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Quantifying Stove Emissions Related to Different Use Patterns for the Silver-mini (Small Turkish) Space Heating Stove

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Abstract

Air pollution levels in Ulaanbaatar, Mongolia's capital, are among the highest in the world. A primary source of this pollution is emissions from traditional coal-burning space heating stoves used in the Ger (tent) regions around Ulaanbaatar. Significant investment has been made to replace traditional heating stoves with improved low-emission high-efficiency stoves. Testing performed to support selection of replacement stoves or for optimizing performance may not be representative of true field performance of the improved stoves. Field observations and lab measurements indicate that performance is impacted, often adversely, by how stoves are actually being used in the field. The objective of this project is to identify factors that influence stove emissions under typical field operating conditions and to quantify the impact of these factors. A highly-instrumented stove testing facility was constructed to allow for rapid and precise adjustment of factors influencing stove performance. Tests were performed using one of the improved stove models currently available in Ulaanbaatar. Complete burn cycles were conducted with Nailakh coal from the Ulaanbaatar region using various startup parameters, refueling conditions, and fuel characteristics. Measurements were collected simultaneously from undiluted chimney gas, diluted gas drawn directly from the chimney and plume gas collected from a dilution tunnel above the chimney. CO, CO_2, O_2, temperature, pressure, and particulate matter (PM) were measured. We found that both refueling events and coal characteristics strongly influenced PM emissions and stove performance. Start-up and refueling events lead to increased PM emissions with more than 98% of PM mass emitted during the 20% of the burn where coal ignition occurs. CO emissions are distributed more evenly over the burn cycle, peaking both during ignition and late in the burn cycle. We anticipate these results being useful for quantifying public health outcomes related to the distribution of improved stoves and to identify opportunities for improving and sustaining performance of the new stoves.

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Executive Summary

Air pollution levels in Ulaanbaatar, Mongolia's capital, are among the highest in the world. A primary source of pollution is traditional coal-burning heating stoves used in the Ger (tent) regions around Ulaanbaatar. Significant investment has been made to replace traditional heating stoves with improved low-emission high-efficiency stoves. Field observations and lab measurements indicate that heating stove performance is impacted by how the stoves are actually being used in the field. The combination of stove design, fuel characteristics, and operational factors, leads to significant levels of variability and uncertainty in emissions measurements. The objective of this project is to identify key factors that influence stove emissions in the field and to quantify the impact of these factors.

A highly-instrumented stove testing facility was constructed to allow for rapid and precise adjustment of factors influencing stove performance. Tests were performed using a high-efficiency stove that is currently available around Ulaanbaatar. Tests included complete burn cycles where a number of factors such as startup parameters, refueling events, and fuel characteristics were modified. All tests were run with internal chimney draft and ceiling height calibrated to represent winter conditions experienced within a Ger in Ulaanbaatar. Redundant emissions measurements were collected from both diluted chimney gas and plume gas mixed with air above the chimney. Using Nailakh coal from the Ulaanbaatar region for testing, CO, CO_2, oxygen, temperature, pressure, and a variety of particulate matter (PM) measurements in the 2.5 micron size range were collected.

Initial results from laboratory experiments focused on the emissions impact of refueling events. Ignition of coal during refueling resulted in larger PM emissions than emission during cold starts. The duration of the elevated PM emission event depended on when during the burn cycle the refueling occurred and how much fuel was added. Finally, the moisture content and size of coal added during refueling seemed to impact the emissions profile but the number of replicate measurements was not sufficient to clearly characterize the effect.

We found that 99% of PM emissions occur during the ignition phase of a burn including cold start (operator lit fuel) and refueling events (residual ember/flame lit coal) which amounts to approximately 20% of the total duration of a burn. Approximately 4 times more PM emissions occur during a refueling ignition event compared to a cold start ignition where an average of 60 grams are emitted during refueling (range 9 – 110 g) compare to an average of 15 g emitted during a cold start (range 1 – 28 g). In addition, the majority of PM emitted during a cold-start was from wood/paper used to start the fuel with only a moderate addition from ignition of the coal. All PM emitted during a refueling event are from ignition of coal. CO emissions were more variable across different stages of the burn with 20 grams of CO emitted per kg coal consumed (range 11 – 27 g/kg).

There are two major implications of these findings. First, ignition events contribute almost all of the PM emissions during a burn and ignition related to a refueling event contribute more than the cold start. Although there was variation in the measured emissions, a good approximation of total PM emissions can be derived as a function of emission events rather than time integrated emission rates over the duration of the burn. Focusing on the number of cold starts and refueling events dramatically simplifies the process of estimating PM source terms related to stove use. The other

major implication of our findings is related to continued field monitoring efforts. Given results from the stove emission testing facility, it should not be necessary to monitor stove use continuously over a full day to get reliable estimates of PM emissions. Rather, one could design the monitoring study to focus primarily on ignition events (cold starts and refueling events), which could reduce the amount of time spend monitoring at a given site by up to 80%.

Additional experimental replicates are needed to reduce uncertainty in the estimates in emission factors and better characterize the variability in emissions related to different operational behaviors. Specific tests of a range of refueling behaviors and air intake settings could provide sufficient data, in combination with activity pattern data, to estimate source strength for pollutant emissions from the improved space heating stoves that are being deployed around Ulaanbaatar. This information could support air quality modeling and impact assessment to quantify the effectiveness of efforts to replace traditional stoves.

Introduction

Air pollution levels in Ulaanbaatar, Mongolia's capital, are among the highest in the world (World Bank, 2011). The primary source of particulate matter pollution in and around Ulaanbaatar is wind-blown dust and combustion products related to transportation, energy, and home heating/cooking (Davy et.al, 2011; Lodoyasamba and Pemberton-Pigott, 2011). The traditional coal-fired space heating stoves used in the Ger (tent) regions around Ulaanbaatar are a major source of particulate matter pollution during the winter months (Iyer et.al, 2010). Significant investment has been made to replace traditional space heating stoves with improved low-emission high-efficiency stoves. Testing protocols for selection of these high-performance heating stoves are often based on manufacturers' recommended operating procedures and/or for identifying ideal operating conditions (Pemberton-Pigott, 2011). These idealized test conditions demonstrate the stoves' optimal performance but may not be representative of true field performance and emissions.

Pemberton-Pigott (2011) summarized a large number of stove performance tests conducted at the SEET Laboratory in Ulaanbaatar noting that stove performance was impacted by a number of factors, including method of operation, i.e., how the stoves are actually being used versus recommended use. Lobscheid et al (2012) report that field operation of improved stoves varies significantly from manufacturers recommended operating method but emission measurements for the range of operating methods observed are not readily available. The combination of stove design, fuel characteristics, and operational factors, along with the inhomogeneous nature of the combustion process, leads to significant levels of variability and uncertainty in emission measurements. Therefore, in addition to performance testing conducted during stove development and roll-out, a combination of field and laboratory tests are needed to quantify emissions under actual operating conditions.

To address this need, Lawrence Berkeley National Laboratory designed and constructed a testing facility specifically for measuring emissions from space heating stoves under field-use conditions. The overall objective of the test facility is to identify and quantify the range of factors that influence emissions from space heating stoves under typical operating conditions as observed in Ulaanbaatar, Mongolia. Although the facility constructed at LBNL was designed to explore a range of factors that may contribute to emissions, the focus of this phase of the project is limited to differences related to startup and refueling scenarios.

Although stove operating procedures can include a continuum of conditions and behaviors they have been grouped into three main operating scenarios based on observations in the field (Lobscheid et al, 2012). These operating scenarios have been defined for convenience as 1) a cold start where the stove is empty and cold prior to loading and lighting the fuel; 2) a warm refueling where there may be embers present in the combustion chamber but they are not sufficient to ignite the coal so the fuel is lit by the operator (i.e., user-ignited refueling event); and 3) a hot refueling where sufficient embers and flames are present in the combustion chamber at the time of refueling to ignite the fresh fuel so coal is simply added to the stove (i.e., residual-fuel-ignited refueling event). Emission factors for these different scenarios are needed to reconstruct real-world emissions and to identify opportunities for additional improvement to stove design.

This report describes the LBNL test facility and summarizes initial results of emission testing for a Silver-mini (small Turkish) top-lit up-draft (TLUD) stove under several start-up and refuel scenarios using Nailaka coal from Ulaanbaatar. In addition to finding significantly elevated PM emissions during cold start and refueling events, we also found issues related to the characteristics of the fuel and the time when refueling occurred. Given time constraints, we included only open (proper) air intake settings, but we expect that different air inlet settings are used in the field and will influence emissions during a burn, leading to changes in PM emissions. We conclude with a summary of recommendations for future experiments that can be performed using the LBNL test facility to quantify actual emissions from improved space heating stoves and relate them to traditional stove emissions. We anticipate that this type of information will be valuable for modeling the impact and improvements related to the transition from traditional stoves to improved space heating stoves in Ulaanbaatar, Mongolia.

Materials and Methods:

The experimental apparatus and tests described in this report build on experience from a number of researchers (Iyer et.al., 2010; Pemberton-Pigott, 2011; Dalkia Energy Services, 2011) working with space heating stoves in the field and lab in addition to prior research at LBNL working with cooking stoves (Kirchstetter et.al., 2010). The main challenge to measuring emissions from space heating stoves is the extreme range of pollutant concentrations in the emissions stream during different phases of a burn. Additionally, reducing moisture in the sample lines to prevent condensation upstream of the analyzers and samplers poses additional challenges. The main tool for dealing with these challenges is dilution of the sample stream. By diluting the flue gas with a controlled amount of dry air, it is possible to simultaneously reduce humidity in the sample lines and dramatically increase the range of flue gas concentrations over which measurements can be made.

There are a range of options available for diluting and drying the gas from the chimney before analyzing. Gullett et al. (2003) and Purvis et.al. (2000) use a dilution tunnel approach for testing combustion appliances. In the dilution tunnel, the entire chimney gas stream is diluted with room air prior to sampling a portion of the diluted gas stream for analysis. For some experiments, an additional dilution step was used with cooled dilution air prior to sampling. Pettersson et.al (2010) and Bowman et al. (2005, 2011a, 2011b) used a full flow dilution tunnel where the chimney flow was diluted with filtered air and the entire exhaust stream was sampled continuously without further dilution. In both approaches, undiluted gas from the chimney is analyzed for carbon (CO_2 and CO), oxygen and sometimes other gas-phase pollutants where the undiluted gas is typically dried using inert drying agent prior to analysis. Iyer et.al (2010) sample directly from the chimney then use a metered amount of dry air to dilute the gas prior to analyzing. Pemberton-Pigott (2011) uses a similar dilution approach but the dilution occurs inside the chimney by injecting a metered amount of dry particle free air into the chimney in a way that the dilution air is drawn directly into a receiving tube so that a fixed amount of diluted, dry and cooled chimney gas is entrained in the sampling stream for analysis. The Pemberton-Pigott approach is used at the Sustainable energy Technology and Research (SeTAR) Centere and the Stove Emissions and Efficiency Testing (SEET) Lab in Ulaanbaatar.

Finally, it was necessary to simulate conditions in the field that influence the draft in the chimney and, subsequently, stove performance. Simulated chimney draft was needed to provide a

standardized testing protocol that was independent of meteorology and indoor/outdoor temperature gradients. The pressure at the top of the chimney was set to a predetermined value to ensure that the draft inside the chimney at ceiling height was representative of meteorological conditions during an Ulaanbaatar winter.

Experimental Apparatus

The experimental apparatus shown in Figure 1 and the schematic of the overall system shown in Figure 3 combine both the Pemberton-Pigott gas dilution approach and the larger dilution tunnel approach into a single test facility. A metal frame (Unistrut®) is constructed to mount the dilution tunnel directly over the stove so the chimney can be supported with cables to the base mounted on a scale (Cardinal model RW-1000) providing real-time mass measurements during emission testing. A rack for the instrumentation is setup to the left of the framework and protected by a double-wall fire curtain. Details of the different system components are described below.

