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Renewable Energy, 2020; 149:1261-1270

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Final publication at: <http://dx.doi.org/10.1016/j.renene.2019.10.120>

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14 April 2022

<http://hdl.handle.net/2440/124625>

¹ Small-Scale Autothermal Thermochemical Conversion of Multiple
² Solid Biomass Feedstock

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⁷ **Abstract**

⁸ The thermochemical conversion of four types of biomass in a batch-fed reverse downdraft
⁹ process for heat generation in cookstoves is investigated. Fuel switching is widely considered
¹⁰ inefficient because many combustion devices do not respond well to changes in fuel. Here, the
¹¹ use of agricultural by-products, represented by wheat straw, sheep manure, cow manure, and
¹² wood pellets is addressed. Two air supply rates within the oxygen-limited regime, where the
¹³ fuel consumption is linearly dependent on the air supply, are investigated. At higher air supply
¹⁴ rates, in the reaction-limited regime, low carbon yields lead to the exposure of the ash fraction
¹⁵ to high temperatures, such that the resultant ash melting has detrimental effects on the process.
¹⁶ Generally, no detrimental impact of the ash content on the conversion process within the oxygen-
¹⁷ limited regime could be identified. The release of gaseous products, evaluated through cold gas
¹⁸ efficiency, increases linearly from 24–54% with higher air flow, corresponding to increasing pro-
¹⁹ cess temperatures from 690–980°C, and is largely fuel type independent. The char produced from
²⁰ all feedstocks fall within the highest classification for biochars, based on its elemental composition
²¹ and determined by international protocols. This emphasises the potential of the investigated pro-
²² cess for a combined production of producer gas and biochar from a variety of low-value biomass
²³ feedstocks.

²⁴ **Keywords:**

²⁵ Thermochemical conversion, Pyrolysis, Biomass, Gasification, Cookstove

²⁶ Declarations of interest: none

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27 **1. Introduction**

28 The difficulty with achieving fuel flexibility in any combustion system is evident even in highly
29 advanced systems, such as internal combustion engines [30] and gas turbines [49]. These need to
30 be adapted significantly to achieve acceptable efficiency when switching between different ho-
31 mogeneous fuels. This inherent difficulty is much more pronounced when using inhomogeneous
32 solid fuels. However, it remains common practice for users of small-scale domestic combustion
33 systems for cooking and heating to alternate their fuel source according to availability and/or
34 season [63]. This includes nearly half the world's population who rely on such systems for basic
35 survival [9]. In order to minimise adverse health and environmental implications of incomplete
36 combustion, a deeper understanding of the combustion properties of a wide range of utilised fuels
37 is necessary to enable more efficient combustor design for small-scale solid fuel systems.

38 Small-scale combustion systems that use a batch-fed autothermal reverse downdraft process,
39 called gasifier stoves, have been shown to exhibit high potential to reduce emissions of incomplete
40 combustion, compared with similar sized conventional systems [34, 48]. In these improved sys-
41 tems, the fuel batch is lit on its top surface, leading to the formation of a reaction front that moves
42 in the opposite direction from the air supply down the fuel bed [12, 39]. The propagation veloc-
43 ity of the reaction front for a specific fuel is mainly dependent on the air supply. Three regimes
44 are identified, dependent on the air flux, namely; the oxygen-limited regime, the reaction-limited
45 regime, and the regime where the process is cooled by convection [17, 44, 21, 40, 41, 38, 56]. In-
46 creasing the air supply rate results in increasing process temperatures, which in turn influences
47 the product composition of the thermochemical conversion in the reaction front. The products
48 are a complex mixture of gases, liquids (forming an aerosol with the gases, called producer gas)
49 and solid char, with increasing yields of gases and decreasing yields of liquids and char, at higher
50 temperatures [16]. The aerosol which is produced is subsequently burned with secondary air for
51 heat generation, while the char can be extracted at the end of the process [28, 29]. This process
52 is widely used with limited understanding of the biomass conversion process and especially the
53 influence of using various fuels with different compositions. Previously wood fuels and rice hulls
54 have been studied [22, 23, 50, 54], however, a more comprehensive investigation with a variety of
55 fuels is needed to provide insight of the key parameters and to influence future designs.

56 The need for an assessment of fuel types and characteristics for small-scale applications has
57 previously been identified [53]. However, most previous investigations in comparable systems

have focussed on woody biomass [27, 23, 50, 51, 35, 32, 19, 33]. Other widely used fuels in practice include agricultural residues and animal manures, which are burned in great quantities per household in countries such as India [37], but have not been studied extensively. Generally, the appropriacy of manure as fuel is debatable since it reduces its availability as a fertiliser. In the autothermal reverse downdraft reactor, this effect is minimised because biochar is produced which can be used as an alternative soil amendment. Few studies have focussed on the use of manure as fuel [59, 3, 7]. High emissions have been reported for direct combustion [52], while the thermochemical conversion process has not specifically been addressed. Continuous downdraft gasification of cow manure has not been found feasible, because of the low heating value of the product gas and a satisfactory process could only be achieved when mixed with sawdust [45]. The use of agricultural residues, such as wheat straw, which are often disposed of by field burning [63, 43] could provide another widely available fuel source. Continuous gasification of straw has been indicated to only be possible with pelletised fuel, as chopped straw led to air blockages [20]. The main difference between woody biomass, agricultural residues and animal manures are the bulk density (depending on particle size and density), the ash content and the related energy density. While wood has a low ash content, it is greater in agricultural residues and is generally much higher in manure. The main topics discussed when dealing with high ash content fuels are melting, fusing and slag formation [61, 24]. It has been suggested that the high K and Si concentrations in straw ash could lead to slag formation [25], but when producing char the high unburned carbon concentrations and the integrity of the initial particle structures could minimise these possible effects. Therefore, general concerns with the use of high ash content fuels in thermochemical conversion processes do not necessarily apply to the presented system. The influence of a high ash content in fuels on the thermochemical conversion process and the quality and efficiency of combustible gas production is not well understood.

The combined production of char and clean-burning combustible gases could provide beneficial implications for the process as well as the environment. Not only can the solid char lower the concentration of tars in the producer gas and retain a large fraction of the ash [29] to reduce particulate emissions, the biochar is also a product that can be used for a variety of subsequent applications. The specific characteristics of the biochar, such as surface area and high carbon content, make it a particularly valuable for soil amendment purposes [62]. Biochar is widely produced in a variety of systems, such as earth pits or rotary kilns, where only a portion of the released volatile

89 products from the biomass feedstock are utilised to sustain the thermochemical conversion pro-
90 cess and the remainder vented [10]. A process that combines the production of biochar with the
91 full utilisation of the volatiles for heat generation could substantially increase the efficiency of the
92 system. The quality of char produced from small-scale reverse downdraft gasification is seldom
93 assessed though and needs further investigation for the application as a soil amendment, espe-
94 cially when utilising unconventional non-woody biomass feedstock.

95 The focus of this article is to assess various value biomass feedstocks for the combined produc-
96 tion of producer gas and solid char in small-scale applications. Four fuels, namely wood pellets,
97 wheat straw, sheep manure and cow manure, have been investigated at two air supply rates. The
98 novelty of the paper is the insight of the producer gas composition and evolution in a small-scale
99 batch-fed autothermal reverse downdraft system, with a specific focus on fuels with a high ash
100 content. In contrast, similar papers typically omit measurements and analysis of the influences of
101 the producer gas, instead looking only at the emissions from the subsequent combustion process.
102 By isolating the products of the thermochemical conversion through the continuous measurement
103 of the producer gas, the results are related to the main constituents of the supplied fuel and the
104 produced char. These measurements enable an in-depth analysis of the thermochemical conver-
105 sion process, the release of combustible products, as well as the quality of produced char. The
106 analysis is extended for multiple fuels, with a wide range of ash contents. The resultant deeper
107 understanding of the influence of biomass fuel composition, especially the ash content, on the au-
108 tothermal thermochemical conversion process and its products, provides valuable information for
109 downstream applications of the producer gas, as well as the char.

