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Supplementary Materials

THE IMPROVED ENERGY EFFICIENCY OF APPLIED SMOULDERING SYSTEMS WITH INCREASING SCALE

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1 Present address
S.1. Global Energy Balance

A global energy balance following Rashwan et al., [1] was conducted on the robust experiments in Table 1. Equation (1) was employed assuming $E_{\text{out}} = 0$, since the time of specific interest was before the smouldering front reached the top of the fuel bed at the end of propagation ($t_f$) (see further discussion below in Section S.10), and neglecting $E_{\text{pyr}}$, which has been shown to be minor relative to the other terms in other applied smouldering systems [2] and neglected here because of the simple GAC degradation (see further discussion in Sections 2.1 and S.2). However, this is not valid in all cases as pyrolysis will be more relevant for other fuels under various smouldering conditions [3]. Here, the chemistry is only approximated, and some pyrolysis reactions evolving CO or CO$_2$ may be embedded within the estimated $E_{\text{oxid}}$ term. Nevertheless, the following analysis will be structured as if the net heat released from smouldering is from an effective oxidation term, which aligns with previous smouldering research using similar fuels [4-7].

The fundamental chemistry steps within smouldering systems, even simple systems using pure carbon, GAC, or charcoal, presents an opportunity for future research [3]. Isolating the unknown term in Eq. (1) on the left:

$$
\dot{E}_{\text{loss}} = \dot{E}_{\text{in}} + \dot{E}_{\text{oxid}} - \frac{dE_{\text{net}}}{dt}
$$

(S.1)

for $0 < t < t_f$

Integrating each of these terms in time from time zero (turning on the heater) solves $E_{\text{loss}}$ during propagation:
\[ E_{\text{loss}}(t) = E_{\text{in}}(t) + E_{\text{oxid}}(t) - E_{\text{net}}(t) \]  
\hspace{1cm} (S.2)

\[ for \ 0 < t < t_f \]

The total energy accumulated from the heater at every measurement time, \( E_{\text{in}}(t + \Delta t) \), was estimated by:

\[ E_{\text{in}}(t + \Delta t) = E_{\text{in}}(t) + \int_{T_{\text{amb}}}^{T_{\text{air}}(t)} \dot{m}_{\text{air}}(t) C_{\text{pa}}(T_{\text{air}}) dT \Delta t \]  
\hspace{1cm} (S.3)

\[ for \ 0 < t \]

where the air mass flow \( (\dot{m}_{\text{air}}) \) was known, the ambient temperature \( (T_{\text{amb}}) \) was taken as the background plenum temperature, and the injected air temperature \( (T_{\text{air}}) \) was taken as the plenum temperatures nearest to the fuel bed (0.04 m and 0.05 m below the LAB and DRUM fuel bed, respectively). The timestep \( (\Delta t) \) was the time between temperature measurements, and a quadratic approximation for the heat capacity of air was used

\[ (C_{\text{pa}}(T_{\text{air}}) = -3 \times 10^{-5} T_{\text{air}}^2 + 0.2261 T_{\text{air}} + 940.35) \ [8, 9] \]

The total energy generated from smouldering, \( E_{\text{oxid}}(t) \), was estimated from the GAC ignition time \( (t_{\text{ig}}) \) to \( t_f \) as:

\[ E_{\text{oxid}}(t + \Delta t) = E_{\text{oxid}}(t) + v_{\text{oxid}}(t)(1 - \phi) \rho_s \frac{m_{\text{GAC}}}{m_s} \Delta H_{\text{oxid}} \Delta t \]  
\hspace{1cm} (S.4)

\[ for \ t_{\text{ig}} \leq t < t_f \]

The mean smoulder velocity \( (v_{\text{oxid}}) \) and GAC bulk density \( ([1 - \phi] \rho_s m_{\text{GAC}}/m_s) \), which assumes the GAC occupied a fraction of the sand pore space, were specific to each experiment. The radial extent covered by TCs was used to calculate the column cross-
sectional area \( (A) \), i.e., the full 0.080 m radius in the LAB analysis and 0.295 m of the 0.300 m radius in the DRUM analysis (Fig. 1). Equation S.4 estimates the cumulative energy released over time based on the extent of propagation. Furthermore, \( v_{oxid} \) was observed to be relatively steady after turning off the heater in each robust experiment when the air flux was fixed (see Fig. S.3(b)). Additional discussion on steady smouldering and the methods used to identify \( t_{ig} \) and \( t_f \) are detailed in Section S.4.

