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THE IMPROVED ENERGY EFFICIENCY OF APPLIED SMOULDERING SYSTEMS WITH INCREASING SCALE

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One file containing supplementary material is available.
Abstract: Smouldering combustion has been demonstrated to be a highly energy efficient approach towards waste-to-energy. The benefits of smouldering are principally due to the matching of the energy generation and transfer time scales as well as its low quenching temperature (< 400°C). This enables effective energy extraction of problematic wastes (e.g., because of low-volatility or high-moisture content). As the engineering applications of smouldering combustion expand, there is a growing interest in designing systems to best house a propagating smouldering reaction. Through a series of experiments, this work quantified the role of radial heat losses in the energy efficiency of different sized reactors. Thermocouples were placed throughout radial and axial coordinates and integrated to estimate the net stored energy throughout the column volume with time. The impact of heat losses was normalized as the system energy efficiency, by dividing the net stored energy by the energy added into the column for ignition and released from smouldering. The results revealed that the system energy efficiency increased from 65 ± 3% to 86 ± 5% with column radius increasing from 0.080 m to 0.300 m, respectively. As a result, scenarios that were not self-sustaining in the thin column were demonstrated to be self-sustaining in the wider column. Thus, increased system energy efficiency increased the robustness of the reaction to quenching. Altogether, this work underscores the importance of scale as a crucial design parameter enabling a smouldering system to be used as an effective waste-to-energy approach.

Keywords: Smoldering combustion; Energy balance; Heat losses; Waste-to-energy; Porous media; Process scale-up.
Nomenclature

Abbreviations
BSS    Borderline-self-sustaining
CEMS   Continuous emissions monitoring system
FID    Flame ionization detector
GAC    Granular activated carbon
DF     Dilution factor
DRUM   Oil-drum sized column
LAB    Laboratory column
MAD    Median absolute deviation
NSS    Non-self-sustaining
MC     Wet mass basis moisture content
R      Robust experimental conditions
SS     Self-sustaining
TC     Thermocouple
W      Weak experimental conditions
WC     Wood chips

Latin Letters
A      Cross sectional area, m²
\( C_p \)  Specific heat capacity, J kg⁻¹ K⁻¹
\( \frac{dm}{dt} \)  Mass loss rate, kg s⁻¹
E      Energy, J
\( \dot{E} \)  Energy rate, J s⁻¹
\( f_{\text{CO}} \)  Fraction of carbon oxidized to carbon monoxide
L      System length, m
M      Molar mass, kg mol⁻¹
\( \dot{M}'' \)  Molar flux, mol m² s⁻¹
\( m/m \)  Mass fraction
\( \dot{m} \)  Mass flow rate, kg s⁻¹
R      System radius, m
\( t_{\text{ig}} \)  Ignition time, s
\( t_f \)   Final time, s
\( T_{\text{peak}} \)  Maximum temperature, K
\( T_{\text{amb}} \)  Initial ambient temperature, K
\( v_{\text{oxid}} \)  Smouldering front velocity, m s⁻¹
\( \dot{V} \)  Volumetric flow rate, m³ s⁻¹
X      Molar fraction

Greek Symbols
\( \Delta H \)  Heat of reaction, MJ kg⁻¹
\( \Delta t \)  Time between measurements, s
\( \phi \)    Porosity
\( \rho \) Density, kg m\(^{-3}\)

\( \tau \) Characteristic time, s

Subscripts

- \( air \): Gas phase / air / emissions
- \( amb \): Ambient
- \( down \): Downstream
- \( eff \): Effective
- \( f \): Final
- \( i \): Initial/entering
- \( in \): Into control volume
- \( ig \): Ignition
- \( j \): Radial position from centre
- \( J \): Radial position nearest to the wall
- \( loss \): Lost from control volume
- \( net \): Net stored
- \( oxid \): Oxidation
- \( out \): Out of control volume
- \( pyr \): Pyrolysis
- \( s \): Solid phase / sand
1. Introduction

1.1. Applied Smouldering Combustion

Applications of smouldering combustion are solving a wide range of engineering challenges. Applied smouldering systems represent a simple, economical, and robust thermal conversion option in many contexts, including soil remediation [1-3], biomass energy conversion [4], wastewater sludge treatment [5], resource recovery [6-8], and sanitation in the developing world [9, 10]. Thus, smouldering is emerging as a viable waste-to-energy option. Most of these applications require the design and construction of engineered smouldering systems (e.g., batch or continuous reactors). The scale of the reactor is intimately related to the waste-to-energy process, with some systems favouring small reactors [6, 10] while other larger systems [3, 11]. Therefore, scale is a key variable defining the potential of smouldering as a waste-to-energy treatment. Meanwhile, most smouldering research – central to elucidating the underlying principles and parameter sensitivities of the technology – exists only at the laboratory (bench) scale. As a result, there is limited understanding of the effects of scale on smouldering behaviour, and this knowledge gap impairs the link between laboratory research and technology applications, as well as limits the predictive capacity of current numerical models (e.g., [12, 13]).

