1	Complete	70	?	N/A	0.00	0.00	0.00	0.00	?	N/A	0.00	0.00	0.58	0.12	23.0	4.6
8	20130725	chimney	h22	v2	Complete	76	1.09	0.27	0.00	0.00	0.00	0.00	0.00	0.00	1.13	0.63	0.56	0.16	21.1	6.0
10	20130726	portable	h4	v2	Complete	84	0.27	0.07	0.00	0.00	0.00	0.00	0.00	0.00	0.21	0.09	0.57	0.13	21.7	5.0
11	20130727	portable	h5	v2	Complete	152	0.78	0.19	0.00	0.00	0.00	0.00	0.00	0.00	0.47	0.09	0.56	0.11	21.8	4.3
12	20130727	portable	h5	v2	Complete	86	?	N/A	0.00	0.00	0.00	0.00	0.19	0.05	0.68	0.08	0.59	0.12	22.4	4.5
13	20130728	chimney	h6	v3	Incomplete	35	?	N/A	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.65	0.13	25.0	5.0
14	20130728	chimney	h6	v3	Complete	104	1.90	0.47	0.00	0.00	0.00	0.00	0.16	0.08	0.37	0.08	0.63	0.14	24.7	5.7
15	20130729	portable	h7	v3	Complete	94	0.31	0.08	0.00	0.00	0.00	0.00	0.00	0.00	0.68	0.11	0.51	0.08	19.6	3.1
16	20130729	portable	h7	v3	Complete	112	?	N/A	0.00	0.00	0.00	0.00	0.09	0.04	0.94	0.11	0.58	0.12	22.7	4.5
17	20130730	chimney	h8	v3	Complete	67	?	N/A	0.00	0.00	0.00	0.00	0.00	0.00	0.50	0.16	0.61	0.12	23.5	4.7
18	20130730	chimney	h8	v3	Complete	84	0.63	0.16	0.00	0.00	0.00	0.00	0.00	0.00	0.69	0.15	0.56	0.10	21.0	3.9
19	20130731	chimney	h9	v4	Complete	130	?	N/A	0.00	0.00	0.00	0.00	0.00	0.00	0.34	0.07	0.64	0.13	25.3	5.1
20	20130731	chimney	h9	v4	Complete	70	1.15	0.29	0.00	0.00	0.00	0.00	0.00	0.00	0.47	0.09	0.61	0.13	23.9	5.2
21	20130801	chimney	h10	v5	Complete	115	0.00	0.00	2.46	0.45	0.00	0.00	0.51	0.09	0.00	0.00	0.48	0.08	17.7	3.1
22	20130801	chimney	h10	v5	Complete	98	0.00	0.00	1.53	0.32	0.00	0.00	0.84	0.10	0.00	0.00	0.48	0.08	17.9	2.8
23	20130808	chimney	h11	v6	Complete	182	3.67	0.92	0.00	0.00	0.00	0.00	0.00	0.00	0.57	0.10	0.64	0.16	25.4	6.2
25	20130809	chimney	h12	v6	Complete	99	0.00	0.00	3.08	0.48	0.00	0.00	0.60	0.10	3.14	0.45	0.47	0.06	17.5	2.1
