Characterization of Gaseous and Particulate Pollutants from Gasification-based Improved Cookstoves

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ABSTRACT

Cookstove studies have reported pollutant concentrations (mainly PM_{2.5}, black carbon and CO) without routinely associating it with the design and operating principles of the stoves. Extensive characterization of pollutants from cookstoves and the effect of different operating conditions are required for a better understanding of the mechanisms of pollutant formation. In this study, a forced draft (FD) and a natural draft (ND) gasification-based improved cookstove were tested under controlled conditions. Real-time pollutant concentrations, both particulate (PM_{2.5}, lung-deposited surface area and particle number size distribution) and gaseous (CO, CO₂ and NO_x), from these stoves using three types of fuel (applewood chips & chunks, cowdung cake and coal) along with different cookstove operating conditions (air flow rates and with or without a cooking pot) were measured and compared. The FD cookstove tended to exhibit higher concentrations of emissions compared to the ND cookstove. Increasing airflow through the FD stove decreased flame length and the residence time of VOCs inside the flame zone, which in turn increased pollutant concentrations. An optimum air flow producing the lowest particulate matter (PM) concentrations was established for the FD cookstove. The CO-CO₂ ratio, an indicator of combustion efficiency, demonstrated strong correlations with PM_{2.5} ($r = 0.857$), particle geometric mean diameter ($r = 0.900$), and the total surface area concentration ($r = 0.908$) indicating that CO-CO₂ ratio may be used as a proxy for these PM metrics. Results reported in this study will facilitate further improvements in the design of future cookstoves.

1. Introduction

Two million tons of biomass, including animal dung and agricultural residue, are burned daily in cookstoves in the developing world [1], and along with coal, provides energy for space heating and cooking to almost three billion of the world's poorest people [2]. High concentrations of pollutants such as particulate matter (PM), carbon monoxide (CO), and organic compounds are emitted due to incomplete combustion of these solid fuels in cookstoves. Inhaled ultrafine particles (UFP) generated from solid fuel combustion can evade the body's mucocilliary defense system and deposit deep in the alveolar region of the lungs leading to short-term inflammation and oxidative stress [1], and long-term chronic obstructive pulmonary disorders (COPD) [3]. Other diseases from exposure to biomass emissions include acute respiratory infections (ARI), cataracts, and tuberculosis [3, 4]. The World Health Organization reported that the exposure to air pollution led to 7 million deaths in 2012, making it the world's largest single environmental health risk [5]. Household air pollution accounted for more than half of these deaths.

The majority of those who rely on solid fuels use traditional three-stone cookstoves and open fires to cook their food [6]. Studies have reported concentrations exceeding 10 mg/m^3 and 300 ppm for $\text{PM}_{2.5}$ and CO, respectively [7-10]. Because most of the improved cookstoves have better combustion efficiency, they produce fewer emissions, thus renewed impetus to promote and distribute these cookstoves has seen the number of people using them rise to approximately 800 million [6, 11]. However, combustion efficiency differs widely between these improved cookstoves with gasification-based cookstove among the best performing stoves. Laboratory studies comparing both the forced-draft (FD) and less frequently the natural-draft (ND) gasification-based stove, to other cookstoves have repeatedly demonstrated their improved

efficiency and reduced emissions [12-15]. For example, Kar et al. [12] reported a 77% reduction in black carbon (BC) emissions for the FD Philips stove compared to a traditional mud cookstove, while Jetter et al. [15] reported lower CO and PM_{2.5} emissions per unit energy delivered to the cooking pot compared to a three-stone stove. With the exception of a few studies [14, 16], correlating pollutant concentrations with the design and operating principles of a particular cookstove has rarely been done. Such correlations are critical for further improvements to cookstove design.

Another important aspect is a protocol for testing the stove which will allow comparisons. Many researchers have proposed a Water Boiling Test (WBT). However this test has limitations which are defined in the WBT protocol itself [17]. The WBT does not prescribe the exact pot to be used for the tests. This introduces another variable (size and material of the pot) potentially changing the emission characteristics. L'Orange et al. [18] showed that the pot temperature has a significant effect on PM characteristics. For example, a hot pot resulted in lower PM₁₀ with lower smaller particle sizes. Other tests, such as the Controlled Cooking Test (CCT) and Uncontrolled Cooking Test (UCT) better capture cookstove performance when cooking actual food in both laboratory and field settings. Arora et al. [19] followed the CCT to demonstrate that emissions characteristics depend on the type of meal cooked, and not just the type of meal but even the thickness of a roti (a type of Indian bread) altered emissions. They found CO fluctuated by up to 49% depending on different cooking styles. Because recently published studies are increasingly demonstrating the effect that small changes in cooking (pot / food / water temperature) alter emission characteristics, it is clear that the WBT and other similar laboratory tests fail to represent field conditions. Although it is true that cookstoves are rarely used without a pot, their performance in the absence of a pot must be investigated to gain a

fundamental understanding of the combustion processes to avoid complexity and reduce uncertainty especially in controlled laboratory studies. Such combustion studies are often lacking due to the emphasis of establishing emission factors (EF) which vary significantly based on the field cooking styles, and also do not accurately correspond to eventual health or climate impacts.