The heat of smouldering \( (\Delta H_{oxid}) \) was estimated as if the GAC was pure carbon, where \( \Delta H_{oxid} \) varied with the fraction of C oxidized to CO \( (f_{rCO}) \) [4-7]:

\[
C + \left( \frac{f_{rCO}}{2} + [1 - f_{rCO}] \right) O_2 \rightarrow f_{rCO}CO + (1 - f_{rCO})CO_2
\]  

\( f_{rCO} \) has a strong impact on \( \Delta H_{oxid} \), i.e., \( \Delta H_{oxid} = 110.5 \) to 393.5 kJ mol\(^{-1}\) for \( f_{rCO} = 1 \) to 0, respectively [6, 10]. A \( f_{rCO} = 0.27 \) was determined from experiments LAB R1 and R2 (Table 1). Like \( v_{oxid} \), \( f_{rCO} \) was relatively steady throughout each experiment (see Figs. S.3(a-b)). This provides a GAC \( \Delta H_{oxid} = 24.9 \) MJ kg\(^{-1}\), adjusted to account for the inert ash and water vaporization in GAC (see additional commentary below in Section S.6). \( \Delta H_{oxid} \) was assumed constant between all robust experiments.

To confirm Eq. (S.5) using \( f_{rCO} = 0.27 \) was accurate, it was used in mass balance calculations to predict the concentrations of CO, CO\(_2\), and O\(_2\) in the emissions from robust experiments and compared to measurements. For experiments LAB R1 and R2, in which mass loss data was not available, the rate of C oxidation was approximated from each experiment’s \( v_{oxid} \). For experiments DRUM R0, R1, and R2, the rate of C oxidation was additionally approximated from each experiment’s mass loss rate profile. In all cases, the measured O\(_2\), CO, and CO\(_2\) concentrations were accurately calculated. Details are
included in Section S.5 (with the key results in Fig. S.5). These results confirm that the stoichiometries in all robust smouldering experiments were very similar and it is appropriate to assume the entire combustible fraction of GAC oxidized from C to CO and CO_2 using a constant \( f_{rc_o} = 0.27 \).

As a wood chip (WC) layer was used to ignite DRUM R1 and R2, an additional energy generation term \( (E_{oxid\,WC,e\,ff}) \) was used to estimate the energy released from smouldering the WC layer, which was similar to Eq. (S.4) but used the WC properties. The details on estimating and implementing \( E_{oxid\,WC,e\,ff} \) are included below in Section S.6.

The net stored energy \( (E_{net}) \) was estimated by integrating the change in thermal energy in the sand measured by the TCs. The energy stored in the air-filled porosity was neglected and the measured temperatures were assumed to represent the sand temperatures since \((1 - \phi)\rho_s C_p_s \gg \phi C_p_g \rho_g\). This was demonstrated in a modelling study focusing on the role of local thermal non-equilibrium with smouldering bitumen mixed with sand [11]. \( E_{net} \) was estimated from temperatures measured throughout the system’s volume at every measurement time with the embedded TCs:

\[
E_{net}(t) = \iiint_V \int_{T_{amb}}^{T_s(l,r,t)} (1 - \phi)\rho_s C_p_s(T_s) dT dV
\]

for \( 0 < t \)

The temperature was assumed to vary linearly between the measurement points. The linear expression for sand heat capacity, \((C_p_s(T) = 1.75T_s + 340.32)\) and sand bulk density \(([1 - \phi]\rho_s = 1670 \text{ kg m}^{-3})\) from [9] was used, as the same coarse grain sand was
used in all experiments here. The integration in Eq. (S.6) was completed in two steps at every temperature measurement time following the methodology from Rashwan et al., [1], which built upon the $E_{net}$ calculation method in [9]. Further details on how this was implemented are included below in Section S.7.

The system energy efficiency was defined to normalize the impact of heat losses on the LAB and DRUM systems (or in other words, a process energy efficiency). The system energy efficiency as a function of time was calculated by:

$$System\ Energy\ Efficiency(t) = \frac{E_{net}(t)}{[E_{in}(t) + E_{oxid}(t)]}$$  \hspace{1cm} (S.7)

As the bottom and top sections of the columns were the least instrumented, the system energy efficiencies were only estimated when they could be calculated reliably. The valid times were heuristically determined as when the front propagated from 0.30 to 0.50 m and 0.30 to 0.70 m in the LAB and DRUM experiments, respectively. Examination of gas exit temperatures during these periods confirmed it was accurate to neglect $E_{out}$ in the analyses (see Section S.10 and Fig. S.11).