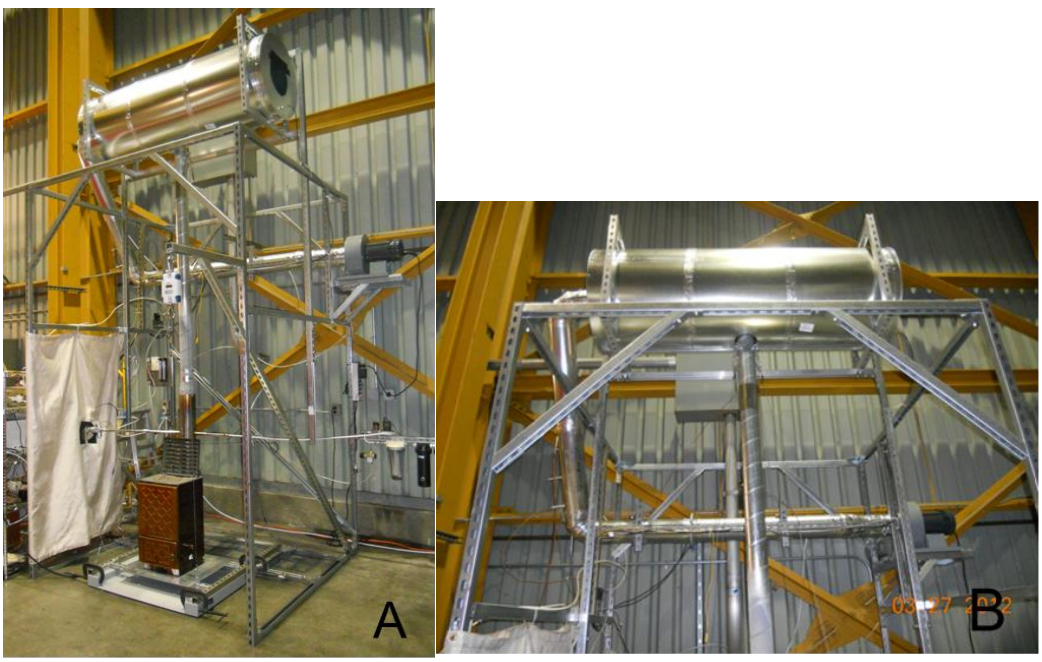

Figure 1: Panel A shows the complete system including stove mounted over a weight cell with chimney extended into the exhaust dilution tunnel then ducted down behind the apparatus to a ventilating exhauster. Panel B shows the detail of the upper dilution tunnel and exhaust ventilation ducting system. The instrumentation rack is mounted to the left behind the fire curtain and the particle free dry air is generated by a compressor mounted to the right of the system (not shown).

Exhaust management and chimney dilution: Smoke from the stove is vented to a 5 inch diameter by 24 inch tall stainless steel section of chimney shown in Figure 3. Two stainless steel sleeves are mounted opposite each other at 12 inches above the stove. Two half inch heavy-walled stainless steel pipes are mounted in the sleeves (adjustable) so they meet near the center of the chimney. The tips of the pipes are milled so that the pipe delivering particle free dry air has a small orifice and the receiving pipe has an inverted cone shape. The pipes are mounted approximately 1 mm apart to

allow chimney gas to be drawn into the sampling line. The difference in flow rate in the delivery line and the sample line are used to control the dilution. The design is based on the design used at the SEET lab (Pemberton-Pigoot, 2010) where chimney gas is rapidly extracted from the chimney and diluted with dry particle free air to prevent moisture/aerosol condensation in the sample line and to bring gas concentrations within operational limits of the analyzers. Particle free dry air is provided by a continuous flow of air from a compressor (Dewalt model D55146) that passes through a coalescing filter, two drying cartridges (Parker/Watt dryrite model DD15), a HEPA particle filter then a mass flow controller for continuous metered flow (Alicat 0-20 LPM).

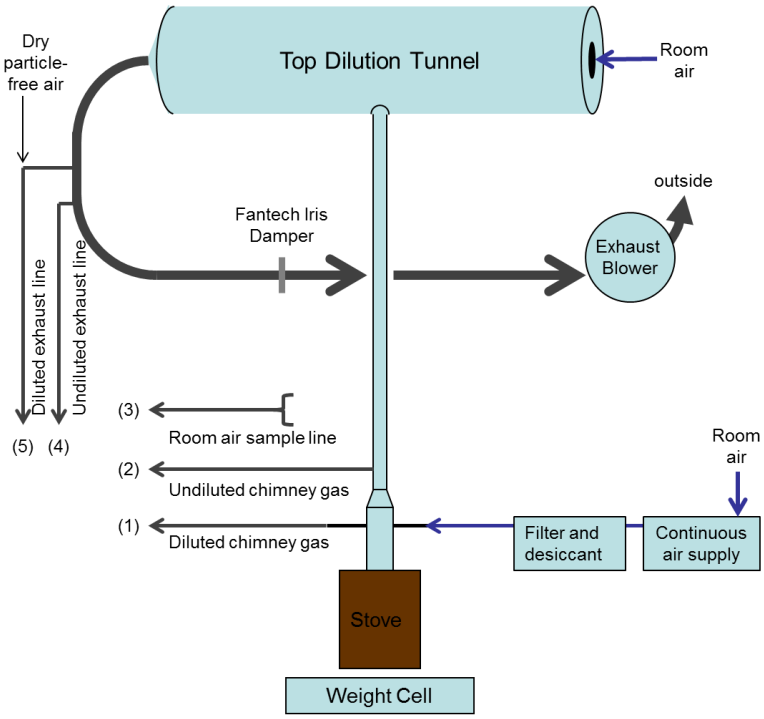

Figure 2: Schematic of LBNL Stove Testing Facility. The arrows indicate sampling lines as described in the text. The continuous supply of room air is provided by a compressor (Dewalt model D55146). Sample lines are exhausted outside along with the diluted stove emissions.

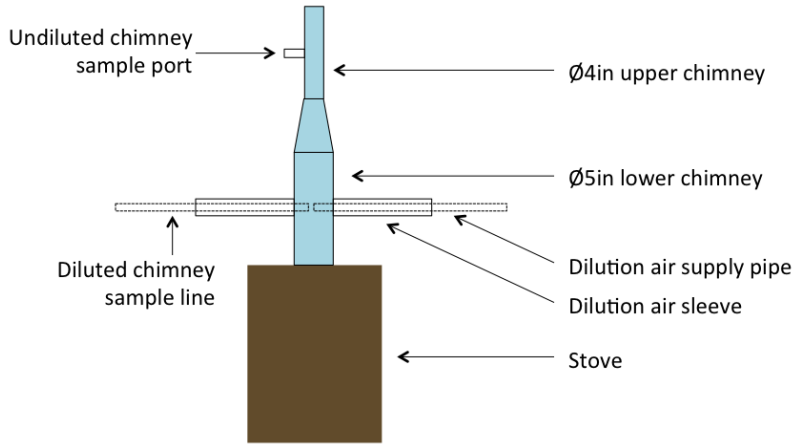

Figure 3: Detailed figure of the chimney sampling system

Above the 5 inch section of chimney pipe, the chimney diameter reduces to 4 inches and continues 113 inches above the stove. The top ten inches of the chimney is inserted through a 5 inch diameter hole into the exhaust dilution tunnel. The dilution tunnel consists of a 6-foot-long section of 24-inch-diameter duct with a 12-inch-diameter opening at one end to allow room air to enter the tunnel. The exhaust end of the tunnel is reduced to a 6-inch duct that transfers the diluted exhaust to the outside. The chimney is supported by guy wires and is not in contact with the dilution tunnel. The segregation of the stove and chimney system is important to allow for accurate real-time mass readings during stove testing. Room air is drawn into the dilution tunnel and mixed with the chimney gas before it enters the 6 inch exhaust duct. The 6 inch duct flows down and across behind the chimney through a calibrated FanTech iris damper to control and measure air flow using the pressure differential across the damper. Finally, the air flows through a fume exhauster (model M8-38-EP13 rated for 700 cfm continuous duty) vented through a spark arrestor screen to the outside of the building. The combination of exhaust flow rate (controlled by the iris damper setting) and the size of the opening to the dilution tunnel are used to set the vacuum above the chimney and subsequently the internal chimney draft (described below).

Sampling lines: There are five sample lines built into the system identified with numbers 1-5 in Figure 2. Each line is used for a different measurement using a range of instruments identified in Figure 4. The different sample lines allow measurement of diluted and undiluted gas from the chimney, diluted and undiluted gas from the dilution tunnel, and room air. In addition to the indicated sample lines in Figure 3, the temperature is measured continuously in the room, inside the chimney just above the stove, inside the chimney at ceiling height (8 foot off floor), and in the 6 inch exhaust duct downstream from the upper dilution tunnel. Pressure is measured in the chimney just above the stove, in the chimney at the ceiling height and in the exhaust duct on each side of the FanTech iris damper.

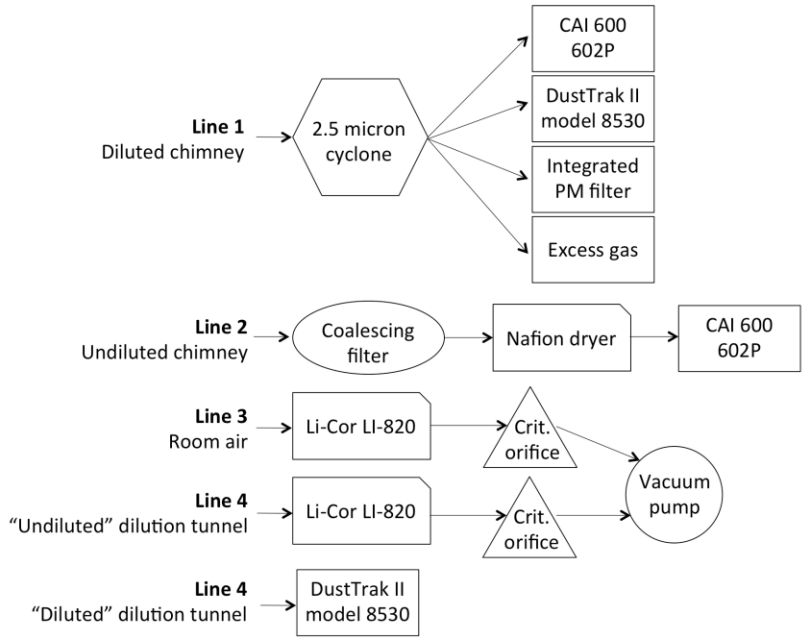

Figure 4: Sample lines and instruments

Instrumentation: The instrumentation and associated sample lines are illustrated in Figure 4. Sample line 1 provides diluted chimney gas that passes through a 2.5 micron cyclone before being split to a gas analyzer (CAI 600 Series Model 602P – $CO_2/CO/O_2$), real time PM mass sensor (TSI DustTrak II model 8530), an integrated PM mass measurement (25 mm Teflon filters) and excess flow for additional lines to mount other instruments or gas samples as needed. Sample line 2 is undiluted chimney gas drawn through a coalescing filter filled with glass beads to reduce static volume followed by a Nafion drying column (MD-110-125-4) with dry air counter current flow fed by a 0 – 16 LPM rotometer (Key Instruments) at ~ 5 LPM continuous then to a second gas analyzer (CAI 600 Series model 602P – $CO_2/CO/O_2$). Line 3 samples room air through a CO_2 analyzer (Li-Cor model LI-820). Sample line 4 is drawn directly from the exhaust after it exits the upper dilution tunnel and is sampled through a second CO_2 analyzer (Li-Cor model LI-820). The flow through both Li-Cor samplers is controlled by constant vacuum and critical orifice. Sample line 5 provides a secondary dry particle free source of air for additional dilution of the exhaust before running through a second real-time PM mass sensor (TSI DustTrak II model 8530).