Although Jetter and Kariher [14] compared cookstove performance for different types of biomass, few studies have incorporated dung and coal into their analysis, a key gap given that: 1) dung features low on the energy ladder and is therefore used by the poorest and most vulnerable members of society [2, 20]; and 2) coal continues to dominate in certain communities and is especially heinous. For example, in the Damodar Valley, India, 2.55 million tons of coal are consumed domestically each year [21]. Households that burn coal consistently demonstrate higher pollutant concentrations compared to those that burn biomass [22]. Impaired immune system, CO poisoning, COPD, and lung cancer have been reported from exposure to coal combustion emissions [1, 22]. Finally, with the exception of a handful of studies that have incorporated multiple particulate metrics, including particle number size distributions, into their study [23-27], most studies focus on PM_{2.5} and CO. Also, with the exception of a couple of studies [8, 10], lung-deposited surface area has not been investigated.

The overall objective of this study was to conduct an extensive characterization of particulate and gaseous pollutants from biomass and coal burned in two gasification-based improved cookstoves; FD Philips (HD 4012) stove and ND Quad, a Top Lit Updraft (TLUD) stove. Correlations between concentrations of different pollutants were investigated to evaluate the feasibility of using one pollutant as a proxy for others. In addition, the influence of a cooking pot on pollutant concentrations was assessed. Finally, the Philips stove was operated at 3

different air flows to examine the influence on particle number size distribution (PNSD). Results obtained in this study were explained based on the operating principles of gasification-based cookstoves. Comparisons between the FD and ND stoves enhanced the understanding of pollutant formation which should facilitate further improvements in design of the stoves.

2. Materials and Methods

Different system components are described in the following sections. An overall test plan is provided in Table 1.

2.1 Cookstoves Studied

Two gasification-based cookstoves, the Philips (Model No. HD4012 LS) and the Quad TLUD were investigated. As illustrated in Figure 1, primary air passes through the inner cylinder, where the biomass is placed. Biomass gasification occurs in the primary oxidation zone (lower region) in the presence of limited oxygen. Producer gas, consisting mainly of CO, lower hydrocarbons and volatiles, is then oxidized by the air in the secondary oxidation zone at the top of the cookstove. A more detailed description of design and operating principles of the stoves is discussed in Section 3.

‘Insert Figure 1 Here’

2.2 Fuels

The three fuels investigated were applewood, cowdung cake, and coal. Each of these fuels are used by different income groups. Cowdung cake is a popular fuel in rural India, especially among households with low and medium income [20]. Fresh dung from grass-fed cows was collected from a farm near St. Louis (MO) USA, shaped into patties and left to dry in the open for two weeks during the summer season. Chemically untreated applewood was

purchased locally. Two sizes of applewood, chips (1-3 cm long thin pieces) and chunks (4-6 cm cubes), were tested to examine the effect of fuel size. Bituminous coal from Brilliant (Alabama) was also tested. Fuels were procured from the same batch and stored at constant ambient conditions to minimize variability. The properties of the various fuels used are reported in the literature and vary from reference to reference: proximate analysis, ultimate analysis and heating value for applewood [25, 28, 29], dung cakes [30-32] and bituminous coal [33, 34].

2.3 Experimental Setup

Experiments were conducted in a fume hood in a controlled environment to facilitate comparisons by minimizing the variability between different runs. A schematic diagram of the experimental setup is shown in Figure 2. A hood with a 0.37 m² area was installed above the cookstove. Flow from the hood was sampled using an aspiration-based sampling system [35, 36]. The aspiration-based sampling system creates a suction using low pressure generated from the flow of dilution air, therefore no pump downstream is required. A dilution flow of 0.094 cubic meters per minute was used to achieve a dilution ratio of 4. After the dilution unit, a fraction of the sample was collected by an isokinetic sampling probe while the rest was vented out. The sample then passed through a diffusion dryer to remove any moisture to prevent interference with readings. Copper and conductive tubing was used throughout the sampling train to minimize particle loss during transport.