26	20130809	portable	h12	v6	Incomplete	89	0.00	0.00	0.00	0.00	?	N/A	0.00	0.00	0.00	0.00	0.20	0.04	7.0	1.4
27	20130810	portable	h13	v6	Complete	212	0.00	0.00	0.00	0.00	?	N/A	0.20	0.05	0.23	0.06	0.25	0.05	8.9	1.8
28	20130810	portable	h13	v6	Incomplete	42	0.00	0.00	0.00	0.00	?	N/A	0.00	0.00	0.00	0.00	0.20	0.04	7.0	1.4
29	20130811	portable	h14	v7	Complete	149	?	N/A	0.00	0.00	0.00	0.00	0.12	0.05	0.59	0.07	0.61	0.12	24.0	4.8
30	20130811	portable	h14	v7	Complete	170	0.91	0.23	0.00	0.00	0.00	0.00	0.24	0.06	0.28	0.06	0.60	0.12	23.4	4.7
31	20130812	chimney	h15	v7	Complete	183	?	N/A	0.00	0.00	0.00	0.00	0.08	0.04	0.48	0.05	0.57	0.11	22.5	4.5
32	20130812	chimney	h15	v7	Complete	109	0.62	0.13	0.00	0.00	0.00	0.00	0.15	0.05	0.49	0.06	0.54	0.08	21.0	3.1
33	20130813	portable	h16	v7	Complete	112	?	N/A	0.00	0.00	0.00	0.00	0.04	0.00	0.38	0.04	0.59	0.12	23.3	4.7
34	20130813	portable	h16	v7	Complete	140	?	N/A	0.00	0.00	0.00	0.00	0.12	0.01	0.32	0.04	0.59	0.12	23.3	4.7
35	20130815	chimney	h17	v8	Complete	96	1.24	0.31	0.00	0.00	0.00	0.00	0.18	0.05	0.41	0.08	0.57	0.11	22.3	4.6
36	20130815	chimney	h17	v8	Complete	107	2.16	0.54	0.00	0.00	0.00	0.00	0.18	0.05	0.41	0.08	0.58	0.13	23.0	5.2
37	20130816	chimney	h18	v8	Complete	111	2.04	0.51	0.00	0.00	0.00	0.00	1.25	0.18	0.00	0.00	0.57	0.11	22.2	4.3
40	20130817	chimney	h19	v8	Complete	158	1.92	0.48	0.00	0.00	0.00	0.00	0.00	0.00	0.65	0.19	0.57	0.13	22.7	5.1
41	20130818	chimney	h20	v9	Incomplete	171	?	N/A	0.00	0.00	0.00	0.00	?	N/A	0.00	0.00	0.62	0.12	24.0	4.8
42	20130819	chimney	h21	v9	Complete	132	1.68	0.37	0.00	0.00	0.00	0.00	0.00	0.00	0.51	0.18	0.62	0.13	24.4	5.2
43	20130819	chimney	h21	v9	Complete	161	?	N/A	0.00	0.00	0.00	0.00	0.00	0.00	1.19	0.14	0.62	0.12	24.5	4.9

