

THE UNIVERSITY of EDINBURGH

# Edinburgh Research Explorer

# Systematic evaluation of pyrolysis processes and biochar quality in the operation of low-cost flame curtain pyrolysis kiln for sustainable biochar production

## Citation for published version:

Jayakumar, Å, Morrisset, D, Koutsomarkos, V, Wurzer, C, Hadden, RM, Lawton, L, Edwards, C & Mašek, O 2023, 'Systematic evaluation of pyrolysis processes and biochar quality in the operation of low-cost flame curtain pyrolysis kiln for sustainable biochar production', *Current Research in Environmental Sustainability*, vol. 5, 100213. https://doi.org/10.1016/j.crsust.2023.100213

# **Digital Object Identifier (DOI):**

10.1016/j.crsust.2023.100213

## Link:

Link to publication record in Edinburgh Research Explorer

**Document Version:** Publisher's PDF, also known as Version of record

**Published In:** Current Research in Environmental Sustainability

## Publisher Rights Statement:

© 2023 The Authors. Published by Elsevier B.V

## **General rights**

Copyright for the publications made accessible via the Edinburgh Research Explorer is retained by the author(s) and / or other copyright owners and it is a condition of accessing these publications that users recognise and abide by the legal requirements associated with these rights.

## Take down policy

The University of Édinburgh has made every reasonable effort to ensure that Edinburgh Research Explorer content complies with UK legislation. If you believe that the public display of this file breaches copyright please contact openaccess@ed.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.



Contents lists available at ScienceDirect



Current Research in Environmental Sustainability



journal homepage: www.sciencedirect.com/journal/current-research-in-environmental-sustainability

# Systematic evaluation of pyrolysis processes and biochar quality in the operation of low-cost flame curtain pyrolysis kiln for sustainable biochar production

Anjali Jayakumar<sup>a,b,\*</sup>, David Morrisset<sup>c,1</sup>, Vasileios Koutsomarkos<sup>c,1</sup>, Christian Wurzer<sup>a</sup>, Rory M. Hadden<sup>c</sup>, Linda Lawton<sup>d</sup>, Christine Edwards<sup>d</sup>, Ondřej Mašek<sup>a</sup>

<sup>a</sup> UK Biochar Research Centre, School of GeoSciences, University of Edinburgh, Edinburgh, UK

<sup>b</sup> School of Engineering, Newcastle University, Newcastle Upon Tyne, NE17RU, UK

<sup>c</sup> Edinburgh Fire Research Centre, School of Engineering, University of Edinburgh, Edinburgh, UK

<sup>d</sup> CyanoSol, School of Pharmacy and Life Sciences, Robert Gordon University, Aberdeen, UK

ARTICLE INFO

Keywords: Flame curtain pyrolysis Low-cost homogeneous biochar Combustion science Emissions Operating guidelines

#### ABSTRACT

Low-cost pyrolysis units such as flame curtain pyrolysis kilns are gaining popularity for biochar production. However, the processes that govern the working of such units are not well understood. Here, emissions, temperatures and mass loss are monitored in real-time during kiln operation, followed by extensive biochar sampling. We found that by adjusting the layering rates of feedstock during kiln operation, we can obtain a biochar yield (28 wt% with a fixed carbon content of 65 wt%) comparable to that produced from the same feedstock in a continuous-scale pyrolysis unit, highlighting the importance of systematic guidelines for optimal kiln operation.

#### 1. Introduction

Biochar is a carbon-rich product resulting from the thermochemical conversion of carbonaceous materials, such as biomass, in an oxygendeficient environment at temperatures above 350 °C (Igalavithana et al., 2017; Lehmann and Joseph, 2015; Woolf et al., 2010). Biochar's long-term carbon-sequestration potential has gained it significant attention as a Carbon Dioxide Removal (CDR) technology and is recognised by the Intergovernmental Panel for Climate Change as a carbonnegative technology (Coninck et al., 2018). Biochar has high recalcitrance - the ability to highly resist both biotic and abiotic degradation making it possible for biochar to stay buried in soil for thousands of years as an efficient carbon sink (Woolf et al., 2010). Biochar can also be used in additional application scenarios prior to its use in soil as a carbon sink, ranging from water filtration, gas storage, and electrochemical applications (Jayakumar et al., 2021; Liu et al., 2019; Lonappan et al., 2020; Rodriguez-Narvaez et al., 2019). Biochar production is also effective as a sustainable waste valorisation technology, particularly in countries such as India, Indonesia, and China, where agricultural residue and organic wastes are often burnt each year (Chen et al., 2017; Goswami et al., 2019; Mathur and Srivastava, 2019). Biochar production thus allows waste-valorisation and use in multiple applications making it even more promising and cost-effective, with added advantages from its carbon sequestration potential (Yang et al., 2021).

Industrial biochar production is gaining more traction due to the climate emergency (Fawzy et al., 2021). However, low-cost processes such as flame curtain pyrolysis, TLUDs (Top-lit updraft gasifiers), double-chamber kilns and rotary cavity kilns could be of greater benefit to enable cost-effective biochar production without large capital inputs, especially in Low and Middle Income Countries (Schmidt et al., 2014; Sparrevik et al., 2013). Flame curtain pyrolysis in particular has gained a lot of interest for biochar production in the last few years due to its simplicity in terms of operation and equipment (a conical shaped kiln or soil pit) compared to the industrial pyrolysis or gasification units commonly used in large-scale biochar production (Schmidt et al., 2014). This cheaper option for household or community-scale biochar production has been especially appealing in locations without access to the infrastructure necessary to support more advanced technologies, or where costs of such technologies are prohibitive.

Existing literature demonstrates and compares the successful use of low-cost pyrolysers such as 'kon-tiki' flame curtain pyrolysers, TLUDs, traditional brick-made and earth-mound kilns to produce well-

https://doi.org/10.1016/j.crsust.2023.100213

Received 19 December 2022; Received in revised form 8 March 2023; Accepted 14 March 2023 Available online 27 March 2023 2666-0490/© 2023 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).

<sup>\*</sup> Corresponding author at: School of Engineering, Newcastle University, Newcastle Upon Tyne, NE17RU, UK.

