HEAT LOSSES IN A SMOULDERING SYSTEM:
THE KEY ROLE OF NON-UNIFORM AIR FLUX

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One file containing supplementary material is available.
Abstract

Smouldering combustion is emerging as a valuable tool for energy conversion purposes. However, the effects of radial/lateral heat losses, while critical to its viability, are not well understood. It is known that heat losses weaken the smouldering reaction near the walls. It is less known that these losses generate non-uniform air flux across the system cross-section, potentially changing conversion rates and quenching limits. This study integrated: (i) highly instrumented smouldering experiments across numerous scales, (ii) a novel method of estimating non-uniform air flux in the experiments, (iii) analytical modelling to predict non-uniform cooling, and (iv) energy balance calculations to quantify the non-uniform heat of smouldering. Altogether, this work demonstrates that heat loss-induced non-uniform air flux is significant, affecting key smouldering propagation and cooling characteristics. The uniform air flux injected at the base became redistributed with a ~50% decrease at the centreline and a ~50% increase at the wall. This was shown to cause a concave (in the direction of air flow) smouldering front and a concave cooling front. The former was shown to cause radial heat transfer inwards, leading to super-adiabatic heating towards the centre of the reactor. The latter was shown to inhibit cooling along the centreline, which progressed ~40% slower than expected during propagation. Altogether, the multiple and integrated analyses used reveal the magnitude and significance of heat losses in smouldering systems. This insight is valuable to better harness smouldering for engineering applications.

Keywords: Energy conversion; Smoldering combustion; Local thermal non-equilibrium; Heat losses; Porous media; Super-adiabatic.
# Nomenclature

## Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>CEMS</td>
<td>Continuous emissions monitoring system</td>
</tr>
<tr>
<td>GAC</td>
<td>Granular activated carbon</td>
</tr>
<tr>
<td>DRUM</td>
<td>Oil-drum sized reactor</td>
</tr>
<tr>
<td>LAB</td>
<td>Laboratory reactor</td>
</tr>
<tr>
<td>LTE</td>
<td>Local thermal equilibrium</td>
</tr>
<tr>
<td>LTNE</td>
<td>Local thermal non-equilibrium</td>
</tr>
<tr>
<td>MC</td>
<td>Wet mass basis moisture content</td>
</tr>
<tr>
<td>TC</td>
<td>Thermocouple</td>
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## Latin Letters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>(a_s)</td>
<td>Specific surface area, m(^{-1})</td>
</tr>
<tr>
<td>(A_{L}/A_C)</td>
<td>Lateral to cross-sectional area ratio, -</td>
</tr>
<tr>
<td>(C_p)</td>
<td>Specific heat capacity, J kg(^{-1}) K(^{-1})</td>
</tr>
<tr>
<td>(d_p)</td>
<td>Particle diameter, m</td>
</tr>
<tr>
<td>(h_{sg})</td>
<td>Interfacial heat transfer coefficient, W m(^{-2}) K(^{-1})</td>
</tr>
<tr>
<td>(f_{rCO})</td>
<td>Fraction of C oxidized to CO</td>
</tr>
<tr>
<td>(k)</td>
<td>Thermal conductivity, W m(^{-1}) K(^{-1})</td>
</tr>
<tr>
<td>(k_p)</td>
<td>Intrinsic permeability, m(^2)</td>
</tr>
<tr>
<td>(m/m)</td>
<td>Mass ratio, -</td>
</tr>
<tr>
<td>(\dot{m})</td>
<td>Mass flux, kg m(^{-2}) s(^{-1})</td>
</tr>
<tr>
<td>(\dot{m}'')</td>
<td>Effective mass flux, kg m(^{-2}) s(^{-1})</td>
</tr>
<tr>
<td>(Nu)</td>
<td>Nusselt number, -</td>
</tr>
<tr>
<td>(Pr)</td>
<td>Prandtl number, -</td>
</tr>
<tr>
<td>(\dot{q}')</td>
<td>Heat flux, W m(^{-2}) s(^{-1})</td>
</tr>
<tr>
<td>(r_o)</td>
<td>Outer radius, m</td>
</tr>
<tr>
<td>(Re)</td>
<td>Reynold’s number, -</td>
</tr>
<tr>
<td>(t_c)</td>
<td>Characteristic time, s</td>
</tr>
<tr>
<td>(t_{ig})</td>
<td>Ignition time, s</td>
</tr>
<tr>
<td>(t_f)</td>
<td>Final time, s</td>
</tr>
<tr>
<td>(T_{peak})</td>
<td>Maximum temperature, K</td>
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<tr>
<td>(T_{amb})</td>
<td>Ambient temperature, K</td>
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<tr>
<td>(u_g)</td>
<td>Darcy flux, m s(^{-1})</td>
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<tr>
<td>(v_{oxid})</td>
<td>Smouldering propagation velocity, m s(^{-1})</td>
</tr>
<tr>
<td>(v_{cool})</td>
<td>Cooling velocity, m s(^{-1})</td>
</tr>
<tr>
<td>(W)</td>
<td>Volumetric mass loss, kg m(^{-3}) s(^{-1})</td>
</tr>
<tr>
<td>(x_c)</td>
<td>Characteristic distance, m</td>
</tr>
<tr>
<td>(Y)</td>
<td>Oxygen mass fraction, -</td>
</tr>
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### Greek Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>δ</td>
<td>Small dimensionless parameter, -</td>
</tr>
<tr>
<td>μ</td>
<td>Dynamic viscosity, Pa·s</td>
</tr>
<tr>
<td>ν</td>
<td>Kinematic viscosity, m² s⁻¹</td>
</tr>
<tr>
<td>ξ</td>
<td>Dimensionless distance, -</td>
</tr>
<tr>
<td>ρ</td>
<td>Density, kg m⁻³</td>
</tr>
<tr>
<td>σ</td>
<td>Stephan Boltzmann constant, W m⁻² K⁻⁴</td>
</tr>
<tr>
<td>τ</td>
<td>Dimensionless time, -</td>
</tr>
<tr>
<td>φ</td>
<td>Porosity, -</td>
</tr>
<tr>
<td>Δt</td>
<td>Difference between measurement times, s</td>
</tr>
<tr>
<td>ΔT</td>
<td>Temperature difference, K</td>
</tr>
<tr>
<td>Δθ</td>
<td>Dimensionless difference between gas and solid temperatures, -</td>
</tr>
<tr>
<td>ΔH</td>
<td>Heat of reaction, MJ kg⁻¹</td>
</tr>
<tr>
<td>ΔHₐₒᵩₐₓ</td>
<td>Heat of smouldering, MJ kg⁻¹</td>
</tr>
<tr>
<td>Λ</td>
<td>Constant, -</td>
</tr>
</tbody>
</table>

