ACCEPTED VERSION

Thomas Kirch, Paul R. Medwell, Cristian H. Birzer, Philip J. van Eyk

Feedstock dependence of emissions from a reverse-downdraft gasifier cookstove

Final publication at: http://dx.doi.org/10.1016/j.esd.2020.02.008

http://hdl.handle.net/2440/124655

Feedstock Dependence of Emissions from a Reverse-Downdraft Gasifier Cookstove

Thomas Kirch[a,b,*], Paul R. Medwell[a,b], Cristian H. Birzer[a,b], Philip J. van Eyk[c]

[a]*School of Mechanical Engineering, The University of Adelaide, S.A. 5005, Australia*
[b]*Humanitarian and Development Solutions Initiative, The University of Adelaide, South Australia 5005, Australia*
[c]*School of Chemical Engineering, The University of Adelaide, S.A. 5005, Australia*

Abstract

The present study investigates the combustion process of the producer gas from a gasifier cookstove, for four solid biomass fuels: wood pellets (WP), wheat straw (WS), sheep manure (SM) and cow manure (CM). It was found that more primary air and/or a deeper fuel bed reduces tars in the producer gas and increases the combustion efficiency, especially from low-ash-containing WP. At higher air supply rates, indications of a strong influence from the fuel ash content on the emissions were found. Although more combustible gases and fewer tars are produced in the conversion process, a substantial increase in particulate matter (PM) emissions is noted. At low air supply rates, the emissions of particulates with an aerodynamic diameter ≤ 2.5 μm (PM$_{2.5}$) released from the combustion process are in the range of 6–30 mg·MJ$^{-1}_{released}$ (WP<WS<SM<CM), low when compared with similar devices. However, when the air supply is increased by a factor of three, the PM$_{2.5}$ emissions almost double for WP and increase more than ten fold for CM. At lower air supply rates, low emissions of both PM and CO are achieved. This is likely due to lower peak temperatures (reducing ash devolatilisation) and larger char yields (to retain ash particulates) from the thermochemical solid biomass conversion process. This shows that low air supply rates and the combined production of heat, for cooking, and char, for subsequent application, may achieve substantial benefits for the emissions of pollutants from gasifer cookstoves.

Keywords:
Combustion, Emissions, Biomass, Particulate Matter, Cookstove

Declarations of interest: none

*Corresponding author, currently at Leibniz-Institut for Agricultural Engineering and Bioeconomy
Email address: `TKirch@atb-potsdam.de` (Thomas Kirch)

1. Introduction

The emissions from cookstoves burning solid biomass contribute considerably to anthropogenic emissions, causing a multitude of adverse effects on the present-day climate [1] as well as human health [2, 3]. These effects are mainly caused by products from incomplete combustion. Variation in feedstock has been linked to increased emissions from incomplete combustion, with animal manures tending to be worse than agricultural residues and woody biomass [4]. These feedstocks are widely being used to fuel cookstoves, and are known to produce high levels of pollutant emissions. Therefore, investigating approaches to mitigate pollutant emissions when using a variety of fuels is an important area to improved combustion performance.

One particular type of cookstove, called gasifier or semi-gasifier stove, is recognised as having potential for efficient and fuel flexible applications [5, 6]. In this type of stove, the thermochemical conversion of the solid fuel is separated from the combustion process of the released products. They tend to be batch-fed systems, where the fuel bed is lit on its top surface, while limited air is supplied from beneath the bed. This leads to the formation of a reaction front in which an autothermal reverse downdraft process converts the solid biomass to form gases and liquids, which are released from the bed as producer gas, leaving solid char as residue [7]. The producer gas is combusted with additional air downstream of (above) the fuel bed. Many studies have investigated the overall efficiency of unique experimental cookstoves [8, 9, 10, 11] and single [5, 12, 13] or multiple [6, 14, 15] commercially available cookstoves while emulating user practices (i.e. performing cooking tasks) . When emulating user practices, the analysis of the combustion process itself is limited as there are additional influences on the process, such as flame quenching on the surface of the pot or inconsistent user fire tending, which impede the study of the underlying combustion process and their quantification is problematic [16]. Whilst such testing methodologies are critical for establishing the suitability of a particular stove for practical application, fundamental studies using well-controlled environments and conditions are needed to provide a deeper understanding of the combustion processes in such systems—in particular, by focusing only on the thermochemical conversion and combustion processes [17].

Investigations of the thermochemical conversion process within the fuel bed have shown that small changes of process parameters, such as reactor diameter or air supply [18], and fuel, including wood [19], miscanthus [20] or rice hulls [21], can lead to a large variation in the producer gas composition. The influence of the fuel bed depth or the utilisation of high ash content fuels, such as manures, has yet to be evaluated. The complexity of producer gas combustion in small-scale applications stems from the presence of a multitude of chemical compounds, including gases, complex organic compounds (tars) and ash constituents, in the thermochemical conversion products [22, 23]. The combustion process of such producer gas from biomass fuels is highly complex, particularly due to the presence of a wide variety of tars, which have also been identified as soot precursors [24], and due to ash, which can have an influence on the combustion chemistry [25]. To enable cookstove improvement, establishing a possible relationship between the producer gas combustion products and organic compounds in the producer gas, as well as ash constituents in the fuel, is necessary.

Previous work by the authors has investigated the thermochemical conversion process and product composition for wood pellets [26] as well as wheat straw, and sheep and cow manure [27] as fuels, in a particular research gasifier cookstove. The air supply was varied for all fuels and additionally the fuel bed depth was varied for the wood pellets. The release of producer gas was the specific focus in those studies. In gasifier cookstoves, the producer gas released from the thermochemical conversion process provides the fuel for a secondary combustion process, which

2

occurs in the form of a non-premixed jet flame when secondary air is added. In the present study the focus of the analysis is placed on the secondary combustion process of the products from the thermochemical conversion process.

The emissions released from the combustion process from a small-scale gasifier stove are analysed in this study. Four biomass fuels, with a wide range of ash contents, up to four air supply configurations and for one fuel, also four fuel bed depths are investigated. Investigations of the products from the thermochemical conversion process of the solid fuel, which provide the fuel for the combustion processes investigated in this study, have previously been published [26, 27]. The main focus is on CO and particulate matter emissions, because of their particular interest for human health and environmental pollution. A deeper understanding of the producer gas combustion process and the influence of gaseous, organic and ash constituents and their contribution to the combustion emissions is the aim of this study. The influence of the combined production of heat, for cooking, and biochar, for subsequent utilisation, on the combustion emissions when utilising a variety of biomass fuels is investigated. The novelty of this work consists of the analysis of the secondary combustion process while considering the composition of the combusted producer gas as well as the produced char.

2. Materials and Methods

2.1. Fuels

Four biomass fuels, were utilised in the present investigation, namely wood pellets, wheat straw, sheep manure and cow manure. These have previously been analysed and described in detail [26, 27]. The proximate and ultimate analyses of all fuels, as well as their calorific value are provided in Table 1.

Wood pellets (WP), with a nominal diameter of 6.5 mm and length of 40 mm, were produced by Pellet Heaters Australia and were purchased from Barbeques Galore (Adelaide, Australia). These pellets consist of compressed hammer-milled wood shavings and saw dust. The wheat straw (WS) was from Reynolds, South Australia, Australia, and had been cut to a nominal length of 5 to 7 mm. Sheep manure (SM) was collected at a shearing station at Mallala, South Australia, Australia. Cow manure (CM) was provided by the Minko North Dairy Farm at Korunye, South Australia, Australia. Prior to testing, both manures were dried at 105 °C overnight, before being stored for the experiments, to achieve a comparable low moisture content.