E-mail address: anjali.jayakumar@ncl.ac.uk (A. Jayakumar).

<sup>&</sup>lt;sup>1</sup> These authors contributed equally.

Current Research in Environmental Sustainability 5 (2023) 100213

characterised biochar while successfully demonstrating its application as a soil additive (Cornelissen et al., 2016; Pandit et al., 2017; Schmidt et al., 2017). A certification standard for artisan biochar production using Kon-Tiki kilns already exists and is used by many farmers across the world (European Biochar Certification, 2022). However, there is still a significant knowledge gap concerning the pyrolysis processes, which underpin the operation of these low-cost biochar production technologies. As a result, there are no systematic guidelines to determine optimum and reproducible operating procedures and process conditions, which consider different feedstock, biochar yield and quality, or the overall emissions, especially pertaining to methane (CH<sub>4</sub>) and carbon monoxide (CO) emissions. This makes the safe replication of such studies challenging, especially when operated by personnel with little or no specific training. Non-regulated processes have the danger of leading to large quantities of emissions and biochar products that do not meet the international biochar standards, which would make its effective use in applications such as soil and water remediation challenging or infeasible.

While the progress found in current literature are welcome developments, it is necessary to establish a firm basis for assessing the efficiency and environmental performance of low-cost biochar production processes. This becomes more relevant when considering that voluntary carbon removal markets start to include biochar as a viable carbon offset technology for carbon sequestration, as recently shown by large purchases of biochar carbon offsets by Multi-National Corporations such as Microsoft (Microsoft, 2022). Such developments highlight the need for data to inform biochar production to produce good quality biochar, including from low-cost production units. Similarly, organisations such as Verra, who are working towards creating global standards for climate change mitigation, are currently developing accounting methodologies to include biochar in the world's largest voluntary CDR program (Verra, 2022). However, studies to properly account for greenhouse gas (GHG) emission reductions of low-cost pyrolysis, such as flame curtain pyrolysis, are still scarce. The conduction of such studies is vital to avoid incentivising potentially harmful practices, with negative environmental and health impacts on a local and/or global level.

Motivated by the above, this work aims to examine flame curtain pyrolysis through an expanded perspective using systematic evaluation protocols developed for combined characterisation of the pyrolysis processes and biochar products. This systematic evaluation allows study of the underlying mechanisms of a flame curtain kiln, and thereby identify some key operating parameters affecting biochar quality and process emissions. There are four major objectives towards this aim:

- a) to comprehensively monitor process conditions during the flame curtain pyrolysis process with real-time measurement of pyrolysis temperature regimes, gaseous emissions (GHG and others), and mass loss in a laboratory specially equipped for fire research,
- b) to systematically sample biochar from different parts of the kiln and characterise it for biochar quality and homogeneity,
- c) to compare quality of biochar produced using the same feedstock between flame curtain pyrolysis and a continuous screw reactor at the UK Biochar Research Centre,
- d) to evaluate the results from the first 3 objective, examining the efficacy and limitations of the adopted methods, and lay out opportunities and challenges for optimised kiln operation for future work.

The systematic evaluation methods that are developed here can easily be adopted to study other low-cost pyrolysis processes such as rotary cavity kilns and TLUDs. The success of the current rapidly expanding carbon removal market for biochar is going to be highly dependent on accurate carbon accounting for which there is currently very little data, especially for low-cost pyrolysis units. This makes the focus of this study timely and impactful.

#### 2. Materials and methods

#### 2.1. Theory- working principles of flame curtain pyrolysis kilns

To generate biochar, it is necessary to thermochemically convert the biomass feedstock (White et al., 2011). In many applications, the energy required for this thermochemical conversion, such as pyrolysis and gasification, is provided from external sources i.e. the separate combustion of fuel, to heat the feedstock to the pyrolysis temperature. To prevent oxidation of the hot biochar, it is necessary that this operation be performed in an inert (low oxygen) atmosphere (Morrisset et al., 2021). A flame curtain pyrolysis kiln seeks to reproduce these conditions by operating the kiln under conditions such that the pyrolysis gases produced are oxidised by a flame, which is sustained over the surface of the kiln (Schmidt et al., 2014). The kiln design ensures that oxygen is consumed in the flame and therefore cannot reach the biochar. A portion of the energy released by the flame is returned to the surface of the feedstock and promotes further pyrolysis (Morrisset et al., 2021; Quintiere, 2006; Torero, 2016). Thus, the pyrolysing biomass is heated from both the hot layer below and from heat transfer associated with the flame sheet above. The relative contributions of each heat transfer mechanism will vary with time as the biomass chars (Cornelissen et al., 2016; Schmidt et al., 2014). After some time, the pyrolysis rate of the top surface of the fuel will reduce to the point where the flame can no longer be sustained (Emberley et al., 2017). At this point, it is necessary to add additional feedstock, often called the 'layering process', to provide more fuel that generates pyrolysis gases such that the flame is sustained across the whole surface of the kiln.

Sustaining the flame across the surface of the kiln is an essential aspect of the operation of these devices. Since the kiln is closed on the sides and bottom, oxygen can only reach the hot char from the top. The flame must therefore be continuous across the surface to ensure that the oxygen cannot reach the hot char (by definition the oxygen concentration 'inside' a diffusion flame is zero), as shown in Fig. 1a (Burke and Schumann, 1928). In addition, maintaining a constant flame sheet limits the emission of pyrolysis products (e.g. CO and CH<sub>4</sub>) to the atmosphere as these are oxidised in the flame to  $CO_2$  and  $H_2O$ .

The successful optimal operation of the flame curtain pyrolysis reactor then relies on achieving an appropriate rate of application of the feedstock in the layering process (Cornelissen et al., 2016). This requires optimising against the characteristic time of the feedstock pyrolysis under the heating conditions of the kiln. The application rate should be high enough that a flow of pyrolysis gases is sustained such that a flame can exist on the whole surface, while allowing complete pyrolysis of the feedstock to ensure the quality and consistency of the biochar produced. High feeding rates also introduce the potential of smothering the flame.