^a Question marks indicate the fuel type was consumed, but the fuel mass was not measured. The associated confidence interval σ is N/A because it could not be calculated.

Table 5: Fuel mass and fuel energy based emission factors for each cooking event.

Test #	Fuel Mass Based Emission Factors (g/kg _{fuel})										Fuel Energy Based Emission Factors (g/MJ)									
	EF _{m,CO}		EF _{m,CO2}		EF _{m,PM2.5}		EF _{m,EC}		EF _{m,OC}		EF _{E,CO}		EF _{E,CO2}		EF _{E,PM2.5}		EF _{E,EC}		EF _{E,OC}	
	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ
3	71	20	1981	349	21.6	5.3	1.2	0.3	14.8	3.6	3.12	1.04	87.3	21.9	0.95	0.29	0.05	0.02	0.65	0.20
4	194	93	1891	367	30.8	14.4	1.4	0.7	24.4	11.3	8.10	4.14	79.1	20.5	1.29	0.64	0.06	0.03	1.02	0.50
5	91	52	2025	436	14.6	7.4	5.1	2.5	9.3	4.6	3.95	2.40	87.7	26.5	0.63	0.35	0.22	0.12	0.40	0.22
6	138	51	1895	386	2.5	1.0	1.0	0.4	2.9	1.0	5.98	2.54	82.4	23.5	0.11	0.05	0.04	0.02	0.12	0.05
7	77	26	1983	398	6.6	2.0	1.5	0.4	4.4	1.3	3.34	1.30	86.2	24.4	0.29	0.10	0.07	0.02	0.19	0.07
8	149	48	1753	507	17.8	5.6	1.6	0.5	14.2	4.5	7.07	3.03	83.0	33.7	0.84	0.36	0.08	0.03	0.67	0.28
10	92	27	1943	450	4.5	1.3	0.3	0.1	2.2	0.6	4.26	1.59	89.4	29.2	0.21	0.08	0.01	0.00	0.10	0.04
11	190	44	1721	339	9.2	2.1	0.6	0.1	5.4	1.2	8.72	2.66	78.8	22.1	0.42	0.13	0.03	0.01	0.25	0.08
12	198	45	1821	366	12.5	2.8	0.3	0.1	8.0	1.8	8.83	2.66	81.3	23.0	0.56	0.17	0.01	0.00	0.36	0.11
13	213	58	2040	413	0.0	0.7	0.8	0.2	0.9	0.2	8.53	2.88	81.6	23.2	0.00	0.03	0.03	0.01	0.03	0.01
14	126	33	2044	466	17.0	4.4	3.0	0.8	12.0	3.1	5.09	1.78	82.9	26.8	0.69	0.24	0.12	0.04	0.49	0.17
15	143	52	1570	264	26.1	9.1	4.1	1.4	20.0	6.9	7.29	2.87	80.0	18.4	1.33	0.51	0.21	0.08	1.02	0.39
16	113	30	1898	381	11.6	3.0	0.3	0.1	8.0	2.0	5.00	1.67	83.6	23.7	0.51	0.17	0.01	0.00	0.35	0.11
17	95	21	2075	415	8.1	1.8	5.8	1.2	1.7	0.4	4.03	1.20	88.3	25.0	0.35	0.10	0.25	0.07	0.07	0.02
18	96	19	1865	341	6.5	1.3	3.9	0.8	1.6	0.3	4.57	1.24	88.7	23.0	0.31	0.08	0.19	0.05	0.08	0.02
19	150	44	1998	406	29.7	8.6	10.0	2.9	19.4	5.6	5.96	2.11	79.1	22.6	1.18	0.41	0.40	0.14	0.77	0.27