'Insert Figure 2 Here'

'Insert Table 1 Here'

The test plan for this study is outlined in Table 1. Unlike the water boiling test (WBT), which assesses thermal efficiency [14, 15] under controlled settings, this work focused on the

pollutant characteristics (concentrations and size distribution in case of PM) as a result of the gasification/combustion processes in different cookstoves. All tests were done without a cooking pot as the focus was on the gasification-combustion process. However, one test (ID 9, Table 1) was performed with a pot to examine its influence and also to highlight that a pot is an important variable that impacts emissions. The TLUD stove was fired using applewood chips and a pot with 5 liters of water. The quantity of water was selected to ensure that it hovered just below boiling point, thus minimizing any interference with instrument readings from added moisture. The impact on emissions on use of a pot is however more complex (e.g. size, shape and material of the pot) and was not the scope of this study. The Philips stove, fueled with applewood chips, was tested at three different air flow rates: low, medium and high, to assess its effect on particulate emissions. Each experimental test was repeated a minimum of three times. Observations on the flame structure (length and intensity) and smoke characteristics were also noted.

2.4 Characterization of Particulate and Gaseous Pollutants

Total number concentrations and PNSD were measured using a scanning mobility particle sizer (SMPS 3080, TSI) and a condensation particle counter (CPC 3022, TSI). The SMPS operates by charging particles to a known charge distribution by a bipolar charger in an electrostatic classifier. Charged particles are then classified based on their electrical mobility in an electric field and an optical sensor in the CPC measures the number concentration. Real-time measurements of lung-deposited surface area were made using a nanoparticle surface area monitor (AeroTrak 9000, TSI). This instrument can provide surface area concentrations of particles deposited in either the tracheobronchial (TB) or alveolar (A) regions of the lung based on deposition curves obtained from models developed by the International Commission on

Radiological Protection (ICRP) [37]. The surface area concentration of particles deposited in the TB region was recorded in this study. Details about the operating principles of the AeroTrak 9000 are provided in Leavey et al. [38].

A DustTrak II (Model 8532, TSI) was used to monitor $PM_{2.5}$. This portable instrument works on the principle of light scattering. A fraction of the sample flow, to be used as sheath flow, is passed through a HEPA filter. Particles scatter the light, as they pass a laser diode. The degree of light scattering depends on the size, shape and density of the particles. The scattered light falls on a spherical mirror which focuses it on a photo detector resulting in a voltage which is proportional to mass concentration.

The EL-USB-CO EasyLog®, manufactured by LASCAR Electronics, was used to record CO levels. It oxidizes CO to CO_2 producing a current proportional to the CO concentration. The range of the instrument is 0-300 ppm with the smallest data logging interval of 10 seconds. A portable gas analyzer (HORIBA PG-250) was used to measure CO_2 and NO_x levels. The NO_x unit uses a cross-flow modulation chemiluminescence, while the CO_2 unit operates on the principle of the non-dispersive infrared (NDIR) method. A LabVIEW® program was created for data acquisition, and so that data could be stored in one second intervals.

2.5 Analysis

The burn cycle of the stoves was divided into three phases: the ignition, steady-state and extinguishing phase (Figure 3). This study focused on data collected whilst the cookstove was operating under steady-state burning conditions, as this represents the major part of the total operation time, and is the phase during which cooking occurs. The steady-state phase was defined as the time period during which concentrations of CO and CO_2 were relatively stable, and a flame with constant intensity and length was observed. The ignition phase was marked as

the time between the ignition and start of the steady-state phase. Similarly, the extinguishing phase was marked from end of the steady-state phase until flame extinction. The duration of steady-state varies with stove, fuel type, and experimental settings. In a field study, Sahu et al. [8] reported the start of steady-state at approximately 20 minutes post ignition, while Leavey et al. [10] reported an interval of 2-15 minutes between ignition and the start of steady-state phase in an another field study. In this laboratory study, steady-state began within 2-5 minutes from ignition, depending on stove and fuel type. A plausible explanation for this disparity is the optimal conditions under which experiments were conducted in the laboratory and the difference in the type of cookstoves used. In the case of coal, two steady-states were defined, the flame phase characterized by flames and heavy smoke and the smoldering phase when flames dies out leaving only red hot glowing coal. The duration and the heat generation rate of both phases were found to be comparable but pollutant characteristics during these two phases differed significantly.

‘Insert Figure 3 Here’

This study reports average concentrations over the steady-state phase only. While most of the PM metrics measured directly, mass and surface area concentrations were calculated using PNSD data from the SMPS. To analyze the correlation between concentrations of different pollutants, a correlation test was performed using R Statistical Software (version 2.101).