At the end of production, steps must be taken to preserve the biochar and prevent further oxidation. This can be achieved in various ways such as flooding the reactor with water, or by covering the reactor with an impermeable layer of soil or a metal plate to prevent oxygen ingress and allowing the char to cool naturally (Pandit et al., 2017; Schmidt et al., 2014). Given the complexity of the processes outlined above (Roberts, 1971) and the heterogeneity of feedstock materials used, this study serves as a starting point to systematically investigate the production of biochar from a flame curtain reactor.

#### 2.2. Feedstock

This study utilised approximately rectangular-shaped hardwood woodchips obtained from Thistle Timber and Building Materials Ltd. Edinburgh, sized 2–4 cm long, 1–3 cm wide and 0.3–0.7 cm thick for all the experiments (**Fig. S1**). The woodchips were dried overnight at 105 °C prior to the experiments. Wood logs 15–20 cm long and 6–9 cm thick were used in the initial wood crib used for the start-up of the kiln.

We recognise that most feedstock obtained on the field for use in flame curtain pyrolysis would be bulkier and contain larger amounts of



**Fig. 1.** a) Illustrative energy balance of a flame curtain pyrolysis kiln, b) schematic of the experimental setup, c) top view and cross-section of the kiln, d) sampling spots in the kiln across the top, middle and bottom layers, e) photographs showing i) the complete kiln setup, ii) the initial wood crib iii) the initial wood crib alight, iv) the initial wood crib burning to form an initial hot layer upon which layers of feedstock are added, v) the 'flame curtain' when a new layer is added; the new layer's feedstock is pyrolysed by the heat transferred from both the flame and the hot layer beneath, vi) the kiln after the flame is quenched.

moisture. But considering that it was the first time such a study was undertaken, we used woodchips that would allow us to simplify the system, allowing us to study the pyrolysis processes and biochar quality in detail. This is thus a proof-of-concept that would allow us to establish a sound methodology to systematically study the flame curtain pyrolysis process and the resulting biochar.

#### 2.3. Experimental setup and operation

Trial runs were performed using woodchips to determine the start-up experimental protocol, mass of feedstock per run, type and positions of monitoring equipment such as an FTIR probe and thermocouples as shown in Fig. 1b. A steel isosceles trapezoidal prism was chosen as the shape of the kiln with dimensions as shown in Fig. 1c. The kiln was constructed from 3 mm thick steel sheets welded together at the joints. This kiln size is typical of a small-scale kiln used in household and field applications (Cornelissen et al., 2016). Visual observations from all the experiments were noted. The start-up process was kept consistent across all runs with the creation of an initial wood crib (Fig. 1e-ii) made of 750 g dried woodchips, 2.5 kg wood logs and lit using two propane blowtorches. Five to six minutes after the fire is lit, the initial wood crib collapses and the wood logs are spread evenly by the operator. Woodchips (400 g) were then added approximately every two minutes for a total of five times before layering began.

During the layering process, the layering rate was varied by keeping the loading interval stable at approximately 60 s, while the mass of feedstock added per layer was either 200, 400, or 600 g, weighed each time for consistency. This was done to identify the effect of different layering rates on the biochar production and overall burning behaviour observed in the kiln. These runs were referred to as WC 200, WC 400 and WC 600, respectively to represent the woodchips (WC) used as feedstock and the layering rate used for each run. Feedstock layers were manually dispersed over the surface of the kiln to distribute the feedstock across the kiln surface as evenly as possible. Fig. 1e- i to v outline the stages of an experimental run, and Table 1 below shows the details of each run.

After quenching the flames with water at the end of every run, sampling of biochar was done manually by taking five samples from the top layer and three samples from the middle and bottom layer following the pattern shown in Fig. 1d. The different layer depths (bottom layer, middle layer, top layer) from which the samples have been drawn are given in Table 1 for each run. All experiments were conducted to achieve a final pile depth of >10 cm, but <70% of the total kiln depth (~19 cm), and a run duration of 25–32 min.

#### 2.4. Measurements

During the operation of the kiln, temperature measurements were made using 16 Type K (1.5 mm diameter) sheathed thermocouples inside the kiln. The location of the thermocouple tree and thermocouple positions on the tree are shown in Fig. 1b. The mass of the kiln was recorded (including the addition of feedstock layers) to allow determination of the mass loss (pyrolysis) rate. Gaseous emissions directly above the kiln were measured using Fourier Transform Infrared Spectroscopy (FTIR) with a Gasmet Dx4000 with a heated sampling line. The concentrations of methane, carbon monoxide, benzene and formaldehyde were measured with a 20 s sampling period directly above the kiln. The exhaust gases from the extraction hood were sampled to measure O<sub>2</sub>, CO, and CO<sub>2</sub> concentrations. Combining these measurements with the flow measurement in the exhaust duct allowed the Heat (energy) Release Rate (HRR) to be calculated (Janssens, 1991). The concentration of the exhaust gases can be used to determine emission mass flow rates. which can be used in combination with mass measurements to calculate emission yields (g/g feedstock) to be calculated.

#### 2.5. Biochar characterisation

#### 2.5.1. Biochar yields from initial wood crib and layering

Biochar samples obtained from all runs were crushed using a blender and sieved to <1 mm for all characterisations. Due to real-time logging of weights, approximate yields are calculated for both the initial wood crib (biochar from bottom layer) and the layering process (biochar from top and middle layers), represented as Yield or FC yield (Bottom) and Yield or FC yield (Layers) respectively and all values are on a moisturefree, dry basis (d.b) (Eqs. S1-S6). The biochar yield (Bottom) was calculated using weights of initial wood crib feedstock and weight of biochar in the kiln recorded just before the start of layering process in Eq. 1. Similarly, biochar yield (Layers) was calculated using Eq. 1 with the weight of feedstock used in layering process and the weight of biochar obtained from the layers; this being calculated as the difference of weights of biochar in the kiln at the end of the process and weight of char in the kiln just before the start of the layering process. It should be noted that this calculation may overestimate the bottom yields since the pyrolysis in the bottom layer continues pyrolysing even during the layering.

$$Biochar yield (\% feedstock) = \frac{Biochar (Kg)}{Feedstock (Kg)}$$
(1)

#### 2.5.2. Proximate analysis by thermogravimetric analysis

Volatile matter (VM), Fixed Carbon (FC), and Ash content were

6

22

13-17

6-12

9-0

16-17

0.03

35

ē.