**S.2. Supplementary Information on the Experimental Procedure and Assumptions**

Figure S.1 shows the unburned hydrocarbon profiles from the flame ionization detector from all DRUM experiments (see the main manuscript for a description of the LAB and DRUM experiments). This figure shows negligible amounts of unburned hydrocarbons produced when smouldering granular activated carbon (GAC), as the two significant peaks are due to smouldering wood chips (WC) for ignition in DRUM R1, R2, and W1 (see Table 1). As a comparison, DRUM smouldering experiments with similar mass fractions of wastewater sewage sludge (i.e., a fuel that exhibits more complicated
degradation chemistry) typically releases 2000 – 20 000 ppm of unburned hydrocarbons throughout a similar smouldering experiment [12]. The first peak in Fig. S.1 near ignition shows unburned hydrocarbons released when smouldering the WC layer. The second peak near the end of propagation shows residual unburned hydrocarbons from WC ignition, which condensed in cooler regions ahead of the smouldering front and then released as the smouldering front reached the end of the column and drove out all condensed hydrocarbons. These condensed hydrocarbons were the tar by-products from pyrolysis reactions when smouldering the WC layer for ignition [13-15]. This release of accumulated condensable by-products at the end of smouldering propagation [13, 16] has been observed in similar experimental smouldering studies, e.g., water from high moisture content feces [17], shale oil [5], oil sludge [18], and canola oil [19]. Altogether, Fig. S.1 substantiates that minimal pyrolysis reactions occurred during GAC smouldering evolving compounds other than CO/CO₂, and nearly all GAC mass loss was due to carbon combustion and a small amount of water vaporization (discussed below in Section S.3).
Fig. S.1. Flame ionization detector results from all DRUM experiments showing the unburned hydrocarbons released throughout propagation (expressed as equivalent ppm of CH₄). The measurement time is expressed as the smouldering front distance travelled using each experiment’s mean propagation velocity \( \text{propagation distance} = \text{ignition distance} + (\text{measurement time} - \text{ignition time}) \times \text{smouldering velocity} \). The first peak labelled ‘Ignition’ is when the smouldering front was near 0 m and the second peak labelled ‘End of Propagation’ is when the smouldering front was near the end of the column, around 0.83 m. The profiles were not corrected for air dilution (shown in Fig. 1).

Figure S.2 presents an example photo of the GAC and sand mixture, which shows that the GAC and sand exhibited similar grain sizes. Also, the effects of mixture heterogeneity were not noticed in any data acquired.
Fig. S.2. The 23.3 g\textsubscript{GAC} kg\textsuperscript{-1} mixture of GAC and sand used in DRUM R2.

Figure S.3 shows the smouldering front characteristics reported in Table 1 measured throughout propagation (note that the mean fraction of carbon oxidized to carbon monoxide is abbreviated as \( f_{r_{\text{CO}}} \)). Though there is data scatter among the key front characteristics, Fig. S.3 shows that they are generally steady throughout propagation, i.e., there no persistent upwards or downwards trends (except for the long-lasting initial effects in DRUM W2).
Fig. S.3. The (a) $f_{rCO}$, (b) propagation velocities, and (c) peak temperatures measured throughout propagation from all self-sustaining and borderline-self-sustaining experiments normalized to their mean values reported in Table 1. The measurement time is expressed as the smouldering front distance travelled using the experiment’s mean propagation velocity ($propagation\, distance = ignition\, distance + (measurement\, time - ignition\, time) \times smouldering\, velocity$). The legend in (c) is common to all frames, where DRUM R0, R1, and R2 are not included in (a) because the resulting CO concentration was beyond the DRUM-CEMS range (see Table 1). The diamond on the DRUM W2 curve in (a) highlights the beginning of the $f_{rCO}$ averaging due to long-lasting initial effects.
S.3. Method to Make Fig. 2 Mass Loss and Time Non-Dimensional

The raw mass loss profiles in Fig. 2 were made non-dimensional by defining the end of the ignition procedure when the air flux was fixed as Non-Dimensional Time = 0 and the end of propagation ($t_f$) as Non-Dimensional Time = 1 for the self-sustaining and borderline-self-sustaining experiments. For these experiments, $t_f$ indicated when the smouldering front reached the top of the fuel bed, which was estimated from each experiment’s $v_{oxid}$ and remaining fuel bed length after the ignition procedure (i.e., the same non-dimensional time definition as [20]). For the non-self-sustaining experiments, $t_f$ was defined as the time the heat wave from ignition reached the last thermocouple (TC) in the fuel bed (i.e., the time the last TC reached 50% of its maximum temperature). The mass loss was made non-dimensional by normalizing to the smoulderable mass remaining in the column after the ignition procedure (i.e., combustible GAC and moisture content). Though Fig. 2 does not include the results LAB R1 and R2 due to measurement challenges (see Table 1), their behaviour is expected to align with the mass loss observed in DRUM R0, R1, and R2. Like DRUM R0, R1, and R2, no GAC was observed upon excavating LAB R1 and R2. Examples of robust mass loss from experiments in a LAB sized column are presented in [12, 21, 22]. Figure 2 highlights characteristic mass loss behaviour. A completely non-reactive experiment would show no mass lost, and therefore be a flat line at Y=0 (i.e., similar to LAB W1 and W2). Robust self-sustained smouldering that is steady and without initial and end effects would be a straight linear line from [X, Y] [0, 0] to [1, -1] (i.e., similar to DRUM R0, R1, and R2).
S.4. Discussion on Robust and Weak Smouldering, Preferred Operating Conditions, Steady Smouldering, and Estimating Ignition

In applied smouldering, robust conditions are preferred so that the system is highly controllable and with minimal risk of quenching. Quenching requires a complex reignition procedure that demands a significant energy input. However, for some applications (e.g., using sewage sludge [22, 23] or human faeces [15-17, 24, 25]) weaker smouldering is unavoidable due to the high water content in the fuel. Nevertheless, weak smouldering still destroys these problematic fuels in a self-sustaining manner when the system is managed properly, so it is valuable to understand both weak and robust smouldering.