110 **2. Materials and Methods**

111 *2.1. Reactor*

112 The utilized small-scale thermochemical conversion and combustion reactor has been described
113 previously [29]. Its main features, in the order of air flow are: an air supply chamber, a fuel grate
114 on which the fuel rests, a reactor with ports for inserting thermocouples into the fuel bed, and
115 a probe for the extraction of products above the fuel bed. Volatile products are combusted in a
116 non-premixed flame, open to the environment, downstream of the extraction probe, however, the
117 combustion process is not considered in this study.

118 Air is delivered at a constant flow rate during each of the experiments, resulting in the air mass
119 flux specified in Table 1. The grate on which the fuel bed rests has 67.2% open area. The inner
120 diameter of the reactor is 98 mm and is insulated with a 25-mm-thick thermal blanket. Eight K-
121 type thermocouples (T1-T8) are situated along the length of the reactor. The entire device is placed
122 on a weighing scale, Radwag WLC 20/A2, with a readability as well as repeatability of 10^{-4} kg
123 and a maximum capacity of 20.0 kg.

124 The probe used to extract volatile products is situated at the top of the initial fuel bed. This
125 probe is connected to a tar trap, which is used for the retention of all non-gaseous products. Sub-
126 sequently, the gas sample is analysed, described in §2.2.

127 2.2. *Gas Analyser*

128 The gas stream was sampled with a MRU Vario Plus analyser. It measures CO₂, CO and CH₄
129 up to 30% (vol./vol.) and an accuracy $\pm 3\%$ of the reading, using NDIR sensors. The measurements
130 of O₂ and H₂ are measured with electrochemical sensors with a range up to 21% (vol./vol.) and an
131 accuracy $\pm 0.2\%$ of the absolute value, and a range up to 100% (vol./vol.) and an accuracy $\pm 0.02\%$
132 of the reading, respectively. N₂ is determined by subtraction. The analyser was calibrated on a
133 daily basis.

134 2.3. *Fuels*

135 Wood pellets, wheat straw, sheep manure and cow manure were tested in the present study.
136 The results of the proximate and ultimate analyses for all fuels are presented in Figure 1. The
137 ternary plots each enable the presentation of three constituents: (a) volatile matter (VM), fixed
138 carbon (FC) and ash; and the three main elements: (b) carbon (C), hydrogen (H) and oxygen (O).
139 Further information about the fuel and the tabulated values of the proximate and ultimate analy-
140 ses are available in the Supplementary Material in Section S1.2. Due to the fuels bulk density 2.1 kg
141 of wood pellets, 0.5 kg of wheat straw and 0.9 kg of manure were used for individual experiments.

142 2.4. *Procedure*

143 Prior to performing the experiments, the reactor was preheated and subsequent tests were
144 started at inner reactor temperatures of <100 °C. Preheating was performed to avoid an influence
145 from the large thermal mass of the reactor and reactor temperatures of <100 °C were chosen to
146 minimise the influence of moisture evaporation when re-fuelling. Fuel was supplied in batches

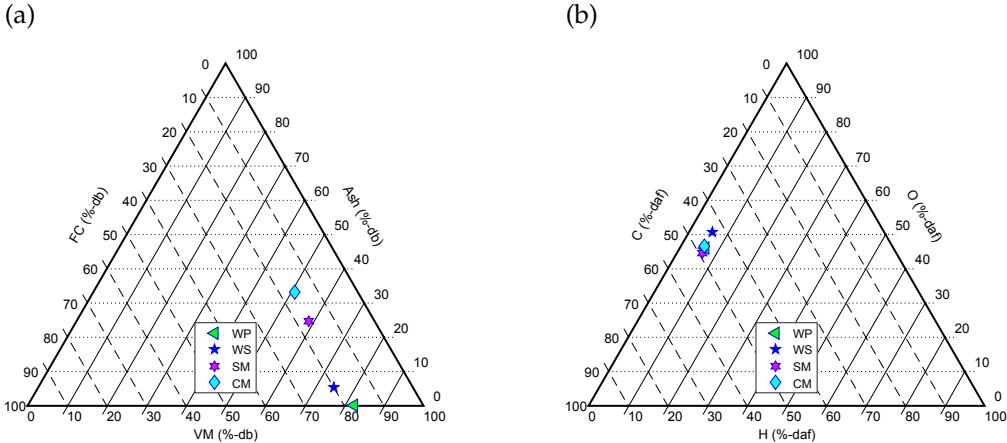


Figure 1: The fuel composition in terms of proximate analysis on a dry basis and ultimate analysis on a dry ash-free basis. The fuel types are: wood pellets (WP), wheat straw (WS), sheep manure (SM) and cow manure (CM). Axes show the three constituents of (a) the proximate analysis, fixed carbon (FC), ash, and volatile matter (VM), and (b) the ultimate analysis, carbon (C), oxygen (O), and hydrogen (H).

and the air mass flux was pre-set on the flow meters, prior to each experiment. Lighting was performed with the aid of 10 mL of methylated spirits (96% ethanol, CAS # 64-17-5) and a paper towel supplied to the top surface of the fuel bed. The temperature within the fuel bed was recorded throughout the process. The reaction front velocity for each configuration was determined in preliminary experiments (determined by the time taken between subsequent thermocouples reaching 600 °C) and the process was quenched once the reaction front reached the grate at the bottom of the fuel bed. Quenching was achieved by introducing ice water into the reactor from the top and by the provision of nitrogen (>99.99% N₂) instead of air to cool and prevent reactions inside the fuel bed. The remaining char was subsequently extracted. Multiple repeats for each tested fuel were performed at the two air supply rates, as presented in Figure 1. Air supply rates of 0.025 kg·m⁻²·s⁻¹, for wood pellets, wheat straw and cow manure, and of 0.03 kg·m⁻²·s⁻¹ for sheep manure, will be referred to herein as “low” and for all fuels 0.075 kg·m⁻²·s⁻¹ as “high”. These two air supply rates were chosen on the basis of preliminary experiments and a previous study [29], which have shown that these represent a high and low value within the oxygen limited regime. The exception of 0.030 kg·m⁻²·s⁻¹ had to be made for sheep manure, since at lower flow rates no gaseous product measurements were possible, because excessive release of tar lead to repeated clogging of the tar trap.

Table 1: Experimental configurations, the number of repetitions performed and the experimental code.

Fuel type	Air Mass Flux (kg·m ⁻² ·s ⁻¹)	Repetitions	Code
Wood pellets	0.025	5	WP-L
	0.075	5	WP-H
Wheat straw	0.025	4	WS-L
	0.075	4	WS-H
Sheep manure	0.030	4	SM-L
	0.075	5	SM-H
Cow manure	0.025	3	CM-L
	0.075	4	CM-H

164 2.5. Analysis

165 For each experiment, the reactor is placed on a weighing scale to measure the fuel mass loss
 166 during the conversion process. The fuel mass loss was expected to display a linear profile with
 167 changes to air supply, based on previous research [40, 55]. The two air supplies at the focus of this
 168 study are 0.025 and 0.075 kg·m⁻²·s⁻¹. These two flow rates were chosen on the basis of air supply
 169 regimes determined from mass loss measurements during preliminary experiments in the range
 170 0.010–0.200 kg·m⁻²·s⁻¹. Only mass loss was measured during the preliminary experiments, whilst
 171 gas sampling was performed during all subsequent tests. It should be noted that comparable
 172 values found in the literature [41] are not based on weight measurements but calculated on the
 173 basis of thermocouple data, represented by the reaction front velocity and the fuel bulk density
 174 ($\dot{m}_{Fuel} = v_{Front} \cdot \rho_{Fuel}$). All figures presenting measured values include error bars that display the
 175 standard error of the mean [8].