### Subscripts

<table>
<thead>
<tr>
<th>Subscript</th>
<th>Description</th>
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<tbody>
<tr>
<td>0</td>
<td>Initial</td>
</tr>
<tr>
<td>amb</td>
<td>Ambient</td>
</tr>
<tr>
<td>app</td>
<td>Approximate</td>
</tr>
<tr>
<td>avg</td>
<td>Average</td>
</tr>
<tr>
<td>bulk</td>
<td>Volume averaged</td>
</tr>
<tr>
<td>c</td>
<td>Characteristic</td>
</tr>
<tr>
<td>cool</td>
<td>Cooling</td>
</tr>
<tr>
<td>eff</td>
<td>Effective</td>
</tr>
<tr>
<td>f</td>
<td>Final</td>
</tr>
<tr>
<td>g</td>
<td>Gas/air</td>
</tr>
<tr>
<td>i</td>
<td>Discretized position</td>
</tr>
<tr>
<td>l</td>
<td>Final discretized position</td>
</tr>
<tr>
<td>ig</td>
<td>Ignition</td>
</tr>
<tr>
<td>inj</td>
<td>Injected</td>
</tr>
<tr>
<td>loss</td>
<td>Lost from control volume</td>
</tr>
<tr>
<td>obs</td>
<td>Observed</td>
</tr>
<tr>
<td>oxid</td>
<td>Oxidation</td>
</tr>
<tr>
<td>pyr</td>
<td>Pyrolysis</td>
</tr>
<tr>
<td>r</td>
<td>Radial</td>
</tr>
<tr>
<td>s</td>
<td>Solid/sand</td>
</tr>
<tr>
<td>sf</td>
<td>Surface</td>
</tr>
<tr>
<td>x</td>
<td>Axial</td>
</tr>
<tr>
<td>¯x</td>
<td>Generic median parameter</td>
</tr>
<tr>
<td>¯x</td>
<td>Generic mean parameter</td>
</tr>
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</table>
1. Introduction

Smouldering combustion is being harnessed for environmentally-beneficial and waste-to-energy processes [1-4]. Specific applications include: treating lagoons of hazardous organic liquids [5-7], remediating contaminated soil [8, 9], improving sanitation in the developing world [2, 10, 11], and recovering valuable resources from waste streams [3, 12, 13]. Engineers are making strides in controlling and optimizing smouldering applications based on new research but also on a history of research in the fire safety community [14].

Smouldering is driven by flameless, heterogeneous combustion as gaseous oxygen reacts with the surface of a condensed phase fuel [14, 15]. In many contexts, smouldering can be self-sustaining for temperatures much lower than those possible for homogeneous combustion. This is the case because the energy generated at the reaction front exceeds the energy lost locally for a wide range of low temperature chemical reactions [14]. Though this property can lead to problematic fires within porous solid fuels (e.g., peat [16] and upholstery [17]), it may also be harnessed for environmental benefit [2, 3, 14, 18]. Moreover, applied smouldering systems often mix fuel within inert porous media, which stores and recycles much of the released energy and thereby provides a further buffer from extinction, i.e., improves the smouldering system’s robustness [14, 19].

Smouldering has a unique property: its set of reactions propagate through space; this will be referred to as propagation. Characteristic zones develop around the reactions, which are dynamic in space and time. Figure 1 (which is further described below) illustrates the zones common to many smouldering systems: inert heating zone, reaction zone, and cooling zone [14]. The inert heating zone, propagating ahead of the smouldering front, is
characterized by endothermic heating and phase change processes (e.g., water boiling/evaporation) but no chemical reactions. All chemistry occurs within the reaction zone including competing endothermic pyrolysis and exothermic oxidation reactions [20, 21], which are typically sequenced in that order when smouldering propagates in the same direction as the airflow (i.e., in forward smouldering) [20-24]. The reaction zone is quite thin in most applied smouldering systems \([0(0.001-0.01 \text{ m})]\) [19, 25-27]. The cooling zone, propagating behind the smouldering front, is dominated by heat transfer (i.e., local thermal non-equilibrium, LTNE) associated with the incoming air flow that is cooler than the hot, fuel-free porous media. In most applied smouldering systems, this region is longer than the other zones \([\geq 0(0.1 \text{ m})]\) and stores much of the reaction energy. Therefore, this zone is most susceptible to radial/lateral heat losses. Indeed, numerical simulations by Zanoni et al., [19, 28] demonstrated that 90-95% of the heat losses from a laboratory-sized smouldering reactor emanated from the cooling zone. Therefore, predicting heat losses in applied smouldering systems depends on predicting the evolution of the cooling zone.

The effect of radial heat losses has received some consideration in the reaction zone and limited attention in the cooling zone. Considering the reaction zone, analytical approximations of smouldering propagation usually follow the methodology established by Dosanjh et al., [29, 30], which integrates the governing equations over the reaction and inert heating zones from ambient to peak smouldering temperatures \((T_{amb} \rightarrow T_{peak})\). Because the fuel load is in excess in porous solid fuels, propagation in many fire safety contexts (e.g., polyurethane foam smouldering) is often limited by the oxygen supply.
Accounting for heat losses from the reaction zone, Bar-Ilan et al., [31] described forward one-dimensional forward smouldering as:

\[
v_{\text{oxid}} = \frac{Y_{O_2} \rho_g u_g \Delta H_{O_2} - \dot{q}_{\text{loss}}'' (A_L/A_C)}{(1 - \phi) \rho_s c_{p_s} + \phi \rho_g c_{p_g} (T_{\text{peak}} - T_{\text{amb}}) - (1 - \phi) \rho_s \Delta H_{\text{pyr}} + Y_{O_2} \rho_g \Delta H_{O_2}}
\]

where \(v_{\text{oxid}}\) is the propagation velocity, \(Y_{O_2}\) is oxygen mass fraction, \(\rho_g\) and \(\rho_s\) are the gas and solid densities, respectively, \(u_g\) is the Darcy air flux, \(\Delta H_{O_2}\) and \(\Delta H_{\text{pyr}}\) are the heat released per mass of oxygen consumed and heat consumed per mass of solid pyrolyzed, respectively, \(\dot{q}_{\text{loss}}''\) is the heat flux lost laterally, \(A_L/A_C\) is the lateral to cross-sectional area ratio, \(\phi\) is the porosity, \(c_{p_g}\) and \(c_{p_s}\) are the gas and solid heat capacities, respectively.