2.2. Test-facilities

Figure 1(a) shows an outline of the facilities, consisting of a square enclosure of 600 mm by 600 mm with a height of 1800 mm, and a 45° inclined hood that contracts into a 150 mm diameter duct in which two baffles are inserted. Isokinetic sample extraction was performed at 12 diameters (1800 mm) downstream of the second baffle. These facilities adhere to specifications provided in ISO 19867-1: 2018. The validity of gaseous measurements at the measurement location has been investigated using computational fluid dynamics and shown to be accurate in the current configuration [28].

2.3. Reactor

The reactor is presented in Figure 1(b) and has previously been described, by Kirch *et al.*, in detail [26]. It has an inner diameter of 98 mm (1D) and a variable fuel bed height of 100–400 mm

Table 1: Proximate and ultimate analyses results (%) and the calorific value of the fuels. All measurements are reported on a mass basis and moisture, volatile matter (VM), fixed carbon (FC) and ash reported on a dry basis. The ultimate analysis (CHNO) and the higher heating value (HHV) are reported on a dry ash free basis. For the proximate and ultimate analyses the standard error of the mean is provided. The mean bulk density is also included.

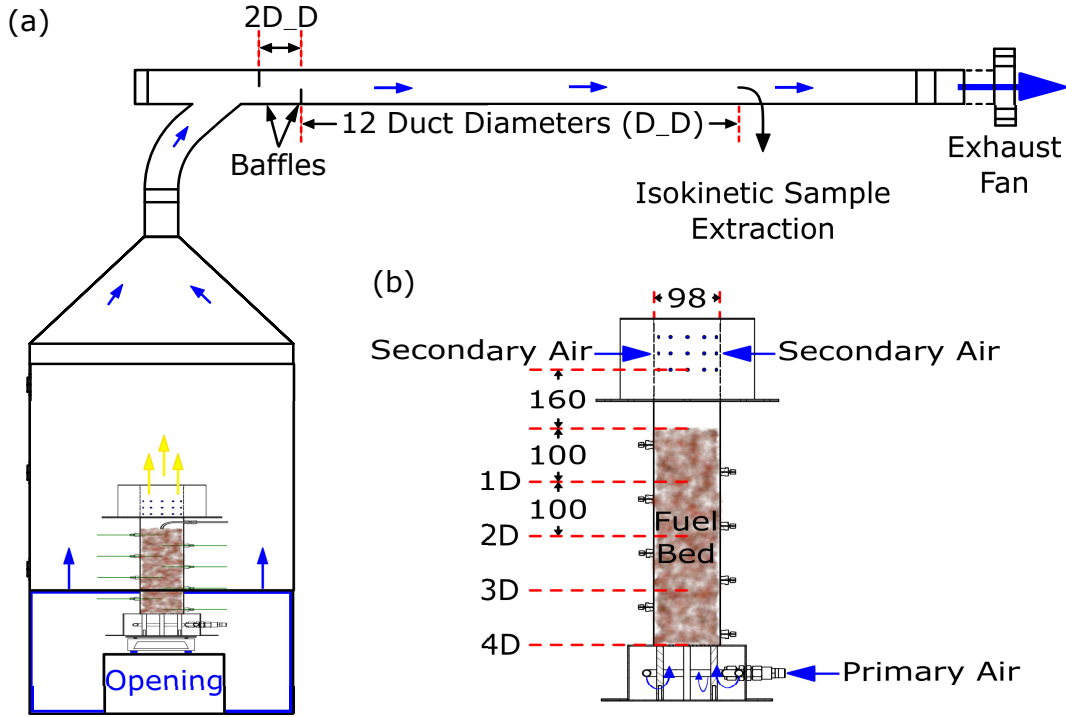| Fuel	Moisture	VM	FC	Ash	HHV	Bulk Density
	O	H	C	N	$(MJ \cdot kg^{-1})$	$(kg \cdot m^{-3})$
Wood Pellets	7.1 ± 0.3	82.4 ± 1.2	17.3 ± 0.5	0.3 ± 0.0	18.8	696
	45.9	6.4 ± 0.1	47.5 ± 0.0	0.1 ± 0.0		
Wheat Straw	7.9 ± 0.3	74.7 ± 3.1	20.0 ± 2.8	5.3 ± 0.4	20.1	166
	45.9	6.4 ± 0.1	43.5 ± 0.4	1.5 ± 0.1		
Sheep Manure	8.3 ± 0.5	58.6 ± 0.7	16.8 ± 0.3	24.6 ± 0.6	21.1	300
	41.3	6.3 ± 0.1	49.0 ± 0.5	3.3 ± 0.0		
Cow Manure	6.2 ± 0.4	50.8 ± 0.9	15.8 ± 0.3	33.3 ± 2.3	20.6	300
	42.5	5.9 ± 0.0	47.5 ± 0.1	4.1 ± 0.0		

Figure 1: (a) The schematic diagram of the exhaust system is presented, including the emissions monitoring set-up. Distances in the exhaust system are provided in relation to the duct diameter (D_D). (b) The experimental gasifier stove is shown. Bed depths are provided in relation to the reactor diameter (D) and dimensions are provided in mm.

(1D–4D). A primary air flow is provided from below and secondary air is supplied downstream of the fuel bed, by dry compressed air. The secondary air inlet consists of a total of 36×6-mm diameter holes, evenly spaced around the circumference in three vertically-aligned rows 30 mm apart. Previous studies [27, 26] have presented measurements upstream of the secondary air inlets,

4

to focus on the characterisation of the producer gas only.

2.4. Analysis Equipment

The gas analyser utilised was a Testo 350XL. This analyser uses an infra-red sensor, with a resolution of 0.01%, for the measurement of CO_2. CO is measured via an electrochemical sensor with a resolution of 1 ppm for low emission levels (<2000 ppm) and 5 ppm for high emission levels (>2000 ppm). Both CO and CO_2 emissions concentrations were recorded at 1 Hz, on a dry basis. The analyser was calibrated daily.

Optical particulate concentration measurements of the fraction with an aerodynamic diameter (d) 0.1<d<2.5 μm were performed using a TSI DustTrak II aerosol monitor model 8531. This provides real-time particulate concentrations in the exhaust flow at 1 Hz, with a readability of 0.001 mg·m^{-3}, a resolution of 0.1% of the reading and a range <400 mg·m^{-3}. Blank measurements showed a time-weighted average ambient particle concentration of 0.031 mg·m^{-3} and real-time concentration measurements were calibrated to the gravimetric particle measurement (more details are provided in Section S 1.1 in the Supporting Information).

2.5. Gravimetric Particulate Measurements

Gravimetric measurements of all particulates and the fraction with an aerodynimc diameter ≤2.5 μm were performed. Sample extraction occurred isokinetically at 12 duct diameters downstream of the second baffle, as indicated in Figure 1(a). An external filter holder upstream of the Testo 350XL gas analyser was used to collect all particulates, while an internal filter holder in the DustTrak II provided the fraction ≤2.5 μm. In both cases, PTFE membrane filters with a pore size of 0.22 μm and a diameter of 37 mm were used. The filter media was desiccated and weighed with a readability of 0.01 mg. After the experiment, the filter media was desiccated for >24 h, weighed, returned to the desiccator for >24 h and weighed again. If the second measurement was within ±0.05 mg of the first, the first measurement was accepted, in accordance with ISO-19867-1:2018. Blank measurements of the ambient particle concentration were performed and the measurements were found to be below the gravimetric limit of detection of 0.01 mg.

2.6. Procedure

The test procedure has been described previously [26] and is only briefly outlined here. To avoid influences from the large thermal mass of the reactor when performing replicate tests, the reactor was initially preheated and all tests were started with reactor wall temperatures <100 °C. Fuel was supplied to the reactor in batches. To ignite the fuel batch, 10 mL of methylated spirits (96 % ethanol, CAS # 64-17-5) and a paper towel were supplied to the top of the fuel bed. When the entire fuel batch was converted to char, the process was quenched by adding water ice from the top of the reactor and nitrogen (>99.99 % N_2), instead of air, from below the fuel bed to cool the process and end all ongoing reactions. Multiple repeats for each tested fuel were performed at two to four air supply mass flux and multiple fuel bed depths in the case of wood pellets, as presented in Table 2.