The operating principle of smouldering is heterogeneous, flameless combustion resulting from gaseous oxygen reacting with a condensed phase fuel comprising, or embedded within, a porous medium [14]. In most contexts, smouldering is limited by the transport of oxygen to the surface of the reacting fuel [14, 15]. A common example is glowing red charcoal in a traditional barbeque. As is recognized in barbeque design, the rate and
direction of air flow through the porous bed has a strong impact on the reaction. Moreover, as seen in barbeques, smouldering will propagate as a reaction wave through the bed until the fuel is exhausted or the air flow is eliminated. There is a wide range of fuels that smoulder, from natural materials such as peat, coal, and forest litter to anthropogenic materials like polyurethane foam, oil-soaked insulation, and coal tar-contaminated soil [14-16]. In uncontrolled smouldering scenarios, such as underground coal seam fires, smouldering proceeds in a runaway manner since the air flow cannot be eliminated, fuel load is in excess, and the system geometry is unbounded [17, 18]. In applied engineering scenarios, air flow is strictly managed, and fuel load and system geometry are carefully controlled to effectively destroy wastes and capture the released energy [6, 9, 15].

In most incinerators (e.g., fluidized bed), energy is delivered very quickly by the combustion reaction and thereby drives all other processes. Therefore, heat transfer processes can be simplified, but efficient use of the energy released is difficult. In contrast, a smouldering system is comprised of multiple zones, each characterized by different heat and mass transfer processes and chemical reactions, where no single process is dominant over the others. In the reaction zone, including the smouldering front itself, competing pyrolysis and oxidation reactions govern the fuel conversion into primarily CO$_2$, CO, H$_2$O, and heat [14]. In the inert heating zone, ahead of the smouldering front, the untreated fuel will undergo heat absorption (known as “preheating”) and drying. The cooling zone behind the smouldering front is also reaction-free, where the hot porous material experiences heat dissipation. This set of coupled, interdependent
zones propagate and vary in thickness and intensity throughout space and time. Consequently, heat transfer processes are complex, but efficiency is potentially very high. Thus, smouldering is a dynamic thermal system, the fundamentals of which continue to be studied primarily in laboratory and theoretical research.

Smouldering, because of its potential for being an energy efficient form of combustion, allows for utilizing a wide array of combustible materials for effective energy recovery [6, 9, 15]. One reason for this is that the rates of fuel conversion and heat transfer are all driven by the porous medium and therefore have compatible characteristic time scales [15]. A second reason is that the porous medium acts as a heat storage reservoir. The fuels in engineered smouldering systems are typically embedded in an inert porous medium, e.g., hydrocarbons in soil [19, 20], sludges or digestates in sand [5, 21, 22], and human faeces in zirconium oxide beads [10], which can store and recycle energy like flaming porous burners [23]. Finally, quenching temperatures for smouldering reactions are generally lower than 400°C [6, 21, 24], which enables the reduction of heat losses and propagation of the reaction with much lower effective energy generation [15, 25] (see Supplementary Materials, Section S.4 for additional discussion on smouldering operating conditions).

If the airflow feeding the reaction is driven in the same direction as the smouldering front propagation, forward heat transfer from cooling the burnt region and the reaction zones is deposited in the inert heating zone [15]. This efficient convective recycling of heat allows the treatment of wastes that are otherwise problematic for incinerators, such as those with low-volatility (e.g., tank bottom oil sludge) or high moisture content (e.g.,
wastewater sludge) [6, 9, 15]. Currently, nearly every incineration technology uses flaming-based reactors [26], but for all problematic wastes it is necessary to either pre-process the waste (e.g., moisture removal, chemical treatment) or add supplemental fuel. In these cases, it has been shown that these wastes are better handled in smouldering-based reactors [3, 4, 6, 8, 9, 11, 20, 22, 27, 28].

As a result, smouldering is self-sustaining under a wide range of conditions. Self-sustaining means that, after an initial small and local input of energy, the process will continue without external energy input indefinitely as long as sufficient fuel and air are present [14]. This is commonly observed after igniting charcoal in a barbeque. The underlying causes are positive energy balances both locally (at the reaction front) and globally (across the fuel bed), where the rate of heat generated reaches thermal equilibrium with the heat lost (e.g., to endothermic processes and to the external environment) at temperatures above quenching [14, 29]. The self-sustaining nature of smouldering is what makes natural smouldering problems so intractable, such as peat forest and underground coal fires [17, 18], and what makes applied smouldering waste-to-energy systems so green and sustainable [6, 9, 15, 16].