Equation (1) indicates that increased heat losses in the reaction zone (i.e., decreasing the numerator) weaken smouldering and slow propagation. While Eq. (1) assumes one-dimensional behaviour, actual radial heat losses are most severe at the system walls. In smouldering experiments, this can manifest as partially burned crust or unreacted fuel near the reactor wall and complete fuel combustion in the reactor centre (e.g., [31-36]). Hereafter, this phenomenon – where radial heat losses in the reaction zone cause a decrease in smouldering strength from the centre to the wall – will be referred to as non-uniform reactions.

Considering the cooling zone, heat losses cause air flow variations across the radius. This has been revealed by numerical [25, 37] and experimental studies [26, 27, 38], with considerable insight provided by Pozzobon et al., [25] and Martins et al., [26]. In the cooling zone, radial heat losses cause a lateral gradient in stored heat from the reactor centre to the wall. This lateral temperature difference causes air to flow preferentially
near the reactor walls because lower temperature air corresponds to less flow resistance (lower air viscosity, higher air density). A physical analogue is the air flow divergence associated with heterogeneity of porous media permeability (e.g., [39-43]). Higher air flux near the wall in the cooling zone corresponds to increased oxygen supply and forward heat transfer into the reaction zone. This in turn may promote stronger smouldering and faster propagation near the wall than in the centre [25, 26]. Hereafter, this phenomenon – where radial heat losses in the cooling zone cause an increase in smouldering strength from the centre to the wall – will be referred to as non-uniform air flux.

Therefore, there are two competing phenomena causing lateral differences in the strength of smouldering associated with radial heat losses. Non-uniform reactions lead to weaker smouldering at the wall, while non-uniform air flux leads to stronger smouldering at the wall. It is hypothesized that the relative influence of these two phenomena will dictate the shape of the smouldering front. This hypothesis and associated conceptual model are illustrated in Figure 1. It is expected that non-uniform reactions will dominate in less robust smouldering systems, such as those exhibiting low permeability to air and/or low energy density, and that this will result in a convex smouldering front. This regime has been given significant attention in the context of smouldering of polyurethane foam and fire safety [14, 31, 44]. It is expected that non-uniform air flux will dominate in more robust smouldering systems, such as those exhibiting high permeability to air and/or high energy density materials, and that this will result in a concave smouldering front. This regime, typical for applied smouldering systems, has received very little attention. Moreover, there is very limited research on the competition between non-uniform air flux and non-uniform reactions.
Few tools are available to study non-uniform air flux in the cooling zone behind a smouldering front. Analytical models, which can be very useful, have not been developed in this context because the combination of complex flows in porous media with combustion has been deemed intractable. Furthermore, detailed numerical models require a deep understanding of the governing processes to circumvent the need for many parameters that cannot be properly quantified.

As the cooling zone behaviour is driven by LTNE heat transfer, it is this fact that emphasizes the controlling nature of phase temperature differences in any modelling. Kuznetsov [45, 46] reviewed models that account for LTNE in a cooling porous medium. These models often follow the Schumann [47] formulation, which simplifies the problem by neglecting conduction in both phases. Kuznetsov [46] developed an alternative formulation that treated the heat transfer between phases as a perturbation and showed the degree of LTNE dissipates over time. Though the model from Kuznetsov [46] was intended for unsteady cooling within a one-dimensional porous system with no energy generation, it is here proposed as a screening tool to diagnose two-dimensional cooling behaviour within the cooling zone in applied smouldering systems.

This study explores the consequences of non-uniform air flux resulting from radial heat losses in smouldering systems. This was achieved by integrating: (i) highly instrumented experiments across numerous scales, (ii) a novel method of estimating the evolution of non-uniform air flux in the experiments, (iii) analytical modelling to predict cooling as a function of axial and radial distance under non-uniform air flux in the cooling zone, and (iv) energy balance calculations to quantify the heat of smouldering as a function of radius under non-uniform air flux in the reaction zone. Non-uniform air flux governed the front
behaviour in all experiments. Data from these experiments were used to populate the two developed analytical models for investigating non-uniform air flux. Altogether, these results provide a novel, comprehensive picture revealing the complicated relationship between heat losses and non-uniform air flux, which has numerous consequences for smouldering systems including: (i) the shapes of the smouldering and cooling fronts, (ii) the distribution of reaction intensity over the reactor radius, (iii) the rates of smouldering propagation and cooling, and (iv) the overall effectiveness and controllability of smouldering treatment.
2. Methodology

2.1. Smouldering Experiments

Five forward smouldering experiments were conducted with a constant injected air flux in highly instrumented reactors (Table 1). The reactors were either 0.08 m radius (LAB) or 0.30 m in radius (DRUM). These experiments included continuous measurement of axial and radial temperatures, mass loss, emissions (CO, CO₂, and O₂), and axial pressures (see the experimental layout in Supplementary Materials, Section S.1). The fixed beds comprised granular activated carbon (GAC) mixed within coarse grain sand. The GAC and sand properties are detailed in [48]. All mixtures were prepared with a mechanical mixer and lightly packed into the reactors. After ignition at the base of the reactor through convective means, the heater was turned off and all experiments supported self-sustained propagation. These experiments are further detailed in [48, 49] and additional details on the established experimental methods are available in [32, 50-53].