3. Construction and Operating Principle of the Gasification-based Cookstoves

The typical construction of a gasification-based cookstove includes two concentric cylinders open at each end (Figure 1). Design parameters of the two stoves are presented in Table 2. Fuel is placed in the inner cylinder and air flowing through it is labelled as the primary

air flow. The annular region between the two cylinders acts as a channel for the secondary air flow. Unlike a fan installed at the bottom of FD cookstoves, a temperature gradient along the height of the stove creates an updraft in ND cookstoves. Thus ND cookstoves are generally taller than FD cookstoves to create a comparable updraft. Also, as most of the FD cookstoves require a power source to recharge the batteries for the fan, ND cookstoves may have an advantage for regions not on the grid.

One ND (Quad TLUD) and one FD (Philips) stove were investigated in this study. Both cookstoves are top-lit, which means that the biomass is initially ignited from the top and the high temperature layer propagates downwards through the packed bed. The top-ignition method produces significantly less PM, CO and NO_x compared to the bottom-ignition method with no significant difference in cookstove efficiency [39] and this might explain why the cookstove manufacturers recommend the top-ignition method. Convection is the major mode of heat transfer to the fuel during bottom-lit ignition, whereas conduction and radiation dominate the heat transfer for the top-lit ignition. In the top-lit ignition method, fuel is ignited at top and the heat is transferred to the fuel beneath through conduction. The packed bed can be divided into three zones starting from top to bottom: the gasification zone characterized by the highest temperature, the devolatilization zone and the drying zone.

The primary air flow is much lower than the secondary air flow due to the cookstove design and resistance of the packed bed. Oxygen deficiency, due to low primary air flow rate combined with high temperature in the primary oxidation zone, leads to the gasification of fuel generating producer gas consisting mainly of CO and N₂. The producer gas then mixes with the secondary air at the top of the stove producing a more stable and smokeless flame compared to traditional stoves. This process of transferring carbon from solid fuel to gaseous fuel (producer

gas) followed by its combustion in a controlled sequential manner is what makes combustion in a gasification-based cookstoves efficient. This modular function (primary and secondary oxidation zone) ensures flame generation at the top of cookstove irrespective of the fuel level in the stove. Producer gas also contains tar, but in much lower concentrations compared to CO and N₂, which participates in particle formation if not oxidized completely in the secondary oxidation zone. Since CO is intentionally produced in the primary oxidation zone, it is of utmost important to oxidize it in the secondary oxidation zone to ensure user's safety.

'Insert Figure 4 Here'

'Insert Table 2 Here'

The primary and secondary air flow rates are key parameters to tune the performance of gasification-based cookstoves. Air flow rate can be controlled in FD cookstoves by adjusting the fan speed but the primary and secondary air flow rates cannot be controlled individually in most of the single fan models. Very few FD gasification-based cookstove models come with two separate fans to control both the primary and secondary air flow rate. The airflow control mechanism in ND stoves, if available, usually does not offer an airflow range as wide as in FD stoves and hardware modifications may be necessary. Too low secondary air flow rates result in incomplete oxidation of the gasification products in the secondary oxidation zone due to insufficient oxygen. Too high secondary air flow rates lower residence time of the producer gas in the secondary oxidation zone resulting in incomplete combustion. High flow rates also lower the temperature of the oxidation zone which decelerate the oxidation reactions.

Experiments were performed to study the effect of air flow rate on particulate emission characteristics of the Philips stove. The Philips stove fueled with applewood chips was tested at a high, medium, and low air flow rate. The PNSD are presented in Figure 4. The lowest particle

number concentrations were observed for a medium airflow indicating that there is an optimum airflow which leads to the most efficient combustion. For this reason, the Philips cookstove was operated at a medium airflow setting for all further experiments. Previous modeling studies [40, 41] also reported an optimum air flow rate for maximum thermal efficiency and lowest gaseous emissions. A low air flow rate leads to incomplete combustion and a high air flow rate leads to heat loss because of excess air, thus decreasing the temperature of oxidation zone.

4. Results and Discussions

In this study, characterization of gaseous and PM emissions from two gasification-based improved cookstoves was performed. Three fuels; applewood, cowdung cake and coal were tested. Real-time measurements of PNSD, PM_{2.5}, lung-deposited surface area, CO, NO_x and CO₂ were collected and the average levels during steady-state phase are reported. Combustion efficiency was characterized by the CO-CO₂ ratio. Correlation tests were also performed on all pollutants and the CO-CO₂ ratio.