2.7. Analysis

The total amount of CO released was calculated on the basis of the concentration and the total duct flow rate. The total amount of CO released as well as the gravimetric PM measurements are mean values of all replicate tests (refer to Table 2) and were normalised to the amount of

Table 2: Experimental configurations: the fuel type, the fuel bed depth (BD, reported in (mm) and in relation to the reactor diameter (D)), the primary air supply (PA), the secondary air supply (SA), the total air supply (A), the number of repetitions performed.

Fuel type (Abbreviation)	BD (mm)	(D)	PA (kg·m^{-2}·s^{-1})	SA (kg·m^{-2}·s^{-1})	A (kg·m^{-2}·s^{-1})	Repetitions
Wood pellets	100	1	0.025	0.100	0.125	2
(WP)	100	1	0.050	0.200	0.250	3
	100	1	0.075	0.300	0.375	8
	100	1	0.125	0.500	0.625	4
	200	2	0.075	0.300	0.375	5
	300	3	0.075	0.300	0.375	4
	400	4	0.025	0.100	0.125	6
	400	4	0.050	0.200	0.250	7
	400	4	0.075	0.300	0.375	6
	400	4	0.125	0.500	0.625	7
Wheat straw	400	4	0.025	0.100	0.125	4
(WS)	400	4	0.075	0.300	0.375	4
Sheep manure	400	4	0.025	0.100	0.125	4
(SM)	400	4	0.075	0.300	0.375	7
Cow manure	400	4	0.025	0.100	0.125	3
(CM)	400	4	0.075	0.300	0.375	4

energy released from the fuel ($E_{released}$ = E_{fuel} - E_{char}), as described previously [26, 27]. Gravimetric measurements of $PM_{2.5}$ and total PM were collected and $PM_{>2.5}$ was calculated as the difference of the two measurements.

To account for dilution in the exhaust system and determine a quantitative value for the combustor efficiency, the measured gaseous emissions were normalised. The utilised normalisation relates the carbonaceous products of complete combustion to all gaseous carbonaceous combustion products. It must be considered that the only gaseous carbonaceous species considered in the normalisation were CO and CO_2, because the hydrocarbon emissions were below the detection limit of 100 ppm. The nominal combustion efficiency (NCE), which is also referred to as modified combustion efficiency (MCE), has been previously established for the evaluation of cookstoves [29] and was calculated using the mole fraction (x) of CO and CO_2 via Equation 1:

$$NCE = \frac{x_{CO_2}}{x_{CO_2} + x_{CO}} \tag{1}$$

Average profiles of the NCE for each configuration were calculated. To achieve this, real time NCE values for each individual test were calculated over the duration of the process and mean values established for each configuration. The NCE results are presented in the Supporting Information.

3. Results

3.1. Particulate Emissions

Figure 2 reports profiles of the $PM_{2.5}$ mass concentration in the exhaust stream over the duration of the experiments, for a range of fuels and primary air supply flux. A representative plot of one experiment for each configuration is shown, although all cases were repeated multiple times (refer to Table 2). The $PM_{2.5}$ mass concentration ($mg \cdot m^{-3}$) in the exhaust gas flow is presented as a function of time for the four fuels in (a) WP, (b) WS, (c) SM and (d) CM. Measurements of multiple primary air supply mass flux, 0.025–0.125 $kg \cdot m^{-2} \cdot s^{-1}$, are shown as indicated in the respective legends. The air supply mass flux is provided on the basis of the reactor cross-sectional area. Spikes at the end of each test indicate the start of increasing char gasification and decreasing fuel conversion within the thermochemical conversion process of the solid fuel.

For wood pellets (WP), the $PM_{2.5}$ concentration decreases as a function of time for all air supply flux. Previously reported findings of the thermochemical conversion process in the solid fuel bed have shown that as the reaction front propagates down the bed and char accumulates downstream of the reaction front, the produced tars can crack [26]. This transforms tars into simpler combustible gases upstream of the secondary air inlet as the reaction front moves down the bed over time. Therefore, the noticeable decrease of $PM_{2.5}$ emissions along the temporal axis could be due to lower tar concentrations in the producer gas. Increasing primary air supply flux have been shown to lead to greater conversion temperatures throughout the fuel bed and a reduction in the overall tar yield [26]. Simultaneously higher temperatures will also cause a change in the composition of tar constituents. It is well understood that the tar fractions which have been identified as a soot precursor are polycyclic aromatic hydrocarbons (PAH) [30, 24, 31]. PAHs are formed within the solid fuel bed at temperatures of >800°C with increasing complexity at higher temperatures [32]. For all fuels investigated, these temperatures only occur at air supply flux of >0.050 $kg \cdot m^{-2} \cdot s^{-1}$ [26, 27]. Therefore, the similar real-time concentrations of $PM_{2.5}$ at 0.025 $kg \cdot m^{-2} \cdot s^{-1}$and 0.050 $kg \cdot m^{-2} \cdot s^{-1}$and the subsequent slight increases may be related to an increasing PAH concentration in the producer gas.

Wheat straw (WS) exhibits a slightly different profile in Figure 2, compared with WP. While at 0.025 $kg \cdot m^{-2} \cdot s^{-1}$primary air supply, similar to WP, a reduction of the $PM_{2.5}$ over time can be noticed, which is not the case at 0.075 $kg \cdot m^{-2} \cdot s^{-1}$. This suggests that either tar cracking in the char bed occurs only at low primary air supply flux or more likely that at high air supply, other factors, such as the fuel ash content, become more influential.