Table 1 summarizes key experimental conditions and results that were used in the analyses below. All experiments experienced varying degrees of non-uniform air flux and therefore represent a spectrum of behaviour. Bulk characteristics (i.e., integrated throughout the reactor volume) of the experiments were presented and discussed in [48]. In this work, these experiments are analyzed for differences observed across the reactor diameter in a manner not previously investigated.
Table 1. Experimental Conditions and Key Smouldering Front Results

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fuel/sand mass fraction [g_{fuel} kg^{-1}]</th>
<th>Darcy air flux (^1) [cm s^{-1}]</th>
<th>Mean centreline propagation velocity [cm min^{-1}]</th>
<th>Mean centreline peak temperature [°C]</th>
<th>Mean (fr_{CO} CO(%))</th>
<th>(CO(%) + CO_2(%))</th>
</tr>
</thead>
<tbody>
<tr>
<td>DRUM R0</td>
<td>20.0</td>
<td>5.0</td>
<td>0.46</td>
<td>804</td>
<td>-(^a)</td>
<td></td>
</tr>
<tr>
<td>2 DRUM R1</td>
<td>20.0</td>
<td>7.5</td>
<td>0.47</td>
<td>765</td>
<td>-(^a)</td>
<td></td>
</tr>
<tr>
<td>2 DRUM R2</td>
<td>23.3</td>
<td>5.0</td>
<td>0.44</td>
<td>834</td>
<td>-(^a)</td>
<td></td>
</tr>
<tr>
<td>3 LAB R1</td>
<td>20.0</td>
<td>7.5</td>
<td>0.61</td>
<td>766</td>
<td>0.27(^b)</td>
<td></td>
</tr>
<tr>
<td>3 LAB R2</td>
<td>23.3</td>
<td>5.0</td>
<td>0.49</td>
<td>874</td>
<td>0.27(^b)</td>
<td></td>
</tr>
</tbody>
</table>

Experimental Conditions’ errors are estimates of experimental equipment error. Smouldering Front Results’ errors estimate experimental variability as the normalized standard deviations from three DRUM and LAB repeat experiments from smouldering wastewater sewage sludge, a highly variable fuel [53]. These errors align with similar experimental studies [43, 53-57].

\(^1\) At standard temperature and pressure (21.1°C at 1 atm).
\(^2\) Used a thin bottom layer of wood chips for ignition and recycled sand, detailed in [48].
\(^3\) Mass balance results were spurious because the instrumentation caused erratic physical vibrations.
\(^a\) CO measurement throughout propagation exceeded DRUM’s continuous emissions measurement system calibration range.
\(^b\) This \(fr_{CO}\) was representative of DRUM R0, R1 and R2 and results in a \(\Delta H_{obs} = 24.9 \text{ MJ kg}^{-1}\). See detailed justification in [48].
2.2. Developing a simplified description of non-uniform air flux

A novel methodology was developed to approximate the flow redistribution in the smouldering reactor at different radial positions throughout time using temperature data and the thermophysical properties of air. These values were then applied to a series of concentric cylinders to estimate the distribution of air flux across the reactor cross-sectional area. This methodology and its derivation are presented below. Essentially, this flow redistribution problem was idealized as if it were a channelling phenomenon, where axial flow resistances were assumed to vary sharply between each concentric cylinder. Therefore, this analysis does not resolve the actual flow field in the smouldering reactor but instead highlights the main trends in flow redistribution because of non-uniform temperatures via a simplified analysis.

Non-uniform air flux can result from sharply changing, heterogeneous permeability (which leads to air channelling) or from more gradual temperature differences that drive heterogeneous thermophysical properties, namely viscosity and density (which leads to flow redistribution) [58, 59]. To fully capture the effect of varying thermophysical properties leading to flow redistribution, the conservation equations need to be analyzed throughout the reactor space. Two-dimensional, radial gas phase continuity equation in a saturated porous medium is [60]:

\[
\frac{\partial (\phi \rho_g)}{\partial t} + \frac{\partial (\rho_g u_{g,x})}{\partial x} + \frac{1}{r} \frac{\partial (\rho_g u_{g,r})}{\partial r} = 0
\]  

(2)

However, given that the objective here is to explain (not fully resolve) the effect of thermophysical properties on the air flux trends, the analysis will be structured under the premise that the non-uniform air flux regime is essentially a perturbation over the one-
dimensional flow. This simplification idealizes non-uniform air flux resulting from thermophysical variations as if it is driven by sharply varying differences in space, e.g., like a channelling problem from permeability heterogeneities. This analysis is therefore limited in that it does not resolve the radial flows. However, this simplification is valuable in that it does allow the complicated 2D problem to be distilled into simpler 1D problems, which can be analyzed together to highlight the key effects of non-uniform temperatures on driving non-uniform air flux. Therefore, it is useful to first simplify Eq. (2) into the one-dimensional gas phase continuity equation and characterize the air flux magnitude only in response to temperature-dependent resistance along the flow path [60]:

$$\frac{\partial (\phi \rho)}{\partial t} + \frac{\partial (\rho u_x)}{\partial x} = 0$$

(3)

The flow is assumed incompressible due to small Mach numbers common to smouldering [42, 61], and the porosity is assumed constant because the GAC (fuel) only comprises a small fraction of the pore space. Therefore, Eq. (3) is simplified assuming the Boussinesq approximation so that the air density only varies with temperature (reasonable here considering the small pressure variations [62-64]) and Darcy’s law (without gravity effects) [65]:

$$m''_{g,x} = - \frac{k_p}{\nu_g(T_g)} \frac{dP_g}{dx}$$

(4)

where $k_p$ is the intrinsic permeability (assumed homogeneous), $\nu_g(T_g) = \mu_g(T_g)/\rho_g(T_g)$ is the kinematic viscosity of air, $dP_g/dx$ is the axial air pressure gradient, $T_g$ is the air temperature, and $m'_{g,x} = \rho_g u_{g,x}$ is the constant air mass flux along the axial direction. Important, the air viscosity increases with increased temperature and this relationship
is essential in evaluating the effects of temperature on gas flow through porous media [58, 59].

The temperature changes along the flow path can be integrated from $x = x_0$, $P(x) = P_0$ to $x = x_f$, $P(x) = P_f$ to deliver an effective air flux that shows the effect of temperature on the resistance to flow. Ahead of the smouldering front, it is expected that the temperature will decrease rapidly, thus showing a much smaller radial variance. By integrating, the effective air mass flux obtained represents a simplified constant magnitude over the flow path (which is reasonable in smouldering systems with low fuel concentrations and minimal permeability heterogeneity, like the GAC experiments in Table 1 [26, 27, 48]).