20	153	40	1844	399	41.8	10.8	9.5	2.4	31.6	8.1	6.41	2.19	77.1	23.6	1.75	0.59	0.40	0.13	1.32	0.44
21	132	35	1536	272	1.8	0.6	0.6	0.1	1.1	0.3	7.48	2.37	86.9	21.6	0.10	0.04	0.03	0.01	0.06	0.02
22	114	26	1583	252	1.7	0.5	0.5	0.1	1.2	0.2	6.39	1.76	88.7	19.9	0.09	0.03	0.03	0.01	0.06	0.02
23	127	36	2094	511	17.3	4.9	0.7	0.2	14.9	4.1	5.01	1.87	82.5	28.6	0.68	0.25	0.03	0.01	0.59	0.22
25	120	30	1500	183	14.0	3.4	1.0	0.2	12.3	2.9	6.82	1.87	85.5	14.4	0.80	0.21	0.06	0.02	0.70	0.18
26	15	5	708	142	0.4	0.1	0.0	0.0	0.1	0.0	2.21	0.80	101.1	28.6	0.05	0.02	0.00	0.00	0.02	0.01
27	70	38	772	171	15.5	8.2	0.2	0.1	9.5	5.0	7.87	4.53	86.3	25.7	1.73	0.98	0.02	0.01	1.06	0.60
28	46	25	661	137	0.0	0.4	0.0	0.0	0.2	0.1	6.51	3.86	94.4	27.2	0.00	0.06	0.00	0.00	0.02	0.01
29	111	32	2047	411	5.8	1.6	0.1	0.0	3.4	0.9	4.64	1.61	85.3	24.2	0.24	0.08	0.01	0.00	0.14	0.05
30	94	26	2049	403	1.7	0.5	0.0	0.0	1.0	0.3	4.00	1.38	87.5	24.5	0.07	0.02	0.00	0.00	0.04	0.01
31	163	74	1806	379	9.2	4.1	0.8	0.4	6.2	2.7	7.27	3.60	80.3	23.3	0.41	0.20	0.04	0.02	0.27	0.13
32	222	106	1577	295	19.8	9.4	1.0	0.5	16.1	7.6	10.54	5.29	75.0	17.9	0.94	0.47	0.05	0.02	0.76	0.38
33	169	57	1823	375	21.3	7.0	0.9	0.3	15.4	5.0	7.24	2.85	78.3	22.5	0.91	0.35	0.04	0.02	0.66	0.25
34	79	23	1992	400	10.3	2.9	0.7	0.2	7.9	2.2	3.39	1.21	85.6	24.3	0.44	0.15	0.03	0.01	0.34	0.12
35	97	30	1874	381	17.0	5.1	2.1	0.6	12.9	3.8	4.33	1.61	84.0	24.3	0.76	0.28	0.09	0.03	0.58	0.21
36	85	25	1945	437	16.5	4.7	0.5	0.1	14.1	4.0	3.68	1.38	84.4	26.9	0.72	0.26	0.02	0.01	0.61	0.22
37	127	42	1857	355	8.2	2.7	1.3	0.4	5.8	1.9	5.70	2.19	83.6	22.6	0.37	0.14	0.06	0.02	0.26	0.10
40	55	15	2008	448	3.7	1.0	1.2	0.3	1.8	0.5	2.42	0.87	88.4	28.1	0.16	0.06	0.05	0.02	0.08	0.03
41	117	36	2087	420	0.7	0.3	0.2	0.1	0.2	0.1	4.86	1.78	87.0	24.7	0.03	0.02	0.01	0.00	0.01	0.00
42	167	72	1992	432	7.2	3.1	1.3	0.5	5.3	2.2	6.82	3.27	81.5	24.7	0.29	0.14	0.05	0.02	0.22	0.10
43	176	78	1991	415	4.9	2.2	0.9	0.4	3.0	1.3	7.17	3.51	81.3	23.5	0.20	0.10	0.04	0.02	0.12	0.06