The optimal temperature and conditions depend on the application. Some applications focused on resource recovery (e.g., [13]) may benefit from lower temperature conditions to acquire higher quality nutrients remaining in the smouldered ash. Other applications may benefit from high temperature smouldering to ensure that harmful recalcitrant compounds (like perfluorinated compounds) are completely destroyed (e.g., [26]).

As forward smouldering supports numerous reaction, phase change, and heat transfer zones ahead of oxidation, the fuel’s physical and chemical conditions can change ahead of the smouldering front throughout propagation [3, 20, 27-29]. Forward smouldering can be unsteady because these zones, especially the pyrolysis and oxidation zones (subdivided within the reaction zone, discussed in the Introduction), can progress at different velocities [27, 30-32]. Unsteady behaviour is often observed as an accelerating smouldering front within a combustible matrix, where all incoming oxygen is depleted and smouldering propagation is therefore oxygen-limited, e.g., polyurethane foam and organic waste [33, 34]. In other cases, e.g., in applied smouldering systems using a
comparatively small amount of fuel mixed within an inert matrix where all incoming oxygen is not depleted and propagation is instead fuel-limited, the differences in these zones' velocities can be negligible and they may travel together. The smoulder velocity ($v_{oxid}$) may therefore be relatively steady away from initial and end effects (e.g., [17, 35]). There is also some evidence that shows, like the reaction zone, the cooling zone may also achieve a steady condition at sufficiently late-times [1, 12, 36]. Here, the experiments did not show significant evidence of pyrolysis reactions when smouldering the GAC (Fig. S.1) so the smouldering front is assumed to have propagated steadily with only an oxidation zone (similar to [6]). Though pyrolysis reactions (evolving only CO or CO$_2$) could have participated in GAC degradation and therefore be lumped into the reaction zone; the chemistry is not completely resolved here. Nevertheless, the proceeding analysis assumed the smouldering front propagated steadily after turning off the heater at a fixed air flux in all robust experiments. In the DRUM experiments, the $v_{oxid}$ during ignition preheating was higher than the steady $v_{oxid}$ because a higher air flux was necessary to reach the ignition temperature, which was sustained until the front travelled approximately 0.1 m. This early $v_{oxid}$ during preheating was assumed to scale linearly with air flux, which the literature suggests is a reasonable assumption for forced forward smouldering over the applied air fluxes (1.0 to 20 cm s$^{-1}$) [3, 15, 22, 24]. The GAC ignition time ($t_{ig}$) was identified when the first TC in the GAC fuel bed experienced a sharp inflection in heating rate near 400°C, a typical ignition temperature for activated carbon [37].
S.5. Estimating the Mass Balance from Smouldering GAC

To confirm Eq. (S.5) is appropriate here, a C, CO, CO₂, and O₂ mass balance was performed on LAB R1 and R2:

\[ \dot{M}_{out}'' = \dot{M}_{in}'' - \dot{M}_{O_2}'' + \dot{M}_{CO}'' + \dot{M}_{CO_2}'' \]  
(S.8)

where \( \dot{M}_{in}'' \) and \( \dot{M}_{out}'' \) are the molar fluxes of air in \( \left( \dot{M}_{in}'' = \dot{m}_{air}/A M_{air} \right) \), and emissions out of the columns \( \left( \dot{M}_{out}'' = \dot{M}_{in}'' + \dot{M}_{C}''[f r_{CO}/2] \right) \), respectively. \( \dot{M}_{O_2}'' \), \( \dot{M}_{C}'' \), \( \dot{M}_{CO}'' \), and \( \dot{M}_{CO_2}'' \) are the molar fluxes of O₂ consumed, C consumed, CO produced, and CO₂ produced from smouldering, respectively. These molar fluxes were calculated from Eq. (S.5) and the applied air mass flow \( (\dot{m}_{air}) \) over the column cross-sectional area \( (A) \) with the molar mass of air \( (M_{air}) \), and expressed as the exiting molar fractions \( (X_f) \) as:

\[
X_{O_2,f} = \frac{1}{\dot{M}_{out}''} \left( \dot{M}_{in}'' X_{O_2,i} - \dot{M}_{C}'' \left[1 - \frac{f r_{CO}}{2}\right]\right)
\]
\[
X_{CO,f} = \frac{1}{\dot{M}_{out}''} \left( \dot{M}_{in}'' X_{CO,i} + \dot{M}_{C}'' f r_{CO}\right) \]  
(S.9)
\[
X_{CO_2,f} = \frac{1}{\dot{M}_{out}''} \left( \dot{M}_{in}'' X_{CO_2,i} + \dot{M}_{C}'' \left[1 - f r_{CO}\right]\right)
\]