t.20

6.15

600

NO

[able]

tails of	the woodchip e	experiment runs	pertormed 11	n the flame curtain pyro	olysis kiin.						
lun	Bottom layer	Wt. in layers	Total feed	Avg. Moisture in feed	Total layers	Final depth of pile	Bottom layer (3 samples)	Middle (3 samples)	Top (5 samples)	Total run duration	Layering duration
	Kg	Kg	Kg	Wt%		cm	cm	cm	cm	min	min
VC 200	6.15	2.80	8.95	0.03	14	13-14	0-6	6-10	10 - 13	32	14
NC 400	6.15	6.80	12.95	0.03	17	16-17	0-6	6-12	13-17	30	17

determined by thermogravimetric analysis using a TGA/DSC 1; Mettler-Toledo, Leicester, UK by the standard methods used for biochar (in quadruplicates), as described by (Crombie et al., 2013).

# 2.5.3. Analysis of biochar samples for polycyclic aromatic compounds (PAHs)

Biochar samples were analysed for PAH content by a commercial laboratory (mas gmbh, Germany, DIN EN 16181 (DIN SPEC 91243):2013–12 in combination with mas house method MAS PA069:2017–12.

#### 2.5.4. Biochar from continuous biochar production units

Woodchip biochar were produced in a continuous flow pyrolysis unit as described in previously work (Buss et al., 2016; Mašek et al., 2018), to compare the quality of biochar produced in flame curtain pyrolysis to biochar produced from the same feedstock in a controlled, lab-scale pyrolysis unit (**Fig. S2**). The woodchips were pyrolysed at 400 °C and 500 °C in an N<sub>2</sub> stream at a flow rate of 1 Lmin<sup>-1</sup> with an average residence time of approximately 40 min and named WC 400 UKBRC and WC 500 UKBRC respectively, with WC (woodchip) representing the feedstock used in the pyrolysing units at the UK Biochar Research Centre (UKBRC), and 400, 500; the temperatures of pyrolysis respectively. The temperature ranges were chosen to reflect the temperatures observed during flame curtain pyrolysis.

#### 3. Results and discussions

We evaluated mass-loss rates, emissions, temperature profiles, and resulting biochar quality obtained from the operation and real-time monitoring of a flame curtain pyrolysis kiln operated with different layering rates. We found that the change in layering rates influences the combustion environment, particularly affecting the temperatures in the kiln and emission profiles. This is evidenced by changes in the mass loss rate and emissions from the kiln. The change in temperatures directly affected the quality and homogeneity of biochar, as evidenced by characterisation of biochar systematically sampled from different parts of the kiln. We also found that it is possible to produce homogeneous biochar meeting international standards from the operation of a flame curtain kiln for the specific conditions used here. This biochar is also comparable in quality to the biochar produced from the same feedstock in a lab-scale continuous screw reactor, with the biochar yield and fixed carbon content slightly lower in the case of flame curtain pyrolysis.

#### 3.1. Mass loss rates and Hood emissions

The combustion conditions in the kiln are characterised by the Mass Loss Rate or MLR (i.e. the pyrolysis rate of the feedstock) (Quintiere, 2006; Drysdale, 2011) and the mass flow of CO and  $CO_2$  as shown in Fig. 2 for each of the feedstock conditions.

The mass data has been processed to show only the period of the layering. For WC 200, the mass loss rate was around 3 g/s for the duration of the experiment with some variation/oscillation observed in synchronisation with the addition of feedstock. The production of CO and  $CO_2$  was relatively constant under these conditions. For WC 400, the mass loss rate did not reach steady state until 10 min into the layering where it then stabilises at 6 g/s. This is reflected in CO and  $CO_2$  mass fluxes that steadily increased over the duration of the layering period and then stabilised as the mass loss rates stabilised after 10 min. For WC 600, the mass loss rate, CO and  $CO_2$  concentrations did not reach a steady state and continually increased after the first two minutes of layering. However, regardless of achieving a steady state condition, the observed MLR behaviour of each layering rate closely matched that of the observed emission trends.

These different trends observed in mass loss rates and emissions as a result of using different layering rates provide insights into the working mechanisms of flame pyrolysis process. Varying the layering rate inherently alters the energy balance and hence the rate of pyrolysis in the kiln (thus influencing the time resolved MLR, CO and  $CO_2$  emissions). Optimum residence times are instrumental for complete pyrolysis or thermochemical conversion of the feedstock. This is evidenced by the increase in mass loss rate between the WC 200 and WC 400 cases while the same trend does not continue for the WC 600 case, as the residence time available for pyrolysis was much lower.

While different layering rates have led to different emission and mass-loss trends (i.e., pyrolysis behaviour), it is also important to closely look at how different mass loss rates and emission trends affect the temperature profiles and the resulting biochar quality and homogeneity. This is important to draw balanced insights on the implications of whether these relatively steady mass loss rates observed in WC 200 and WC 400 can point to a more complete pyrolysis for good yield and quality of biochar compared to WC 600.

#### 3.2. Temperature profiles

The temperature data has been processed to show the period of layering. The average temperature data obtained from 16 thermocouples (4 thermocouples for each height - 5, 10, 15 and 20 cm from the bottom of the kiln) are presented in Fig. 3 **a to c**. These indicate that the temperature of the biochar layer at 5 and 10 cm in WC 200, WC 400 and WC 600 are relatively constant for the duration of the kiln operation. The temperature at 5 cm from the bottom of the kiln were between 300 and 350 °C for each layering rate. The temperature at 10 cm showed some dependence on the layering rate. For the WC 200 case, there is an initial period of fluctuation followed by a quasi-steady temperature between 450 and 500 °C. The WC 400 is characterised by temperatures in excess of 400 °C for an initial period before decreasing to a relatively constant 380 °C for the remainder of the operation. The WC 600 case is characterised by a constantly decreasing average temperature again



Fig. 2. The time resolved MLR data, CO<sub>2</sub> and CO emissions for the layering period for a) WC 200, b) WC 400, c) WC 600, (time zero in all graphs represents the start of layering process).