176 Eight thermocouples recorded the gas phase temperature within the reactor. Mean maximum
 177 temperatures are determined as an average of the highest temperatures of the thermocouples T1–
 178 T7 and the value reported for each configuration was the mean of all repeat tests. Measurements
 179 of the lowest thermocouple T8 (at 20 mm from the fuel grate) were disregarded, since an increase
 180 of temperature due to the proximity to the fuel grate was observed.

181 An elemental balance was performed for carbon (C) and hydrogen (H) in the thermochemical
 182 conversion. The supply of N₂ via air was considered to be conserved, allowing the calculation
 183 of molecular C and H in the measured gas via the relationship between the supplied N₂ and the
 184 analysed N₂, as per Equation 1. Equations 2 and 3 present the overall calculation of molecular C
 185 and H in the measured gas, while the equations can be adapted for individual gas species. The

186 content of tars and water in the producer gas was not measured.

$$\dot{n}_{gas} = \frac{x_{gas}}{x_{N_2measured}} \cdot \dot{n}_{N_2air} \quad (1)$$

$$C_{gas} = \frac{m_{air} \cdot \omega_{N_{air}} / M_{N_2} \cdot (x_{CO_2} + x_{CO} + x_{CH_4}) / x_{N_2}}{(m_{fuel-daf} \cdot \omega_C - m_{char-af} \cdot \omega_C) / M_C} \quad (2)$$

$$H_{2gas} = \frac{m_{air} \cdot \omega_{N_{air}} / M_{N_2} \cdot (x_{H_2} + 2 \cdot x_{CH_4}) / x_{N_2}}{(m_{fuel-daf} \cdot \omega_H - m_{char-af} \cdot \omega_H) / M_H} \quad (3)$$

187 Where \dot{n} is the molecular gas flow and x is the molecular gas concentration. The provided or
188 product mass is represented by m , the mass fraction by ω and the molecular mass by M .

189 To evaluate the process performance, the cold gas efficiency (CGE) was calculated on the basis
190 of the energy content of the produced gases relative to the energy content of the converted fuel, as
191 presented in Equation 4. The measured gas concentrations are considered as the energy content of
192 the producer gas, while other hydrocarbon compounds and carbonaceous particles that might be
193 released from the fuel bed are not included. The higher heating value (HHV) of the fuel and the
194 char were measured using a bomb calorimeter and that of the gaseous species were based on well
195 characterised values found in the literature [60].

$$CGE = \frac{V_{N2-air} / x_{N2} \cdot (x_{CO} \cdot 12.6 + x_{CH_4} \cdot 39.8 + x_{H_2} \cdot 12.8)}{HHV_{fuel} \cdot m_{fuel} - HHV_{char} \cdot m_{char}} \quad (4)$$

196 Fuels, as well as produced char samples from each configuration, were analysed for their ult-
197 imate (CHN), proximate (moisture (M), volatile matter (VM), fixed carbon (FC) and ash content)
198 composition and their HHV. The proximate analysis was performed via thermogravimetric analy-
199 sis (TGA), using a previously established method [47]. For both fuel and char samples, the ash con-
200 tent was also determined following ISO 18122:2015 [1] and the moisture content following ASTM
201 D4442-92(2003) [4]. The reported proximate analyses therefore consist of the moisture content, via
202 the ASTM standard, the VM fraction, via TGA analysis, the ash content, via the ISO standard, and
203 the fixed carbon fraction is calculated via subtraction.

204 **3. Results**

205 *3.1. Mass Flux and Process Temperature*

206 Figure 2 shows the fuel mass flux—the consumption of fuel per time and reactor area—as a
207 function of the supplied air mass flux, for each of the fuels considered in this study. Preliminary
208 experiments were performed over a wide range of conditions, where only one repetition was per-
209 formed. Also shown are the experimental results at the two air supply rates used for the majority
210 of this work, as well as values found in the literature [41] for wood pellets. Previous research has
211 shown that with increasing air flow, under sub-stoichiometric conditions, the fuel consumption
212 increases initially linearly (oxygen-limited regime), then less severely until a further increase does
213 not change the fuel consumption (reaction-limited regime) and at super-stoichiometric air supply,
214 leads to cooling of the process until extinction (refer to §1). Cookstoves generally operate in the
215 initial regime, where the fuel mass flux is linearly dependent on the air mass flux, and this regime
216 is therefore the focus of the present study.

217 All fuels investigated behave similarly and the fuel mass flux increases linearly in the oxygen-
218 limited regime in Figure 2 up to an air supply rate of $\approx 0.1 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. In this regime, the value of
219 the fuel mass flux (WP>WS>CM>SM) is notably lower when the fuel contains a high amount of
220 ash. This lower fuel mass flux can be explained by a lower oxidiser-fuel contact, lower diffusion of
221 gas species and the lower energy content of the bed, due to the high ash content, as hypothesised
222 previously [40]. The lower fuel mass flux at a given air mass flux also results in a higher air to fuel
223 ratio (A/F), which has previously been found to be similar for various biomass types with lower
224 ash content [56]. This shows that the high ash content in manures has an impact on the conversion
225 speed, but also that the conversion regimes are dependent on the superficial velocity (air mass flux
226 / air density) and thus the oxygen availability as well as residence time.

227 For WP and WS, increasing the air mass flux to $> 0.1 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ leads to a transition to the
228 reaction-limited regime and the fuel mass flux increases less rapidly with increasing air mass flux,
229 until it reaches a plateau. For SM and CM, higher air flows exceeding $> 0.125 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ lead to
230 a slight decrease in the fuel mass flux. This decrease is caused by a variety of factors including ash
231 melting, which substantially alters the fuel bed properties. At such high air supply rates little to
232 no char is produced. Therefore, the utilisation of this type of system outside the oxygen-limited
233 regime is not advisable, as further discussed in Sections 3.3 and 3.4.