where the entering molar fractions \( (X_i) \) were taken as 0.21, 0.0, and 0.00040 for O₂, CO, and CO₂, respectively. The \( \dot{M}_{C}'' \) was estimated as:

\[
\dot{M}_{C}'' = V_{oxid}(1 - \phi) \rho_s \frac{m_{GAC}}{m_s} \frac{m_C}{m_{GAC} M_C} \]  
(S.10)
where the mass fraction of carbon in the GAC \( \frac{m_C}{m_{GAC}} \) was estimated from subtracting the moisture and ash contents measured from the proximate analysis (see Section 2.1) and expressed in moles with the molar mass of carbon \( M_C \).

For DRUM R0, R1, and R2, as CO and O\(_2\) were not measured (see Table 1), a mass balance was performed only on the CO\(_2\) using Eq. (S.10) and used each experiment’s mass loss rate \( \frac{dm}{dt} \) profile to better capture subtle variations in reaction intensity throughout time. Because of noise in the mass loss measurements (seen in Fig. 2), the mass loss profiles were smoothed with a locally weighted linear regression method using the ‘lowess’ function from the MATLAB curve fitting toolbox [38] over a span of 26.7 min (i.e., 801 data points), which was identified heuristically. Every tenth smoothed mass loss data point was used to determine the \( \frac{dm}{dt} \) profile in Fig. S.4(b).

![Fig. S.4](image)

Fig. S.4. (a) raw and smoothed mass loss data and (b) mass loss rate profile from DRUM R2. Every 10\(^{th}\) smoothed mass loss data point was used to determine the mass loss rate profile, where the profile from taking all smoothed data is also presented in (b) for comparison.
Figure S.4 shows some interference before ignition due to equipment vibrations, which was common among all DRUM experiments (where ignition was identified from the temperature data near 30 min in DRUM R2 and noted on Fig S.4). However, this interference did not affect the subsequent mass loss measurements after the air flux was fixed, seen in Fig. S.4(a) as the mass loss inflection at 51 min until the end of propagation near 216 min (both times are noted on Fig. S.4).

The CO$_2$ concentration was calculated in DRUM R0, R1, and R2 following:

$$\dot{M}_c''(t) = \frac{m_c}{m_c + m_{H_2O}} \frac{1}{AM_c} \frac{dm}{dt}(t)$$  \hspace{1cm} (S.11)

$$X_{CO_2,down}(t) = \frac{DF(t)}{DF(t) + 1} X_{CO_2,f}(t) + \frac{1}{DF(t) + 1} X_{CO_2,i}$$  \hspace{1cm} (S.12)

$dm/dt$ was corrected for the GAC moisture content ($m_c/[m_c + m_{H_2O}]$) and a dilution factor ($DF(t) = \dot{V}_{out}(t)/[\dot{V}_{down} - \dot{V}_{out}(t)]$) was used to account for downstream dilution in the CEMS-DRUM measurements ($X_{down}(t)$) (see Fig. 1), which was around 0.26 for DRUM R0 and R2 to 0.45 for DRUM R1. In all DRUM experiments, the flowrate downstream of dilution ($\dot{V}_{down}$) was kept constant at 0.07 m$^3$ s$^{-1}$ (at 21.1°C and 1 atm), and the flowrate upstream of dilution was approximated from the ideal gas law (at 21.1°C and 1 atm) ($\dot{V}_{out}(t) = [\dot{M}_{out}''(t)/\dot{M}_{in}''] \times [\dot{m}_{air}/\rho_{air}]$).

Figure S.5 shows the calculated emissions concentrations from Eqs. (S.5, S.8-S.12) applied to all robust experiments and compared to their respective measured emissions profiles.
Fig. S.5. (a) LAB R1, (b) LAB R2, (c) DRUM R0, (d) DRUM R1, and (e) DRUM R2 emissions profiles and calculations assuming the combustible fraction of GAC is pure carbon, where the degradation follows Eq. (S.5), and the destruction rate is approximated from the respective mean smoulder velocity in all experiments and mass loss rate profiles in DRUM R0, R1, and R2. The LAB calculations are presented from the approximate ignition time until the end of propagation and the DRUM calculations are presented when the airflow was fixed to the desired air flux, as noted on (c-e), until the end of propagation.
The mass balance results in Figs. S.5(a-b) confirms that Eq. (S.5) is a reasonable simplification to approximate the GAC degradation. That is, most of the combustible fraction of GAC oxidized from carbon to carbon dioxide and carbon monoxide. In addition, the mass balance results in Figs. S.5(c-e) shows the CO₂ concentration profile is also closely calculated in DRUM R0, R1, and R2 by assuming the average $f_{rCO} = 0.27$. Moreover, the calculations in Figs. S.5(c-e) improve when using the respective $dm/dt$ profiles compared to $v_{oxid}$ values, as the subtle changes in reaction intensity are better captured with the $dm/dt$ profiles.