Table 6: Emission rates, MCE, and optical properties for each cooking event.

Test #	Emission Rates (g/hr)								MCE (-)		Optical Properties							
	ER _{CO}		ER _{PM2.5}		ER _{EC}		ER _{OC}				SSA ₈₈₀ (-)		MSC ₆₃₅ (m ² /g)		MSC ₈₈₀ (m ² /g)		MAC ₈₈₀ (m ² /g)	
	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ	Value	σ
3	128	41	39.1	11.2	2.1	0.6	26.7	7.6	0.95	0.01	0.70	0.05	2.4	0.5	2.2	0.4	16.6	2.3
4	200	101	31.9	15.5	1.5	0.7	25.2	12.2	0.86	0.06	0.86	0.03	5.8	1.2	4.5	0.9	15.8	2.1
5	164	98	26.2	14.1	9.2	4.9	16.6	8.8	0.93	0.04	0.58	0.06	5.6	1.2	4.6	1.0	9.6	1.4
^a 6	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.90	0.03	0.11	0.03	2.8	0.7	1.4	0.4	29.2	4.1
^a 7	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.94	0.02	0.54	0.06	5.4	1.2	3.4	0.7	12.2	1.6
8	263	117	31.4	13.9	2.8	1.3	24.9	11.0	0.88	0.02	0.77	0.04	6.0	1.2	3.7	0.8	12.2	1.7
10	32	12	1.5	0.6	0.1	0.0	0.7	0.3	0.93	0.01	0.65	0.06	3.2	0.7	1.7	0.4	15.3	2.1
11	93	27	4.5	1.3	0.3	0.1	2.7	0.8	0.85	0.02	0.64	0.06	2.5	0.5	1.8	0.4	16.2	2.2
^a 12	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.85	0.02	0.78	0.04	2.2	0.5	1.6	0.3	21.6	3.1
^{a,b} 13	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.86	0.03	0.30	0.06	N/A	N/A	N/A	N/A	6.2	1.0
14	176	59	23.9	7.8	4.2	1.4	16.9	5.5	0.91	0.01	0.52	0.06	3.4	0.7	2.1	0.4	10.8	1.5
15	90	35	16.4	6.1	2.5	0.9	12.5	4.7	0.87	0.04	0.70	0.05	4.0	0.8	2.7	0.5	7.2	1.0
^a 16	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.91	0.02	0.90	0.02	3.0	0.6	2.2	0.5	9.0	1.3
^a 17	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.93	0.01	0.32	0.05	2.6	0.5	1.4	0.3	4.3	0.6
18	90	23	6.2	1.6	3.7	0.9	1.5	0.4	0.93	0.01	0.31	0.05	2.4	0.5	1.3	0.3	4.8	0.7
^a 19	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.89	0.02	0.50	0.07	2.3	0.5	1.8	0.4	5.3	0.9
20	214	69	58.3	18.6	13.3	4.2	44.1	13.9	0.88	0.02	0.66	0.05	3.7	0.8	3.2	0.7	7.2	0.9
21	204	62	2.7	1.0	0.9	0.3	1.6	0.5	0.88	0.02	0.66	0.06	6.7	1.9	5.0	1.4	8.3	1.3
22	166	44	2.5	0.8	0.7	0.2	1.7	0.4	0.90	0.02	0.51	0.07	6.0	1.7	3.8	1.1	12.0	1.9
23	178	63	24.3	8.6	0.9	0.3	20.8	7.3	0.91	0.01	0.80	0.04	5.6	1.2	4.1	0.9	26.6	3.5
^a 25	496	132	58.0	15.1	4.2	1.1	50.9	13.0	0.89	0.02	0.81	0.04	6.2	1.3	4.4	0.9	14.4	1.9
^a 26	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.97	0.01	0.93	0.09	2.1	0.8	1.7	0.6	5.1	7.4
^a 27	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.87	0.07	0.96	0.01	1.7	0.4	3.6	0.7	11.5	2.2
^{a,b,c} 28	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.90	0.05	0.73	0.58	N/A	N/A	N/A	N/A	N/A	N/A
^a 29	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.92	0.02	0.93	0.02	2.5	0.5	2.9	0.6	9.7	1.7
30	47	15	0.9	0.3	0.0	0.0	0.5	0.2	0.93	0.01	0.78	0.10	1.1	0.2	0.4	0.1	8.8	4.3
^a 31	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.88	0.05	0.80	0.04	5.8	1.2	5.1	1.1	15.0	2.2
32	154	76	13.8	6.8	0.7	0.3	11.2	5.4	0.82	0.09	0.91	0.02	6.4	1.3	6.9	1.4	14.0	2.0
^a 33	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.87	0.04	0.77	0.05	2.3	0.6	2.1	0.5	14.1	2.2
^a 34	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.94	0.01	0.65	0.06	2.3	0.5	1.7	0.4	13.9	2.1
35	111	39	19.5	6.8	2.4	0.8	14.7	5.1	0.93	0.02	0.64	0.06	5.1	1.1	3.3	0.7	15.1	2.1
36	131	47	25.5	8.9	0.7	0.2	21.8	7.5	0.94	0.01	0.89	0.02	7.0	1.4	5.0	1.0	22.4	3.3
37	226	84	14.6	5.4	2.3	0.8	10.4	3.7	0.90	0.03	0.71	0.05	6.1	1.3	4.5	1.0	11.6	1.6
40	54	18	3.6	1.2	1.2	0.4	1.8	0.6	0.96	0.01	0.48	0.06	4.5	0.9	3.1	0.6	9.9	1.3
^a 41	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.92	0.02	0.18	0.06	1.5	0.6	0.7	0.4	13.0	1.8
42	166	78	7.2	3.3	1.3	0.6	5.3	2.4	0.88	0.04	0.55	0.06	5.8	1.3	3.3	0.7	14.6	2.1
^a 43	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A	0.88	0.05	0.50	0.06	4.3	1.0	2.3	0.6	12.5	1.8