**Fig. 3.** Average temperature-time profiles recorded for the layering process at different heights from the bottom of the kiln a) WC 200 at 5 and 10 cm, b) WC 400 at 5 and 10 cm, c) WC 600 at 5 and 10 cm, d) WC 200 at 15 and 20 cm, e) WC 400 at 15 and 20 cm, f) WC 600 at 5 and 10 cm (time zero in all graphs represents the start of layering process and standard errors are represented by the shaded regions around the average curves).

indicating that this higher layering rate is altering the energy balance resulting in sporadic and unstable heating from the flame to the unpyrolysed feedstock.

The temperature profile in the kiln at 15 and 20 cm from the bottom of the kiln was determined (Fig. 3 d, e and f). These data show high degrees of fluctuation when the thermocouple is measuring in the gas phase (i.e., hot gasses). The final pile depth in the kiln was 16-17 cm and as a result, the feedstock came in contact with the thermocouple at 15 cm after 10 min, indicated by reduced fluctuations and temperatures averaging around 400–480  $^\circ\text{C}.$  Both WC 200 and WC 400 trials recorded temperatures above 400 °C in the kiln indicating that the temperatures were sufficient for complete pyrolysis of feedstock at these heights. However, the thermocouples in the WC 600 case started recording the feedstock temperatures at a height of 15 cm by 6 min. Recorded temperatures oscillated between 250 and 500 °C, indicating that the combustion conditions were variable possibly due to local quenching of the flame resulting in localised burning. In all cases, the temperatures at 20 cm from the bottom of the kiln exhibited significant fluctuation throughout each experiment, indicating that these are gas phase temperature measurements, and not representative of the biochar production conditions.

The layering rates influenced the temperature profiles by altering the net heat flux interactions between the flame and the feedstock (fuel), this resulted in changes in temperature within the feedstock regions of the kiln, which will directly affect the quantity and quality of biochar produced, as will be discussed in more detail in the Section 3.3.

#### 3.3. Biochar composition, yields and homogeneity

We analysed the effects of different layering rates used in WC 200,

WC 400 and WC 600 on the composition, homogeneity and yield of biochar. We also compare the composition and yield of biochar produced from flame curtain pyrolysis to that obtained from the same feedstock in a continuous scale pyrolysis unit having almost ideal conditions for complete pyrolysis such as uniform temperature, optimum residence time and oxygen-deficient conditions.

#### 3.3.1. Biochar from flame curtain pyrolysis

All the biochar samples taken from the bottom, middle and top layers were analysed for composition using proximate analysis. The biochar compositions along with the digital photographs of the samples from for all the woodchip runs are provided in Fig. 4. Biochar yields and average biochar compositions are provided in Table 2.

For the same feedstock, with increase in Highest Treatment Temperature (HTT) during pyrolysis, there is a higher break down of organic matter, releasing more volatile matter from the system, while increasing the fixed carbon content in biochar (Crombie et al., 2013). This is because of the different extents to which cellulose, hemicellulose, and lignin break down under different temperatures (White et al., 2011).

Despite the almost identical initial wood crib and start-up processes, there is a slight shift in the biochar composition as we move from WC 200 to WC 400 to WC 600. The fixed carbon content of the bottom layer decreased from 69 to 64 to 57 wt% for WC 200, WC 400 and WC 600 respectively. This can be attributed to the changes in temperatures for the different layering (Fig. 3 **a**, **b**, and **c**).

The changes in biochar quality observed from the layering process provide further insight in the various transient phenomena that dictate and describe the transient pyrolysis process. For WC 200, the largely stable mass loss rate and temperatures in the kiln produced homogeneous biochar, as can be seen by the uniform composition of biochar



**Fig. 4.** Photographs of samples taken from top, middle and bottom layers for a) WC 200, c) WC 400, e) WC 600, and biochar composition in weight % on a dry basis (d.b) calculated from proximate analysis of biochar sampled from bottom, middle, top and top-middle layers for b) WC200, d) WC 400, f) WC 600. \* - B- biochar sampled from bottom timber crib, M- biochar sampled from Middle layers, T- biochar sampled from Top, T + M – biochar sampled from top and middle layers representing the total biochar from layering.

across different layers with very small variations (Fig. 4b). There is also a visual confirmation to this finding based on the images of similar-sized biochar samples taken from top and middle layers of the kiln as seen in Fig. 4a. The temperatures observed in the top layers for the WC 400 run displayed slightly higher variation compared to WC 200 (Fig. 3). This consequently resulted in a higher degree of variation in the biochar composition, especially for the biochar from the top layer (Fig. 4d). While the average fixed carbon content of biochar produced from the

#### Table 2

Woodchip biochar yields and average composition.

	Yield		Fixed Carbon		Fixed Carbon Yield		Volatile Matter		Volatile Matter Yield		Ash		Ash Yield	
Run	W	L	w	L	w	L	W	L	w	L	w	L	w	L
	%	%	%	%	%	%	%	%	%	%	%	%	%	%
WC 200	23.7	19.8	69.3	70.1	16.4	13.9	26.2	25.1	6.2	4.9	4.3	4.7	1.04	0.9
WC 400	29.4	27.7	63.6	65.3	18.7	18.1	32.2	30.4	9.51	8.4	4.1	4.1	1.1	1.2
WC 600	27.4	63.5	56.7	41.2	15.5	26.2	38.7	51.9	10.6	33.1	4.4	6.7	1.2	4.3
WC 400C UKBRC	43.01		65.7		28.4		30.2		12.9		3.9		1.7	
WC 500C UKBRC	30.1		75.1		22.6		19.4		5.7		5.3		1.5	

W-Initial Woodcrib, L- Layers.

layering process is higher for WC 200 (70 wt%) compared to WC 400 (65 wt%), the biochar and fixed carbon yields (% feed) are only 20% and 14% for WC 200 compared to 28% and 18% for WC 400 respectively. We also see higher emissions normalised per mass of feedstock and biochar for WC 200 compared to WC 400 (Fig. 5a and b, **Table S1, Table S2**). Moreover, a larger mass of feedstock was consumed for WC 200 compared to WC 400 to produce the same amount of biochar (Fig. 5a). Despite gaining homogeneous biochar with a higher fixed carbon content, much lower biochar yields with higher emissions per Kg of biochar produced indicate lower process efficiency for WC 200.