4.1 Particulate Emissions

Tests 1 to 4 (Philips) and 5 to 8 (TLUD) are for comparison of emissions for different fuels. Figure 5 presents PNSD measured for each cookstove for (A) coal and dung, (B) applewood chips and chunks, with corresponding values of particle geometric mean diameter (GMD), standard deviation and number concentration presented in Table 3. During the flame phase, coal produced higher concentrations (Philips: $2.42 \times 10^9 \text{ \# cm}^{-3}$, TLUD: $1.83 \times 10^9 \text{ \# cm}^{-3}$) of larger particles (Philips GMD: 221 nm, TLUD GMD: 255 nm) compared to the other fuels. In a field study, Zhang et al. [22] reported a much lower mean diameter (70.3-75.7 nm) for honeycomb briquette, made from a mixture of anthracite coal and clay, during first 15 minutes of

combustion. This could be due to improved combustion as the honeycomb shape provides higher surface area and better airflow mixing through the cookstove. For both cookstoves, particle GMD during the coal smoldering phase (Philips:84 nm, TLUD:72 nm) was significantly lower than that of the coal flame phase. This could be attributed to a reduced devolatilization rate and thus lower tar production rate during the smoldering phase.

'Insert Figure 5 Here'

'Insert Table 3 Here'

The second highest particle concentration and mean size was observed for dung cakes for both the Philips (number concentration: $1.17 \times 10^9 \text{ \# cm}^{-3}$, GMD:112 nm) and the TLUD stove (number concentration: $6.84 \times 10^8 \text{ \# cm}^{-3}$, GMD:85 nm). The difference between these two cookstoves can be explained by the higher airflow rate in the Philips cookstove compared to the TLUD stove. Tiwari et al. [42] reported much higher GMD for cowdung (152 nm) burned in a traditional U-shaped cookstove. Applewood, both chips and chunks, emitted smaller particles than dung and coal.

'Insert Figure 6 Here'

Figure 6 displays bar charts of $\text{PM}_{2.5}$, mass concentration, lung-deposited surface area and total surface area concentration. There is no $\text{PM}_{2.5}$ and lung-deposited surface area data for any of the coal runs because the instruments were unable to measure such high concentrations. Therefore, mass concentrations (Figure 6B) were calculated from the PNSD data. As depicted in Figure 5A, a significant fraction of particles generated during the smoldering phase of coal exceeded the detection limit and thus the calculated mass concentration values are an underestimation of actual levels. The most marked difference in $\text{PM}_{2.5}$ levels between cookstoves was for dung cakes. The Philips stove demonstrated a mean concentration of 119.80 mg m^{-3}

compared to 9.08 mg m^{-3} for the TLUD stove. Although the Philips stove also demonstrated a higher $\text{PM}_{2.5}$ for applewood chips and chunks, the difference was less significant. It was observed during the experiments that the TLUD stove flame length was higher compared to the Philips stove with the same fuel type. Smaller flames, combined with higher secondary air flow in the Philips stove, reduced the residence time of gasification products in the secondary oxidation zone. Moreover, increasing differences between the $\text{PM}_{2.5}$ levels of the two stoves, operating with the same fuel, was observed with increasing fuel size (chips:11.1%, chunks:28.1%, dung:1219.4%). The larger size of the dung cake resulted in the lowest packing efficiency, thus providing the lowest resistance for primary air flow which enhanced the effect of a higher flow rate on cookstove combustion efficiency. This observation highlights the issue of fuel compatibility for a given cookstove. . Lung-deposited surface area (Figure 6C) and total surface area concentrations (Figure 6D) demonstrated similar trends to $\text{PM}_{2.5}$. However, lung-deposited surface area may be a better PM metric than $\text{PM}_{2.5}$ to understand health impacts of PM emissions characterized by high number concentration of smaller particles [8].

4.2 Gaseous Emissions

Figure 7 presents data on NO_x , CO and CO-CO₂ ratio. The CO-CO₂ ratio [43], i.e. the ratio of partially oxidized to fully oxidized carbon, may be a better proxy of combustion efficiency from the exposure perspective than the modified combustion efficiency (MCE), which is defined as $\text{CO}_2/(\text{CO}+\text{CO}_2)$ [15]. MCE is less sensitive to change in CO levels compared to the CO-CO₂ ratio on a molar basis.