Setting the Standard Chimney Draft: The goal is to set the draft (at ceiling height) that would occur if the stove and chimney (inside and outside) were cooled to a value that is representative of winter conditions in Ulaanbaatar. We assume that the typical chimney consists of a 10 foot section of 4 inch pipe extended 1 meter above the roof line (ceiling height of a Ger). The air inlet to the stove is open at ~ 6 inches above the floor. Indoor and outdoor temperatures are 20° and -20° C, respectively and a 5 meters-per-second wind speed at the top of the chimney.

The draft in this case is dominated by wind effect where the typical flue wind pressure coefficient is about -0.5 so the dP_{wind} is estimated as

$$dP_{wind} = 0.5\rho * Cp * V^2 \quad (1)$$

Where ρ is the density of air, C_p is the specific heat of chimney gases and V is wind speed at the top of the stack. The resulting draft at ceiling height for wind effect is approximately - 7.5 Pa. For stack effect we assume 1.45 m of chimney is indoors and 1 m is outdoors. With no fire, the temperature of air in the flue is the same as the room temperature therefore the inside section of chimney generates no stack effect. The outside section of chimney generates a draft caused by the temperature gradient calculated as

$$dP_{stack} = \rho \times g \times h \times dT/T \tag{2}$$

Where ρ is the density of air, g is acceleration of gravity (-9.8 m/s^2), h is the height of the section of chimney and dT/T is the temperature gradient relative to room temperature (K). The resulting draft for stack effect is approximately - 1.7 Pa. So the total draft (dP_{wind} + dP_{stack}) at ceiling height at startup is -7.5 - 1.7 = -9.2 Pa. We set the draft, or internal pressure, to a value between -7 and -10 Pa. The draft in the chimney is set by the negative pressure exerted at the top of the chimney, which is controlled by the velocity of the dilution flow air in the dilution tunnel and the size of the air inlet to the dilution tunnel. The velocity of the air flow through the dilution tunnel is a function of the diameter of the dilution tunnel opening and the mass flow rate provided by the exhaust blower and controlled by the iris damper.

Fuel Data Analysis: A domestic coal (Utah) was used initially to fire the stove before testing. The stove is fired by running through a complete burn cycle which burns off any residue or moisture on the stove that would affect total mass during subsequent runs. This is necessary to provide an accurate determination of fuel mass consumed during testing.

All emission tests were performed using Nailakh coal that was shipped to LBNL from Ulaanbaatar. The coal was packed in 25 gallon screw-top plastic drums. Each drum was opened and inspected on arrival at LBNL then allowed to air dry for approximately 48 hours to remove liquid water that had condensed in the containers during shipment. The barrels were then re-sealed and stored at ambient temperature until use. For the initial runs, the coal was scooped directly from the barrel to a weighing bucket. Consequently, changes in coal chunk size as the drum was used up were observed due to the finer material settling to the bottom of the barrel. We recognized that this change in the coal size caused changes in stove performance. To correct for this variation in later runs, the contents of each drum were transferred to a hopper (18 inches by 24 inches by 6 inches deep) and mixed prior to transferring to the weighing bucket. This provided for a more uniform size distribution of coal during testing. A sample of each coal was shipped to Geochemical Testing Environmental and Energy Analysis Lab for analysis. Results of the coal analysis are summarized in Table 1. The Utah coal is included only for comparison. The complete analyses are included in Appendix 1.

Table 1: Summary Results from Analysis of Coal Samples[1]

	Test Date		Nailakh 01/09/2012	Utah 1/30/2012
Proximate	Air Dry Loss	wt%	20.26	2.56
	Residual Moisture	wt%	8.23	1.20
	Moisture	wt%	26.82	3.7
	Ash	wt%	8.00	6.7
	Volatile Matter	wt%	27.81	39.1
	Fixed Carbon	wt%	37.37	50.3
Ultimate	Hydrogen	wt%	6.30	5.5
	Carbon	wt%	48.18	72.5
	Nitrogen	wt%	1.30	1.6
	Sulfur	wt%	0.64	0.32
	Oxygen	wt%	35.58	13.19
Other	Heating Value (Btu/lb)	Btu/lb	8305	13101
	Sulfur	lb/Btu	0.77	0.28
	Chlorine	mg/kg(dry)	100	447

[1] Complete results provided in Appendix 1.

The coal was used as received except that large coal chunks (> 3 inches rough diameter) were broken into smaller chunks prior to burning. We initially included coal powder in the testing but found that adding too much coal powder would plug the air intake ports at the bottom of the combustion chamber so that the coal either would not ignite during refueling or could not be ignited during initial cold starts.

Calculations and Variable Definitions: Many calculations used in this study have been summarized previously in Iyer et.al. (2010). We use actual measured fuel mass from the load cell during the burn. The chimney flow ($F_{_chimney}$) is calculated using the CO_2 balance knowing the total flow (FlowTech iris) in the dilution tunnel exhaust ($Q_{_exhaust}$,, m^3/h), and the concentration of CO_2 in the room ($C_{_CO2_room}$, mg/m^3), concentration of CO_2 in the undiluted chimney gas ($C_{_CO2_chimney}$), and concentration of CO_2 in the exhaust from the dilution tunnel ($C_{_CO2_exhaust}$) we calculate the chimney flow ($F_{_chimney}$, m^3/h) as

$$F_{_chimney} = \frac{Q_exhaust(C_CO2_exhaust - C_CO2_room)}{C_CO2_chimney - C_CO2_room} \quad (3)$$

To get emission rate (mg/hr) for each pollutant we multiply the undiluted chimney concentration (mg/m^3) by the chimney flow (m^3/hr). All reports of emissions concentrations (mg/m^3) herein are reported per volume of undiluted chimney air.

The undiluted chimney concentration of CO_2, CO, and O_2 are measurement directly in the chimney. We use a material balance on CO_2 to estimate the dilution ratio between diluted and undiluted chimney gas. The dilution ratio (*DR*) is used to calculate the undiluted chimney gas concentration of PM such that:

$$DR = \frac{Q2}{Q3} = \frac{C3 - C1}{C2 - C1} \quad (4)$$

where Q is flow (m^3/h) and C is concentration (ppm or mg/m^3) and the subscripts *1*, *2* and *3* refer to the room, chimney and diluted line, respectively. To get the undiluted $PM_{chimney}$ concentration from the measured $PM_{diluted}$ concentration would be:

$$PM_{chimney} = (PM_{diluted} - PM_{room})/DR + PM_{room} \tag{5}$$

The PM concentration in the room, PM_{room} was measured intermittently throughout the study with an average concentration of ~ 0.022 (mg/m^3).

The act of starting a fire in a cold stove and the act of refueling a stove with hot coal are classified as ignition events. An ignition event is defined to last from the moment the fuel is lit (starter wood ignition for startup and fuel addition for refuel) until the ratio of CO/CO2 decreases to less than 2%. These events are referred to as "startup" and "refuel" herein.

The rate of PM emissions (mg/hr) was calculated using the product of the measured chimney PM concentration and the calculated flow rate through the chimney.

$$PM_{chimney_rate} = PM_{chimney} \cdot F_{_chimney} \tag{6}$$

Total or net PM emissions (mg) during the test are determined by an integration of the PM emissions rate over time as shown in Equation 6. In all cases a numerical integration was performed with a time base of one minute.

$$PM_{chimney_net} = \int_{t_0}^{t_f} PM_{chimney_rate}\, \mathrm{dt} \tag{7}$$

The emissions factor for PM or CO (mg/kg or (g/kg)) was calculated per mass of combusted as-received coal and starter wood/paper, if applicable.

$$\mathrm{EF}_{PM} = \frac{PM_{chimney_net}}{m_{combusted}} \tag{8}$$

A time-resolved emission factor was not calculated because batch combustion of coal is not a homogeneous process and the interpretation of the time-resolved factor would change throughout stages of the burn.

Combustion Chemistry of Nailakh Coal: Let us consider the act of combusting 1kg of as-received Nailakh coal. We can imagine taking this coal and separating it into three distinct components: dry ash-free coal, ash, and water. The chemical formula for moisture and ash-free (MAF) coal (ignoring the contribution of nitrogen and sulfur) can be written by considering the mass fractions of carbon, hydrogen, and oxygen (from ultimate analysis) and their respective molar masses:

$$C_{\frac{73.92}{12}} H_{\frac{5.06}{1}} O_{\frac{18.05}{16}} N_{\frac{1.99}{14}} S_{\frac{0.98}{32}} \equiv C_{6.16} H_{5.06} O_{1.13} N_{0.142} S_{0.031} \equiv C_1 H_{0.821} O_{0.183} N_{0.0231} S_{0.005} \tag{9}$$

This result is relatively consistent with Nazaroff and Alvarez Cohen's (2001) finding that lignite coal has a chemical formula of:

$$C_1 H_{0.79} O_{0.073} N_{0.017} S_{0.008} \tag{10}$$

However, it is interesting to note that the oxygen content of the Nailakh coal is roughly twice the value of typical lignite coal. For ease of future calculations, we will ignore the contribution of nitrogen and sulfur and normalize the chemical formula of the Nailakh coal to one mole of oxygen:

$$C_1H_{0.821}O_{0.183}N_{0.0231}S_{0.005} \sim C_{5.46}H_{4.49}O \tag{11}$$

When written in this form, it can be said that this MAF coal has a "molar mass" of 86 g/mol. The mole fraction of carbon in this coal is (5.46)/(5.46+4.49+1)=0.499 and the mass fraction is (73.92)/(73.92+5.06+18.05)=76.2%. From analysis of the Nailakh coal, we know that the lower heating value of dry ash-free coal is 12742 BTU/lb or 29.6 MJ/kg. For every mole of dry ash-free carbon combusted, we can calculate the amount of heat released:

$$HR_{per_mol_carbon} = \left(29.6\ \frac{\text{MJ}}{kg_{MAF_coal}}\right)\left(0.086\frac{kg_{MAF_coal}}{mol_{MAF_coal}}\right)\left(\frac{1}{5.46}\frac{mol_{MAF_coal}}{mol_{carbon}}\right) = 0.466\frac{\text{MJ}}{mol_{carbon}} \tag{12}$$

The mass fraction of ash and water in as-received coal are 8% and 26.82%, respectively. Therefore, for every 1kg of as-received coal burned in the stove, we will burn 652g of dry ash-free coal, vaporize 268g of water, and leave behind 80g of ash. Assuming complete lean combustion of dry ash-free coal in the presence of dry atmospheric air and excess water, the following equation summarizes the combustion stoichiometry of burning one kilogram of as-received coal in dry atmospheric air along with a 26.82% mass fraction of excess water:

$$\begin{aligned} C_\alpha H_\beta O_\gamma + \frac{1}{\phi}\left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right)(O_2 + 3.76N_2) + \psi H_2O \\ \rightarrow \alpha CO_2 + \left(\frac{\beta}{2} + \psi\right)H_2O + \frac{3.76}{\phi}\left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right)N_2 + \left(\alpha + \frac{\beta}{4} - \frac{\gamma}{2}\right)\left(\frac{1}{\phi} - 1\right)O_2 \end{aligned} \tag{13}$$

Note: the ash is omitted from stoichiometric analysis because it does not combust, it is not a significant portion of the flue gas, and has relatively low thermal mass.

Where ϕ is the equivalence ratio (lean combustion occurs when $\phi<1$). In the case of our dry and ash-free coal, $\alpha=5.46$, $\beta=4.49$, and $\gamma=1$. The mole fraction of water (ψ) compared to dry ash-free coal is:

$$\psi = \frac{\frac{268g}{16\frac{g}{mol}}}{\frac{652g}{86\frac{g}{mol}}} = 2.2 \tag{14}$$

Therefore, we can rewrite the stoichiometric balance as follows:

$$C_{5.46}H_{4.49}O + \frac{6.08}{\phi}(O_2 + 3.76N_2) + 2.2H_2O \rightarrow 5.46CO_2 + 4.44H_2O + \frac{22.9}{\phi}N_2 + 6.08\left(\frac{1}{\phi} - 1\right)O_2 \tag{15}$$

A challenge arises when trying to handle the moisture content of coal during combustion: the mole fraction of excess water (ψ) in the coal is not constant throughout the burn. It is clear from Equation 15 that water vapor arises from both the combustion process and from vaporization of liquid water. It is possible for these two water vapor streams to be distinguished by using relative

humidity (RH) to measure the ambient and chimney humidity and to compare the extra water vapor in the chimney to expected values from combustion of MAF coal alone; any excess water will be due to vaporization of liquid water. However, without RH sensors in the instrumentation setup, we were unable to determine the full gas composition of the chimney stream. Without this information, we were unable to determine the time-resolved loss rate of liquid water from the fuel bed.