**S.6. Estimating the Energy Generation from Smouldering Wood Chips ($E_{oxid_{WC,eff}}$) and the GAC Heat of Smouldering ($ΔH_{oxid}$)**

The WC were from construction waste wood (BRQ Fibre et Broyure Inc., Trois Rivieres, QC) grinded down and partially air dried, also used in [39] and similar to the demolition waste reported in [40]. As the WC originated from waste wood, its ash content was relatively high and its properties were quite variable (from six repeats: median ash content = 16%, median absolute deviation (MAD) = 3%; median MC = 11%, MAD = 0.4%). The WC bulk density ($[1 − ϕ_{WC}]ρ_{WC}$) was measured as 160 and 200 kg m⁻³ in DRUM R1 and R2, respectively, which is typical for partially dried WC [41, 42] and variable as the WC were derived from construction waste. The WC ignition time ($t_{ig_{WC}}$) was approximated from the centre 0.02 m TC temperature inflection, which occurred around the ignition temperature ($T_{ig_{WC}}$), 200-250°C, a reasonable temperature range for smoulder ignition in biomass [43, 44]. The smoulder propagation velocity through the WC ($v_{oxid_{WC}}$) was approximated by dividing the WC bed length by the difference in the WC and GAC ignition
times. It was assumed that the WC and GAC smouldered in series, so the end of WC smouldering corresponded with GAC ignition, i.e., $t_{f_{WC}} = t_{i_{GAC}}$. The effective WC heat of smouldering ($\Delta H_{oxid_{WC,eff}}$) was estimated from a local energy balance that assumed all smouldering energy was released into nearby sand:

$$\Delta H_{oxid_{WC,eff}}(1 - \phi_{WC})\rho_{WC} = \int_{T_{i_{GAC}}}^{T_{peak_{WC}}} (1 - \phi)\rho_{s}C_{p_s}(T_s)dT$$

(S.13)

The WC bulk density and temperature data from the centre TC 0.020 m above the plenum in DRUM R1 were used in Eq. (S.13), where the $T_{i_{GAC}}$ was observed as the temperature inflection at 236°C and the WC peak temperature ($T_{peak_{WC}}$) was 1028°C. The linear expression for sand heat capacity ($C_{p_s}(T) = 1.75T + 340.32$) and sand bulk density ($[1 - \phi]\rho_{s} = 1670 \text{ kg m}^{-3}$) from [9] were used, as the same coarse grain sand was used in all experiments here. Equation (S.13) resulted in $\Delta H_{oxid_{WC,eff}} = 9.4 \text{ MJ kg}^{-1}$. Though this underestimates typical calorific values for complete combustion of wood (15-20 MJ kg$^{-1}$ [45]), $\Delta H_{oxid_{WC,eff}}$ should be lower here due to its high ash content, losing volatile components from pyrolysis ahead of smouldering, and fostering high CO production [3, 27, 46].

The GAC heat of smouldering ($\Delta H_{oxid}$) was estimated with Eq. (S.5) and assumed a constant $f r_{CO} = 0.27$, i.e., 26.4 MJ kg$^{-1}$. Assuming the entire combustible fraction (94.6%) of GAC was pure carbon and accounting for the heat of vaporization of the 3.2% water content (using 2.26 MJ kg$^{-1}$ as the heat of vaporization), the $\Delta H_{oxid}$ per mass of GAC was estimated as 24.9 MJ kg$^{-1}$. These assumptions are discussed in Section 2.1.
S.7. Method to Integrate Eq. (S.6)

Zanoni et al., [9] estimated the net stored energy ($E_{net}$) profile in a column of sand with a heat pulse by integrating the centreline temperature-distance profile along a single vertical TC transect and assumed a uniform radial temperature distribution. Here, we build on the method in [9] by integrating multiple vertical transects of TCs at different radial positions ($j$) to account for the radial temperature distribution, so to better estimate $E_{net}$. This same methodology was also used by Rashwan et al., [1] to estimate the predicted $E_{net}$ from simulated temperatures in an analytical modelling study. The first step used a trapezoidal numerical integration method along the vertical TC transects:

$$\frac{E_{net}}{A} (r_j, t) = (1 - \phi) \rho_s \int_0^L C_{ps} (T_s) dT dl$$

for $0 < t$  

$$(S.14)$$

The second step summed the resulting net stored energy per unit area values ($E_{net}/A (r_j, t)$) from each vertical transect of TCs as if they represented vertical heights in a series of stacked frustums from the centre TC position ($r_{j=1} = 0$) to the TC position nearest to the wall ($r_{j=j} = R$):

$$E_{net}(t) = \sum_{2}^{j+1} \frac{\pi}{3} \left[ \frac{E_{net}}{A} (r_{j-1}, t) - \frac{E_{net}}{A} (r_j, t) \right] (r_j^2 + r_{j-1}^2 + r_j r_{j-1})$$