'Insert Figure 7 Here'

NO_x formation rates depend on the temperature and the source of nitrogen which can either be from fuel or ambient air. In a typical packed bed biomass combustion system, nitrogen (N) from fuel is the main source of NO_x formation [44]. Dung has the highest N content, followed by coal and applewood [30, 44-46]. From Figure 7A, it can be clearly observed that the N content of the fuel is not the only factor governing NO_x formation, as NO_x levels of applewood chips with the Phillips stove (47 ppm) are comparable to that of dung (59 ppm) whose N content is around six times higher than applewood. In addition, higher temperatures promote NO_x formation [22, 47] which may explain the higher NO_x concentration measured for the Philips stove compared to the TLUD stove for applewood and coal. The Philips stove combustion chamber is lined with a layer of ceramic material which ensures low heat loss through the walls and thus higher temperatures. The similar level of NO_x observed during dung combustion for each of the stoves may be due to its significantly higher N content. In addition, the lower NO_x levels observed from coal during the flame phase, as compared to the smoldering phase, for both stoves could be due to lower temperatures during the flame phase because of highly endothermic devolatilization reactions and moisture evaporation from the unburned coal.

CO is the major product from biomass gasification in the primary oxidation zone. A gasification-based stove must efficiently oxidize it to CO₂ in the secondary oxidation zone. As demonstrated in Figure 7B, significantly higher CO levels were observed with the Philips stove fueled with coal during the flame phase (800 ppm) and dung cakes (548 ppm), and the TLUD stove fueled with coal during the flame phase (632 ppm). Though these concentrations were measured in the plume, burning coal or dung cakes in similar stoves in a poorly ventilated setting may lead to subtle cardiovascular and neurobehavioral effects at low level chronic concentrations and even death at acute concentrations of CO [48]. The TLUD stove had lower CO

concentrations than the Philips stove for all fuels, especially dung (Philips:548 ppm, TLUD:130 ppm). The CO concentration trends were similar to the PM concentration, thus the same explanation that more gasification products remain unoxidized in the Philips stove due to the lower residence time in the secondary oxidation zone, may be given. A more detailed discussion on the correlations between these metrics is provided in Section 4.3.

To investigate the effect of fuel size on concentrations of gaseous pollutants, two different sizes of applewood (chips and chunks) were tested. CO levels decreased with decreasing fuel size (Philips:213 ppm (chunks) and 65 ppm (chips), TLUD:106 ppm (chunks) and 46 ppm (chips)) while NO_x showed the opposite trend (Philips: 20 ppm (chunks) and 47 ppm (chips), TLUD: 16 ppm (chunks) and 20 ppm (chips)). Similar trends have been reported by Bhattacharya et al.[39] who suggested that the smaller size of the fuel intensified combustion thus promoting higher temperatures, facilitating CO oxidation and NO_x formation.

4.3 Correlation Tests between Different Pollutants

Correlations between concentrations of different pollutants can be explored to see whether one pollutant may be used as a proxy for another. The primary focus was to investigate correlations between gaseous and particulate emission parameters. Table 4 presents a correlation coefficient (r) matrix with p-values for the 95% confidence interval. No significant correlations were observed between NO_x and the PM metrics at the 95% confidence interval, with the exception of lung-deposited surface area (r=0.455). Strong correlations between PM metrics and CO levels were expected, as both are products of incomplete combustion. However, CO demonstrated only moderate positive correlations with particle GMD (r=0.702), surface area concentration (r=0.646) and mass concentration (r=0.645). Also, weak correlations between CO and lung-deposited surface area (r=0.445) and total number concentration (r=0.394) were also

observed. A moderately strong correlation ($r=0.760$) was observed between CO and PM_{2.5} which is similar to correlations reported in the literature by Venkataraman et al. [30] ($R^2=0.71$). Likewise in a field study, Chowdhury et al. [49] reported correlations (R^2) between PM_{2.5} and CO concentrations ranging from 0.605 to 0.705 depending upon the type of cookstove. In a similar study, Commodore et al. [50] correlated 4-hour mean personal PM_{2.5} exposures with personal CO exposures during lunch ($r=0.67$) and dinner ($r=0.72$). Leavey et al. [10] reported a r -value of 0.71 for the correlation between average PM_{2.5} and CO concentrations over the steady-state phase.