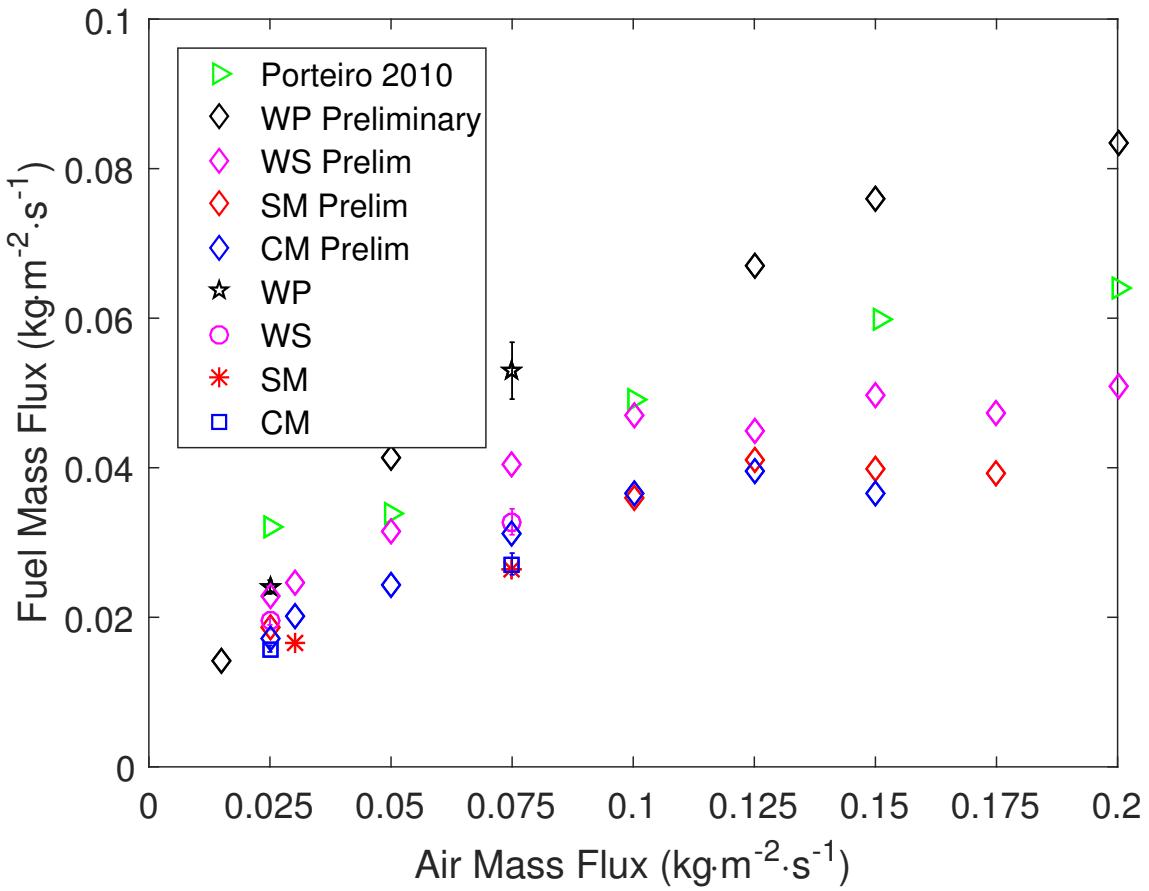


Figure 2: The fuel mass flux, the fuel consumption per time and reactor area, is presented as a function of the air mass flux for different experimental configurations, preliminary data and data found in the literature [41]. The error bars display the standard error of the mean.

The relationship between the mean peak temperature and the bulk density of the fuel bed is shown in Figure 3. The peak temperature is recorded at the centre of the fuel bed. It can be seen that there is a slight bulk density dependence of the mean peak temperature, similar to previous studies [40, 46]. With a lower bulk density, the total heat release per reactor volume is much lower and the thermal mass of the reactor wall and volume will have a greater influence on the peak temperature, compared with fuels with higher bulk density. Previously a higher bulk density has been related to a decreasing reaction front propagation velocity through the fuel bed [40], which also corresponds with a higher heat release per reactor volume. Here, no influence of other potentially related parameters, such as ash content, volatile matter content or particle size was

²⁴³ found.

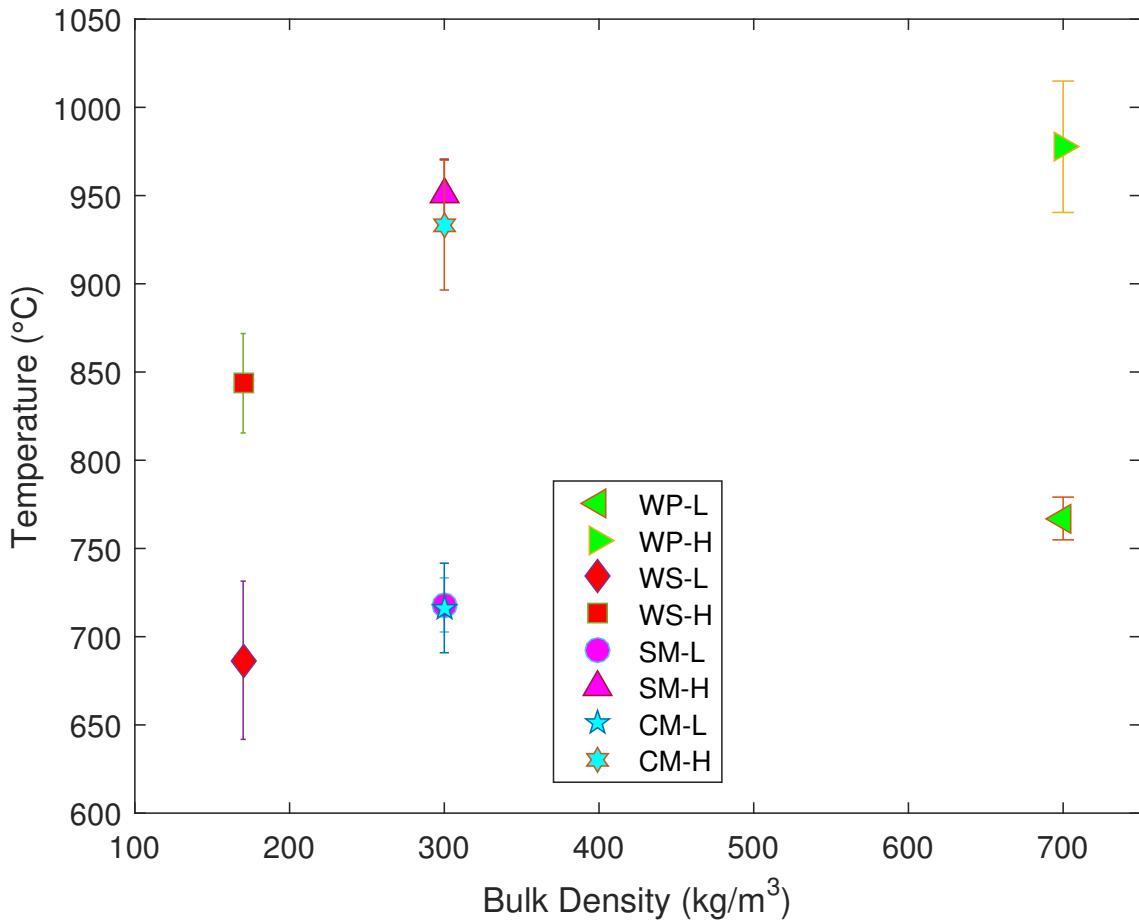


Figure 3: The mean peak conversion temperature at the reactor centre is presented in relation to the initial bulk density of the fuel. The error bars display the standard error of the mean.

²⁴⁴ 3.2. Gaseous Products

²⁴⁵ The time-weighted average species concentration over the duration of the experiment, as well
²⁴⁶ as the calculated HHV of the producer gases (refer to §2.5) are presented in Figure 4. The mean
²⁴⁷ volumetric concentration of the main product species from the thermochemical conversion process
²⁴⁸ and O_2 are shown for all four fuels at high and low air supply. It can be noted that for each
²⁴⁹ configuration, the O_2 concentration is low and re-evaluation of the concentrations on a 0% O_2
²⁵⁰ basis did not change the trends in between configurations. In terms of application in small-scale

251 combustion systems, it needs to be kept in mind that the producer gas presented here is the fuel
252 for the subsequent combustion process.

253 In Figure 4 it can be seen that increasing the air supply has contrary effects for CO₂ and CO.
254 At higher air supply rates, less CO₂ and more CO is produced, which may be explained by a
255 shift in the biomass conversion chemistry to an increasing primary product ratio of CO/CO₂ with
256 increasing process temperatures [31]. Furthermore, previous research suggests that higher pro-
257 cess temperatures [5] as well as higher temperatures in the char layer downstream of the reaction
258 front [29] promote the conversion of tars (hydrocarbon compounds with higher boiling points than
259 benzene [36]) to form mainly CO and H₂ (via reactions R13–R16, presented in the Supplementary
260 Material in Table S3). This conversion of tars contributes to an increasing CO yield with greater
261 air supply. As the combustible gases are more easily burned completely than the tars, greater
262 conversion yields of these products may be beneficial for subsequent combustion.