Experimental Design: The two primary objectives of this study were to develop the facility for testing emissions from space heating stoves with typical "real-world" operating conditions, and to use the test facility to explore different stove use behavior that influence PM emissions. As a result, we included only a limited number of replicate experiments in the initial set of experiments and focused on different combinations of use behaviors.

The emission tests were all performed with a Silver-mini (small Turkish) top-lit up-draft (TLUD) stove. The typical test included a cold start with approximately 10 kg of fuel followed by a refueling event with ½ the fuel used during startup. The refueling events were conducted at different stages of the burn with the earliest refueling event occurring as soon as the fuel bed collapsed and the latest refueling conducted while enough embers remained to ignite the coal. Some of the refueling and startup events used less than the 10 kg of coal to explore the impact on emissions during ignition. A summary of experiments is provided in Table 2.

Table 2: Summary of Experiments

Test Name	Stages of burn	Description of test
	Cold-start	Typical mass of fuel at both ignitions
20-Mar-12	Early hot-refuel	Hot refueling performed just after fuel bed collapse
		Typical mass of fuel at startup
	Cold-start	Extra fuel used at refueling
21-Mar-12	Late hot-refuel	Hot refueling performed late in run
	Cold-start	Typical mass of fuel at both ignitions
27-Mar-12	Late hot-refuel	Hot refueling performed late in run
		Stove did not start initially.
		Typical mass of fuel used at startup
28-Mr-12	Cold start	No refuel event
		Light load of fuel used at startup
		Typical mass of fuel used at refueling
	Cold-start	Refueling performed late in run
13-Jun-12	Late hot-refuel	Only exhaust dilution line used in PM sampling

Results

The results are first presented graphically to illustrate the different stages of the burn for each experiment. These results are quantitatively described and interpreted in the Discussion section.

Figure 5 illustrates results from 21-Mar-12. Each panel in Figure 5 provides different information about the conditions of the burn and/or emissions during the burn as described below. We noticed some inconsistencies with the PM sampling from the dilution line coming from the chimney so all PM data presented in the burn panels provided in Figs. 5-9 are based on the sampling line from the

exhaust dilution tunnel. All data is provided in the summary table (Table 2) that follow but we assign more confidence to the PM data from the exhaust dilution tunnel. The exhaust dilution data are presented in the detailed burn panels.

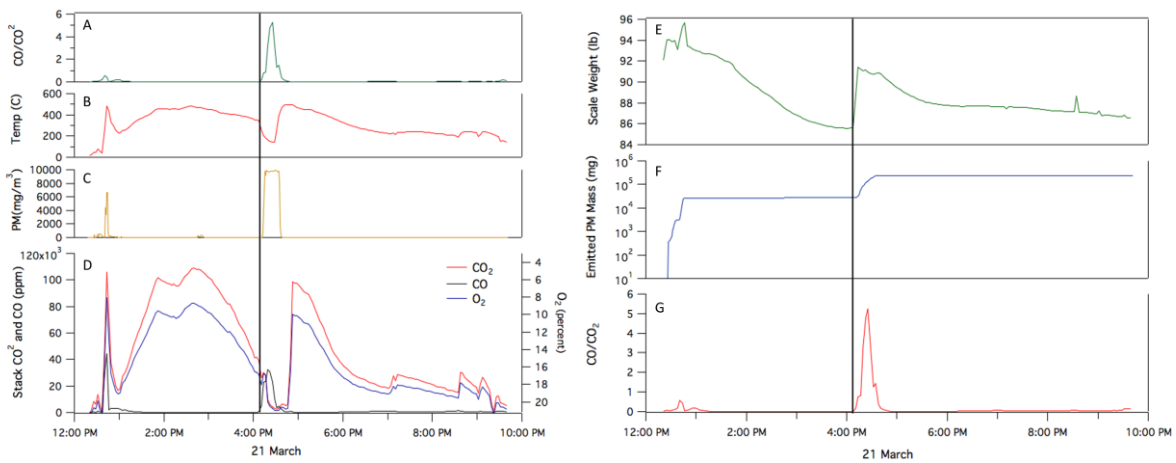

Figure 5 Detailed burn results illustrated for experiment 21-Mar-12.

Referring to Figure 5, the different panels in the figure include the following information.

- Panel A is the CO/CO_2 ratio which is an indication of the stage of the burn. At ambient levels, the CO/CO_2 ratio is below ~ 1% but during ignition the levels will exceed 2% then fall below 2% during the active phase of the burn rising above 2% again late in a burn. The ratio can exceed 10% when the oxygen levels fall either because of poor air flow into the combustion chamber or excess volatilization of moisture, both of which can starve the fire for oxygen.
- Panel B shows the temperature profile inside the chimney measured at the ceiling height (8' off floor). This value is used to calculate stove efficiency but is also an indication of the stage of the burn clearly showing the time of startup and refueling events. A spike in stack temperature always occurs when the wood is first ignited during a cold start. During a refueling event using Nailakh coal, which has high moisture content, we always see a sharp cooling of the chimney gas that corresponds with a spike in the CO/CO_2 ratio caused by the evolution of water vapor. Once the water vapor is gone we typically see a rapid increase in stack temperature as the fresh coal is ignited and volatile organic compounds (VOCs) are burned off.
- Panel C reports the time resolved concentration of PM2.5 measured in the chimney gas. This value is used along with the chimney gas flow rate to estimate an emission factor.
- Panel D shows the time series for CO_2, CO and O_2. The oxygen is reported on the secondary axis with the values plotted in reverse order showing that O_2 is inversely correlated with CO_2. We generally see a spike in CO_2 and CO during cold start with a drop in O_2 as the wood is ignited lasting until the wood is consumed. As the coal lights, the levels of CO_2 again increase with a corresponding drop in O_2 and CO until late in the burn when the CO_2 drops and the O_2 and CO increase again. During refueling events we typically see a drop in CO_2 with associated increase in CO until the coal begins to ignite when CO_2 rapidly increases until the fuel bed is fully lit. The spikes that occur late in the burn are from agitating the grate in the combustion chamber to open the air intake holes.

- Panel E shows the total mass of the stove and fuel during the course of the burn. The spikes that sometimes occur at the beginning of a burn or during refueling are artifacts caused by manipulating the lid when monitoring the status of the burn.
- Panel F reports the integrated PM2.5 mass released during the burn. The mass is plotted on a log scale. The step-wise shape of the curve highlights the contribution to PM emissions during the ignition stages of the burn. The initial spike of PM during the cold start ignition is primarily from the wood and the subsequent increase in PM mass related to coal ignition. During refueling, the entire PM mass loading is due to ignition of coal.
- Panel G is a repeat of the CO/CO_2 ratio for reference.

The burn panels for the remaining experiments are illustrated in Figures 6-9 for experiments 20-Mar-12, 27-Mar-12, 28-Mar-12 and 13-Jun-12, respectively.

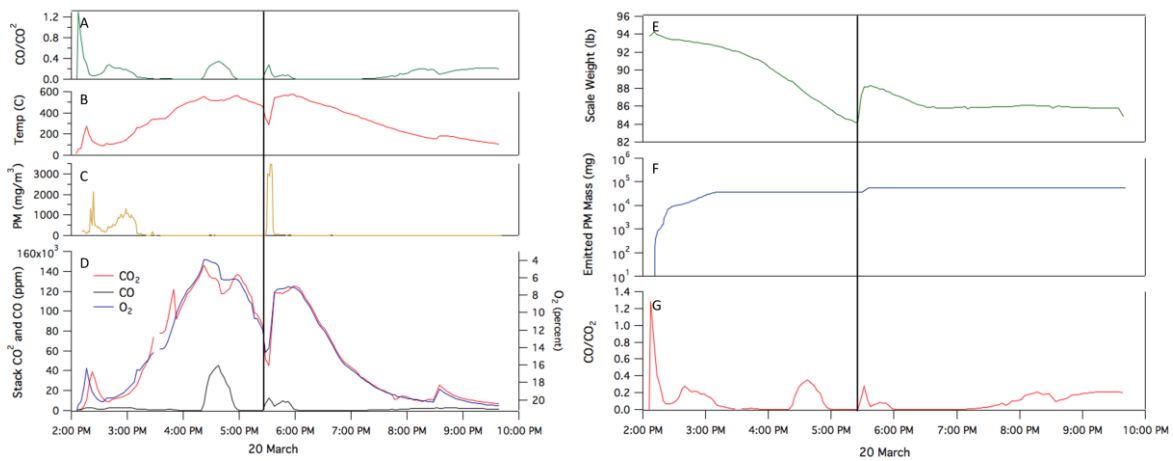

Figure 6 Burn data panels for experiment 20-Mar-12.

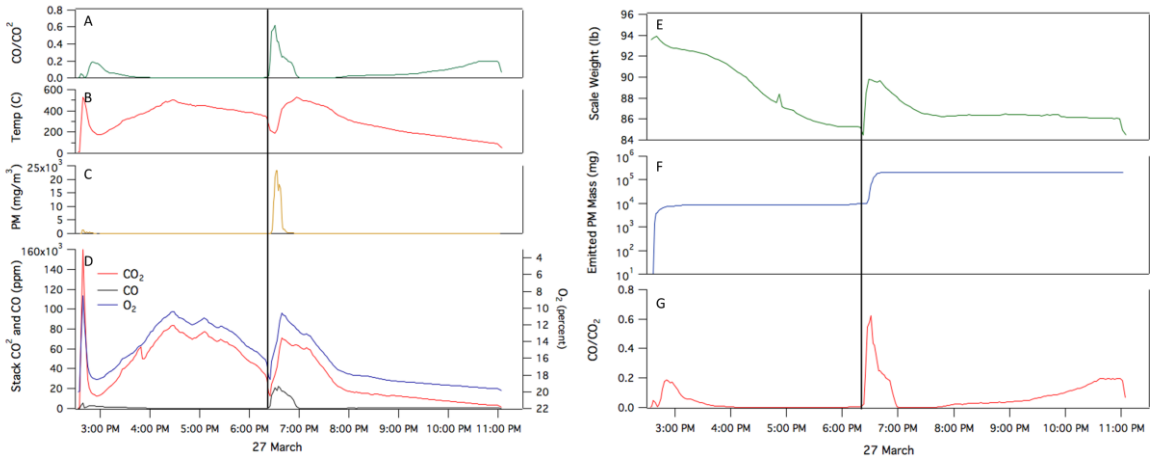

Figure 7 Burn data panels for experiment 27-Mar-12.