'Insert Table 4 Here'

Even weaker correlations were observed between the PM metrics and CO₂, with the exception of total number concentration. This indicates that PM emissions are not only dependent on the combustion rate, but also on the extent of combustion. To demonstrate this, correlation tests were performed between CO-CO₂ ratios and PM metrics. Stronger correlations of PM_{2.5} ($r=0.857$), particle GMD ($r=0.900$), surface area concentration ($r=0.908$) and mass concentration ($r=0.905$) with CO-CO₂ ratio (Figure 8) were observed. This suggests that CO concentration normalized by an indicator of combustion rate (CO₂ in this study) is a better proxy for PM metrics than CO alone. Moderate correlation ($r=0.625$) was found between PM_{2.5} and lung-deposited surface area. Leavey et al. [10] reported a similar correlation ($r=0.64$) between PM_{2.5} and lung-deposited surface area during steady-state. This highlights the possibility of translating the correlations between different pollutants from laboratory to field and vice-versa. The CO-CO₂ ratio demonstrated a weaker correlation with lung-deposited surface area ($r=0.496$) compared to total surface area concentration calculated from PNSD ($r=0.908$) as the two surface area concentrations are interrelated by a deposition efficiency curve which is not linear.

414 *'Insert Figure 8 Here'*

415 Insignificant correlations between NO_x and CO, CO_2 and CO- CO_2 ratio were observed.
416 Mass concentration, surface area concentration and GMD demonstrated a high correlation
417 ($r>0.835$) with each other. Likewise, a strong correlation ($r=0.929$) was observed between $\text{PM}_{2.5}$
418 and mass concentration calculated from PNSD data because only a small fraction of particles
419 was observed around the upper detection limit of SMPS for all fuels except coal flame phase.
420 The total number concentration demonstrated a moderate correlation with particle GMD
421 ($r=0.556$), mass concentration ($r=0.636$), and surface area concentration ($r=0.712$).

422 **4.4 Effect of a cooking pot on emission characteristics**

423 The TLUD cookstove fueled with applewood chips was tested with and without a
424 cooking pot. As presented in Figure 9A, increased concentrations were observed for CO, $\text{PM}_{2.5}$
425 and lung-deposited surface area when a pot was used, but the opposite trend was observed for
426 NO_x . Concentration of CO was almost 10 times higher when a pot was added. This may be due
427 to changes in the flame structure and obstruction of air flow caused by the physical presence of
428 the cooking pot. Also, the addition of a pot may lower the gasification zone temperature which
429 reduces the rate of NO_x formation and rate of oxidation of CO and VOCs. $\text{PM}_{2.5}$ also increased
430 by a factor of 10 from 2.16 mg m^{-3} to 21.4 mg m^{-3} with addition of a pot, demonstrating a similar
431 trend as CO. However, a relatively smaller difference was observed for lung-deposited surface
432 area (with pot: $4256 \mu\text{m}^2 \text{ cm}^{-3}$, without pot: $3816 \mu\text{m}^2 \text{ cm}^{-3}$). This indicates that the addition of a
433 cooking pot promoted the formation of larger particles, for example in Figure 8B, particle GMDs
434 were 69 nm (with pot) and 46 nm (without pot), whereas no significant change was observed for
435 the total number concentration ($5.40 \times 10^8 \text{ \# cm}^{-3}$ with pot and $5.20 \times 10^8 \text{ \# cm}^{-3}$ without pot).
436 Addition of a pot changes multiple factors such as flame temperature and structure, and airflow

characteristics. Therefore it is hard to associate the observed changes in pollutant characteristics to a single cause. This highlights the importance of studying these cookstoves without a pot but understanding the effect of pot is also critical as it reflects the real field conditions.

'Insert Figure 9 Here'

5. Conclusions

This study compared the gaseous and particulate pollutant characteristics from two gasification-based improved cookstoves fed with different types of fuel. Pollutant concentrations from coal and dung combustion were significantly higher than that of applewood. The ND TLUD stove demonstrated lower PM concentration compared to the FD Philips stove which could be attributed to a higher airflow rate and lower height of the Philips stove. Moreover, differences in PM_{2.5} levels between the two stoves, operated with the same fuel increased with increasing fuel size. The trends observed in pollutant concentrations were explained by the design and operating principle of gasification-based cookstoves. Fuel choice is governed by multiple factors such as household income, season and geographical location which leads to fuel stacking. Both cookstoves studied demonstrated significant difference in the emission levels with different types of fuel. Therefore, it is important for an improved cookstove to be compatible with multiple fuels common in the target region to achieve desired health and environmental impacts. High concentrations of pollutants were observed for dung cakes and coal indicating that section of the population lying at the bottom of energy ladder is at a greater risk. Therefore, it is tentatively suggested that a cookstove should be designed specifically for dung cake and coal.

Strong correlations between CO-CO₂ ratio and PM_{2.5x}, particle GMD and surface area concentration were observed. Thus CO-CO₂ ratio may be used as a proxy for the particulate

metrics. The CO-CO₂ ratio is an indicator of combustion efficiency, a higher ratio indicative of a lower efficiency. Since PM is also a product of incomplete combustion, the two metrics correlate well. It is important to note however, that any such quantitative correlation factor may not be applicable with other types of cookstove, fuel, or experimental design. The presence of a pot could also affect the nature of such correlations.