The WC 600 run had far more variations in all aspects; the temperatures, mass loss rates, and emissions were all strongly indicative of deviations from a steady pyrolysis process. The biochar samples were non-homogeneously pyrolysed, as evident from a mix of burnt and woody biochar chips (Fig. 4e). The biochar obtained was nonhomogeneous (large variations in biochar composition) with a significant amount of non-pyrolysed woodchips with high volatile matter and low fixed carbon content (Fig. 4f) indicative of hot and cold spots in the kiln. The average FC content of the biochar obtained from layering process was only 41%, falling below the 50% organic carbon content requirement to be defined as 'biochar' (for woody biomass as per European Biochar Certificate guidelines). The low temperatures led to incomplete, non-steady flow of pyrolysis gases during the layering process. This led to the lack of a stable flame curtain necessary to sustain a steady pyrolysis. Even though the biochar yields and fixed carbon yields calculated for WC 600 in Table 2 are higher compared to WC 200 and WC 400, the low fixed carbon content and non-homogeneous nature of the char in the kiln make this a poor option for the production of good quality biochar using flame curtain pyrolysis. Moreover, with the normalised emissions for CO and CO2 indicate lower emissions compared to WC 200 and WC 400 (Fig. 5a and b), it becomes clear that the WC 600 run serves as an example for how a process that produces pyrogenic carbon of low quality can mask bad reactor efficiency if we only look at the yields or emissions in isolation, especially when larger layering rates (layering done too quickly) are used.

While there have been efforts to assess CO and  $CH_4$  trends for biomass pyrolysis, especially for pyrolysers including industrial units, the current study does not provide enough data to directly link CO and  $CH_4$  emissions (Sørmo et al., 2020). Hence, we recommend future research to assess potential links between CO and  $CH_4$  emissions or monitor  $CH_4$  emissions directly to estimate the full environmental impact of the process.

These results shed light on the practical implications of operating the kiln by highlighting the importance of adding feedstock at an optimum rate. We show that it is possible to adjust the layering rate to achieve steady mass loss rates and stable temperatures. This in turn can ensure steady outgassing necessary for maintaining a stable flame curtain to sustain a steady pyrolysis process capable of producing homogeneous biochar, while minimising emissions and ensuring better yields.

# 3.3.2. Comparing biochar from flame curtain pyrolysis and continuous pyrolysis unit at UKBRC

Woodchip biochar was produced at UKBRC to compare the quality of biochar produced in flame curtain pyrolysis to biochar produced from the same feedstock in a controlled, lab-scale pyrolysis unit. This was done to gain insights on the effectiveness of the two differing approaches and define acceptability of the end product.

It can be seen that the woodchip biochar compositions from WC 200 and WC 400 fall within the composition ranges seen in WC 400C UKBRC and WC 500C UKBRC (Fig. 6 a and b). Most notably, among all the runs



**Fig. 5.** a) Total  $CO_2$  emissions normalised per Kg of feedstock, total  $CO_2$  emissions normalised per Kg of biochar produced and mass of feedstock consumed per Kg of biochar produced, b) Total CO emissions normalised per Kg of feedstock, total CO emissions normalised per Kg of biochar produced, (all graphs show emissions quantified for layering).



Fig. 6. a) Digital photograph and b) composition; both of biochar produced at two different temperatures 400 °C and 500 °C in a continuous screw reactor at UKBRC -WC 400C UKBRC, WC 500C UKBRC.

conducted (we avoid WC 600 in our comparisons due to FC content below international guidelines for biochar) (European Biochar Foundation (EBC), 2016; IBI, 2022), WC 400C UKBRC had the highest average biochar yield of 43% while WC 200 has the lowest at 20%. It is welcoming to see that even though our runs were not optimised, the WC 400 run from an open pyrolysis unit could operate with a biochar yield (layering) of 28% with an average biochar FC content of 65%, comparable to WC 500C UKBRC run with a 33% yield with an average biochar FC content of 75%. However, it is important to note that an open flame curtain kiln makes it more difficult to utilise the waste heat generated during its operation.

Pyrolysis, especially at lower temperatures ( $\sim$ 350–500 °C) can sometimes lead to unfavourable production of toxic compounds such as Polycyclic Aromatic Hydrocarbons (PAHs) that stick to biochar surfaces (Buss et al., 2016; Buss et al., 2022). To check for PAH levels in our biochar, we also tested all our biochar samples, both from flame pyrolysis and our UKBRC unit for US EPA 16 PAHs by protocols recommended by IBI (International Biochar Initiative) guidelines. All samples had PAHs concentrations below the IBI recommended limits of 6–20 mg Kg<sup>-1</sup>, except the woodchip biochar produced at 400 °C UKBRC with slightly elevated PAH levels (Table 3). This unit's inherent design usually leads to more PAHs for low temperature biochar for certain types of feedstock, due to issues with separation of solid and vapour streams, making it more suitable for higher temperature (> 500 °C) biochar production. It is also worth noting that the low PAHs in WC 600 was because of the largely non-pyrolysed wood and not because of a better

#### Table 3

PAH concentrations in biochar.

Sample	16 EPA PAHs (mg Kg- <sup>1</sup> )
WC 200 (biochar sampled from top and middle layers) WC 400 (from biochar sampled from top and middle layers)	1.98 1.44
WC 600 (from biochar sampled from top and middle layers)	0.576
WC 400C UKBRC	25.3
WC 500C UKBRC	5.84

quality of biochar with low PAHs.

While the results of the study do not provide sufficient information that would allow optimisation of operating conditions for flame curtain pyrolysis, they do provide important guidance on parameters affecting the efficacy and environmental impact of the process and help to narrow down the operating envelope of the process. It also shows the importance of evaluating the performance of flame curtain pyrolysis and other low-cost pyrolysis processes by taking in to account various relevant and often inter-playing parameters such as biochar homogeneity in the kiln, temperature profiles, quantity and quality of emissions, yields, and biochar composition. It is also important to note that while conclusions from this study couldn't be directly applied to bulkier feedstock usually used on the ground, the results from this study will be relevant to feedstock such as coconut shells, coconut husk, corn cob and other agricultural crop residue that are available in abundance in farm and large agricultural settings.