For both manures, sheep (SM) and cow (CM), the trends appear similar in Figure 2. There is no reduction of the $PM_{2.5}$ concentration over time at 0.025 $kg \cdot m^{-2} \cdot s^{-1}$, which suggests that tar cracking with an increasing char layer downstream of the reaction does not notably influence the emissions. Similarly to WS this suggests that other factors, such as the fuel ash content, are more influential. The overall trend of lower $PM_{2.5}$ concentrations over time at all flow rates for WP, lower $PM_{2.5}$ concentrations over time only at low primary air supply flux for WS and no reduction in $PM_{2.5}$ concentration for the manures, suggests that an increasing ash content (CM>SM>WS>WP) has a greater influence on the emissions than the combustion of tars, or potentially the ash could inhibit tar cracking in the produced char layer. It must be considered that with increasing ash content in the fuel, the conversion of fuel carbon to char decreases substantially when increasing the air supply, as presented previously [27]. This leads to a rapid decrease of the fixed carbon content of the produced char. It is possible that either or both mechanisms may influence tar cracking in the char layer, but this is beyond the current scope of work.
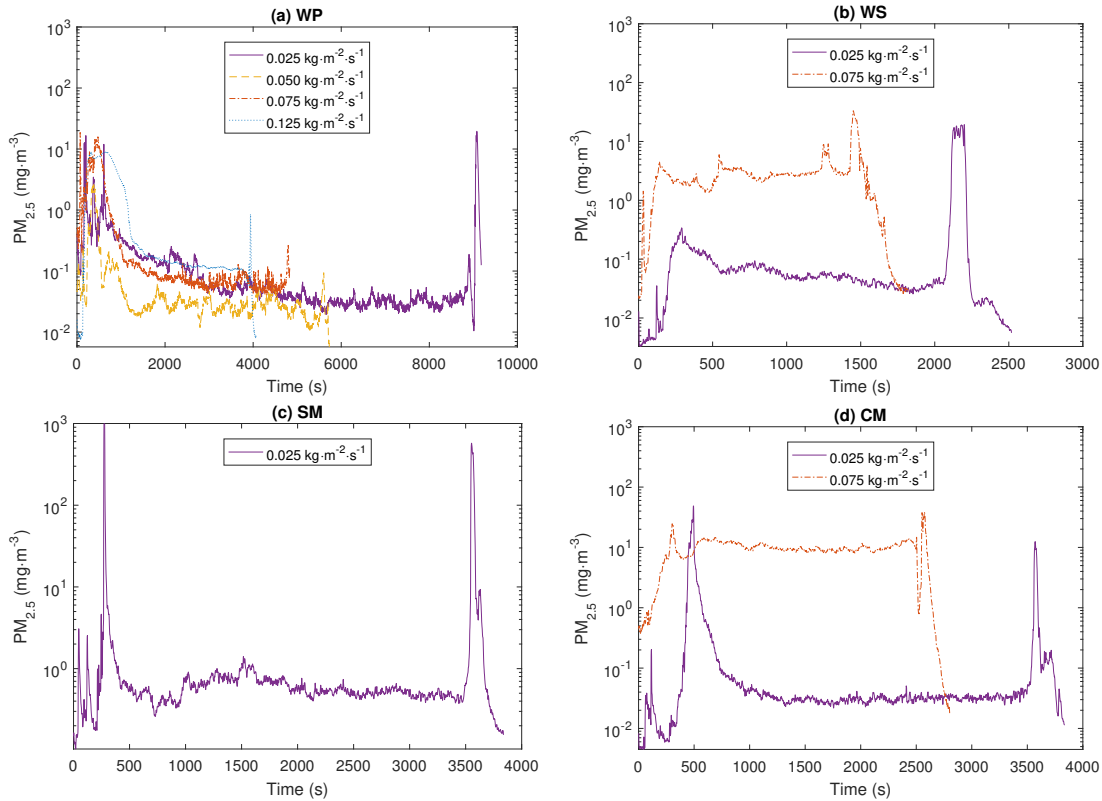
Figure 2: Temporal concentration of $PM_{2.5}$ mass in the exhaust gas stream, for all fuels: (a) wood pellets, (b) wheat straw, (c) sheep manure and (d) cow manure. The fuel bed depth is 4D. Air supply flux of 0.025–0.125 $kg \cdot m^{-2} \cdot s^{-1}$ are indicated in the respective legends.

Figure 3 presents the mean gravimetric measurements of: (a) $PM_{2.5}$ and (b) total PM for each configuration under investigation. For each configuration, the quantity of particulates released has been normalised to the mean energy released from the fuel ($E_{released} = E_{fuel} - E_{char}$) for each configuration. A logarithmic representation of the normalised particulate emissions was chosen to accommodate large variations in between configurations (numerical values, as well as further information such as the total heat release (firepower), are provided in the Supporting Information in Table S1).

In Figure 3(a), the $PM_{2.5}$ emissions from WP are similar at one diameter fuel bed depth (1D) with an increasing air supply flux, while they increase at 4D. The discrepancy of $PM_{2.5}$ between 1D and 4D could be due to a lower influence of transients, during lighting and quenching. With only 1D, transients are particularly influential due to the short length of the overall duration of the process (refer to Table S1), leading to a greater effect on the total emissions. Previous research has also found that during transient events, organic compounds dominate the emissions, while during steady-state combustion, inorganic compounds are of greater importance [33]. At 4D, the increase in the $PM_{2.5}$ emissions from WP with increasing air flow rate could be related to soot formation (which contributes mainly to $PM_{2.5}$ rather than larger particulates). Soot formation
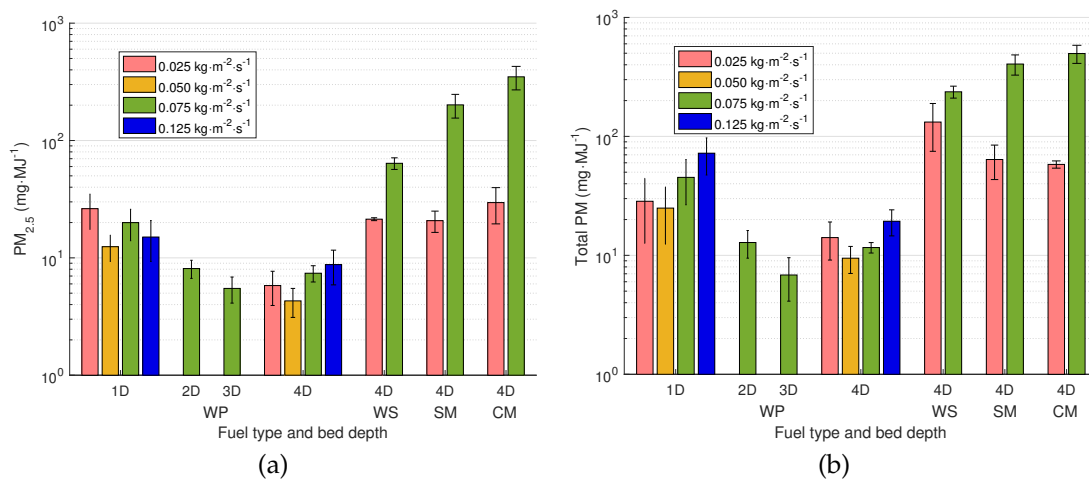
Figure 3: The gravimetric particulate emissions of (a) the $PM_{2.5}$ and (b) the total PM, normalised to the energy released from the fuel, are presented on a logarithmic scale for all investigated configurations and fuels. The error bars present the standard error of the mean.

could be the result of an increasing PAH content in the producer gas, as discussed earlier in this section. The results also suggest that mechanisms apart from soot influence the $PM_{2.5}$ emissions. Similar trends have previously been observed for incomplete combustion emissions from biomass fuels, such as CO and soot [34]. It can be seen that for WP the release of CO decreases while the related nominal combustion efficiency (refer to Figure S1) increases with the increasing air supply, as described in more detail in the subsequent section. Thus more complete combustion, leading to less CO and soot, would be expected, as discussed in more detail in the following section. Therefore, the increase in PM emissions with the air supply could be due to influences other than incomplete combustion. Two mechanisms, namely the entrainment of ash particles in the gas stream and increasing devolatilisation of fuel ash constituents, could be influential [35]. Especially at high air supply flux, when little fixed carbon remains in the char product, ash constituents will become loose on the particle surface, as described previously [27]. These loose ash particles can be entrained more easily by the surrounding gas flow, for which the velocity also increases with the amount of air supplied. Furthermore, an increasing air supply leads to increasing fuel bed temperatures which will cause an enhanced devolatilisation of fuel ash constituents [36, 37, 38].