for $0 < t$

$$(S.15)$$

Here, a dummy value of zero is used at the radial TC position nearest to the wall ($E_{net}/A (r_{j+1}, t) = 0$) to integrate the $E_{net}$ from all TCs initially at $T_{amb}$. Equation (S.15) assumes the temperature distribution is axisymmetric. These calculations were completed using an in-house MATLAB code.
As minimal information was available near the bottom of both LAB and DRUM columns due to the TC placement (Fig. 1), the plenum temperature was used as the 0 m value for all radial TC transects and each transect was integrated up to their respective highest TC (Fig. 1). Consequently, the $E_{net}$ estimates are least reliable when the smouldering front travelled in the bottom and top of the columns, as these regions were least instrumented. Similar integration techniques were also used in [1, 47].

**S.8. Discussion on Errors in $E_{net}$**

Early and late time errors in estimating the $E_{net}$ profiles were due to poor TC resolution near the top and bottom regions of the columns. In Fig. 4(a), the $E_{net}$ curve shows the early-time error as the $E_{net}$ curve exceeds the $E_{net} + E_{oxid}$ curve from 0 to 52 min (which is not physically possible if all energy terms are accounted for). This overestimation results from linearly interpolating over the TC transects to the plenum (0 m). However, the early-time error attenuates as the smouldering front travels away from the plenum. This same early-time error is also seen in Fig. 4(b) from 0 to 78 min, but the error is less pronounced than in Fig. 4(a) as the base of the DRUM was better instrumented than the LAB (see Fig. 1). At late times, i.e., when the front is above the top TCs used for estimating $E_{net}$, the experimental values underestimate the $E_{net}$ in both columns. Both the early- and late-time errors were considered in heuristically defining the reliable range of $E_{net}$ estimates when the smouldering front propagated between 0.3 to 0.5 m and 0.3 to 0.7 m in the LAB and DRUM, respectively (see Section S.1).

In Fig. 4, oscillations in the $E_{net}$ curves do not reflect a physical phenomenon but result from measurement error due to coarse TC placement. This error was examined via
numerical modelling in Fig. S.6, which illustrates that increasing spatial density of temperature measurements minimizes these oscillations.

Fig. S.6. The $E_{\text{net}}$ profiles from [36] base case simulation of forward smouldering bitumen within sand with various TC spacings. The ‘3rd Order Fit During Smouldering’ curve is an arbitrary function used to visualize the main changes in $E_{\text{net}}$ from ignition until the end of propagation, not for quantification purposes.

Figure S.6 presents various $E_{\text{net}}$ curves from the base case simulation result in [36] by following Eq. (S.6), but assuming uniform radial temperature distribution. Figure S.6 shows the same early- and late-time errors from Fig. 4, which vary depending on the TC spacing. Furthermore, the oscillations in $E_{\text{net}}$ are nearly unnoticeable when the TC spacing is reduced to 0.01 m, roughly the smouldering reaction zone thickness [2, 5, 6, 48]. This observation can provide guidance for future studies on smouldering energy
balances. However, the experimenter should be careful that the high TC density does not significantly alter the bulk thermophysical properties of the system.

S.9. Supplementary Information on the Results

A few additional figures are presented in this section to supplement the discussion in Section 3 (where more additional information is included in [12, 47]). Figure S.7 presents the $E_{in} + E_{oxid}$ and $E_{net}$ profiles from LAB R1, DRUM R0 and DRUM R2. The distributions of system energy efficiency measurements per experiment, approximately 1000 per experiment, are presented as violin plots [49, 50] in Fig. S.8. The distributions of the grouped LAB and DRUM system energy efficiencies are also presented as violin plots in Fig. S.9.

In Fig. S.9, the DRUM system energy efficiencies from 0.3-0.7 m (thereby approximating the efficiency when the front propagated ~0.5 m) appear more reliable than the 0.3-0.5 m efficiencies, as the median absolute deviation is lower at 4% compared to 5%, respectively. That is, the signal noise appears to reduce as more data points are used over a longer duration.
Fig. S.7. The $E_{\text{net}}$ and $E_{\text{in}} + E_{\text{oxid}}$ profiles from (a) LAB R1, (b) DRUM R0, and (c) DRUM R1. Ignition and end of propagation are noted on all figures and the dotted blue lines are fitted curves to illustrate the approximate correct $E_{\text{net}}$ profiles without the false oscillations.
Fig. S.8. System energy efficiencies estimated over the middle of the columns when the smouldering front travelled between 0.3 to 0.5 m in the robust LAB and DRUM experiments.