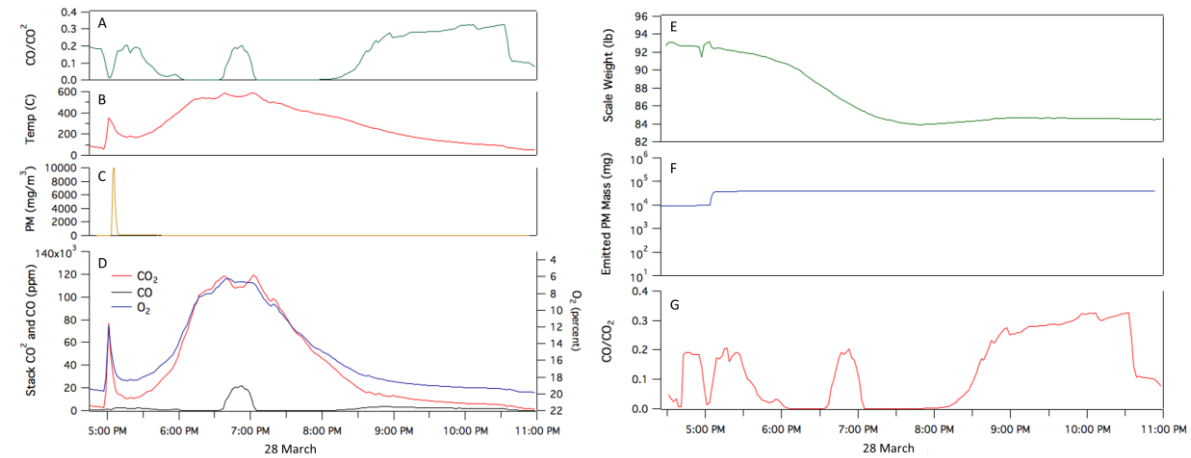

Figure 8 Burn data panels for experiment 28-Mar-12.

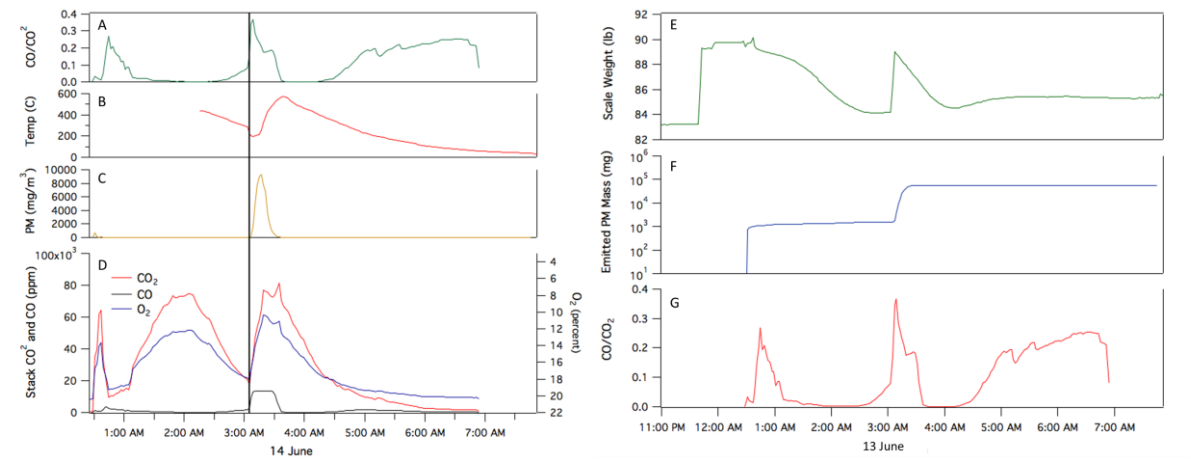

Figure 9 Burn data panels for experiment 13-Jun-12.

The 20-Mar-12 experiment (Figure 6) produced an extended period of emissions during the initial cold start compared to the other experiments. This extended emission period was due to difficulty in getting the coal to light. As was typical with all the burns, the initial spike in emissions was from ignition of the wood followed by a somewhat longer period of lower emissions related to the coal ignition. We also had trouble lighting the coal on 28-Mar-12 (Figure 8) because the coal used on that day was from lower in the storage barrel and had a larger fraction of coal powder. We surmise that when the fine coal was poured into the combustion chamber it restricted air flow through the holes at the bottom of the chamber preventing the coal from lighting. Ultimately, we removed all the coal and re-loaded the stove with coal from a fresh barrel that had been mixed in a hopper before use distributing the fines more evenly throughout the fuel. The unsuccessful ignition events warmed the stove but did not light the coal so when the stove was re-filled and lit, the results simulate what would be seen with a warm start although no ash or embers were present in the stove at the time of ignition.

Although the temperatures at the time of refueling were not significantly different between the runs, the experiment on 20-Mar-12 (Figure 6) included a refuel at the earliest time possible which was

shortly after the bed of coal had collapsed, the mass of coal in the stove was still dropping, and the temperature in the stove was higher than the other refueling events. The other re-fuel events were all conducted late in the run when the remaining embers in the combustion chamber were judged to be just sufficient to ignite the fresh fuel added during refueling. The difference between these two scenarios appears to be a faster ignition for the earlier hot-refuel as compared to the later hot-refueling. Other than the pseudo warm start from the unsuccessful ignition in 28-Mar-12 run (Figure 8), we did not conduct any actual warm refueling events that required reigniting of the fuel from the top while ash and embers were present at the bottom of the fuel bed.

The largest emission event occurred during the hot-refueling on 21-Mar-12 when the measured concentration exceeded the capacity of the instrument as indicated by the flat top on the large peak in Panel B of Figure 5. For subsequent runs, we used a larger dilution flow during ignition events to prevent the instruments from going out of range. Also during this run we had a series of failed ignitions that produced emissions from only the wood. We found that the majority of PM mass emitted during cold-start ignition was from wood where the coal ignition was typically much lower with the only exception being the 20-Mar-12 burn (Figure 6).

Discussion

The primary purpose of this report is to explore how refueling scenarios for an improved stove impacts emissions during a startup and operation. Namely, we are interested in knowing whether or not refueling the stove has a more negative impact on emissions than lighting it and burning from a cold start. For interpretation of results, we divide the total burn cycle (cold start to dead out) into either ignition or burn phases. The ignition phase lasts from the time when the fuel is ignited either by the operator (cold start) or by remaining flames/embers in the fire box (hot start) until the CO/CO_2 ratio drops below 2%. The burn phase includes the time between ignition and either a refuel event or the end of the test. The amount of time in each phase for each test is shown in Figure 10 along with the mass of fuel added for each start and refuel event. A refuel was performed in all tests except the 28-Mar test. The mass of coal fuel added during cold starts ranged between 6 kg and 10.75 kg with an additional amount of wood and paper starter between 0.3 kg and 0.5 kg. The mass of fuel added during refuel events ranged from 4.6 kg to 6.8 kg of coal.

In all instances, the net fuel consumed during the ignition phases (startup or refuel) was significantly less than the fuel consumed during the remainder of the test as illustrated in Figure 11. There was no correlation between the amount of fuel consumed during an ignition phases and the amount of fuel loaded in the fuel prior to the phase. In addition, there was no difference between the amount of fuel consumed during cold start and refueling events. For cold starts, the average fuel consumed (± standard deviation) for all runs was 1.9 ± 0.9 kg and for the refueling events was 1.8 ± 0.6 kg.

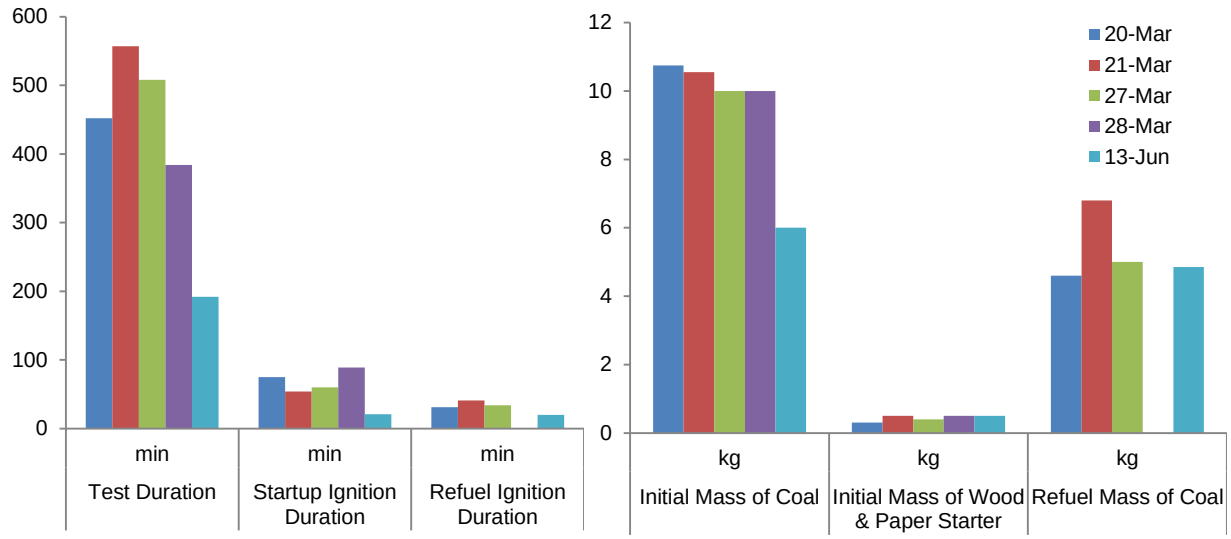

Figure 10: The duration of tests and ignition periods are shown in the left panel and the amount of fuel added during each phase of the burn is shown in the right panel.

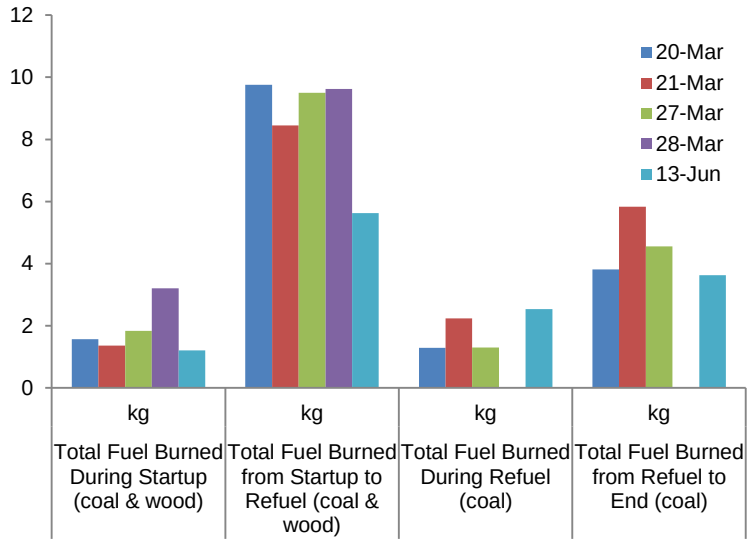

Figure 11: Fuel Consumption during the startup ignition phase, the refuel ignition phase, and the remaining burn period.

The consumption of fuel occurs in a non-linear fashion and the behavior of fuel consumption is distinct between the startup and refuels as shown in Figure 12. During startup, the shape of the fuel bed mass vs. time curve is convex with a negative second time derivate behavior. This behavior indicates a slow initial drive off of fuel and excess water, followed by a rapidly increasing rate of fuel consumption and water vaporization as the temperature in the fire box increases. This phenomenon is expected given the combustion nature of a TLUD device where a downward-migrating combustion front vaporizes water that is within thermal reach of the combustion front, and as the combustion front gains power, its thermal reach and fuel consumption rate increase. In contrast,

the mass consumption behavior of a refuel event is quite different. A refuel event display's a positive second time derivative behavior with a large fraction of the refuel mass being combusted or vaporized within a short timeframe. This again corroborates our intuition where pouring cold and wet fuel onto a hot fuel bed will cause the excess water in the fuel to rapidly vaporize and the fuel to combust as hot combustion gases from the already-lit lower fuel bed rise through the new cold refuel bed.

The fuel consumption follows a similar trend for all burns with the difference in curves related to the time of refueling and the amount of fuel added. The 13-Jun (Figure 9) run started with about half the normal amount of fuel so the refueling event was earlier in the run compared to the other experiments. In addition, for the 13-Jun run, less initial mass during cold start corresponded to less ash and residue in the fire box at the time of refuel allowing for greater air supply during and after refueling leading to the higher rate of fuel consumption after refueling compared to the other tests.

The slight rebound in mass that was observed towards the end of the runs in Figure 12 may have been due to temperature effects on the scale. Future runs should include better shielding for the scale to prevent the temperature from affecting the readings.