For WS, SM and CM a more substantial increase in the PM emissions can be noted with an increasing primary air supply. In Figure 4 the $PM_{2.5}$ and $PM_{>2.5}$ emissions are presented as a function of the fuel ash content. At high air supply, the overall trend of $PM_{2.5}$ can be related to the fuel ash content and follows the relationship WP<WS<SM<CM. As stated previously, ash constituents can be entrained in the gas stream or devolatilised from the fuel bed. Previously it has been suggested that especially S, Cl, K and Na, which are typically contained in many biomass fuels, even if present only in trace amounts in a combustion process, can have an impact on the formation of emissions, including CO and PM [39, 25]. The effects of certain compounds on the combustion chemistry, especially on producer gas combustion are largely unknown. Therefore, an increasing amount of ash in the fuel may lead to higher concentrations of ash constituents participating in the combustion process, which may contribute to the increasing $PM_{2.5}$ emissions.
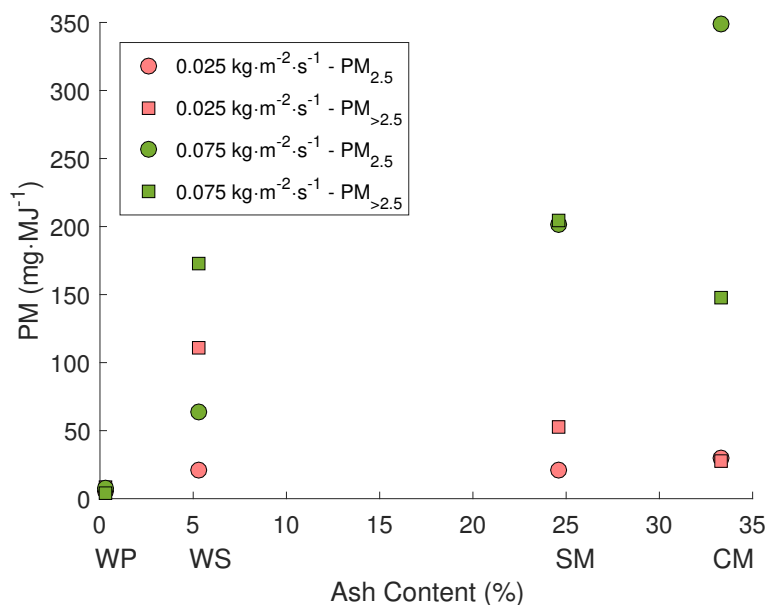
9

Figure 4: The mean $PM_{2.5}$ and $PM_{>2.5}$, normalised to the energy released from the fuel, as a function of the fuel ash content are presented.

The trends of the total PM in Figure 3(b) appear only slightly different, in comparison with $PM_{2.5}$. For all fuels a higher air supply flux leads to greater emissions. This increase in total PM emissions can also be related to greater entrainment of coarse fly ash ($>10\mu$m) particles with higher gas stream velocities. This contribution to total PM has previously been demonstrated for fixed-bed wood pellet combustion at similar primary air supply flux [40] and could affect the results here similarly. In Figure 4 for the $PM_{>2.5}$ emissions it can be noticed that while they are very similar for all non-woody biomass fuels at 0.075 $kg \cdot m^{-2} \cdot s^{-1}$ air supply, WS exhibits the largest emissions at 0.025 $kg \cdot m^{-2} \cdot s^{-1}$.

Overall, it can be noticed that low air supply flux lead to very efficient combustion and low emissions of PM from all fuels. The production of char and the retention of a large fraction of the ash in its structure appears to enable the much cleaner combustion of high ash-content fuels compared with high air supply flux [17, 26] (the elemental composition of the char produced at low air supply from WP, WS and SM is presented elsewhere [41]). Therefore, it would be beneficial to design cookstoves to utilise a low air supply flux, while providing sufficient fire power for cooking, by adjusting the cross sectional area and to aim at producing char as a solid product rather than combusting it.

3.2. Gaseous Emissions

Figure 5 presents the accumulated CO emissions, normalised by the energy released from the fuel, for all investigated fuels and process configurations. CO is the main gaseous emission from incomplete combustion and a significant health concern for users. An analysis of real-time gaseous emissions measurements and their relationship to the efficiency of the combustion process is provided in the Supporting Information in Section S2.

10

Increasing the air supply flux for WP leads to a reduction in CO, however, the opposite trend is apparent for the manures, and no clear trend is measured for WS. Extremely low CO emissions can be seen in all cases for WP, especially when increasing primary air supply flux >0.025 kg·m^{-2}·s^{-1}, as well as with increasing fuel bed depths. A lower tar fraction from the conversion process before secondary combustion and higher cold gas efficiencies with increasing air supply flux as well as fuel bed depths, have been presented previously [27] and the resultant higher gas quality for combustion, leading to lower CO emissions, corroborates those findings here.

The trend of greater CO emissions with increasing air supply flux for the manures, and to some degree for WS, is similar to the PM emissions (refer to Figure 3). This suggests that particulate matter constituents, presumably ash constituents, influence the combustion process and the release of CO emissions. In regard to the producer gas composition, all fuels have been shown to behave similarly, with lower tar yields and higher gas quality at higher primary air supply flux [27]. This strengthens the argument that fuel ash constituents influence the formation of emissions from incomplete combustion. When focusing on the manures, it can be seen that especially low CO emissions are released at 0.025 kg·m^{-2}·s^{-1} from SM but higher emissions are released from CM, while at high air supply flux the values are similar for both manures. The particularly low emissions at low air supply flux highlight the potentially high efficiency of the combustion process, even when utilising high ash content fuels.

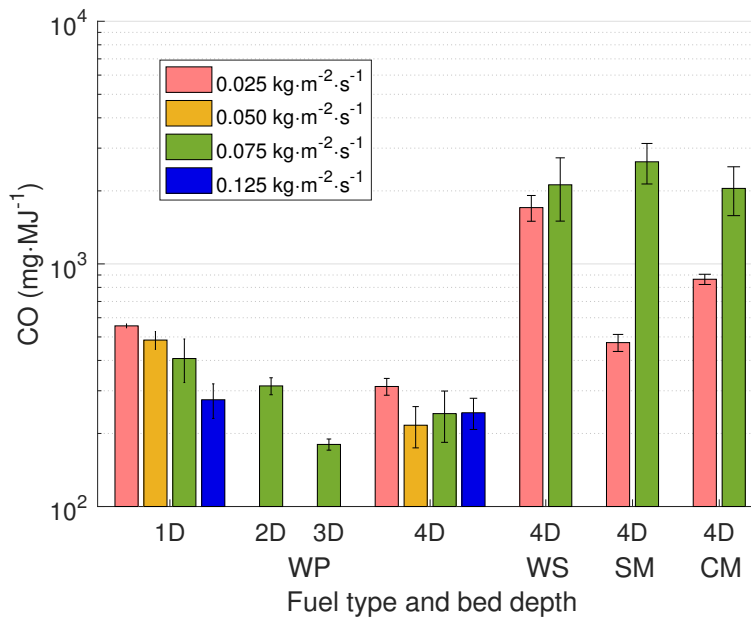Figure 5: The emissions of CO normalised to the energy released from the fuel are presented on a logarithmic scale for all investigated configurations and fuels. The error bars present the standard error of the mean.

4. Discussion

The three mechanisms for PM emissions discussed here are; (1) incomplete combustion, leading to carbonaceous products, and release of ash constituents via (2) devolatilisation, and (3) en-

trainment. To achieve a reduction in PM emissions, each must be considered as individual but interconnected mechanisms. Different approaches are necessary for their mitigation, while currently the main approach for emissions reduction from cookstoves is to achieve more complete hydrocarbon combustion, while often disregarding the influence of ash constituents. As noticed here, a higher primary air supply can reduce tars in the producer gas and reduce incomplete combustion leading to lower CO emissions, notably from low ash content wood pellets. Simultaneously, the emissions of PM in all cases and CO for the higher ash content non-woody biomass fuels increase with the air supply, which can be related to the fuel ash content.