Fig. S.9. System energy efficiencies grouped over all LAB and DRUM experiments when the front travelled 0.3-0.5 m in the LAB experiments and 0.3-0.5 m and 0.3-0.7 m in the DRUM experiments.
Figure S.10 presents a photo of the top surface of DRUM W1 (a borderline-self-sustaining experiment, Table 1) after the experiment cooled. The pink region in the bottom left of Fig. S.10 shows where smouldering occurred, where the sand was relatively free of GAC. The sand in the smouldered region is characteristically red, possibly because of iron oxidation in the sand [51-53]. The yellow region in the top right shows where minimal smouldering occurred, as unburned GAC remained within the yellow sand. This extinction region developed near the heater and grew up to a maximum area at the top of the column (seen in Fig. S.10).

Fig. S.10. Top of DRUM W1 upon excavation.
S.10. Discussion on $E_{out}$

The rate of energy lost from hot emissions exiting the column (i.e., $\dot{E}_{out}$ in Eq. (1)) is important in resolving the full global energy balance, but practically negligible throughout most of propagation in applied smouldering systems [2, 36]. This negligible impact is because hot emissions are not emitted until the smouldering front is near the end of an applied smouldering system, as most of the heat released from smouldering is transferred directly to nearby fuel and inert porous media [3, 12]. Therefore, the time integral of $\dot{E}_{out}$ (i.e., $E_{out}(t) = \int_0^t \dot{E}_{out}(t) dt$) was not included in the system energy efficiency formulation in Eq. (S.7) because it is essentially an end effect.

Figure S.11 plots the exiting air temperatures from all robust experiments (Table 1) measured just above the packed beds 0.58 m and 0.91 m above the plenums in the LAB and DRUM columns, respectively. The time on the x-axis in Fig. S.11 was normalized to the time in each experiment when the front reached 0.5 m up the column, i.e., the end of the system energy efficiency analysis in Section 3.1. This normalization was performed to highlight the cool, ambient temperatures ($\sim$20°C) in the exiting air throughout smouldering propagation. In addition, the $E_{out}$ profiles were estimated from LAB R1 and R2 (which showed slightly elevated exiting air temperatures when the front approached 0.5 m because of their shorter fuel beds, see Fig. 1). These $E_{out}$ profiles were normalized to the cumulative energy released from smouldering and added for ignition from each experiment at the corresponding times (i.e., $E_{in} + E_{oxid}$, where these terms are detailed in Section S.1). $E_{out}$ was estimated following:
\[ E_{\text{out}}(t + \Delta t) = E_{\text{out}}(t) + \int_{T_{\text{amb}}}^{T_{\text{air}}(t)} \left[ \dot{m}_{\text{air}}(t) + v_{\text{oxid}}(t)(1 - \phi)\rho_s \frac{m_{\text{GAC}}}{m_s} \left(1 - \frac{m_{\text{ash}}}{m_{\text{GAC}}} \right) A \right] C_{p_{\text{air}}}(T_{\text{air}}) dT \Delta t \]  

for \( 0 < t \)

where \( \dot{m}_{\text{air}} \) is the mass flow rate of air injected into the column, \( v_{\text{oxid}} \) is the smouldering velocity, \( \rho_s \) is the sand density, \( A \) is the cross-sectional area, \( C_{p_{\text{air}}} \) is the heat capacity of the air (this assumed heat capacity of the emissions out of the column was similar to that of air, which is reasonable as \( \text{N}_2 \) comprised the majority of both incoming air and exiting emissions), \( m_{\text{GAC}}/m_s \) is the mass fraction of GAC to sand, and \( (1 - m_{\text{ash}})/m_{\text{GAC}} \) is the smoulderable fraction of the GAC (i.e., all components except the ash). Most terms in Eq. (S.16) are further discussed in Section S.1. Equation (S.16) is very similar to the \( E_{\text{in}} \) estimation from Eq. (S.3), except the extra mass leaving the column due to smouldering was accounted for in the exiting mass flow (which was less than 5% of the air mass injected into the system to drive smouldering in LAB R1 and R2).

The \( E_{\text{out}}/[E_{\text{in}} + E_{\text{oxid}}] \) profiles highlight the minimal influence of \( E_{\text{out}} \) throughout smouldering propagation. That is, in LAB R1 and R2 (the experiments with the hottest exiting air temperatures), \( E_{\text{out}} \) was less than 0.5% of \( E_{\text{in}} + E_{\text{oxid}} \) at all times throughout the system energy efficiency analysis in Section 3.1. At later times when the columns cooled after smouldering completion, the influence of \( E_{\text{out}} \) would increase; however, these later times were beyond the scope of this analysis.
The exiting air temperatures from all robust experiments (Table 1) and the $E_{out}$ profiles estimated from LAB R1 and R2 normalized to each experiment’s corresponding $E_{in} + E_{oxid}$ profiles. The time is normalized to when each experiment’s smouldering front reached 0.5 m up the column (i.e., Non-Dimensional Time = $t/t_{0.5m}$). The $E_{out}/[E_{in} + E_{oxid}]$ results at early-times (Non-Dimensional Time < 0.05) were not plotted because they showed erratic results due to the small $E_{out}$ and $E_{in} + E_{oxid}$ values.

S.11. References