However, to create more generalised guidelines, it is essential to further assess and quantify the various heat transfer conditions and mechanisms involved in the flame curtain pyrolysis. This would involve, among many other factors, assessment, and quantification of conduction heat transfer rates between layers and the radiant heat feedback from the flame. Understanding these underlying mechanisms will allow us to fine-tune and control the heat transfer rates, which could then be used to optimise the process (quantifying what is 'too slow', 'too quick', 'too much' or 'too little ' in terms of layering of different feedstock types and kiln sizes) for sustainably producing biochar. Future work can then provide simple-to-follow standard operating procedures and instructional videos for reproducible and sustainable biochar production from low-cost pyrolysis processes for users worldwide. Such systematic evaluation will also allow us to obtain enough data necessary for different certification and carbon accounting methodologies or initiatives for climate change mitigation involving biochar. Any attempt to evaluate performance on a single indicator can be misleading. The results presented here are representative of yields and emissions that can be expected, for the specific materials and conditions used.

#### 4. Conclusions

Permissible 16 EPA PAH limits in mg Kg<sup>-1</sup>: IBI guidelines- 6-20.

Our study shows that it is possible to adjust the layering rates during

a flame curtain kiln operation to yield biochar comparable to the quality and yield of biochar produced in a continuous biochar production facility. The study also showed that the true performance of a flame curtain pyrolysis process could be seriously misjudged if only biochar yields or gaseous emissions are considered in isolation. This work serves as a starting point for the creation of guidelines for optimum operation of a flame curtain pyrolysis kiln to optimise biochar quality, biochar yield, and emissions.

#### Funding

This work was supported by the Biotechnology and Biological Sciences Research Council Grant (BB/S011579/1). C. Wurzer received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 721991.

#### Contributions

Conceptualization: AJ, OM, LL, CE, Methodology: AJ, CW, DM, VK, RH, OM, Investigation: AJ, CW, VK, DM, RH, Visualization: AJ, DM, VK, CW, RH, Funding acquisition: LL, CE, RH, OM, Project administration: AJ, RH, OM, Supervision: RH, OM, Writing – original draft: AJ, VK, DM, CW, Writing – review & editing: AJ, DM, VK, CW, RH, OM, LL.

#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Data availability

Data will be made available on request.

#### Acknowledgements

The authors thank Gokul GR for giving useful suggestions for improving the manuscript. The authors are grateful for the support of the staff and students at the Edinburgh Fire Research Centre.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.crsust.2023.100213.

#### References

- Burke, S.P., Schumann, T.E.W., 1928. Diffusion Flames. Ind. Eng. Chem. 20, 998–1004. https://doi.org/10.1021/ie50226a005.
- Buss, W., Graham, M.C., MacKinnon, G., Mašek, O., 2016. Strategies for producing biochars with minimum PAH contamination. J. Anal. Appl. Pyrolysis 119, 24–30. https://doi.org/10.1016/j.jaap.2016.04.001.
- Buss, W., Hilber, I., Graham, M.C., Mašek, O., 2022 May 23. Composition of PAHs in Biochar and Implications for Biochar Production. ACS Sustain. Chem. Eng. 10 (20), 6755–6765. https://doi.org/10.1021/acssuschemeng.2c00952. Epub 2022 May 11. PMID: 35634266; PMCID: PMC9131514.
- Chen, J., Li, C., Ristovski, Z., Milic, A., Gu, Y., Islam, M.S., Wang, S., Hao, J., Zhang, H., He, C., Guo, H., Fu, H., Miljevic, B., Morawska, L., Thai, P., Lam, Y.F., Pereira, G., Ding, A., Huang, X., Dumka, U.C., 2017. A review of biomass burning: emissions and impacts on air quality, health and climate in China. Sci. Total Environ. 579, 1000–1034. https://doi.org/10.1016/j.scitotenv.2016.11.025.
- Coninck, De, et al., 2018. No Title. Strength. Implement. Glob. Response. Glob. Warm. 1.5°C. An IPCC Spec. Rep. impacts Glob. Warm. 1.5°C above pre-industrial levels Relat. Glob. Greenh. gas Emiss. pathways, Context strengthen.
- Cornelissen, G., Pandit, N.R., Taylor, P., Pandit, B.H., Sparrevik, M., Schmidt, H.P., 2016. Emissions and char quality of flame-curtain "Kon Tiki" kilns for farmer-scale charcoal/biochar production. PLoS One 11, 1–16. https://doi.org/10.1371/journal. pone.0154617.

- Crombie, K., Mašek, O., Sohi, S.P., Brownsort, P., Cross, A., 2013. The effect of pyrolysis conditions on biochar stability as determined by three methods. GCB Bioenergy 5, 122–131. https://doi.org/10.1111/gcbb.12030.
- Drysdale, Dougal, 2011. An Introduction to Fire Dynamics. John wiley & sons. https:// doi.org/10.1002/9781119975465.
- Emberley, R., Inghelbrecht, A., Yu, Z., Torero, J.L., 2017. Self-extinction of timber. Proc. Combust. Inst. 36, 3055–3062. https://doi.org/10.1016/j.proci.2016.07.077.
   European Biochar Certification (EBC), 2016. Guidelines for a sustainable production of
- biochar. Eur. Biochar Found. 1–22 https://doi.org/10.13140/RG.2.1.4658.7043.
   European Biochar Certification (EBC), 2022. Guidelines for Carbon Sink Certification for Artisan Biochar Production. https://www.european-biochar.org/media/doc
- /139/artisan-c-sink-guidelines v1\_0.pdf.
  Fawzy, S., Osman, A.I., Yang, H., Doran, J., Rooney, D.W., 2021. Industrial biochar systems for atmospheric carbon removal: a review. Environ. Chem. Lett. 19, 3023–3055. https://doi.org/10.1007/s10311-021-01210-1.

Goswami, S.B., Mondal, R., Mandi, S.K., 2019. Crop residue management options in rice-rice system: a review. Arch. Agron. Soil Sci. 1–17.