263 Similar to the CO and CO₂ results, the hydrogen-containing species, H₂ and CH₄, also follow
264 contrary trends with increased air flow rate. The mean H₂ concentration is higher with increasing
265 air supply and process temperatures, while the CH₄ concentration decreases. This trend can be
266 explained by a combination of the decomposition of hydrocarbon compounds at higher tempera-
267 tures (R13–R16, see Supplementary Material in Table S3) and the interplay of homogeneous gasi-
268 fication reactions (R7–R12, see Supplementary Material in Table S3).

269 When comparing the different fuels with one another it can be seen that the highest concentra-
270 tions of all combustible species is released from the WP, hence also exhibiting the highest HHV.
271 CM exhibits the lowest HHV, while WS and SM are quite similar. Therefore, WP provides the
272 highest quality gas for subsequent combustion. Whilst this supports the previous work that has
273 focussed on woody biomass as a feedstock, it also highlights the challenges associated with the
274 use of lower-grade fuels that are widely used.

275 In a previous study, wheat straw had been tested at an air supply of approximately 0.065 kg·m⁻²·s⁻¹
276 and it was found that it exhibited a similar process temperature, but higher peak CO and CO₂ con-
277 centrations [25]. Although the previous system [25] is very similar to the one presented here, the
278 comparison is limited because the char was consumed, leading to a higher prevalence of heteroge-
279 neous gasification reactions (R2–R5) and higher carbon oxide fractions in the producer gas. Sim-
280 ilarly, higher temperatures and higher concentrations of all combustible gaseous products (CO,
H₂ and CH₄), have been found with complete wheat straw fuel conversion elsewhere [61]. In

282 a similar, but continuous process, with char production, higher H₂ and CO concentrations were
 283 achieved with a simultaneous decrease in tars when using wood chips and wood pellets as fuel
 284 at 0.022 kg·m⁻²·s⁻¹ [11]. In this continuous reactor the product gas passed through a constant
 285 layer of char at elevated temperatures [11], where tar cracking may occur, while here the char
 286 layer thickness increases as the reaction front propagates down the fuel bed and initially no char
 287 is present.

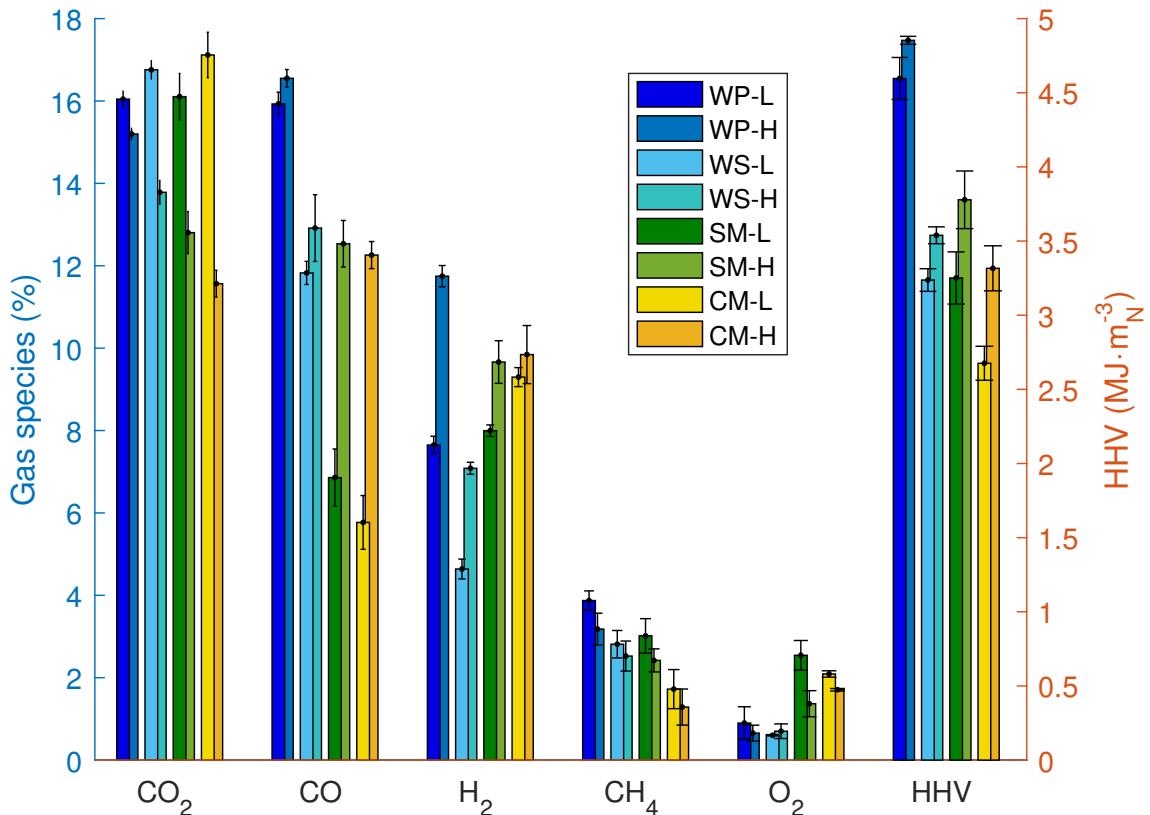


Figure 4: The time-weighted average volumetric producer gas composition, over the duration of the conversion process, as a mean of all replicates and the resulting HHV, of the gaseous products, are presented. The error bars display the standard error of the mean.

288 3.3. Biomass Conversion

289 The molecular conversion balance of carbon from the different fuels into the gaseous products,
 290 solid char and “other” (accounting mainly for tars and released particles) are presented in Fig-
 291 ure 5. With increasing air flow, a general trend of increasing gaseous products accompanied by a

292 reduction of tars and char can be seen. This trend can be explained by higher process temperatures
293 and has been well documented for biomass conversion processes [16, 6].

294 Focussing on a comparison of the conversion to permanent gases it can be seen that the com-
295 bined yield of CO₂, CO and CH₄, at low flow rate follows the relation of WP > WS > CM > SM. At
296 high air supply, the relation changes to CM > SM > WP > WS. The release of combustible gases,
297 CO and CH₄, from the manures are particularly sensitive to changes in the air supply: from <40%
298 at low air supply to >50% at high air supply. The higher increase of gas yield for the manures ap-
299 pears to be due to greater conversion of the fixed carbon fraction, with a simultaneous reduction of
300 the char yield. For WP and WS both the char and other yields change more similarly, between low
301 and high air supply. When considering that all fuels have a very similar carbon content, on a dry
302 ash free basis (see Figure 1), the larger ash content in the manures appears to facilitate a greater
303 conversion of fixed carbon to gaseous products at high air supply.

304 The char-carbon yield is lowest for the manures with high air flow. Thus, there will be less car-
305 bon available to form the structure of the char. This will be even more pronounced when further
306 increasing the air supply, as described in Section 3.1. As char is a desired product of this pro-
307 cess, a further increase of the air supply is not suitable for its production, especially when using
308 manures as fuel—however, the higher proportion of producer gas in the high air supply case is
309 advantageous.

310 The “other” fraction primarily includes tars, which are generally considered an undesirable
311 product because they have been identified as a soot precursor for the subsequent combustion [18].
312 The fraction of tars is notably highest in SM, therefore explaining why this fuel was prone to clog-
313 ging of the sampling line at 0.025 kg·m⁻²·s⁻¹ and instead required an air supply of 0.030 kg·m⁻²·s⁻¹
314 to ameliorate this issue (as described in §2.4). The lowest yields are achieved from WS, supporting
315 the potential of this widely wasted fuel in this type of thermochemical conversion process.