A previous investigation of multiple wood species, crop residues and cow manure, completely combusted in a traditional Indian stove [42], has shown that an elemental analysis of the $PM_{2.5}$ emissions could account for ≈50% of the total mass (the rest is assumed to be oxygen). These ≈50% were made up about half by C—lowest from cow manure—and ash constituents, mainly ions, K and Cl as well as ammonia—all highest from cow manure [42]. This demonstrates the significance of ash constituents in the PM emissions, especially from manures, while their influence on the combustion process [25], as well as their total contribution for a particular fuel, are largely unknown. This fraction of the PM, containing ash constituents, cannot be addressed through an increase in efficiency of the combustion process [43], but necessitates alternative approaches and considerations. Alternative approaches may include air control or the production of char, as investigated here, but also others such as fuel additives or the integration of advanced materials or even filters into the stove design. Here low air supply flux lead to very efficient combustion and low emissions of CO and PM for all fuels. Adapting the reactor cross sectional area to accommodate low air supply flux, while still providing sufficient firepower for cooking, could be an approach to reduce pollutant emissions. This would lead to a larger reactor diameter, while the height might need to be reduced. As it has also been shown here that a greater reactor height leads to lower emissions [26], its reduction might be a drawback. Additionally, the reactor dimensions would then only be adapted to a specific fuel. A modular cookstove design where multiple reactor sizes may be utilised could be a solution, but would substantially increase the systems complexity. Furthermore, it has been shown here that the production of char and the retention of a large fraction of the ash in its structure enables much cleaner combustion of high ash-content fuels. Since the char contains large amounts of energy, it might not be in the user's interest to retain it. Therefore, a clear incentive, most likely an economic benefit, is necessary to make the production of char attractive. Increasing utilisation of biochar as a soil amendment [44] or as additive to anaerobic digestion [41, 45] could provide such incentives. More rigorous investigation and in-depth discussion of the fate of ash constituents in biomass combustion and mechanisms for their mitigation are therefore of importance.

To demonstrate the benefits of avoiding flame quenching by the pot and of the production of solid char in the presented gasifier cookstove these results are compared with values found in the literature from similar gasifier cookstoves, which were tested while performing cooking tasks. Figure 6 presents a comparison of the CO and $PM_{2.5}$ emissions of the present investigation and results found in the literature from similar devices with similar fuels. One study investigated the combustion process in a medium-size gasifier stove and reports the emissions on the basis of energy released from the fuel [46], similar to the present study. The remaining studies utilise standardised test protocols to evaluate the stove performance, either through the international [10, 47, 48, 6] or the Chinese [14, 12, 13] Water Boiling Test, where the emissions are reported on the basis of energy delivered to a cooking vessel contents. Since in most studies the results are reported on the basis of energy delivered to a cooking vessel, rather than on the basis of the energy

12

released from the fuel (see Figures 3, 4 and 5), a moderate heat transfer efficiency of 35% [47] is assumed for the results from the present investigation, for better comparability.
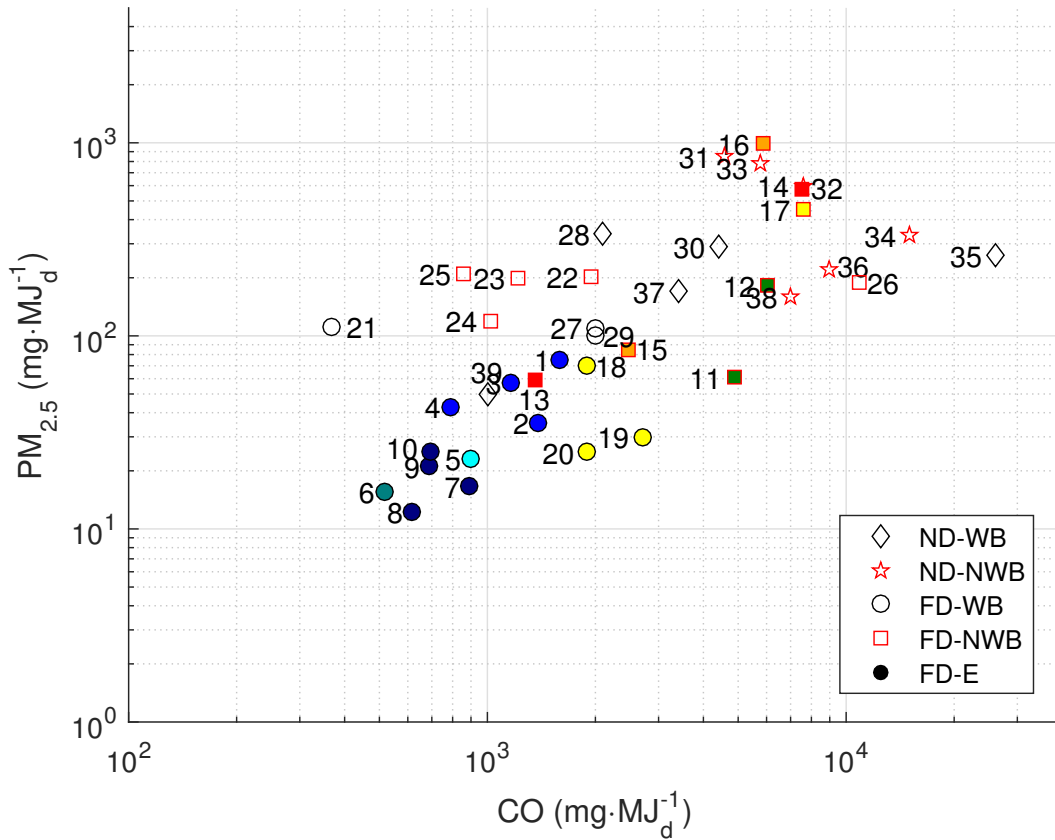

Figure 6: Results from the present investigation, under the assumption of a moderate thermal efficiency of 35%, as well as values found in the literature for $PM_{2.5}$ and CO emissions normalised to the energy delivered to the cooking vessel contents are presented. Three types of gasifier stoves: forced draft stoves (FD), forced draft experimental reactors (FD-E, face colours reflect different reactor configurations) and natural draft stoves (ND), using woody biomass (WB, red edge colour) or non-woody biomass (NWB, black edge colour) are included. Multiple stoves and fuels are investigated: Present investigation, (1–4, blue) 1D, wood pellets (WP); (5, cyan) 2D, WP; (6, teal) 3D, WP; (7–10, navy) 4D, WP; (11–12, green) 4D, wheat straw; (13–14, red) 4D, sheep manure; (15–16, orange) 4D, cow manure; (17–20, yellow) [10]; (21–22) [46]; (23–25) [14]; (26–27) [47]; (28–29) [48]; (30) [12]; (31–33) [13]; (34–39) [6].

It can be seen in Figure 6 that very low emissions of PM and CO are only reached with wood fuels in literature, mostly pelletised, while here this can also be achieved using non-woody biomass. Lowest emissions are achieved in stoves specifically designed for experimental purposes with >1D fuel bed depth. Very consistent conditions in the experimental stoves and a reduction of the influence of transients, due to the larger depth, will contribute to their better performance. Furthermore, the present investigation is the only case where insulation material surrounds the combustion chamber, leading to lower heat loss and presumably higher efficiency of the thermochemical conversion. The apparent direct relationship between the $PM_{2.5}$ and CO emissions, as they increase simultaneously and not independently, in Figure 6 should be noted. This further

suggests that both CO and PM emissions cannot only be related to incomplete combustion of the carbonaceous fuel, but also that CO emissions may be influenced by the same mechanisms that lead to the release of particulates, as described in the Gaseous Emissions Section. Additionally, two general trends can be noticed in Figure 6: (1) forced draft stoves perform better in terms of emissions than natural draft stoves and (2) woody biomass achieves lower emissions than non-woody biomass.

For both CO and PM emissions, better results are achieved using forced draft, particularly in well-controlled experimental stoves. Similar to previous studies, this suggests that a constant, rather than buoyancy driven, air supply achieves better mixing of air with the combustible gases to aid more complete combustion [17]. While the emissions of PM can be due to multiple influences, as discussed earlier in this section, CO is released as a result of inefficiency of the combustion process. Lower emissions achieved in the present study (Figure 6 1–16), when compared with investigations emulating user practices (Figure 6 17–22 and 26–39), could also be due to the absence of a pot in the secondary combustion zone. In theory, the combustion process should be completed upstream of a pot and only hot gases would get in contact with the pot. If at any time the secondary combustion zone extends to the surface of the pot, local flame quenching will occur leading to the release of increasing products from incomplete combustion, which is avoided in the present investigation.