Due to these benefits, applied smouldering has been recently upscaled into a commercial technology. A pilot smouldering reactor demonstrated the ex situ treatment of coal-tar contaminated soil [30], which was similar in size as another smouldering reactor studied for deriving liquid fuel from waste tires [7]. Following [30], a modular, scalable, batch treatment system was developed for contaminated soils and organic sludges [3, 11, 31]. This batch smouldering system has been applied in numerous cases, including in
Southeast Asia for treating crude oil lagoon sludge [3, 11] and in China for oil sludge mixed with contaminated soil [16]. In addition, another pilot smouldering reactor was developed that represented a 300-fold scale-up of a typical laboratory column [16]. In all these cases, it has been assumed that large scale smouldering systems behave similarly to laboratory systems, although almost no research exists on this topic.

Significant research – all at the laboratory scale – has explored how to maximize the envelope of self-sustaining behaviour in applied smouldering, including identifying and extending the quenching limits [12, 20, 28-30, 32], optimizing the fuel/inert ratio [21, 24], examining inert-free systems [4, 22], and considering supplemental, low-volatility fuels for high-volatility wastes [33]. In all these cases, the focus – whether explicitly acknowledged or not – was shifting the energy balance in the smouldering system to more positive values [15]. Though there are many components of a comprehensive system energy balance (e.g., energy for ignition, emissions treatment, compressed air), a key aspect of the energy balance is radial heat losses to the external environment. Given that the reaction front is very thin and the effective thermal conductivity of the porous medium is low in applied smouldering systems [15, 29, 32, 34], radial heat losses have not been considered relevant to the smouldering stability. Therefore, this aspect has received little attention, and the link between system scale losses and smouldering stability has never been directly studied in applied smouldering systems.

Though heat transfer near the reactions closely match the heat generation time scales, heat transfer at the system scale is often much slower than smouldering propagation [15]. Therefore, the cooling zone temperatures are unsteady in most applied smouldering
systems and the link between system scale losses and improved smouldering performance is not simple. Consequently, heat losses as a function of scale have never been quantified in applied smouldering experiments. Furthermore, this problem is not easily rectified using existing data sets, since most smouldering experiments are not adequately instrumented, and few methodologies have attempted to quantify their heat losses or energy efficiency.

1.2. The Energy Balance in Applied Smouldering Systems

Zanoni et al. [12, 29] provides insight into how heat losses affect smouldering propagation by developing a global energy balance around a one-dimensional smouldering system:

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

(1)

where $\dot{E}_{\text{in}}$ is the rate energy is added from the igniter, $\dot{E}_{\text{oxid}}$ is the rate energy is released by exothermic oxidation, $\dot{E}_{\text{pyr}}$ is the rate energy is removed by endothermic pyrolysis, $\dot{E}_{\text{loss}}$ is the rate energy is removed by radial heat losses, $\dot{E}_{\text{out}}$ is the rate energy is lost due to convection at the system boundary, and $dE_{\text{net}}/dt$ is the net rate of energy accumulation. At self-sustaining conditions away from the inlet and outlet boundaries $\dot{E}_{\text{in}} = \dot{E}_{\text{out}} = 0$; moreover, $\dot{E}_{\text{pyr}}$ has been shown to be negligible in many applied smouldering conditions [15, 29]. Therefore, $dE_{\text{net}}/dt$ is primarily governed by $\dot{E}_{\text{oxid}}$ and $\dot{E}_{\text{loss}}$, and the balance between them will dictate whether a smouldering system is self-sustaining ($dE_{\text{net}}/dt \geq 0$), or trending towards extinction ($dE_{\text{net}}/dt < 0$). Whether the smouldering reaction exists or not depends on the local energy balance at the reaction zone. If the
conditions are such that the Damköhler number (i.e., of the second type based on local mass transfer) exceeds the critical value, then smouldering persists; otherwise, extinction will occur [15]. However, integrating Eq. (1) over the system (i.e., reactor bed) can provide measure of the robustness of the smouldering system [29]. Positive $dE_{\text{net}}/dt$ indicates the smouldering system is increasing robustness, as the accumulating stored energy acts as a buffer against extinction [29]. At sufficiently late times with enough energy accumulated, modest changes in heat losses do not significantly affect smouldering characteristics [13, 29, 32, 34]. However, if this buffer against extinction is relatively small, but the Damköhler number remains above the critical value, weak smouldering occurs [29, 35] (see a practical discussion on weak and robust smouldering in the Supplementary Materials, Section S.4). This happens, for example, due to low oxygen flux or low fuel concentration [12, 21, 24, 29, 32, 35]. Under such conditions, slight changes in $E_{\text{loss}}$, can strongly affect weak smouldering characteristics, dropping peak temperatures and propagation velocities [35]. Though weakened smouldering may persist, if $dE_{\text{net}}/dt$ becomes negative for a sufficiently long time the Damköhler number will fall below the critical value and the system will trend to extinction [12, 29]. Altogether, numerical models [13, 29] and experiments [34] suggest that 30-50% of the energy generated from laboratory applied smouldering systems escape as heat losses. However, these values have not been well-quantified, and no measurements investigate how this fraction may decrease with increasing system scale, nor are there clear demonstrations of the expected increase in robustness.
To address the major gap between smouldering research and technology applications, this work quantifies the influence of scale on the system energy efficiency and demonstrates its implications on the smouldering reaction stability. The fraction of energy lost radially to the environment was determined for smouldering experiments using columns at a variety of diameters. The experiments spanned the spectrum from robust smouldering to weak smouldering to extinction conditions. The experiments were instrumented with radial and axial thermocouples and mass loss and emission products were measured. The methodology from Rashwan et al., [36] was applied to the robust experiments to permit global energy balances that quantified energy efficiency. While developed for applied smouldering systems, this methodology could be adapted for other thermal systems using porous media, e.g., for pyrolysis, gasification, or energy storage purposes [37, 38]. This work provides the first measurements of the reduced heat losses, and therefore increased robustness, of applied smouldering systems as scale increases. Altogether, this work underscores the importance of scale as a crucial design parameter in optimizing an applied smouldering system and provides a valuable framework to estimate heat losses and energy efficiency at any scale.
2. Methodology