316 Figure 6 presents the molecular balance of the supplied hydrogen in the fuel, into the mea-
317 sured gases and solid char, as well as “other”, which accounts for mainly tars and water. For
318 both air supply rates, WS presents the lowest conversion to gaseous products. It can be assumed
319 that the low gas yield is due to lower peak process temperatures, when compared with the other
320 fuels (see Figure 3). For both low and high air supply, it should also be noted that while the gas
321 yields are similar for WP and SM, they are highest for CM. This trend cannot be explained by a
322 temperature influence, as WP achieved the highest process temperature. Similar to the conversion

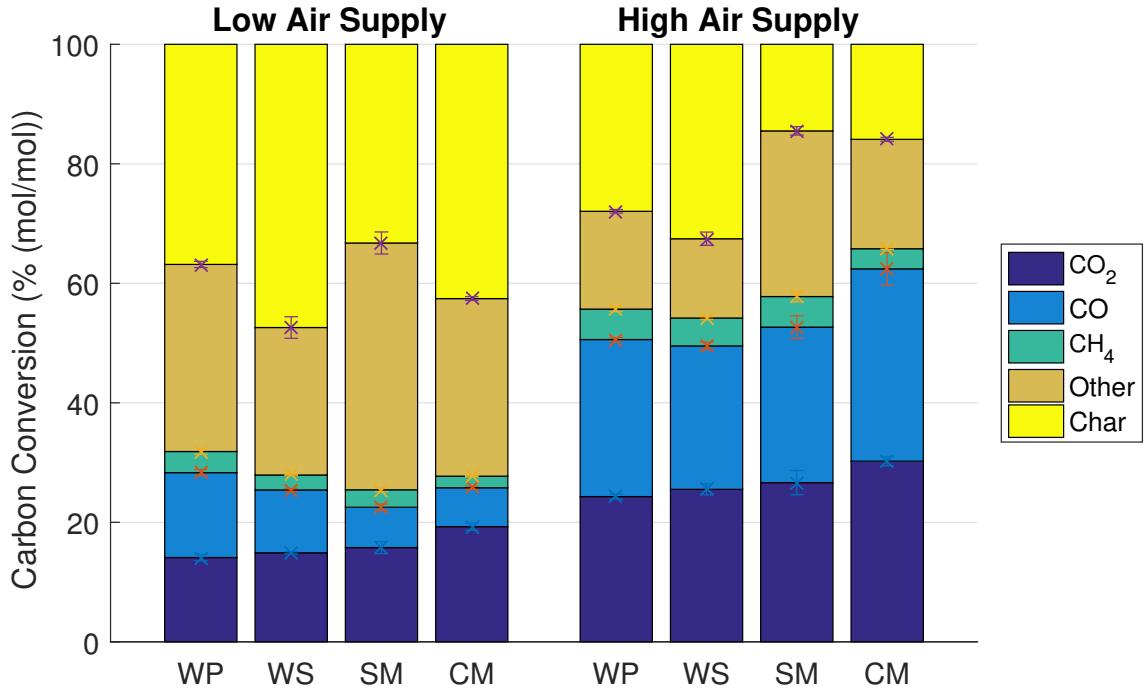


Figure 5: Conversion of fuel carbon to the gaseous products, CO_2 , CO and CH_4 , “other”, accounting mainly for tars but also for carbonaceous particles that could be ejected from the fuel bed, and the solid char. The error bars display the standard error of the mean.

of carbon, the relatively high conversion yield of hydrogen-containing gases from the manures could be caused by the influence of the ash constituents. While the release of CH_4 appears to be consistent in between fuels at low and high air supply, the release of H_2 seems fuel dependent. This could be due to the presence of ash, since H_2 is highest in the manures.

The thermochemical conversion process can be evaluated through the cold gas efficiency (CGE), which provides a measure of the energy in the produced gas relative to that consumed from the supplied fuel (see §2.5). In Figure 7 the CGE is presented in relation to the mean peak process temperature. The efficiency appears to have a near-linear response with temperature, irrespective of fuel type.

In a similar-sized continuous downdraft gasifier using woody biomass pellets, at an air supply rate of $\approx 0.050 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, a CGE of over 70% was reported [26]. Although, the peak process temperature is higher at $0.075 \text{ kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ here and the producer gas composition appears similar, the CGE is more than 20% lower. Complete conversion of the char and a larger fraction of tars to

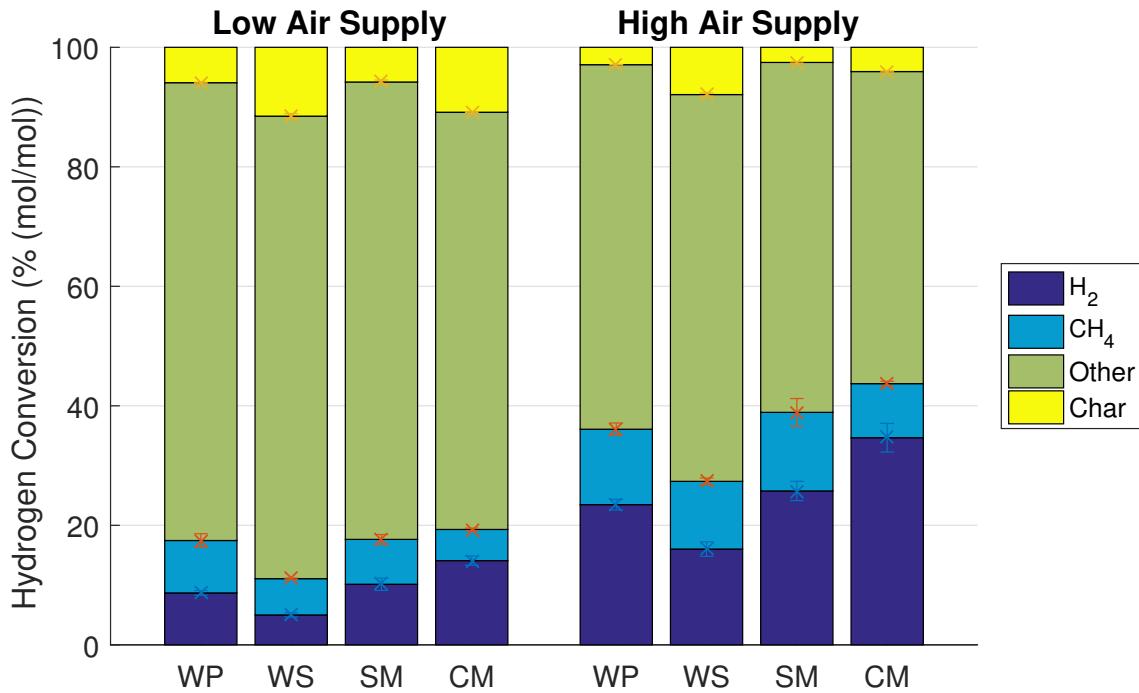


Figure 6: Conversion of fuel hydrogen to the gaseous products, H₂ and CH₄, “other”, accounting mainly for water and tars, and the solid char. The error bars display the standard error of the mean.

³³⁶ producer gas in the continuous gasifier will contribute to this higher CGE.

³³⁷ *3.4. Biochar*

³³⁸ In Figure 8(a) the proximate-, and in 8(b) the ultimate-, analyses of the produced chars are
³³⁹ shown as ternary plots (the respective values are provided in the Supplementary Material in Ta-
³⁴⁰ bles S4 and S5). The ternary plots enable the presentation of (a) the three constituents; volatile
³⁴¹ matter (VM), fixed carbon (FC) and ash; and (b) the three main elements: carbon (C), hydrogen
³⁴² (H) and oxygen (O).