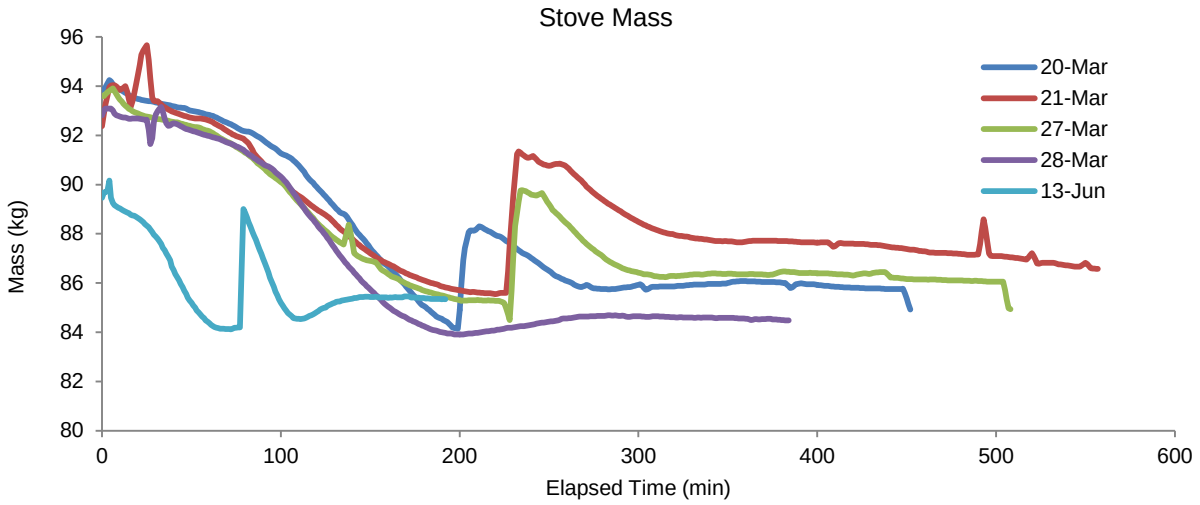

Figure 12: The mass of stove and fuel over each burn.

The temperature of the stove during the tests is plotted in Figure 13. Temperature was measured with a thermocouple mounted in the chimney just above the stove. There is typically an initial spike in temperature corresponding to ignition and combustion of wood followed by a steady increase in temperature as the coal ignites. We observed a dramatic decrease in temperature upon introduction of cold and wet fuel during refuel ignition events. This cooling is due to the evaporation of water from the fuel and the combination of reduced temperature, excess water vapor and reduced oxygen may contribute to increased PM and CO emissions during refuel. The duration of the dip in temperature depends on the temperature in fire-box when refueling occurred where earlier refueling had a higher temperature and shorter period of depressed temperature compared to later refueling. However, the duration of the ignition period (value reported in Table 2) did not correlate with the temperature of the stove at refueling.

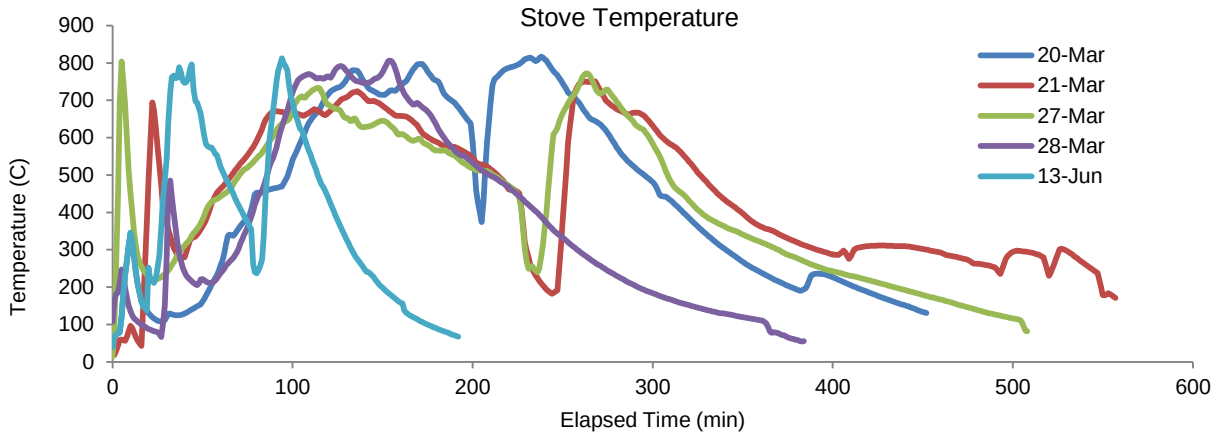

Figure 13: Stove Temperature vs. Time

Total PM and CO emissions increase in a nearly stepwise fashion during ignition events (Figure 14). This effect is likely due to lowered combustion temperatures owning to the thermal mass of the fuel bed and the energy required to vaporize liquid water in the fuel. CO, however, has an additional increase in emission rate when the fuel remaining in the fuel bed becomes small compared to the initial or refuel load. This effect of increased CO with diminishing fuel could be related to ash and residue buildup around remaining fuel, and/or blockage of air inlet pathways into the combustion box as dense layers of ash and coke accumulate on the grate, both of which would decrease fuel/air mixing late in the run. During the 21-Mar run, the grate was agitated vigorously late in the run to maintain open air intake ports remove loose ash from the fire-box and this seemed to reduce the CO emissions relative to the runs where the fires was allowed to go out naturally. This supports the premise that reduced fuel/air mixing can lead to higher CO emissions during burn-out.

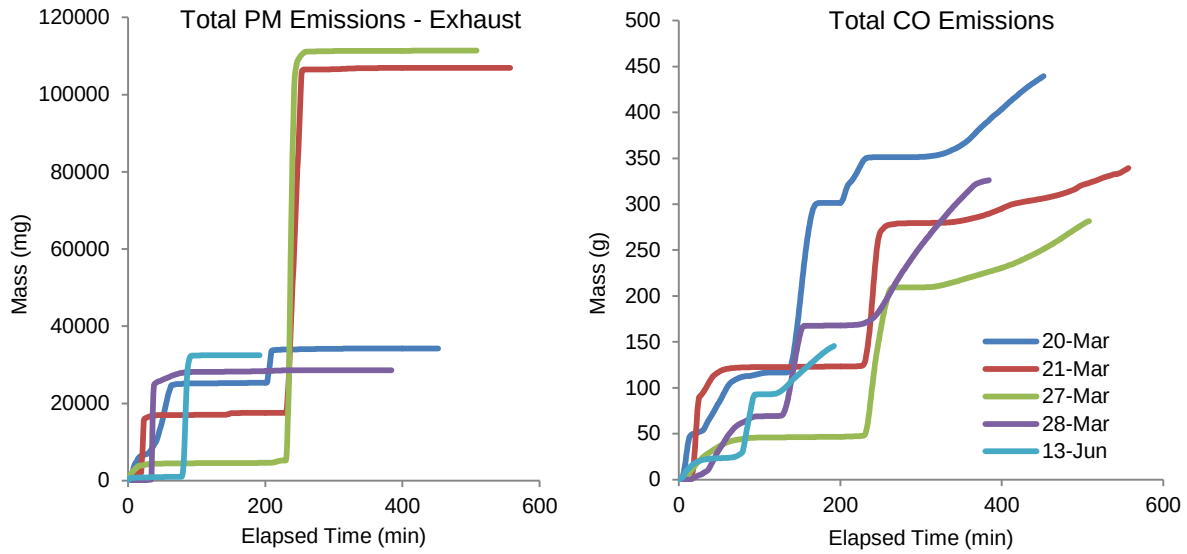

Figure 14: Total emissions of PM (left panel) and CO (right panel) vs. time for each test

The stepwise behavior of PM and CO emissions during ignition events translates to large fractions of total emissions occurring within the relatively short timeframe of ignition. In the cases where refueling events occurred, total PM and CO emissions were higher during refuel than during startup except in the case of the 20-Mar test. This is potentially because the 20-Mar refueling occurred earlier in the burn and at a stove temperature that was more than 250 C° higher than the other tests. The higher temperatures (shown in left panel of Figure 15) may have helped more-fully combust particles and oxidize CO and more rapidly achieve the efficient burn phase compared to the later refueling. Another difference between cold start and refuel events is that a significant portion of PM emissions during cold start is from wood/paper combustion while the refuel events are entirely related to coal combustion. This is easiest to see in Figure 8 for the 28-Mar test which shows that the initial PM emissions occur during wood combustion and the lower slope for the 28-Mar PM emission mass in Figure 14 is from ignition of coal. Additional testing would be needed to conclusively separate emission from wood and coal during the cold start but these results highlight the opportunity for reducing emission through improved startup procedures.

In addition to the finding that ignition events during refuel have higher PM emissions than cold starts, another significant observation is that 99% ± 0.4% of PM emissions occur during the relatively short time period related to ignition of any kind as illustrated in Figure 15 and Figure 16. Considering the contribution of wood/paper to PM emission during cold start ignition, ignition during refuel events is clearly a significant contribution to PM during a burn cycle for the improved space-heating stoves.

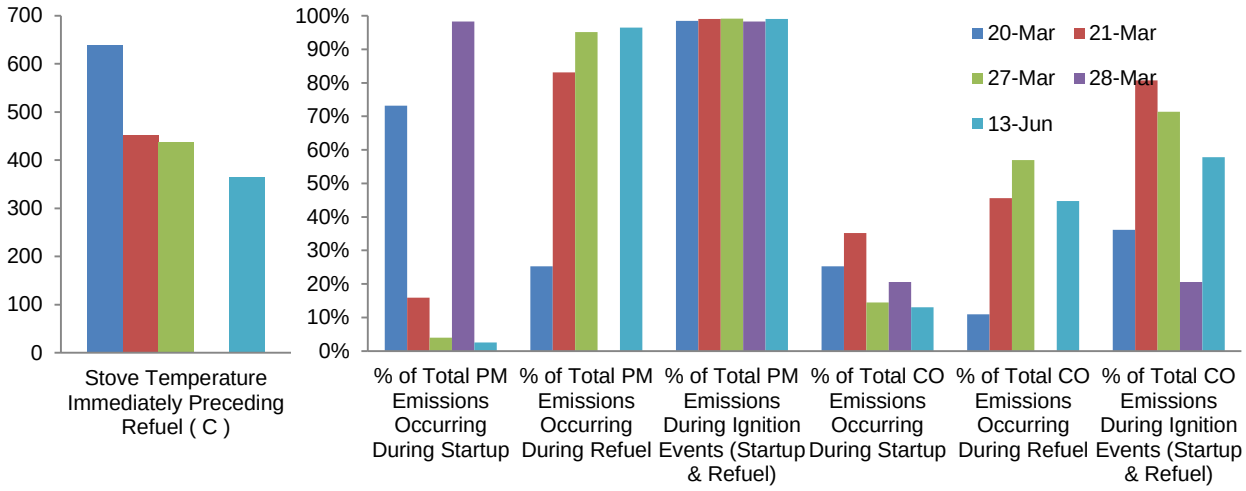

Figure 15: Refuel temperature (left panel) and % emissions during each ignition event