2.1. Experimental Equipment and Setup

All experiments followed established methods [20, 21, 24, 28] slightly adapted to use a convective ignition method in the laboratory column (LAB) and the oil-drum sized column (DRUM), illustrated in Fig. 1. Like recent studies [39-41], granular activated carbon (GAC, at least 90% particles larger than 80 mesh, DARCO® 12X40, Cabot Corp.) was used as a model fuel for this study because: 1) simple oxidation chemistry, 2) lack of pyrolysis reactions, 3) ease of mixing and experimental preparation, and 4) there is an emerging, practical interest in smouldering spent GAC from adsorption treatment [41]. Proximate analysis indicated the GAC wet basis moisture content (MC) was 3.2% (ASTM-D2867-17), volatile matter content was 3.2% (ASTM-D5832-98), ash content was 2.2% (ASTM-D2866-11), and fixed carbon content was 91.4% (calculated by the difference). The GAC higher heating value was measured as 30.9 MJ kg\(^{-1}\) (IKA C 200 bomb calorimeter), which normalized to the combustible fraction (i.e., the sum of the volatile matter and fixed carbon) is 32.7 MJ kg\(^{-1}\) and compares well to the theoretical value for pure carbon, 32.8 MJ kg\(^{-1}\) [25, 42, 43] (see additional discussion on GAC characterization in the Supplementary Materials, Section S.2).

Table 1 presents the experimental conditions, illustrating that the variables adjusted were column diameter, fuel concentration, and applied air flux. The experimental conditions reflect both common values used in applied smouldering research (e.g., [21, 33, 41, 44, 45]) and application (e.g., [3, 19, 31]). The GAC was mechanically mixed with coarse grain sand (1.180 ≤ mean grain diameter ≤ 2.000 mm, porosity (\(\phi\)) = 0.37, bulk density
\((1 - \phi)\rho_s = 1670 \text{ kg m}^{-3}\), MC between 0.04% and 0.4%, (Number 12, Bell & Mackenzie) to desired fuel concentrations (example photo in the Supplementary Materials, Fig. S.2). The GAC and sand exhibited similar grain sizes and were mixed and packed into the columns with sufficient care that heterogeneity was not apparent in any data acquired. All DRUM experiments had a 0.054 to 0.098 m clean sand cap on top of the fuel bed to lower the exiting air temperature when the smouldering front approached the top of the column (for safety purposes). Experiments DRUM R1, R2, and W2 also included a loosely packed, 0.027 to 0.030 m layer of wood chips (see Supplementary Materials, Section S.6 for details) on top of a 0.010 to 0.040 m bottom layer of clean sand to facilitate ignition at lower temperatures. Smouldering the GAC led to a small drop in the fuel bed height (3% to 5%) in all experiments; the three experiments with an ignition wood chip layer incurred an additional height drop equal to the layer’s thickness.
Fig. 1. Illustration of the LAB and DRUM experimental setups. LAB thermocouples (TCs) are placed 0.080 m, 0.043 m, 0.026 m, 0.012 m, 0.006 m, and 0.000 m deep from the column wall into the column centre. DRUM TCs placed 0.300 m, 0.165 m, 0.073 m, and 0.005 m deep from the column wall into the column centre.
Table 1. Experimental Conditions and Key Smouldering Front Results