- IBI, 2022. https://biochar-international.org/certification-program/.
- Igalavithana, A.D., Mandal, S., Niazi, N.K., Vithanage, M., Parikh, S.J., Mukome, F.N.D., Rizwan, M., Oleszczuk, P., Al-Wabel, M., Bolan, N., Tsang, D.C.W., Kim, K.H., Ok, Y. S., 2017. Advances and future directions of biochar characterization methods and applications. Crit. Rev. Environ. Sci. Technol. 47, 2275–2330. https://doi.org/ 10.1080/10643389.2017.1421844.
- Janssens, M., 1991. Piloted ignition of wood: a review. Fire Mater. 15, 151–167. https:// doi.org/10.1002/fam.810150402.
- Jayakumar, A., Wurzer, C., Soldatou, S., Edwards, C., Lawton, L.A., Mašek, O., 2021. New directions and challenges in engineering biologically-enhanced biochar for biological water treatment. Sci. Total Environ. 796, 148977 https://doi.org/ 10.1016/j.scitotenv.2021.148977.
- Lehmann, J., Joseph, S., 2015. Biochar for Environmental Management: Science, Technology and Implementation. Routledge.
- Liu, W.-J., Jiang, H., Yu, H.-Q., 2019. Emerging applications of biochar-based materials for energy storage and conversion. Energy Environ. Sci. 12, 1751–1779. https://doi. org/10.1039/C9EE00206E.
- Lonappan, L., Liu, Y., Rouissi, T., Brar, S.K., Surampalli, R.Y., 2020. Development of biochar-based green functional materials using organic acids for environmental applications. J. Clean. Prod. 244 https://doi.org/10.1016/j.jclepro.2019.118841.
- Mašek, O., Buss, W., Roy-Poirier, A., Lowe, W., Peters, C., Brownsort, P., Mignard, D., Pritchard, C., Sohi, S., 2018. Consistency of biochar properties over time and production scales: a characterisation of standard materials. J. Anal. Appl. Pyrolysis 132, 200–210. https://doi.org/10.1016/j.jaap.2018.02.020.
- Mathur, R., Srivastava, V.K., 2019. Crop residue burning: effects on environment. In: Greenhouse Gas Emissions. Springer, pp. 127–140.
- Microsoft, 2022. FY22 white Pap. 30–31. https://dl10erj175o600.cloudfront.net/wpcontent/uploads/2022/03/14135114/2021-Environmental-Sustainability-Report. pdf.
- Morrisset, D., Hadden, R.M., Bartlett, A.I., Law, A., Emberley, R., 2021. Time dependent contribution of char oxidation and flame heat feedback on the mass loss rate of timber. Fire Saf. J. 120, 103058 https://doi.org/10.1016/j.firesaf.2020.103058.
- Pandit, N.R., Mulder, J., Hale, S.E., Schmidt, H.P., Cornelissen, G., 2017. Biochar from "Kon Tiki" flame curtain and other kilns: effects of nutrient enrichment and kiln type on crop yield and soil chemistry. PLoS One 12, 1–18. https://doi.org/10.1371/ journal.pone.0176378.
- Quintiere, J.G., 2006. A theoretical basis for flammability properties. Fire Mater. 30, 175–214. https://doi.org/10.1002/fam.905.
- Roberts, A.F., 1971. Problems associated with the theoretical analysis of the burning of wood. Symp. Combust. 13, 893–903. https://doi.org/10.1016/S0082-0784(71) 80090-5.
- Rodriguez-Narvaez, O.M., Peralta-Hernandez, J.M., Goonetilleke, A., Bandala, E.R., 2019. Biochar-supported nanomaterials for environmental applications. J. Ind. Eng. Chem. 78, 21–33. https://doi.org/10.1016/j.jiec.2019.06.008.
- Schmidt, H., Taylor, P., Eglise, A., Arbaz, C., 2014. Kon-Tiki flame curtain pyrolysis for the democratization of biochar production. Biochar J. 14–24.
- Schmidt, H.P., Pandit, B.H., Cornelissen, G., Kammann, C.I., 2017. Biochar-based fertilization with liquid nutrient enrichment: 21 Field trials covering 13 crop species in Nepal. L. Degrad. Dev. 28, 2324–2342. https://doi.org/10.1002/ldr.2761.
- Sørmo, E., Silvani, L., Thune, G., Gerber, H., Schmidt, H.P., Smebye, A.B., Cornelissen, G., 2020. Waste timber pyrolysis in a medium-scale unit: emission budgets and biochar quality. Sci. Total Environ. 718 https://doi.org/10.1016/J. SCITOTENV.2020.137335.
- Sparrevik, M., Field, J.L., Martinsen, V., Breedveld, G.D., Cornelissen, G., 2013. Life cycle assessment to evaluate the environmental impact of biochar implementation in conservation agriculture in Zambia. Environ. Sci. Technol. 47, 1206–1215. https:// doi.org/10.1021/es302720k.
- Torero, J., 2016. Flaming ignition of solid fuels. In: Hurley, M.J., Gottuk, D., Hall, J.R., Harada, K., Kuligowski, E., Puchovsky, M., Torero, J., Watts, J.M., Wieczorek, C. (Eds.), SFPE Handbook of Fire Protection Engineering. Springer, New York, New York, NY, pp. 633–661. https://doi.org/10.1007/978-1-4939-2565-0\_21. Verra, 2022. https://verra.org/.

#### A. Jayakumar et al.

White, J.E., Catallo, W.J., Legendre, B.L., 2011. Biomass pyrolysis kinetics: a

comparative critical review with relevant agricultural residue case studies. J. Anal.

- Appl. Pyrolysis 91, 1–33. https://doi.org/10.1016/j.jaap.2011.01.004. Woolf, D., Amonette, J.E., Street-Perrott, F.A., Lehmann, J., Joseph, S., 2010. Sustainable biochar to mitigate global climate change. Nat. Commun. 1, 56. https://doi.org/ 10.1038/ncomms1053.
- Yang, Q., Zhou, H., Bartocci, P., et al., 2021. Prospective contributions of biomass pyrolysis to China's 2050 carbon reduction and renewable energy goals. Nat. Commun. 12, 1698. https://doi.org/10.1038/s41467-021-21868-z.