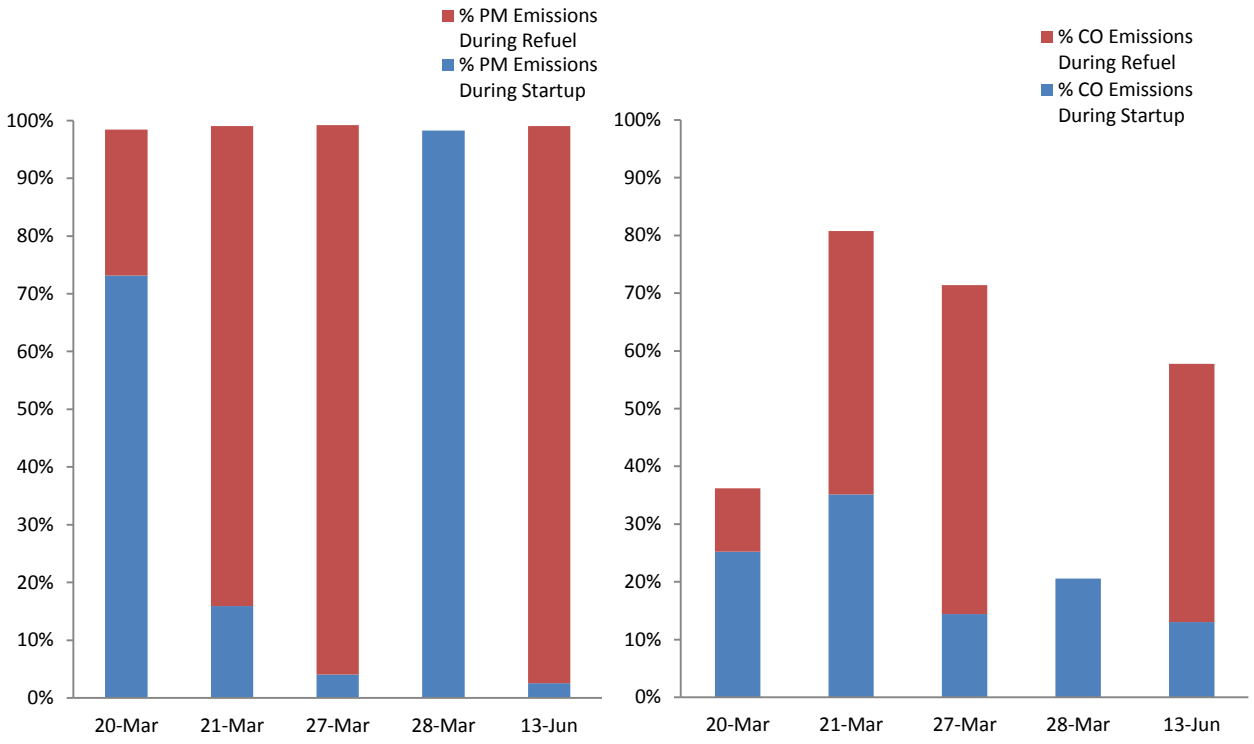

Figure 16: Stack bar presentation of percent PM and CO emissions that occur during ignition events including both cold start and refueling.

All relevant data from the experiments along with calculated emission rates are summarized in Table 2. The first section of the table reports information related to fuel addition and consumption along with duration of test and ignition events. PM data are reported in the second section of the table for both the chimney dilution sample line and the exhaust dilution tunnel sample line during each test. As indicated earlier, we observed several anomalies with the chimney dilution sample line during the testing and as a result, we do not consider those values to be reliable. They are reported only for comparison and completeness. Several times we noted a significant drop in the measured concentration in the chimney dilution line. The concentration measured in the chimney dilution line converted to chimney concentration was always lower than the value measured with the exhaust dilution tunnel sample line. This occurred even after switching the DustTrak particle sampler used on the two lines. The gas phase measurements from the chimney dilution line did not indicate complete plugging of the line but particles may have been lost to the walls of the collection cone in the chimney or the sample transfer line from chimney to instrument. Additional testing would be needed to determine why the chimney dilution line was under-estimating the chimney concentration of PM during the testing.

Time resolved emission factors are not reported because the batch combustion of coal is not a homogeneous process and the interpretation of the time-resolved factor would change throughout stages of the burn. In addition, we found that that 99% of PM emissions (by mass) occur during ignition so we suggest that a more relevant statement of PM emissions may be on mass per ignition event basis. As such, for the stove and conditions in this study, we found PM2.5 emission rate of 15

grams per cold start event (± 80% coefficient of variation) and 60 grams of PM2.5 per refuel event (± 80% coefficient of variation).

Table 2: Summary of Test Data

Metric		Unit	20-Mar	21-Mar	27-Mar	28-Mar	13-Jun
Test Duration		min	452	557	508	384	192
Initial Mass of Coal		kg	10.75	10.55	10.00	10.00	6.00
Initial Mass of Wood & Paper Starter		kg	0.30	0.50	0.40	0.50	0.50
Refuel Mass of Coal		kg	4.60	6.80	5.00	0.00	4.85
Total Fuel Burned During Startup (coal & wood)		kg	1.57	1.37	1.83	3.21	1.21
Total Fuel Burned from Startup to Refuel (coal & wood)		kg	9.76	8.45	9.50	9.62	5.63
Total Fuel Burned During Refuel (coal)		kg	1.29	2.24	1.31	0.00	2.54
Total Fuel Burned from Refuel to End (coal)		kg	3.81	5.83	4.56	0.00	3.63
Startup Ignition Duration		min	75	54	60	89	21
Refuel Ignition Duration		min	31	41	34	NA	20
Stove Temperature Immediately Preceding Refuel		C	638	451	438	NA	364
Total PM Emissions During Full Test	chimney	mg	2.0E+04	3.3E+04	3.8E+02	3.2E+03	NA
	exhaust	mg	3.4E+04	1.1E+05	1.1E+05	2.9E+04	3.3E+04
Net PM Emissions During Startup	chimney	mg	1.9E+04	1.2E+04	0.0E+00	3.1E+03	NA
	exhaust	mg	2.5E+04	1.7E+04	4.5E+03	2.8E+04	8.4E+02
PM Emissions During Refuel	chimney	mg	5.1E+02	2.1E+04	3.8E+02	0.0E+00	NA
	exhaust	mg	8.6E+03	8.9E+04	1.1E+05	0.0E+00	3.1E+04
PM Emissions During Ignition Events (Startup & Refuel)	chimney	mg	2.0E+04	3.3E+04	3.8E+02	3.1E+03	NA
	exhaust	mg	3.4E+04	1.1E+05	1.1E+05	2.8E+04	3.2E+04
PM Emissions Rate During Startup	chimney	mg/hr	1.5E+04	1.3E+04	0.0E+00	2.1E+03	NA
	exhaust	mg/hr	2.0E+04	1.9E+04	4.4E+03	1.9E+04	2.2E+03
PM Emissions Rate During Refuel	chimney	mg/hr	9.5E+02	3.0E+04	6.5E+02	0.0E+00	NA
	exhaust	mg/hr	1.6E+04	1.3E+05	1.8E+05	0.0E+00	9.0E+04
PM Emissions Rate During Ignition Events (Startup & Refuel)	chimney	mg/hr	1.2E+04	2.0E+04	2.4E+02	1.9E+03	NA
	exhaust	mg/hr	1.2E+04	6.6E+04	6.8E+04	1.6E+04	1.0E+03
PM Emission Factor (PM emissions/fuel consumed) During Full Test	chimney	mg/kg	1.4E+03	2.3E+03	2.7E+01	3.3E+02	NA
	exhaust	mg/kg	2.5E+03	7.5E+03	7.9E+03	3.0E+03	3.5E+03
PM Emission Factor (PM emissions/fuel consumed) During Startup	chimney	mg/kg	1.2E+04	8.7E+03	0.0E+00	9.8E+02	NA
	exhaust	mg/kg	1.6E+04	1.2E+04	2.4E+03	8.8E+03	6.9E+02
PM Emission Factor (PM emissions/fuel consumed) During Refuel	chimney	mg/kg	3.9E+02	9.4E+03	2.9E+02	NA	NA
	exhaust	mg/kg	6.7E+03	4.0E+04	8.1E+04	NA	1.2E+04
% of Total PM Emissions Occurring During Startup	chimney	%	96.9%	35.9%	0.0%	98.1%	NA
	exhaust	%	73.2%	15.9%	4.0%	98.3%	2.6%
% of Total PM Emissions Occurring During Refuel	chimney	%	2.6%	63.7%	99.4%	0.0%	NA
	exhaust	%	25.3%	83.1%	95.2%	0.0%	96.5%
% of Total PM Emissions During Ignition Events (Startup & Refuel)	chimney	%	99.5%	99.7%	99.4%	98.1%	NA
	exhaust	%	98.5%	99.0%	99.2%	98.3%	99.1%
Total CO Emissions During Test		g	439	339	282	326	145
CO Emissions During Startup		g	111	119	41	67	19
CO Emissions During Refuel		g	48	155	160	0	65
CO Emissions During Ignition Events (Startup & Refuel)		g	159	274	201	67	84
CO Emissions Rate During Startup		g/hr	88	130	40	45	50
CO Emissions Rate During Refuel		g/hr	91	221	276	0	189
CO Emissions Rate During Startup & Refuel		g/hr	61	170	124	53	26
% of Total CO Emissions Occurring During Startup		%	25.2%	35.2%	14.4%	20.6%	13.1%
% of Total CO Emissions Occurring During Refuel		%	10.9%	45.6%	57.0%	0.0%	44.7%
% of Total CO Emissions During Startup & Refuel		%	36.2%	80.8%	71.4%	20.6%	57.8%

Conclusion

A carefully designed and constructed stove testing facility at Lawrence Berkeley National Laboratory was used to measure the impact of operational behavior on emissions for an improved space heating stove from Mongolia. Field evidence suggests that the improved stoves are not being used according to manufacturer's recommendation and that variation in startup and refueling practice could lead to significantly elevated emissions. We tested a Silver-Mini (small Turkish) top-load up-draft space heating stove and found that PM emissions occurred almost entirely during ignition events (i.e. when the fuel is lit by the operator or by residual embers/flame in the fire box) while CO emission were more distributed over different phases of the burn. For the ignition events, we found that refueling produced up to five times more PM emissions than initial cold-starts with the one exception being an early refueling when the temperature in the fire-box was much higher. For the cold-starts, it appears that the majority of PM is from ignition and combustion of wood and paper with only a moderate amount of PM from the ignition of coal. However, for the refueling events, all of the PM emissions are from the coal.

PM emission factors integrated over the entire burn range from 3 – 8 grams per kg fuel consumed while emission factors for the ignition period range from 1 – 16 g/kg fuel consumed for cold-start conditions and from 7 – 81 g/kg fuel consumed for refuel events. The duration of ignition phases varied from run to run (range 21 – 89 minutes for cold starts and 20 – 41 minutes for refueling events). In addition, while fuel consumption was measured directly during this study, measuring fuel consumption in the field during different phases of a burn are not feasible. Fortunately, emissions are almost entirely from ignition events and are relatively consistent regardless of the amount of fuel lit, the duration of the ignition phase for the conditions tested in this study. Therefore, we recommend reporting emissions per ignition period which could greatly simplify the calculation of source terms for air quality modeling. We found that cold-start ignition events produced approximately 15 grams of PM (range 1 – 30 g) while refuel ignition events produced approximately 60 grams of PM (range 9 – 110 g).

Uncertainty and Variability: The large variance (standard deviation or coefficient of variation) in the results is due to the relatively small number of experiments in which we combined several factors that influence emissions. The variance could be reduced with experiments that focus on a small number of covariant factors. For example, we combined differences in operational behaviors with differences in fuel characteristics (size, moisture). We also used different mass of coal loading and varied the amount of ash removed from the fire-box prior to refueling. In addition, the variance may be inflated because it includes both variability (heterogeneity across different conditions) and uncertainty (lack of true knowledge about factors influencing the outcome). In short, uncertainty is the component of variance that can be reduced with additional and/or improved measurements while variability can only be better characterized through additional measurements.

The lack of experimental replicates limits our ability to provide a reliable estimate of uncertainty but several qualitative statements of uncertainty can be made along with opportunities to improve the experiments. First, there is an opportunity to reduce uncertainty in the estimates of emission rates by confirming the calibration of flows with particular focus on the iris flow control valve in the exhaust line. We used the calibrated values that came from the manufacturer to interpret flows in the exhaust dilution tunnel, which was in turn used to estimate chimney flow based on the carbon mass balance.