Equation (4) then becomes:

$$\dot{m}_{g,x} = \frac{k_p (P_0 - P_f)}{\int_{x_0}^{x_f} v_g(T_s) \, dx} \tag{5}$$

If non-uniform air flux is occurring due to thermophysical variations, and the system is axisymmetric, then the reactor can be simplified into a series of concentric cylinders where Eq. (5) can be used to provide an estimate of the air mass flux through each cylinder. As described above, this assumption idealizes the thermophysical properties varying sharply across the boundary of each concentric cylinder. By applying Eq. (5) at various radial locations, the effect of temperature non-uniformity on driving flow non-uniformity can be approximated. In actuality, radial flows will lead to this redistribution and will evolve throughout space and time (e.g., [59, 66, 67]). However, the main trends can be highlighted without fully resolving the changing flow field. That is, Eq. (5) may be used to estimate the effective axial air mass flux across the reactor length $(x = x_0 \rightarrow x_f)$ at a specific radial location ($r_i$) to maximize the temperature data over the integration:
\[ \dot{m}''_{g,x}(r_i) = \frac{k_p(P_0 - P_f)}{\int_{x_0}^{x_f} v_g(T_g, r_i) dx} \]  

(6)

This analysis simplifies the real two-dimensional problem into a set of idealized one-dimensional problems and therefore requires a few assumptions: 

(i) the radial flow at the boundaries of the integration are negligible (i.e., at \(x_0\) and \(x_f\), \(\partial P / \partial r \sim 0\), which physically correspond to air spaces with no porous media, as seen in Fig. S.1); 

(ii) the axial flow variations are assumed much faster than the temperature variations so that a quasi-steady state assumption may be used to approximate the air mass fluxes independent of heat transfer; 

(iii) the \(\dot{m}''_{g,x}(r_i)\) estimated at various radial positions are assumed to act in parallel, therefore, the constant injected air mass flux into the reactor \(\dot{m}''_{g,\text{inj}}\) may be estimated by the area-weighted averages of the \(\dot{m}''_{g,x}(r_i)\) from \(r_i = 0 \rightarrow r_0\). As described above, these assumptions essentially treat the flow redistribution problem as if it is a channelling problem, where flow resistances between different concentric cylinders are assumed to vary sharply at the concentric cylinders’ interfaces. Figure 2 presents a conceptual model of this simplified problem:

With the kinematic viscosity varying with temperature across space \(v_g(x, r_i)\) the integration procedure for Eq. (6) over the reactor cross-section may be described as:

\[ \dot{m}''_{g,\text{inj}} = \frac{1}{\pi r_0^2} \iint_A \dot{m}''_{g,x}(r_i) r dr d\theta = \frac{k_p(P_0 - P_f)}{\pi r_0^2} \times \iint_A \left[ \frac{1}{\int_{x_0}^{x_f} v_g(x, r_i) dx} \right] r dr d\theta \]  

(7)

Equations (6-7) can then be used to infer the axial air mass flux normalized to the injected mass flux at various radial locations, using only the temperature-dependent kinematic viscosity field \(v_g(x, r_i)\), as:
\[
\frac{\dot{m}_{g,x}(r_i)}{\dot{m}_{g,\text{inj}}} = \frac{1}{\int_{x_0}^{x_f} v_g(x,r_i)dx} \times \pi r_i^2 \int_A \left[ \frac{1}{\int_{x_0}^{x_f} v_g(x,r_i)dx} \right] rdrd\theta
\]  

Equation (8) is presented only as a function of temperature to cancel out the uncertainty of the intrinsic permeability (discussed in the Supplementary Materials, Fig. S.3) and to follow a similar integration method from [48, 68], which is detailed in the Supplementary Materials, Eqs. (S.1-S.2). Equation (8) encapsulates physics of the non-uniform air flux concept articulated in the Introduction and in Fig. 1(b).

The measured temperatures and pressures in the experiments, along with the values and functions listed in Table 2, were used to solve Eqs. (6-7). Equation (7) was solved at every measurement time \(t\) to approximate the evolution of non-uniform air flux as a function of radius \(\dot{m}_{g,x}(r_i, t)\) in DRUM R0, R1, and R2, which had the necessary spatial resolution of temperature measurements (see the experimental layout in Fig. S.1).
Table 2. Temperature-Dependent Model Input Parameters.

<table>
<thead>
<tr>
<th>Par.</th>
<th>Details</th>
<th>Value(^1)</th>
<th>Unit</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(T_{amb})</td>
<td>Varied slightly between experiments</td>
<td>16-22</td>
<td>°C</td>
<td>Measured</td>
</tr>
<tr>
<td>(d_p)</td>
<td></td>
<td>2.00E-3</td>
<td>m</td>
<td>[65]</td>
</tr>
<tr>
<td>(\rho_s)</td>
<td></td>
<td>2650</td>
<td>kg m(^{-3})</td>
<td>[65]</td>
</tr>
<tr>
<td>(\phi)</td>
<td>Constant in all experiments</td>
<td>0.37</td>
<td>-</td>
<td>[65]</td>
</tr>
<tr>
<td>(\sigma)</td>
<td></td>
<td>5.67E-8</td>
<td>W m(^{-2}) K(^{-1})</td>
<td>[69]</td>
</tr>
<tr>
<td>(k_p)</td>
<td></td>
<td>8.0E-10</td>
<td>m(^2)</td>
<td>Measured (^2)</td>
</tr>
<tr>
<td>(M)</td>
<td></td>
<td>0.0290</td>
<td>kg mol(^{-1})</td>
<td>[69]</td>
</tr>
<tr>
<td>(R_g)</td>
<td></td>
<td>8.31</td>
<td>m(^3) Pa K(^{-1}) mol(^{-1})</td>
<td>[69]</td>
</tr>
<tr>
<td>(C_{p_s}(T_s))</td>
<td>(1.75T_s + 340.32)</td>
<td>1558</td>
<td>J kg(^{-1}) K(^{-1})</td>
<td>[65]</td>
</tr>
<tr>
<td>(C_{p_g}(T_g))</td>
<td>(-3 \times 10^{-5}(T_g^2) + 0.2261T_g + 940.35)</td>
<td>1081</td>
<td>J kg(^{-1}) K(^{-1})</td>
<td>[65, 69]</td>
</tr>
<tr>
<td>(\mu_g(T_g))</td>
<td>(-9 \times 10^{-12}(T_g^2) + 4 \times 10^{-8}T_g + 6 \times 10^{-6})</td>
<td>2.90E-5</td>
<td>Pa s</td>
<td>[65, 69]</td>
</tr>
<tr>
<td>(v_g(T_g))</td>
<td>(6 \times 10^{-11}(T_g^2) + 7 \times 10^{-8}T_g - 1 \times 10^{-5})</td>
<td>- (^3)</td>
<td>m(^2) s(^{-1})</td>
<td>[69]</td>
</tr>
<tr>
<td>(k_{s_app}(T_s))</td>
<td>(16\sigma d_p(T_s^3)/3 + 6.38 \times 10^{-4}T_s + 0.0915)</td>
<td>0.808</td>
<td>W m(^{-1}) K(^{-1})</td>
<td>[65]</td>
</tr>
<tr>
<td>(k_g(T_g))</td>
<td>(-1 \times 10^{-8}(T_g^2) + 8 \times 10^{-5}T_g + 4.3 \times 10^{-3})</td>
<td>5.46E-2</td>
<td>W m(^{-1}) K(^{-1})</td>
<td>[65, 69]</td>
</tr>
</tbody>
</table>