<table>
<thead>
<tr>
<th>Experiment</th>
<th>GAC/sand (g\text{GAC} kg^{-1}s)</th>
<th>\text{Darcy} air flux (cm s^{-1})</th>
<th>Initial fuel bed height (m)</th>
<th>Smouldering Front Results</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>\text{LAB} \pm 0.3%</td>
<td>\text{DRUM} \pm 2%</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>\text{DRUM} \pm 4%</td>
<td>\text{DRUM} \pm 1%</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>\text{Drum} \pm 2%</td>
<td>\text{DRUM} \pm 1%</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>\text{Lab} \pm 10%</td>
<td>\text{Lab} \pm 1%</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Scenarios Expected to be Robust</th>
</tr>
</thead>
<tbody>
<tr>
<td>DRUM R0 20.0 5.0 0.813 SS 0.46 804</td>
</tr>
<tr>
<td>DRUM R1 20.0 7.5 0.868 SS 0.47 765</td>
</tr>
<tr>
<td>DRUM R2 23.3 5.0 0.865 SS 0.44 834</td>
</tr>
<tr>
<td>LAB R1 20.0 7.5 0.568 SS 0.61 766</td>
</tr>
<tr>
<td>LAB R2 23.3 5.0 0.560 SS 0.49 874</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Scenarios Expected to be Weak</th>
</tr>
</thead>
<tbody>
<tr>
<td>DRUM W1 10.0 5.0 0.719 BSS 0.19 601 0.26</td>
</tr>
<tr>
<td>DRUM W2 20.0 1.0 0.874 SS 0.16 782 0.29</td>
</tr>
<tr>
<td>LAB W1 10.0 5.0 0.253 NSS - -</td>
</tr>
<tr>
<td>LAB W2 20.0 1.0 0.262 NSS - -</td>
</tr>
<tr>
<td>LAB W3 15.0 5.0 0.277 SS 0.33 661 0.18</td>
</tr>
<tr>
<td>LAB W4 20.0 2.0 0.255 SS 0.24 678 0.13</td>
</tr>
</tbody>
</table>

Experimental Conditions’ errors represent conservative estimates of equipment error. Smouldering Front Results’ errors encompass a conservative estimate of experimental variability as they represent the normalized standard deviations from three DRUM and LAB repeat experiments from smouldering wastewater sewage sludge, a highly variable fuel [21]. These errors align well with similar experimental studies [21, 33, 41, 44, 45].

1 At standard temperature and pressure (21.1°C at 1 atm).
2 The percentages represent vol.% values.
3 Used a thin bottom layer of wood chips for ignition and recycled sand.
4 Mass balance results were spurious because the instrumentation caused small erratic physical vibrations.
5 Used a thin bottom layer of wood chips below a bottom 0.128 m layer of 20.0 g\text{GAC} kg^{-1}s layer for ignition.
6 The \( f_{\text{rCO}} \) averaging began after 194 min from turning off the heater because of persistent initial effects and used a manual rotameter to control low air flow (0-0.010 m^{3}s^{-1}, King Instrument Company).
7 Used a less instrumented experimental setup using conductive ignition described in [21].
8 CO measurement throughout propagation exceeded CEMS-DRUM calibration range, which was 0-0.3%.
The LAB and DRUM columns were similarly constructed with stainless-steel and inner radii of 0.080 m and 0.300 m, respectively, and wrapped in 0.051 m thick insulation for safety purposes (ASTM C518 R-Value = 8.7 at 24°C, MinWool®, Johns Manville [LAB]; ASTM C518 R-Value = 9.6 at 24°C, FyreWrap® Elite® Blanket, Unifrax [DRUM]). Both setups used similar mass flux controllers (FMA5400/5500 Series, Omega Ltd. [LAB]; 8290B045PDB67 ASCO Numatics [DRUM]), inline air heaters for convective ignition (F074719 2 kW SureHeat® JET [LAB], F074736 36 kW SureHeat® MAX [DRUM], Osram Sylvania), and mass balances (KCC150 [LAB], KD1500 [DRUM], Mettler Toledo). Thermocouples (Type K, 0.0032 m diameter Omega Ltd. [LAB], 0.0064 m diameter Kelvin Technologies [DRUM]) were placed in various radial and axial positions (Fig. 1). All emissions sampling locations are noted in Fig. 1. Continuous emissions monitoring systems (CEMS) were used in the LAB (CEMS-LAB for O₂, CO₂, and CO, MGA3000C, ADC) and DRUM experiments (CEMS-DRUM, custom assembly with a Uras for CH₄, CO₂, CO, and a flame ionization detector for unburned hydrocarbons, ABB Ltd.). The CEMS-DRUM and mass balances logged directly to a personal computer with equipment specific software every 5 and 2 seconds, respectively. All other instruments were connected to a data logger (Multifunction Switch/Measure Unit 34980A, Agilent Technologies) and personal computer that logged every 2 and 3 seconds for the LAB and DRUM experiments, respectively.