When considering the difference between woody and non-woody biomass, especially the very low ash content of woody biomass in comparison with other biomass fuels must be considered. As the influence of ash constituents on the combustion process is still largely unknown, as described above, this adds complexity to the discussion of fuels with increasing ash content. Many more parameters, such as bulk density, moisture content or fuel particle size could be of influence, but their in depth discussion is outside the scope of this work. For example, the analysis of the thermochemical conversion process of the investigated fuels has shown a slight increase of the peak reaction front temperature with greater fuel bulk density, which in turn affects the distribution across combustible products [27]. It is shown here, though, that in highly controlled conditions, very low emissions may be achieved with all investigated fuels, including the manures and wheat straw at low primary air supply flux. This demonstrates that a high fuel ash content is not necessarily a problem, but it appears that the production of char to retain a large fraction of the ash will be required to achieve low emissions from such fuels. It needs to be stressed that the results from the present study show that low value agricultural residues and even manures can be burned almost as cleanly as high quality wood pellets under controlled conditions in gasifier cookstoves. Therefore, gasifier cookstove designs should endeavour to accommodate low primary air flow rates, maximum reactor heights and promote the production of solid char, which have been shown as effective measures for emissions reduction.

5. Conclusions

The present study investigates the secondary combustion process in a small-scale batch fed cookstove. Multiple fuels, namely wood pellets, wheat straw, sheep manure and cow manure, have been tested, while the producer gas composition upstream of the secondary combustion zone has previously been reported. The products of the combustion process are analysed in light of the fuel, producer gas, and char product composition.

- Low tar concentrations in the producer gas from wood pellets reduces the emissions of CO. This is achieved with increasing air supply flux or greater fuel bed depth.

- Interestingly the previous trend does not extend to the PM emissions. Generally higher PM emissions have been found with increasing air supply, presumably due to the influence of the fuel ash constituents.

- Low air supply flux lead to very efficient combustion and low emissions of CO and PM for all fuels, in comparison with values found in the literature. Low primary air flow rates are therefore desired to minimise pollutant emissions.

- From all non-woody biomass fuels, the higher ash content, leads to increasing emissions of CO as well as PM with increasing air supply.

- A large fraction of CO and PM emissions is released during transient events at start-up and shut-down. In application PM emissions released at shut-down could be avoided by an isolated quenching method. Generally the reduction of emissions during transient events needs further scrutiny.

The strong influence of the ash fraction of the fuel is apparent. Especially at high air supply the influence of devolatilised and entrained particles leads to much greater emissions of PM for all fuels and of CO in case of the non-woody biomass. It was shown that a larger yield of carbon in the produced char retains a greater fraction of the ash and substantially improves the combustion performance. It appears that the production of char, or other measures to retain the ash in the bed, will be necessary to achieve low emissions from cookstoves, especially when using high ash content fuels.

6. Acknowledgements

7. Supporting Information

Additional information, supplementing results and numerical values of the presented results

[1] T. C. Bond, S. J. Doherty, D. W. Fahey, P. M. Forster, T. Berntsen, B. J. Deangelo, M. G. Flanner, S. Ghan, B. Kärcher, D. Koch, S. Kinne, Y. Kondo, P. K. Quinn, M. C. Sarofim, M. G. Schultz, M. Schulz, C. Venkataraman, H. Zhang, S. Zhang, N. Bellouin, S. K. Guttikunda, P. K. Hopke, M. Z. Jacobson, J. W. Kaiser, Z. Klimont, U. Lohmann, J. P. Schwarz, D. Shindell, T. Storelvmo, S. G. Warren, C. S. Zender, Bounding the role of black carbon in the climate system: A scientific assessment, Journal of Geophysical Research: Atmospheres 118 (11) (2013) 5380–5552.

[2] S. Bonjour, H. Adair-Rohani, J. Wolf, N. G. Bruce, S. Mehta, A. Prüss-Ustün, M. Lahiff, E. A. Rehfuess, V. Mishra, K. R. Smith, Solid fuel use for household cooking: Country and regional estimates for 1980-2010, Environmental Health Perspectives 121 (7) (2013) 784–790.

[3] B. N. Young, M. L. Clark, S. Rajkumar, M. L. Benka-Coker, A. Bachand, R. D. Brook, T. L. Nelson, J. Volckens, S. J. Reynolds, C. L'Orange, N. Good, K. Koehler, S. Africano, A. B. Osorto Pinel, J. L. Peel, Exposure to household air pollution from biomass cookstoves and blood pressure among women in rural Honduras: A cross-sectional study, Indoor Air (2018) 130–142.

[4] H. S. Mukunda, S. Dasappa, P. J. Paul, N. K. S. Rajan, M. Yagnaraman, D. Ravi Kumar, M. Deogaonkar, Gasifier stoves – science, technology and field outreach, Current Science 98 (5) (2010) 627–638.

[5] L. Deng, D. Torres-Rojas, M. Burford, T. H. Whitlow, J. Lehmann, E. M. Fisher, Fuel sensitivity of biomass cookstove performance, Applied Energy 215 (2018) 13–20.

[6] J. Tryner, B. D. Willson, A. J. Marchese, The effects of fuel type and stove design on emissions and efficiency of natural-draft semi-gasifier biomass cookstoves, Energy for Sustainable Development 23 (2014) 99–109.

[7] T. B. Reed, R. Larson, A wood-gas stove for developing countries, Energy for Sustainable Development 3 (2) (1996) 34–37.

[8] T. B. Reed, E. Anselmo, K. Kircher, Testing & Modeling the Wood-Gas Turbo Stove, in: A. Bridgwater (Ed.), Progress in Thermochemical Biomass Conversion Conference, Blackwell Science Ltd, Bodmin, 2001, pp. 693–704.

[9] C. Birzer, P. Medwell, J. Wilkey, T. West, M. Higgins, G. Macfarlane, M. Read, An analysis of combustion from a top-lit up-draft (TLUD) cookstove, Journal of Humanitarian Engineering 2 (1) (2013) 1–8.

[10] J. Tryner, J. W. Tillotson, M. E. Baumgardner, J. T. Mohr, M. W. Defoort, A. J. Marchese, The Effects of Air Flow Rates, Secondary Air Inlet Geometry, Fuel Type, and Operating Mode on the Performance of Gasifier Cookstoves, Environmental Science and Technology 50 (17) (2016) 9754–9763.

[11] J. Tryner, J. Volckens, A. J. Marchese, Effects of operational mode on particle size and number emissions from a biomass gasifier cookstove, Aerosol Science and Technology 52 (1) (2018) 87–97.

[12] H. Yibo, H. Li, X. Chen, C. Xue, C. Chen, G. Liu, Effects of moisture content in fuel on thermal performance and emission of biomass semi-gasified cookstove, Energy for Sustainable Development 21 (1) (2014) 60–65.

[13] Z. Zongxi, S. Zhenfeng, Z. Yinghua, D. Hongyan, Z. Yuguang, Y. X. Zhang, A. Riaz, C. Pemberton Pigott, D. Renjie, Effects of biomass pellet composition on the thermal and emissions performances of a TLUD cooking stove, International Journal of Agricultural and Biological Engineering 10 (4) (2017) 189–197.

[14] Y. Chen, G. Shen, S. Su, W. Du, Y. Huangfu, G. Liu, X. Wang, B. Xing, K. R. Smith, S. Tao, Efficiencies and pollutant emissions from forced-draft biomass-pellet semi-gasifier stoves: Comparison of International and Chinese water boiling test protocols, Energy for Sustainable Development 32 (2016) 22–30.

[15] Z. Zhang, Y. Zhang, Y. Zhou, R. Ahmad, C. Pemberton-Pigott, H. Annegarn, R. Dong, Systematic and conceptual errors in standards and protocols for thermal performance of biomass stoves, Renewable and Sustainable Energy Reviews 72 (17) (2017) 1343–1354.