³⁴³ The largest differences between the fuels can be seen in (a) along the fixed carbon and ash
³⁴⁴ axes and in (b) along the carbon and oxygen axes. It can be noted that the carbon and the fixed
³⁴⁵ carbon fractions generally decrease when increasing the air supply and are especially low for the
³⁴⁶ manures. At 0.075 kg·m⁻²·s⁻¹ only approximately 10% and 20% of fixed carbon remain in the
³⁴⁷ char for CM and SM, respectively. At higher flow rates, the fixed carbon fraction is too low to
³⁴⁸ retain a carbon structure, which is the basis of the char. Without the supporting carbon structure

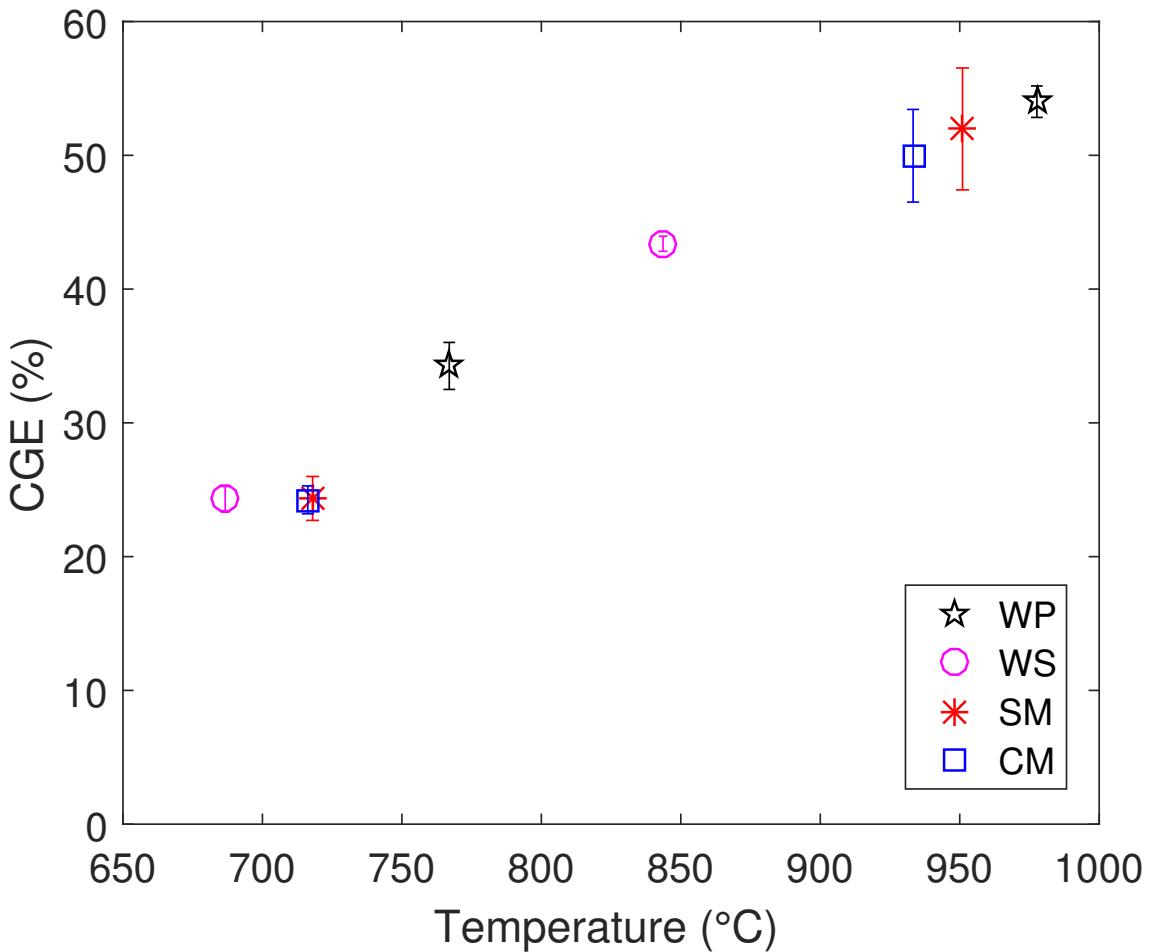


Figure 7: The cold gas efficiency (CGE) versus the mean peak temperature in the reaction front for all tested configurations. The error bars display the standard error of the mean.

and temperatures in excess of 900 °C (refer to Figure 3), ash melting occurs, which substantially alters the porous structure of the fuel bed and has negative process implications (refer to § 1). The impact of ash melting on the solid product can be seen in photographs which are included in the Supplementary Material in Figures S5 and S6.

Table 2 presents the higher heating value, the weight-based yield and the energy yield of the char. At low air supply for wood pellets, the char yield is 20% and the energy yield is 37%, which agree well with literature values under pyrolysis conditions of $\gtrsim 20\%$ and $\gtrsim 40\%$, respectively [16, 62]. Similarly, wheat straw under low air supply conditions gives $\gtrsim 30\%$ char yield and $\gtrsim 40\%$

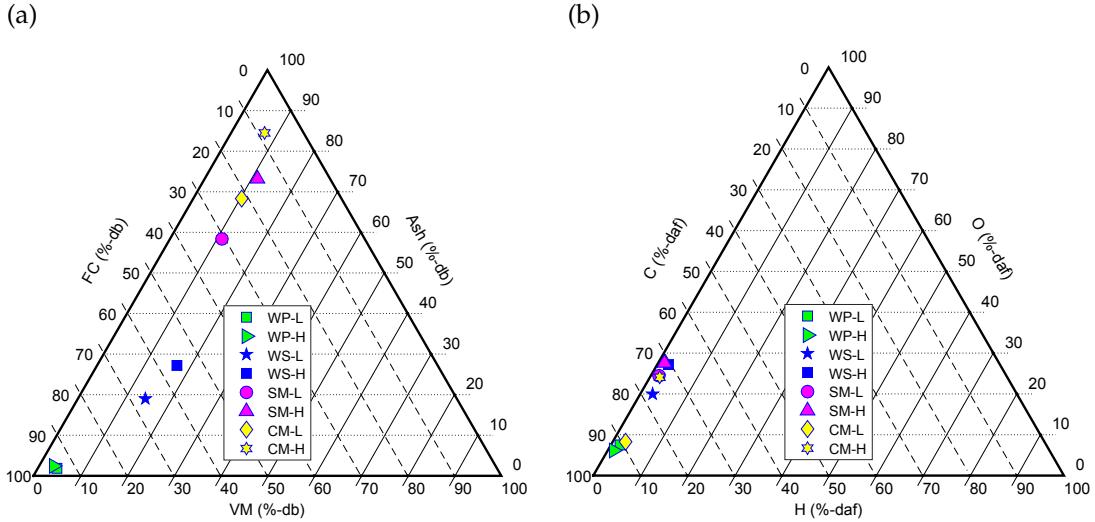


Figure 8: In (a) the proximate analyses, on a dry basis (db), and in (b) the ultimate analyses, on a dry ash free basis (daf), are presented. All values are on a mass basis.

357 energy yield in both Table 2 and pyrolysis literature [16, 62]. Therefore, the results indicate that
 358 at low air supply, pyrolysis conditions of temperatures $>700^{\circ}\text{C}$ are approached. In contrast, at
 359 higher air supply, the yields of both char and energy are lower by nearly a factor of two. Generally,
 360 it can be seen that increasing the air supply leads to a reduction in yields as well as heating value,
 361 because of the increasing ash fraction in the char.

Table 2: The mass yield (g/g), the energy yield (MJ/MJ) and the bulk calorific value (MJ/kg) of the produced char.