Combustion in cookstoves is a complex process. This study demonstrated the importance of operating parameters and cookstove design in reducing the pollutant concentrations. More attention should be given to developing detailed cookstove combustion models that enhance the understanding between cookstove design, operation and performance to facilitate the engineering of the next generation of improved cookstoves. Though significantly cleaner than traditional cookstoves, improved cookstoves still emit high concentrations of pollutants. Therefore they should be viewed only as an interim solution towards the goal of providing cleaner energy for household use.

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Table 1: Experimental plan for laboratory test of cookstoves

Test ID	Cookstove	Fuel type	Fuel load (g)	Air flow	Objective
1	Philips	Applewood chips	210	Medium	Fuel comparison
2		Applewood chunks	280		
3		Cowdung cake	110		
4		Coal	650		
5	TLUD	Applewood chips	860	NA	Fuel comparison
6		Applewood chunks	1200		Stove comparison
7		Cowdung cake	450		Impact of pot
8		Coal	2500		
9		Applewood chips (with pot)	860		
10	Philips	Applewood chips	210	Low	Impact of airflow (with Test 1)
11		Applewood chips	210	High	

Table 2: Design parameters of the stoves

	Philips	Quad TLUD
Height (cm)	30	43
Distance between primary and secondary air inlets (cm)	12	21.5
Diameter of inner cylinder (cm)	11.5	18
Diameter of outer cylinder (cm)	18.5	22.5
Fuel chamber capacity (cm ⁻³)	1350	5468
Material of construction	Stainless steel	Sheet metal
Insulation	Ceramic inner walls	None

Table 3: Geometric mean diameter, standard deviation and total number concentration during steady-state operation of the TLUD and Philips stove

Test ID	Experiment	Geo. Mean Dia. (nm)	Geo. Std. Deviation	Total number Conc. (#/cc)
8	TLUD Coal Flame	255	1.51	1.83×10^9
4	Philips Coal Flame	221	1.64	2.42×10^9
8	TLUD Coal Smoldering	72	1.77	2.29×10^9
4	Philips Coal Smoldering	84	1.61	2.18×10^9
7	TLUD Dung	85	1.76	6.84×10^8
3	Philips Dung	112	1.66	1.17×10^9
5	TLUD Applewood Chips	46	1.56	5.20×10^8
1	Philips Applewood Chips	51	1.64	6.00×10^8
6	TLUD Applewood Chunks	49	1.70	3.99×10^8
2	Philips Applewood Chunks	48	1.67	5.56×10^8

Table 4: Correlation coefficient (r) matrix with p-values, corresponding to 95% confidence interval, in parentheses

	CO/CO ₂	CO	CO ₂	NO _x	LDSA	PM _{2.5}	TNC	GMD	SA Conc.
CO/CO ₂									
CO	0.883 (<0.001)								
CO ₂	-0.615 (<0.001)	-0.535 (<0.001)							
NO _x	-0.278 (0.086)	-0.201 (0.21)	0.181 (0.25)						
LDSA	0.496 (0.022)	0.445 (0.038)	0.010 (0.967)	0.455 (0.033)					
PM _{2.5}	0.857 (<0.001)	0.760 (<0.001)	-0.453 (0.020)	0.289 (0.152)	0.625 (0.001)				
TNC	0.613 (<0.001)	0.394 (0.023)	-0.597 (<0.001)	0.001 (0.99)	0.750 (<0.001)	0.943 (<0.001)			
GMD	0.900 (<0.001)	0.702 (<0.001)	-0.541 (<0.001)	0.039 (0.82)	0.568 (0.014)	0.835 (<0.001)	0.556 (<0.001)		
SA Conc.	0.908 (<0.001)	0.646 (<0.001)	-0.510 (0.002)	-0.105 (0.560)	0.681 (0.002)	0.941 (<0.001)	0.712 (<0.001)	0.902 (<0.001)	
Mass Conc.	0.905 (<0.001)	0.645 (<0.001)	-0.464 (0.006)	-0.113 (0.530)	0.641 (0.004)	0.929 (<0.001)	0.636 (<0.001)	0.909 (<0.001)	0.993 (<0.001)

LDSA

TNC

GMD

SA Conc.

Mass Conc.

Lung Deposited Surface Area

Total Number Concentration

Geometric Mean Diameter

Surface Area Concentration

Mass Concentration