[16] C. A. Roden, T. C. Bond, S. Conway, A. B. Osorto Pinel, N. A. MacCarty, D. Still, Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves, Atmospheric Environment 43 (6) (2009) 1170–1181.

[17] T. Kirch, C. H. Birzer, P. J. V. Eyk, P. R. Medwell, P. J. van Eyk, P. R. Medwell, Influence of Primary and Secondary Air Supply on Gaseous Emissions from a Small-Scale Staged Solid Biomass Fuel Combustor, Energy and Fuels 32 (2018) 4212–4220.

[18] Y. Mehta, C. Richards, Gasification Performance of a Top-Lit Updraft Cook Stove, Energies 10 (10) (2017) 1529.

[19] A. M. James R, W. Yuan, M. Boyette, The Effect of Biomass Physical Properties on Top-Lit Updraft Gasification of Woodchips, Energies 9 (4) (2016) 283–296.

[20] X. H. Pham, B. Piriou, S. Salvador, J. Valette, L. Van de Steene, Oxidative pyrolysis of pine wood, wheat straw and miscanthus pellets in a fixed bed, Fuel Processing Technology 178 (2018) 226–235.

[21] A. M. James R, W. Yuan, M. D. Boyette, D. Wang, Airflow and insulation effects on simultaneous syngas and biochar production in a top-lit updraft biomass gasifier, Renewable Energy 117 (2018) 116–124.

[22] T. C. Lieuwen, V. Yang, R. A. Yetter (Eds.), Synthesis Gas, Taylor & Francis, Boca Raton, 2009.

[23] C. Mandl, I. Obernberger, I. R. Scharler, Characterisation of fuel bound nitrogen in the gasification process and the staged combustion of producer gas from the updraft gasification of softwood pellets, Biomass and Bioenergy 35 (11) (2011) 4595–4604.

[24] M. Frenklach, Reaction mechanism of soot formation in flames, Physical Chemistry Chemical Physics 4 (11) (2002) 2028–2037.

[25] P. Glarborg, Hidden interactions—Trace species governing combustion and emissions, Proceedings of the Combustion Institute 31 (2007) 77–98.

[26] T. Kirch, P. R. Medwell, C. H. Birzer, P. J. Van Eyk, Influences of Fuel Bed Depth and Air Supply on Small-Scale Batch-Fed Reverse Downdraft Biomass Conversion, Energy and Fuels 32 (2018) 8507–8518.

[27] T. Kirch, P. R. Medwell, C. H. Birzer, P. J. Van Eyk, Small-scale autothermal thermochemical conversion of various solid biomass feedstocks, Renewable Energy (2019).

[28] T. Kirch, M. J. Evans, P. R. Medwell, V. H. Rapp, C. H. Birzer, A. J. Gadgil, Mixing uniformity of emissions for point-wise measurements in exhaust ducts, Proceedings of the 21st Australasian Fluid Mechanics Conference (2018).

[29] M. Johnson, R. Edwards, V. Berrueta, O. Masera, New approaches to performance testing of improved cookstoves, Environmental Science and Technology 44 (1) (2010) 368–374.

[30] M. Frenklach, D. W. Clary, W. C. Gardiner, S. E. Stein, Detailed kinetic modeling of soot formation in shock-tube pyrolysis of acetylene, Symposium (International) on Combustion 20 (1) (1985) 887–901.

[31] A. Indarto, A. Giordana, G. Ghigo, Formation of PAHs and soot platelets: multiconfiguration theoretical study of the key step in the ring closure – radical breeding polyyne-based mechanism, Journal of Physical Organic Chemistry 23 (2010) 400–410.

[32] A. Molino, S. Chianese, D. Musmarra, Biomass gasification technology: The state of the art overview, Journal of Energy Chemistry 25 (2016) 10–25.

[33] J. M. Jones, A. R. Lea-Langton, L. Ma, M. Pourkashanian, A. Williams, Pollutants Generated by the Combustion of Solid Biomass Fuels, Springer, London, 2014.

[34] K. L. Bignal, S. Langridge, J. L. Zhou, Release of polycyclic aromatic hydrocarbons, carbon monoxide and particulate matter from biomass combustion in a wood-fired boiler under varying boiler conditions, Atmospheric Environment 42 (39) (2008) 8863–8871.

[35] Y. Huang, H. Liu, H. Yuan, X. Zhuang, S. Yuan, X. Yin, C. Wu, Release and Transformation Pathways of Various K Species during Thermal Conversion of Agricultural Straw. Part 1: Devolatilization Stage, Energy and Fuels 32 (2018) 9605–9613.

[36] D. M. Quyn, H. Wu, C. Z. Li, Volatilisation and catalytic effects of alkali and alkaline earth metallic species during the pyrolysis and gasification of Victorian brown coal. Part I. Volatilisation of Na and Cl from a set of NaCl-loaded samples, Fuel 81 (2) (2002) 143–149.

[37] Y. Wang, H. Wu, Z. Sárossy, C. Dong, P. Glarborg, Release and transformation of chlorine and potassium during pyrolysis of KCl doped biomass, Fuel 197 (2017) 422–432.

[38] X. Wei, U. Schnell, K. R. G. Hein, Behaviour of gaseous chlorine and alkali metals during biomass thermal utilisation, Fuel 84 (2005) 841–848.

[39] B. S. Haynes, H. G. Wagner, Soot Formation, Progress in Energy and Combustion Science 7 (1981) 229–273.

[40] H. Wiinikka, R. Gebart, Critical parameters for particle emissions in small-scale fixed-bed combustion of wood pellets, Energy and Fuels 18 (4) (2004) 897–907.

[41] M. Indren, C. H. Birzer, S. P. Kidd, T. Hall, P. R. Medwell, Effects of biochar parent material and microbial pre-loading in biochar-amended high-solids anaerobic digestion, Bioresource Technology 298 (2020) 1 –10.

[42] G. Habib, C. Venkataraman, T. C. Bond, J. J. Schauer, Chemical , Microphysical and Optical Properties of Primary Particles from the Combustion of Biomass Fuels, Environmental Science and Technology (2008) 8829–8834.

[43] T. C. Bond, D. G. Streets, K. F. Yarber, S. M. Nelson, J. H. Woo, Z. Klimont, A technology-based global inventory of black and organic carbon emissions from combustion, Journal of Geophysical Research D: Atmospheres 109 (14) (2004) 1–43.

[44] K. Weber, P. Quicker, Properties of biochar, Fuel 217 (2018) 240–261.

[45] M. Indren, C. H. Birzer, S. P. Kidd, P. R. Medwell, Effect of total solids content on anaerobic digestion of poultry litter with biochar, Journal of Environmental Management 255 (2020).

[46] G. Shen, S. Tao, S. Wei, Y. Zhang, R. Wang, B. Wang, W. Li, H. Shen, Y. Huang, Y. Chen, H. Chen, Y. Yang, W. Wang, W. Wei, X. Wang, W. Liu, X. Wang, S. L. Simonich, Reductions in emissions of carbonaceous particulate matter and polycyclic aromatic hydrocarbons from combustion of biomass pellets in comparison with raw fuel burning, Environmental Science and Technology 46 (11) (2012) 6409–6416.

[47] J. Jetter, Y. Zhao, K. R. Smith, B. Khan, T. Yelverton, P. DeCarlo, M. D. Hays, Pollutant emissions and energy efficiency under controlled conditions for household biomass cookstoves and implications for metrics useful in setting international test standards, Environmental Science and Technology 46 (19) (2012) 10827–10834.

[48] P. Arora, P. Das, S. Jain, V. V. N. Kishore, A laboratory based comparative study of Indian biomass cookstove testing protocol and Water Boiling Test, Energy for Sustainable Development 21 (1) (2014) 81–88.