Air supply	WP	WS	SM	CM
Mass yield (g/g)				
low	0.20	0.32	0.40	0.50
high	0.14	0.24	0.31	0.37
Energy yield (MJ/MJ)				
low	0.37	0.44	0.33	0.39
high	0.26	0.27	0.21	0.12
HHV (MJ/kg)				
low	32.5	22.6	11.1	9.8
high	31.2	18.2	9.2	3.9

362 When considering a molecular balance, rather than a mass balance as presented by the prox-
 363 imate and ultimate analyses, generally a decrease of both the molecular H:C and O:C ratios has
 364 previously been identified with increasing process temperature, for woody biomass as well as

wheat straw [62] (H:C and O:C ratios for produced chars as well as the fuels are presented in the Supplementary Material in Figure S7). This trend can also be found in the present study and similarly applies to the SM and CM chars. The difference in O:C ratio, between the fuel and the char, is less severe in the case of the manures, as oxygen can be expected to be present within the ash [58]. All produced chars fall within the category of Class 1 biochars, based on their molecular composition, with H:C and O:C ratios lower than 0.7 and 0.4 respectively, as proposed by the European Biochar Foundation [2, 15]. Therefore, even the high ash content chars could attain the highest classification biochars for soil amendment purposes, but future work regarding further classification criteria will address this issue in more depth.

4. Discussion

As mentioned in Section 1, three regimes have been identified for the thermochemical conversion of many biomass materials with increasing air supply, namely: oxygen-limited, where the fuel conversion is linearly dependent on the air supply; reaction-limited, where the fuel conversion plateaus with respect to increasing air supply; and the regime where the process is cooled by convection. The present study focusses on the oxygen-limited regime, since char is mainly produced in this regime, however, preliminary experiments were also performed in the reaction-limited regime and the findings may aid in defining ongoing processes in these regimes more clearly.

When little oxygen is supplied to the ignited fuel, the conversion of the solid biomass is dominated by devolatilisation, leading to the formation of solid char (as defined by reaction R1 in the Supplementary Material in Table S3) with a lower influence of heterogeneous gasification reactions R2–R5 of the resultant char. When the air supply is increased, there is a general rise in fuel consumption, accompanied by higher process temperatures (see §3.1), higher yields of permanent gases and a reduction of the tar (see §3.3) and char yield (see §3.4), caused by a stronger influence of reactions R2–R5 (refer to the Supplementary Material in Table S3). This is supported by a comparison with the relevant literature reporting pyrolysis experiments [16, 62] (see §3.4), where similar char yields have been reported at comparable process temperatures for low air supply, while higher yields are reported at high air supply, when heterogeneous gasification reactions are more prominent if oxygen is supplied to the conversion process.

394 At low air supply, it should be noted that the molecular yield of C and H in the char is some-
395 what similar (interestingly WP and SM, and WS and CM, exhibit similar yields) while the re-
396 duction of yields at high air supply is more severe in the manures (see §3.3) compared with the
397 lignocellulose fuels (WP and WS). Previously, it has been suggested that a higher lignin content,
398 the most stable lignocellulose component, promotes the formation of char during pyrolysis [13].
399 While this will contribute to a similar carbon conversion yield of char for WP and WS, which
400 generally have a similar lignin content [57], the fraction in manures is generally very low [42].
401 Furthermore, it is shown that the duration of the conversion per unit mass of supplied fuel was
402 very similar for all fuels, although in the case of the high ash content fuels, much less dry ash-free
403 fuel is available (see §3.2). This shows that the dry ash-free conversion process occurs more slowly
404 in high ash content fuels but a larger fraction of the FC is consumed, leading to nearly complete
405 conversion of FC under oxygen-limited conditions.

406 In the manure and WS chars for high and low air supply rates, more than 50% and 20% (g/g),
407 respectively, of the product is ash. This ash is retained in the solid structure which minimises the
408 influence of elutriation or ash melting, from these high ash containing fuels. In all cases, even for
409 CM at high air supply where the char contains only 10% (g/g) FC (see §3.4), the carbon structure
410 largely remains but loose ash can be noticed on the char surface (refer to the Supplementary Mate-
411 rial in Figure S5). This shows that through the production of the char, not only is a valuable solid
412 (see §3.4) created, but also beneficial process implications are achieved. Furthermore, the clear
413 separation of a devolatilisation and a char conversion phase, which becomes less pronounced as
414 lower char yields are achieved, will cease at higher air supply rates for high ash content fuels.

415 The high ash content in the manures does not appear to have a negative influence on the con-
416 version to permanent gases, within the oxygen-limited regime (see §3.3). The presence of char has
417 previously been shown to increase tar conversion to permanent gases [29] (as per reactions R13–
418 R16, see Supplementary Material in Table S3). While this influence has not specifically been stud-
419 ied for chars with a high ash content, it can be assumed to be influential on the release of gaseous
420 products. Overall, the molecular conversion to permanent gases and its evaluation through the
421 CGE, appears to be mostly process temperature dependent, while the fuel type plays a secondary
422 role.

423 In the reaction-limited regime, the conversion reactions of biomass devolatilisation and het-
424 erogeneous gasification of the produced char body (R1–R5, see Supplementary Material in Ta-

ble S3) occur more concurrently. This leads to a more simultaneous conversion and release of the volatile matter and fixed carbon fractions from the fuel batch. For all fuels, this causes a substantial reduction in the char yield and a change in bed morphology. The absence of the char structure at high air supplies leads to ash either being expelled in the form of fly-ash or exposed to high process temperatures exceeding the ash melting point [14] leading to product melting and particle fusing, which alters the fuel bed properties and causes problems within the reactor (refer to the Supplementary Material in Figures S5 and S6). This is of little importance for low-ash fuels but can be detrimental to high-ash fuels, such as manures. Problems occurring in the reactor include channel forming, which changes the combustion characteristic from a near homogeneous reaction front to locally differing conversion conditions, or ash fusing to walls. Furthermore, the plateau of the fuel conversion with a simultaneous increase of the air supply, in the reaction-limited regime, leads to increased dilution of combustible products.

5. Conclusions

The presented study investigates the conversion behaviour of four different biomass fuels in an autothermal reverse downdraft process, which is often used in gasifier cookstoves. The influence of two air supplies on the biomass conversion within the oxygen-limited regime, where the fuel conversion is linearly dependent on the air supply, is the focus of this study. Process implications of higher air supplies are also addressed.

- The conversion behaviour is similar for all fuels, irrespective of the ash-content, but the fuel conversion is inversely proportional to the ash-content.
- With increasing air supply and increasing process temperatures more fuel carbon is consumed and the possibility of ash-melting increases. By limiting the air supply, where more char is produced and peak temperatures are lower, ash-melting can be avoided.
- The fuel conversion to gaseous products in the oxygen-limited regime is mainly temperature dependent and independent of fuel type. Thus gaseous product estimation from this process could be based on the peak process temperatures.
- All produced chars achieve the highest classification, through international protocols, for soil amendment purposes, based on their elemental composition.

453 Overall, it is shown that the thermochemical conversion of a high value biomass fuel, wood pel-
454 lets, exhibit the best performance, but similar results can be achieved even with the lowest value
455 biomass fuels, manures, and the agricultural by-product, wheat straw. The issue of ash melting
456 and fusing, which is often detrimental to traditional combustion of high ash-content fuels can be
457 avoided here through limitation of the air supply and the production of char. This highlights the
458 potential for utilisation of low value fuels in the presented system for a combined production of
459 producer gas for heat generation and biochar for soil amendment applications.

460 **6. Acknowledgements**

461 The authors wish to acknowledge the support of The University of Adelaide and Marc Simp-
462 son, the laboratory facilities manager. The support provided by the Studienstiftung des Deutschen
463 Volkes is also gratefully acknowledged.

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