\(^1\) Average temperature dependent values are examples over the centreline \(T_{amb}\) to \(T_{peak}\) measured in DRUM R2.


\(^3\) The effective kinematic viscosity was not used in any equation, but its relationship to was used to approximate the evolution of non-uniform air flux, where the quadratic relationship was fitted to \(v_g(T_g)\) values from 250 K – 1300 K.
2.3. Analytical model to predict cooling as a function of radius under non-uniform air flux

The solution for unsteady, one-dimensional, LTNE heat transfer in porous media from Kuznetsov [46] was used to describe the axial distribution of temperature in the cooling zone as a function of time at different radial positions. As in Section 2.2, the real two-dimensional heat transfer problem was simplified into a series of one-dimensional problems at various radial locations to explore the implications of non-uniform air flux. The non-uniform air flux experimental values obtained from the Section 2.2 methodology were used as inputs in this analysis. This methodology and derivations are presented below.

Though heat losses were not modelled explicitly, they were embodied in the observed peak temperature profile \( T_{\text{peak}}(r_i) \) and the evolution of non-uniform air mass flux \( \dot{m}_{g,x}(r_i, t) \). For brevity, only the governing equations, solution, and key distinctions from Kuznetsov [46] are discussed. The development below will consider the cooling at multiple radial locations discreetly (i.e., in each concentric cylinder in Fig. 2). Therefore, a one-dimensional (axial) analytical model is presented. However, in the results, this one-dimensional model will be applied to all concentric cylinders across the radius using \( \dot{m}_{g,x}(r_i, t) \), estimated from experimental observations, to approximate two-dimensional cooling behaviour. Assuming the sand can be represented as a homogeneous continuum and saturated with air, the one-dimensional, volume-averaged energy equations for the air and sand phases are [70]:

\[
\phi \rho_g C_p \frac{\partial T_g}{\partial t} + \dot{m}_{g,x} C_p \frac{\partial T_g}{\partial x} = \phi k_g \frac{\partial^2 T_g}{\partial x^2} + h_{sg} \alpha_{sg} (T_s - T_g) \tag{9}
\]
$$\rho_s C_p \frac{\partial T_s}{\partial t} = (1 - \phi)k_{s_{app}} \frac{\partial^2 T_s}{\partial x^2} - h_{sg} a_{sg} (T_s - T_g)$$  \hspace{1cm} (10)$$

Radiation heat transfer was included in the approximated solid conductivity following the Rosseland approximation, $k_{s_{app}} = k_s + 16\sigma d_p T_s^3/3$ [65], and the specific surface area was estimated assuming the sand grains were spherical, $a_{sg} = 6(1 - \phi)/d_p$ [46, 65]. The heat transfer coefficient between phases, $h_{sg}$, was approximated following Zanoni et al., [65], which is valid for $Pr = 0.72$, $0.5 \leq Re \leq 31$, and $0.125 \leq d_p \leq 2.000$ mm:

$$Nu = h_{sg} d_p / k_g = 0.001(Re^{1.97} Pr^{1/3})$$  \hspace{1cm} (11)$$

The $h_{sg}$ correlation in Eq. (11) is different than used by Kuznetsov [46], who used a correlation from Dixon and Cresswell [71]. The correlation from [71], along with other commonly used correlations (e.g., [72]), overestimates the heat transferred between phases at low Reynold’s numbers relevant for many smouldering scenarios ($1 \leq Re \leq 40$) [56, 65]. Zanoni et al., [56] presents the justification for Eq. (11) and Rashwan et al., [68] demonstrated Eq. (11) is appropriate here, as it was shown compatible with a similar LTNE solution from Kuznetsov [70].

The initial condition assumed the air temperature along a specific radial position reached the peak smouldering temperature up through the reactor [46]:

$$T_g(x, 0) = T_{peak}(r_i)$$  \hspace{1cm} (12)$$

In other words, Eq. (12) assumed the smouldering front proceeded with constant intensity as it propagated up the reactor at a radial location. This assumption is not always true but is often observed in robust smouldering (far from extinction) within inert media (e.g.,
Furthermore, the extinction near the wall was neglected, which is also often observed in robust smouldering experiments as a thin ring of char inside the reactor wall from non-uniform reactions [32, 33, 35, 36].

Following Kuznetsov [46], the boundary conditions were:

\[ T_g(0,t) = T_{amb} \]  \hspace{1cm} (13)
\[ \frac{\partial T_g}{\partial x} (\infty, t) = 0 \]  \hspace{1cm} (14)

Equation (13) assumed ambient temperature air was constantly injected into the reactor, driving the cooling. Equation (14) assumed that the gas temperature did not change axially far ahead from the cooling front, which is often assumed to develop analytical solutions of a travelling smouldering front (e.g., [29, 68, 76]).