2.2. Experimental Procedure

Ignition was achieved via injecting hot air (300-400°C in the DRUM and 400-500°C in the LAB, the maximum air temperatures using the respective setups) into the column plenum.
until the thermocouples (TCs) located 0.1 m above the plenum reached peak smouldering temperatures, 700-900°C. The convective heater was then turned off and ambient temperature air was injected at the desired air flux to sustain propagation. Experiments LAB W1, W2, W3, and W4 used a conductive ignition method with a less instrumented setup as detailed in [21]. Though there were differences in the ignition procedure, all experiments showed signs of strong ignition in the temperature histories (i.e., very high peak temperatures near the heater) and upon excavation (i.e., no fuel remaining around the heater). Moreover, the ignition method is not expected to affect the results presented in this work, which focuses on front propagation away from the boundaries [15]. The smouldering front characteristics reported in Table 1 were averaged after turning off the heater and fixing the ambient air flux until the front reached the last centre TC, at least 0.01 m from the end of the fuel bed. The mean centreline propagation velocity, centreline peak temperature, and $f_{rCO}$ were all relatively steady during this time, and the most data scatter was observed in the weak experiments (see Supplementary Materials, Fig. S.3 and additional data in [25, 43]). The propagation velocities were calculated by dividing the axial spacing between thermocouples by the time between front arrivals at successive thermocouples, following the method in [27].

The experiments in Table 1 are separated into two groups based on their experimental conditions: “Scenarios Expected to be Robust” and “Scenarios Expected to be Weak”. Within these groups, the experiments were identified as robust or weak self-sustaining (SS), borderline-self-sustaining (BSS), and non-self-sustaining (NSS) by following the criteria in [21] and by considering mass loss behaviour. The effect of reactor size on the
fate of smouldering is seen in the second group of experiments, “Scenarios Expected to be Weak”. In this group, LAB W1/DRUM W1 and LAB W2/DRUM W2 used the same experimental conditions, respectively, but showed different smouldering front results. That is, LAB W1 and W2 were NSS and DRUM W1 and W2 were BSS and SS, respectively. The measurements in Fig. 2 show the overall normalized mass removed from the reactors over time and provides insight into smouldering robustness. Figure 2 shows the spectrum of mass loss behaviour in these experiments from NSS to weak BSS to robust SS smouldering, where the shallower slopes indicate a weak or weakening reaction, and the end point of each curve near Non-Dimensional Time = 1 indicates the effectiveness of the treatment process (i.e., the fraction of fuel removed from the system by smouldering, where Non-Dimensional Mass Loss = -1 means all the fuel was smouldered).
Fig. 2. Non-dimensional mass loss profiles from all DRUM and weak LAB experiments (where the mass was normalized to the total smoulderable mass in each experiment). The graphs plot every 100th data point for clarity and are bounded by the end of ignition (Non-Dimensional Time = 0) and the end of propagation (Non-Dimensional Time = 1) and Non-Dimensional Mass Loss = -1 means no fuel remaining. The non-self-sustaining (NSS: LAB W1 and W2), borderline-self-sustaining (BSS: DRUM W2), weak self-sustaining (Weak SS: DRUM W2 and LAB W3), and robust self-sustaining (Robust SS: DRUM R0, R1, and R2) experiments are noted. The time was made non-dimensional using the reactor length and each experiment’s propagation velocity, where the methods for making the mass and time non-dimensional are detailed in the Supplementary Materials, Section S.3.

The global energy balances for the robust experiments (i.e., Eq. (1) terms integrated over the whole system and time) were determined from the experimental data following the methodology provided in [25, 36] and summarized in the Supplementary Materials, Section S.1.
3. Results and Discussion

3.1. The Effects of Heat Losses on Robust Smouldering Systems

Figure 3 visualizes the temperatures at various times from experiments LAB R2 and DRUM R2, which were representative of the patterns in all robust experiments.
Fig. 3. (a-c) LAB R2 and (d-f) DRUM R2 colour contour maps show the temperature distribution across the column radius and axis at specific times (plotted using MATLAB’s `contourf` function). The black circles mark the thermocouple (TC) locations and the black dashed lines bound the fuel bed region upon excavation. The red lines show the approximate smouldering front positions identified from the centreline TCs: (a, d) just after ignition, (b, e) part-way up the column, and (c, f) near the end of the column. The plenum air temperatures were assumed uniform over the column radius.
Figure 3 reveals the temperature gradients from the column centre to the wall, driven by radial heat losses. Much of the energy released from smouldering accumulated in the cooling zone behind the smouldering front, which is characteristic of applied smouldering systems [15, 29, 36]. Because nearly all heat losses from the DRUM and LAB experiments were drawn from this cooling zone [29, 36], the temperatures needed to be well resolved in this region to conduct a global energy balance. Figure 3 shows that by spacing TCs across radial and axial locations, the temperature distribution was sufficiently captured to estimate $E_{\text{net}}$ profiles.