While the manufacturer's calibration data is likely to be correct, the value should be confirmed using an alternative duct flow measurement device or approach. Second, CO_2 measurements in the undiluted chimney gas exceeded the highest calibration gas available at the time of the tests. Several tests were conducted to confirm that the concentration/response relationship was linear but future tests should use a higher calibration gas concentration to insure accurate measurements of CO_2 in the un-diluted chimney gas, which is used in the carbon mass balance to estimate dilution rates in the chimney dilution line and in the exhaust dilution tunnel. The higher concentration calibration gas was available and used for the last test. Third, during the early tests, we were simultaneously constructing and verifying performance of the test facility and measuring emission performance from the stove. As we used the facility and gained experience, we identified opportunities to improve the design, for example, initial runs used a rotometer flow control for dilution flow that was subsequently changed out for a mass flow controller providing more precise controls in the dilution line. These improvements and experience simplified the operation, reducing the chance of making an error during the burn thereby reducing uncertainty in results. Finally, calibration of the DustTraks using integrated filter samples could reduce uncertainty in the PM emission estimates. We did include filter measurements but it took several experiments to work out the sample time, duration and volume to prevent filter overload and provide reliable measurements. Future work should include targeted filter measurements to allow for source specific calibration of the DustTraks as described in Kirchstetter et.al. (2010).

Recommendations for Future Work: The tests facility used in this work has a number of advantages including redundancy in measurements (emission factors measured in multiple locations using different instruments) that increase confidence in the results and provide backup data in case of instrument problems; heavily instrumented stove, stack and exhaust lines that providing ancillary data for interpretation of results; and an ability to make rapid and very precise modifications to test conditions without inconveniencing occupants. These design and operational characteristics provide high quality data on the performance of improved space heating stoves under relevant field operating conditions. However, we learned in this work that the combination of stove design, fuel characteristics and operational parameters along with the inhomogeneous nature of the combustion process, leads to a significant amount of variability in emissions. In addition, the limited number of tests performed to date doesn't allow us to sufficiently characterize uncertainty in emission factor measurements. Several opportunities exist where the LBNL test facility could contribute to improved knowledge or support other projects aimed at reducing pollution with improved space heating stoves.

Verify initial emission test results: Our initial set of experiments was developed to screen a range of stove use scenarios and identify those that were most likely to lead to elevated emission events for the Mongolian heating stoves. Given the time constraints of the study, we used lessons learned from each completed experiment to modify the design of subsequent experiments. As we gained experience with the stove, a number of the initial scenarios that we initially considered to be important were deemed to be unlikely to occur and excluded from further testing.

For example, our initial experimental design included a number of test looking at the impact of loading the stove in the reverse order (paper and wood on bottom with coal on top and then lit from bottom) but given the design of the stove and fire box, this was considered an unlikely behavior for a top load up draft design. In addition, the refueling event with hot embers on the grate

of the fire box closely resembled the case where a fresh bed of coal was lit from the bottom. Therefore, all of the fueling order experiments were excluded.

The lessons learned from early experiments quickly focused our effort on refueling events and coal characteristics. Both of these factors influenced PM emissions and stove performance. Therefore, the primary focus of this report was on refueling. Refueling events led to increased PM emissions.

These initial results should be verified with replicate tests designed to not only quantify the expected emission factor but the variability associated with different factors in the burn. This is critical for using the numbers to support air quality models and policy decisions. Quantifying variability and uncertainty typically requires a large number of experiments. The actual number of replicate tests needed for each combination of factors will depend on the amount of variability observed from conditions during the burn and the level of control that can be achieved in the factors influencing the results. The LBNL stove testing facility is designed to provide a very high degree of control during each test. This highly controlled experimental system combined with redundancy in the instrumentation is expected to provide consistent and repeatable results but replicates are still necessary to characterize uncertainty.

Measure emissions across a broader range of operating behaviors: Our initial experiments focused specifically on refueling events that lead to elevated emission events. We clearly found that realistic stove use scenarios that deviated from the manufacturer's recommended operating procedures can result in significant increase in emission. However, given the variability in stove design, fuel characteristics and use patterns, additional experiments should be completed to characterize and quantify the influence of different combinations of factors on emissions.

Focusing on a limited number of experiments based on how people are actually using these stoves will generate results that can provide simple guidance that reduce some of the problems associated with real use scenarios. More important, the suite of experiments can produce emission factors that can readily support air quality modeling in combination with activity pattern data (i.e., how many cold-start and refueling events per day)

Specific recommendations for additional testing include:

1. Refueling amount – we have initial results indicating that the magnitude of the elevated emission event is correlated with the amount of fuel added.
2. Refueling time – we can clearly identify different parts of the burn so we should test what effect we get when the refueling takes place during different parts of the burn starting at fuel bed collapse and going through the point where re-ignition from embers fails.
3. Refueling fuel characteristics – it appears that the size of the coal chunks can influence the magnitude/duration of emission event so we should identify the worst case refuel amount and time from 1 and 2 then at that condition use carefully segregated coal chunks starting from very large pieces and going down to very fine and include some well characterized combinations of the size fractions. (large medium and small)
4. Fuel moisture content – there seems to be a moisture effect but we have not characterized it very well so we should start with the worst case identified in 1 and 2 then using a typical coal size mixture, run a series of experiments with differing moisture content.

5. Air intake setting – initial results indicate that the primary air intake in the TLUD design can influence the stove performance and emissions and this is most likely to be important following a refueling event. A series of refueling experiments should be performed with the primary air inlet at different settings from fully open to fully closed.
6. Air intake during the burn period – we maintained fully open air inlets during all phases of the study but it is possible that some users shut down the air supply to extend the duration of a burn. The influence of reduced air intake during extended burns should be tested.

It would also be important to extend our experiments to include the traditional stove to understand the relative importance of these emission events during refuel. In addition, we can test other improved stove designs that are currently in use and/or scheduled for distribution so that ultimately, we can provide a complete PM source term profile for stoves in Ulaanbaatar.

Support teams measuring emissions in the field: Impacts of the improved stove will almost certainly be measured in the field in terms of air quality measurements and stack emission measurements. However, the LBNL test facility can provide a useful test-bed for developing field sampling tools and protocols to validate performance and debug the process. We know that there are a range of use behaviors, air intake settings, and fuels and each combination will have slightly (or sometimes dramatically) different characteristics that need to be considered when sampling emissions in the field. For example, if the results found in this study can be verified, it may only be necessary to measure PM emissions during start-up and refueling events and not monitor for extended periods. This could reduce the amount of time by up to 80% with potential savings of resources in the field.

The premise is that by doing some initial testing in collaboration with LBNL before shipping out to Ulaanbaatar, field teams will be able to streamline and optimize their sampling design and collect data that is more likely to be relevant to the program. In addition, if issues come up in the field, having a working test facility to interact with and troubleshoot could reduce down-time and increase the amount and quality of data from the field study.

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Appendix 1: Results of Coal Analysis

1A. Geochemical Testing, Coal Analysis Report: Nailakh

GEOCHEMICAL TESTING
Environmental and Energy Analysis

2005 N. Center Ave.
Somerset, PA 15501

814/443-1671
814/445-6666
FAX: 814/445-6729

COAL ANALYSIS REPORT

Client: BERKELEY LAB

Sampled by: Agnes Lobscheid

Sampling Date: 01/09/2012

Analyzed on: 01/11/2012

Description: Sample ID: Nailakh Mongolian Coal

LAB NO. 12-138241

Air Dry Loss..... 20.26 %

Residual Moisture..... 8.23 %

	As Received	Dry	Dry Ash-Free
PROXIMATE ANALYSIS D3172			
Moisture.....	26.82		
Ash.....	8.00	10.93	
Volatile Matter.....	27.81	38.01	42.67
Fixed Carbon.....	37.37	51.06	57.33
	---	---	---
	100.00	100.00	100.00
ULTIMATE ANALYSIS			
Hydrogen.....D5373..	6.30	4.51	5.06
Carbon.....D5373..	48.18	65.84	73.92
Nitrogen.....D5373..	1.30	1.78	1.99
Sulfur.....D4239-02	0.64	0.87	0.98
Oxygen.....D3176..	35.58	16.07	18.05
Ash.....D3174-02	8.00	10.93	
	---	---	---
	100.00	100.00	100.00
Heating Value(BTU/Lb) D5865	8305	11349	12742
FORMS OF SULFUR D2492			
Sulfate Sulfur.....	0.02	0.02	0.02
Pyritic Sulfur.....	0.02	0.03	0.04
Organic Sulfur.....	0.60	0.82	0.92

Free Swelling Index D720-91 0.0

Equilibrium Moisture D1412-03 26.88

Robert L. Stull
Director of Coal Services

GEOCHEMICAL TESTING
Environmental and Energy Analysis

2005 N. Center Ave.
Somerset, PA 15501

814/443-1671
814/445-6666
FAX: 814/445-6729

COAL ANALYSIS REPORT

Client: BERKELEY LAB

Sampled by: Agnes Lobscheid

Sampling Date: 01/09/2012

Analyzed on: 01/11/2012

Description: Sample ID: Nailakh Mongolian Coal

LAB NO. 12-138241

Ash Fusion (Reducing Atmosphere) ASTM D-1857

	Initial D.	Softening T.	Hemi T.	Fluid T.
Temp o F	2060	2100	2140	2220

Lbs Sulfur/Million Btu 0.77

Lbs SO2/Million Btu 1.54

Chlorine.....D4208-02 < 100 mg/kg, dry

Robert L. Stull
Director of Coal Services

1B. Geochemical Testing, Coal Analysis Report: Utah

GEOCHEMICAL TESTING
Environmental and Energy Analysis

2005 N. Center Ave.
Somerset, PA 15501

814/443-1671
814/445-6666
FAX: 814/445-6729

COAL ANALYSIS REPORT

Client: BERKELEY LAB

Sampled by: Agnes Lobscheid

Sampling Date: 01/30/2012

Analyzed on: 02/01/2012

Description: Sample ID: Utah

LAB NO. 12-140233

Air Dry Loss..... 2.56 %

Residual Moisture..... 1.20 %

	As Received	Dry	Dry Ash-Free
PROXIMATE ANALYSIS D3172			
Moisture.....	3.73		
Ash.....	6.77	7.03	
Volatile Matter.....	39.11	40.63	43.70
Fixed Carbon.....	50.39	52.34	56.30
	---	---	---
	100.00	100.00	100.00
ULTIMATE ANALYSIS			
Hydrogen.....D5373..	5.50	5.28	5.68
Carbon.....D5373..	72.50	75.31	81.01
Nitrogen.....D5373..	1.67	1.73	1.86
Sulfur.....D4239-02	0.37	0.38	0.41
Oxygen.....D3176..	13.19	10.27	11.04
Ash.....D3174-02	6.77	7.03	
	---	---	---
	100.00	100.00	100.00
Heating Value(BTU/Lb) D5865	13101	13608	14638
FORMS OF SULFUR D2492			
Sulfate Sulfur.....	0.01	0.01	0.01
Pyritic Sulfur.....	0.01	0.01	0.01
Organic Sulfur.....	0.35	0.36	0.39

Free Swelling Index D720-91 4.0

Equilibrium Moisture D1412-03 3.88

Robert L. Stull
Director of Coal Services

GEOCHEMICAL TESTING
Environmental and Energy Analysis

2005 N. Center Ave.
Somerset, PA 15501

814/443-1671
814/445-6666
FAX: 814/445-6729

COAL ANALYSIS REPORT

Client: BERKELEY LAB

Sampled by: Agnes Lobscheid

Sampling Date: 01/30/2012

Analyzed on: 02/01/2012

Description: Sample ID: Utah

LAB NO. 12-140233

Ash Fusion (Reducing Atmosphere) ASTM D-1857

	Initial D.	Softening T.	Hemi T.	Fluid T.
Temp o F	2260	2320	2390	2540

Lbs Sulfur/Million Btu 0.28

Lbs SO2/Million Btu 0.56

Chlorine.....D4208-02 447 mg/kg, dry

Robert L. Stull
Director of Coal Services