Towards making Eqs. (9-10) non-dimensional, a small dimensionless parameter was defined as [46]:

\[ \delta = \phi \rho_g \dot{m}'' \bar{c}_p^3 / h_{sg} a_{sg} (\rho C_p)_{bulk} k_{bulk} \]  \hspace{1cm} (15)

where \( \phi \rho_g C_{p_g} + (1 - \phi) \rho_s C_{p_s} = (\rho C_p)_{bulk} \) and \( \phi k_g + (1 - \phi) k_{s_app} = k_{bulk} \) [46]. By balancing axial bulk energy diffusion and storage with convection, the axial characteristic length, \( x_c \), and time, \( t_{c,x} \), were identified [46]:

\[ \left( \rho C_p \right)_{bulk} \Delta T \frac{\Delta T}{t_{c,x}} \sim \dot{m}_g'' C_{p_g} \Delta T \frac{\Delta T}{x_c} \sim k_{bulk} \Delta T \frac{\Delta T}{x_c^2} \]  \hspace{1cm} (16)
Equation (16) implies \( x_c = k_{\text{bulk}}/\dot{m}_g'' C_{pg} \) and \( t_{cx} = (\rho C_p)_{\text{bulk}} k_{\text{bulk}}/(\dot{m}_g'' C_{pg})^2 \), and the dimensionless distance and times were defined as \( \xi = x/x_c \) and \( \tau = t/t_{cx} \) [46]. Dividing the characteristic distance from time leads to the cooling velocity [46]:

\[
v_{\text{cool}} = \dot{m}_g'' C_{pg}/(\rho C_p)_{\text{bulk}}
\]

As shown in Eq. (17), \( v_{\text{cool}} \) is directly proportional to \( \dot{m}_g'' \). Therefore, unlike the competing processes governing smouldering strength in the reaction zone (as described in the Introduction, Fig. 1), non-uniform air flux across the radius is predicted to directly correspond to non-uniform cooling across the radius in the cooling zone.

The dimensionless temperature was described as \( \theta = (T - T_{peak})/(T_{amb} - T_{peak}) \) and the degree of LTNE was assumed small and treated as [46]:

\[
\theta_s = \theta_g + \delta \Delta \theta
\]

Kuznetsov [46] obtained the solutions to Eqs. (9-10) by using Laplace transform methods. Further details on the non-dimensional forms of Eqs. (9-10) and boundary conditions in Eq. (12-14) are included in the Supplementary Materials, Eqs. (S.3-S.4) and Eqs. (S.5-S.7), respectively. The solution for the dimensionless air temperature is [46]:

\[
\theta_g(\xi, \tau) = \frac{1}{2} \text{erfc} \left( \frac{\xi - \tau}{2\sqrt{\tau}} \right) + \frac{1}{2} \exp \xi \cdot \text{erfc} \left( \frac{\xi + \tau}{2\sqrt{\tau}} \right)
\]

The difference between phase temperatures is [46]:
\[
\Delta \theta = \frac{1 - A_2}{4 \tau \sqrt{\pi \tau}} \left\{ \left( \xi + \tau \right) \exp \left\{ -\left( \frac{\xi + \tau}{2 \sqrt{\tau}} \right)^2 \right\} + \left( \xi - \tau \right) \exp \left\{ \xi - \left( \frac{\xi + \tau}{2 \sqrt{\tau}} \right)^2 \right\} \right\} +
\]

\[
(A_1 - A_2) \left\{ \frac{1}{2} \exp \xi \cdot \text{erfc} \left( \frac{\xi + \tau}{2 \sqrt{\tau}} \right) \right\}
\]

\[
- \frac{1}{2 \sqrt{\pi \tau}} \left[ \exp \left\{ -\left( \frac{\xi - \tau}{2 \sqrt{\tau}} \right)^2 \right\} + \exp \left\{ \xi - \left( \frac{\xi + \tau}{2 \sqrt{\tau}} \right)^2 \right\} \right]\]
\]

(20)

where \( A_1 = (\rho C_p)_{\text{bulk}} / \phi \rho g C_p g, A_2 = k_g (\rho C_p)_{\text{bulk}} / \rho g C_p g k_{\text{bulk}} \).

Following the methodology from [68], the temperature-dependant effective thermophysical parameters were estimated over \( T_{amb} \) to \( T_{\text{peak}}(\rho_i) \) at various radial locations, e.g., \( (\rho C_p)_{\text{eff}}(\rho_i) = \int_{T_{amb}}^{T_{\text{peak}}(\rho_i)} \rho C_p(T) dT / (T_{\text{peak}}(\rho_i) - T_{amb}) \) (see Table 2).

2.4. Quantifying the heat of smouldering as a function of radius under non-uniform air flux

A traditional solution for steady, one-dimensional smouldering propagation was rearranged to quantify an effective heat of smouldering at different radial positions. In so doing, the effective heat of smouldering embeds phenomena not previously considered: upstream radial heat transfer because of non-uniform air flux. Like in Sections 2.2 and 2.3, the real two-dimensional propagation was simplified into a series of one-dimensional problems at various radial locations. The methodology and derivations are presented below.

Towards understanding the implications of non-uniform air flux on the smouldering front, the one-dimensional energy balance over the reaction and inert heating zones in Fig. 1 (i.e., the local energy balance [14, 19, 77]) was integrated following a traditional energy balance (e.g., [27, 29, 62, 76, 78, 79]), where the starting energy equation is:
\[
(\rho C_p)_{\text{bulk}} \frac{\partial T}{\partial t} + \dot{m}'_{\text{fg}} C_{p,\text{g}} \frac{\partial T}{\partial x} = k_{\text{bulk}} \frac{\partial^2 T}{\partial x^2} + \Delta H_{\text{oxid,eff}} \cdot W
\]

Equation (21) assumes local thermal equilibrium (LTE). Though the differences in phase temperatures are necessary to resolve the temperature profiles in smouldering systems [56], they are not necessary to estimate rates of smouldering propagation in many contexts (e.g., [27, 56, 76, 78-83]). As propagation here was assumed fuel-limited, which is common to many applied smouldering systems (e.g., [27, 36, 50, 76, 78, 79]), the generation term rate is normalized to the fuel destruction rate. Therefore, \( W = -\frac{\partial}{\partial t} \left\{ (1 - \phi) \rho_s m_{\text{fuel}} / m_s \right\} \) is the mass lost per volume, where the bulk density of fuel is described using the mass fraction of fuel to sand \( (m_{\text{fuel}} / m_s) \).

It is important to highlight that fuel-limited propagation here results from the global processes driving smouldering propagation, not the local processes that govern the rate of smouldering [49]. That is, though smouldering propagation is fuel-limited in many applied systems, the local processes driving smouldering are probably limited by oxygen transport from the bulk pore space to the reacting surface (i.e., diffusion-limited) [14]. Williams [22] argued that self-sustaining smouldering is likely only limited by chemistry (i.e., kinetic-limited) in an unstable state near extinction. The interplay between local mass transfer and chemistry in smouldering remains an open question, as there is no clear consensus in the literature [14]. Therefore, though it is unclear how to describe the local rate of smouldering, propagation in most applied smouldering systems is clearly fuel-limited, which is used here in simplifying Eq. (21).