^a Emission rates could not be calculated because fuel consumption was not measured^b MSC could not be calculated because no PM mass was detected on the filter^c MAC could not be calculated because no EC was detected on the filter

APPENDIX G: MEASUREMENT UNCERTAINTY ESTIMATES

Measurement uncertainty was estimated for each input parameter as 68% confidence intervals and then propagated to the final output metrics using first order error propagation assuming uncorrelated variables and random errors. Input uncertainties are listed in Table 7. The total uncertainty of a measured value is calculated as:

$$\text{total uncertainty} = \text{measured value} * \text{relative uncertainty} + \text{absolute uncertainty} \quad (7)$$

Table 7: Input measurement uncertainties

Input	Relative uncertainty (%)	Absolute uncertainty	Absolute units	Note
CO and background CO	5	3	ppm	
CO ₂ and background CO ₂	5	200	ppm	
light scattering	20	20	Mm ⁻¹	
light absorption	20	0	Mm ⁻¹	Plus propagated MicroAeth flow uncertainty
filter flows and MicroAeth flow	3	0	cm ³ /min	Plus difference in pre and post flow checks
gravimetric filter mass	0	5	µg	
EC and OC loading density	0	Output by OC/EC analyzer	µg/cm ²	
fuel mass	10	0	kg	
fuel carbon fraction and heating value	10 for measured values, 20 for assumed values	9	MJ/kg	
fuel moisture content	5		%MC	

APPENDIX H: EMISSION FACTOR VARIANCE

The following method was used to determine the contribution of devolatilization and solid combustion phases to the emission factor variance.

Let E_n be an emission characteristic, such as emission factor, within phase n , and let w be the weight describing contribution of the phase to the total. The weight must be consistent with the emission characteristic—for example, for emission factor in g/kg, w_1 must be the fraction of fuel burned in phase 1. E and w can also be defined for energy consumed in a phase or time spent in a phase.

For a situation with two phases

$$E_t = w_1 E_1 + w_2 E_2 \quad (8)$$

However, the weights are not independent:

$$E_t = w_1 E_1 + (1 - w_1) E_2 \quad (9)$$

Define uncertainty in the weights as σ_{wn} , and that in the emission characteristics as σ_{En} .

Assuming that weights and emission characteristics are independent, squared uncertainty in the total emission is:

$$\sigma_{Et}^2 = \sigma_{w1}^2 (E_1 - E_2)^2 + \sigma_{E1}^2 w_1^2 + \sigma_{E2}^2 (1 - w_1)^2 \quad (10)$$

The first term on the right hand side is the effect of the uncertainty in the relative fraction of phase 1. If the two emission characteristics E_1 and E_2 are identical, this uncertainty has no effect. The second and third terms show the contribution of the uncertainty of the emission characteristic in phases 1 and 2, respectively.

Contributions of the phase fraction and the emission characteristics to variance are summarized in Table S.7.

Table 8: Contributions of burn phases to emission factor variance. The ignition phase contributes most of the variance.

	Units	EF _{PM}		EF _{CO}	
		Value	Uncertainty	Value	Uncertainty
w ₁ ^a	none	0.65	0.14	0.65	0.14
EF, ignition phase ^b	g/kg	29.6	21.6	154	125
EF, solid phase ^b	g/kg	0.5	0.8	53	84
Contribution to variance		Absolute ^c	Relative	Absolute ^c	Relative
Term 1 (phase fraction)		16.5	8%	16.4	1%
Term 2 (ignition phase)		197	92%	1177	57%
Term 3 (solid phase)		0.086	0%	860	42%

^a Estimated from CO₂ concentration; uncertainty is standard deviation over all tests

^b Value for each event is average over the relevant phase; average given here is over all events; uncertainty is standard deviation over all events

^c Units are (g/kg)²

APPENDIX I: BOOK OF FUMITRON

The supplementary file *The_Book_of_Fumitron_v9.pdf* is a manual for the Fumitron emission equipment that includes operating instructions, diagrams, theory of operation, maintenance and repair, and parts lists.