Other details are revealed from Fig. 3. For example, the temperatures ahead of the smouldering fronts show the regions near the walls heated before those near the centre (most clearly seen above the red lines in Figs. 3(b) and 3(e)). This aligns with other studies that have shown the smouldering front is not typically flat but often curved across the radius in the direction of propagation because the cooler air temperatures near the column wall (behind the smouldering front) facilitates enhanced axial convective heat transfer [13, 32, 34, 43]. Here, the smouldering front position was drawn flat for simplicity, as the detailed shape could not be resolved with the data resolution. Furthermore, though the TCs capture the temperature profiles behind the smouldering front well, they do not resolve the sharp temperature change in the reaction zones ahead of the smouldering front (just above the red lines in Fig. 3). Others have shown that applied smouldering in similar configurations facilitates a thin reaction zone (~0.01 m, where temperatures vary from the ignition to peak temperatures) [13, 29, 32, 34]. As the vertical spacing was larger here (see Fig. 1), these steep gradients were not well resolved. Because only a thin
region in the system is impacted by these measurement errors, the key trends from the global energy balances are largely unaffected.

Figure 4 presents the $E_{\text{net}}$ profiles for experiments LAB R2 and DRUM R2 (plots for the other robust experiments are included in the Supplementary Materials, Fig. S.7). This figure reveals how the net stored energy grew in both experiments from when the smouldering front ignited until it reached the top of the fuel bed, reaching peaks of approximately 7 and 200 MJ in LAB R2 and DRUM R2, respectively. Beyond these times, $E_{\text{net}}$ decreased reflecting the column cooling due to radial and convective losses. The jaggedness in the $E_{\text{net}}$ curves are an artefact of the TC spacing in the experiments. Numerical modelling of $E_{\text{net}}$ calculations as a function of TC spacing revealed that: (1) the true $E_{\text{net}}$ does increase monotonically as expected, (2) the $E_{\text{net}}$ estimate becomes more smooth as vertical discretization becomes finer, (3) achieving a monotonic result would require experiments with TC spacing < 0.01 m, and (4) the intersections in $E_{\text{net}}$ and $E_{\text{in}} + E_{\text{oxid}}$ are artefacts, with sufficient TC spacing, $E_{\text{in}} + E_{\text{oxid}}$ is always greater than $E_{\text{net}}$ (see Supplementary Materials, Section S.8 and Fig. S.6). An approximation of the true $E_{\text{net}}$ function is illustrated in Fig. 4 for qualitative interpretation.
Fig. 4. The $E_{\text{net}}$ and $E_{\text{in}} + E_{\text{oxid}}$ profiles from (a) LAB R2 and (b) DRUM R2. Ignition and end of propagation are noted on both figures and the dotted blue lines are fitted curves to illustrate the approximate correct $E_{\text{net}}$ profiles without the false oscillations.
The sum of $E_{in} + E_{oxid}$ are also plotted in Fig. 4, with the first term mainly contributing the energy prior to ignition and the latter contributing thereafter. If the columns were perfectly insulated, all of the energy added would remain stored in the hot sand until convective losses began near the end of smouldering [12, 29], and $E_{net}$ would equal $E_{in} + E_{oxid}$. Thus, the differences between $E_{in} + E_{oxid}$ and $E_{net}$ in Fig. 4 reveal $E_{loss}$. Figure 4 demonstrates that radial losses are proportionally less in DRUM R2 than in LAB R2.

Figure 5 focuses on the influence of scale on smouldering performance by plotting the system energy efficiencies, $E_{net}/(E_{in} + E_{oxid})$, for all robust experiments when the smoulder front travelled approximately 0.3 to 0.5 m. It reveals that the median system energy efficiency was 65 ± 3% for the LAB experiments and 86 ± 5% for the DRUM experiments when the front travelled ~0.4 m; the uncertainty values indicate the median absolute deviations (see the Supplementary Materials for the grouped results, Fig. S.9). A two-sample Kolmogorov-Smirnov test showed the DRUM experiments system energy efficiencies are larger than those of the LAB at the 0.01% significance level [49].
Fig. 5. Boxplots of the 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.

The LAB system energy efficiency estimated here compares well with other laboratory estimates of radial heat losses from smouldering columns [12, 13, 34, 36], which provides confidence in this approach. Furthermore, Rashwan et al., [36] has shown that these measurements agree well with theoretical predictions in both the LAB and DRUM columns, by assuming the system energy efficiencies are governed by transient heat transfer in the cooling zone.

### 3.2. The Effects of Heat Losses on Weak Smouldering Systems

As articulated in the Introduction, while moderate heat losses do not significantly affect robust smouldering propagation, they may lead to extinction in weak smouldering
systems [15, 29]. It was therefore hypothesized that reducing the radial heat losses, here by increasing scale, could transform a NSS scenario into a SS scenario. Indeed, this is demonstrated in Fig. 6. Figures 6(a) and 6(b) show LAB W1 and W2, two NSS experiments that quenched due to critically low GAC concentration (10 g\textsubscript{GAC} kg\textsuperscript{-1}s\textsuperscript{-1}) and critically low air flux (1 cm s\textsuperscript{-1}), respectively. Note that the LAB column limits (i.e., LAB W3 using 15 g\textsubscript{GAC} kg\textsuperscript{-1}s\textsuperscript{-1} at 5 cm s\textsuperscript{-1} and LAB W4 using 20 g\textsubscript{GAC} kg\textsuperscript{-1}s\textsuperscript{-1} at 2 cm s\textsuperscript{-1} were SS, see Table 1) are consistent with applied smouldering literature, with minimum air fluxes around 0.5 – 1.4 cm s\textsuperscript{-1} [20, 29, 50] and minimum fuel mass fractions around 16 – 28 g\textsubscript{fuel} kg\textsuperscript{-1}s\textsuperscript{-1} [20, 28, 29, 32, 35]. Figures 6(c) and 6(d) show that DRUM W1 and W2, with the same experimental conditions as LAB W1 and W2, respectively, exhibited SS behaviour. This demonstrates that the reduced radial heat losses improved the global energy balance such that the smouldering behaviour shifted towards more robust conditions. These robust conditions meant that energy was generated faster than it was lost near the reactions, and quenching was avoided.

The ability for increasing scale to shift the extinction limits for smouldering to lower fuel concentrations and lower air flux values is confirmed by the mass loss curves for these four experiments (LAB W1 and W2, DRUM W1 and W2) in Fig. 2. This figure also reveals that, even though these two DRUM cases were SS, the fuel was not entirely oxidized. Though DRUM W1 fostered SS smouldering along the centre of the column, material near the column walls experienced extinction as a wedge of unburned material increased in thickness with height but did not progress into the centre of the column (see a photo of the excavation in the Supplementary Materials, Fig. S.10). Therefore, DRUM W1 is
qualified as borderline-self-sustaining, which was more robust than LAB W1 but would require further intervention to be fully self-sustaining. A rich discussion on various adjustments to improve smouldering robustness is presented in [12, 29]. However, it is also important to point out that the dynamics that lead to quenching in Fig. 6 are currently not well understood. Torero et al., [15] discusses this knowledge gap and points out there is room for a harmonized explanation of extinction in smouldering systems.
Fig. 6. Centreline temperature-time profiles showing: (a) Non-self-sustaining smouldering in the LAB due to critically low fuel concentration, LAB W1; (b) Non-self-sustaining smouldering in the LAB due to critically low air flux, LAB W2; (c) Borderline-Self-sustaining smouldering in the DRUM at low fuel concentration, DRUM W1; and (d) Self-sustaining smouldering in the DRUM at low air flux, DRUM W2, which had the same thermocouple layout as DRUM W1. All thermocouples were embedded in the fuel bed.
4. Summary and Conclusions

As the applications of smouldering combustion expand, especially those related to waste-to-energy processes, there is a growing interest in designing systems to best house a propagating smouldering reaction. To effectively operate a smouldering system, it is necessary to determine the conditions that will result in self-sustained propagation. Radial heat losses have been demonstrated to play a major role on the stability of the smouldering reaction; therefore, understanding of the relationship between stability and reactor dimensions is critical. The results from a novel approach to quantify the decreasing fraction of total energy lost radially from increasing the column radius has been presented. Through a global energy balance, the impact of heat losses was normalized as the system energy efficiency by dividing the net stored energy from the energy added into the column for ignition and released from smouldering. This approach facilitated a valuable estimate of the system energy efficiency. The energy efficiency was found to increase with column radius from 65 ± 3% to 86 ± 5% in 0.080 m to 0.300 m radii columns, respectively. Weak smouldering experiments using low air fluxes and low fuel concentrations also showed that increased system energy efficiency increased the robustness of the reaction to quenching. Essentially, these results highlight how processes affecting global energy storage in the system, largely behind the forward smouldering front here, impact the local energy balance at the reaction zone under extreme conditions. Altogether, this work underscores the importance of scale as a crucial design parameter in optimizing a smouldering system.
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6. References