The effective heat of smouldering per mass of fuel \( (\Delta H_{\text{oxid,eff}}) \) used here also encompasses any radial heat transfer ahead of the smouldering front that is not described
in Eq. (21), e.g., radial convective currents, radiation, and conduction. By assuming the smouldering front is steady in a frame of reference travelling with the smouldering front \( v_{\text{oxid}} \), Eq. (21) becomes:

\[
-v_{\text{oxid}}(\rho C_p)_{\text{bulk}} \frac{\partial T}{\partial x} + \dot{m}_g' C_{pg} \frac{\partial T}{\partial x} = k_{\text{bulk}} \frac{\partial^2 T}{\partial x^2} + \Delta H_{\text{oxid,eff}} \cdot W \tag{22}
\]

where, the mass loss rate relative to the smouldering front is now \( W = -v_{\text{oxid}} \partial \frac{1 - \phi}{\partial x} \rho_s m_{\text{fuel}} / m_s \)|\( \partial x \).

Equation (22) can be integrated from the reaction front \( (x \to -\infty) \) where \( T = T_{\text{peak}}(r_i) \) to far ahead of the reaction front \( (x \to \infty) \) where \( T = T_{\text{amb}} \). The temperatures at these boundaries are assumed approximately constant along the axis, i.e., \( dT/dx \sim 0 \), and the fuel is assumed completely consumed. Furthermore, it is also assumed that: (i) the increase in gas flux due to smouldering is small, which is reasonable as a mass balance from Rashwan et al., [48] showed the gas mole flux entering a GAC and sand smouldering reactor is approximately 99% of the gas mole flux exiting (representative of many applied smouldering conditions, based on common fuel concentrations and propagation velocities), (ii) \( u_g \gg v_{\text{oxid}} \), (iii) \( (1 - \phi) \rho_s C_{ps} \gg \phi \rho_g C_{pg} \), and (iv) the heat capacity of the smouldering emissions is comparable to the heat capacity of air \( (C_{pg}) \). Altogether, the resulting expression is used to estimate the \( \Delta H_{\text{oxid,eff}} \) from experimental parameters and observations.

\[
\Delta H_{\text{oxid,eff}} = \frac{v_{\text{oxid}} (1 - \phi) \rho_s \int_{T_{\text{amb}}}^{T_{\text{peak}}} C_{ps}(T) dT - \dot{m}_g' \int_{T_{\text{amb}}}^{T_{\text{peak}}} C_{pg}(T) dT}{v_{\text{oxid}} (1 - \phi) \rho_s \frac{m_{\text{fuel}}}{m_s}} \tag{23}
\]
Equation (23) is very similar to other descriptions of smouldering propagation, e.g., Eq. (1) [27, 29, 62, 76, 78, 79], but uses the temperature-dependent heat capacities of: (i) coarse sand measured from Zanoni et al., [65], as the same sand was used in all experiments analyzed here, and (ii) air [65, 69]. All parameters used in Eq. (23) are detailed in Tables 1 and 2. Furthermore, heat losses are neglected in Eq. (23), as the reaction zone is very thin in most applied smouldering systems and assumed to be nearly adiabatic [19, 25-27].

By applying Eq. (23) at multiple radial locations, the area-weighted, average heat of smouldering ($\Delta H_{\text{oxid,avg}}$):

$$\Delta H_{\text{oxid,avg}} = \frac{1}{\pi r_0^2} \int \int \Delta H_{\text{oxid,eff}}(r_i) r dr d\theta$$  \hspace{1cm} (24)

Like Eq. (8), the method used to integrate Eq. (24) is similar as the methods in [48, 68] and detailed in the Supplementary Materials, Eq. (S.8). The average heat of smouldering can be compared to the heat of smouldering observed from experiments ($\Delta H_{\text{oxid,obs}}$) [84]:

$$\Delta H_{\text{oxid,obs}} = f r_{CO} \Delta H_{\text{oxid,0.5O}_2+\text{C} \rightarrow \text{CO}} + (1 - f r_{CO}) \Delta H_{\text{oxid,O}_2+\text{C} \rightarrow \text{CO}_2}$$ \hspace{1cm} (25)

where $f r_{CO}$ is the fraction of C oxidized to CO measured from experiments. Additional discussion on the use of Eq. (25) in applied smouldering systems is provided in [25-27, 48, 78, 79, 84].
3. Results and Discussion

3.1. Experimental observations of the smouldering front shape and temperature distribution

Figure 3 presents the temperature histories from DRUM R2 from various thermocouples (TCs) throughout space, plotted until their respective temperatures peaked (i.e., the front arrived). Figure 3 shows the smouldering temperatures across the radius (i.e., near the centreline, half-radius, and wall) when the smouldering front travelled 0.2 m at ~60 minutes, 0.4 m at ~100 minutes, 0.6 m at ~150 minutes, and 0.8 m at ~200 minutes. First, the figure reveals that smouldering was robust. The temperatures at the centreline increase rapidly as the front arrives, the peak temperatures at the centreline are consistent in time, and the propagation velocity at the centreline is nearly steady. Moreover, the average propagation velocity over the reactor radius and mass loss rate were steady after ignition effects (as discussed in the Supplementary Materials, Fig. S.6 and [48]). These are all characteristics of robust smouldering, common to all experiments. Additional evidence of steady, robust smouldering propagation is discussed in Section 3.4.

Second, Fig. 3 shows how the smouldering front was curved, since front curvature can be inferred by observing which radial TC heats first at a given axial location. Like previous applied smouldering studies [25-27], Fig. 3 shows the smouldering front in DRUM R2 was curved upwards (concave) (Fig. 1(b)). This front shape was observed in all the Table 1 experiments. Additional plots from DRUM R0 and R1 are included in the Supplementary Materials, Figs. S.4(a) and S.4(b), respectively.
Third, Figure 3 illustrates how the smouldering front curvature evolved throughout propagation. The concave shape had not yet established when the smouldering front propagated 0.2 m. However, from 0.4 m to 0.8 m, the smouldering front became concave – as heating first occurred near the wall, then half-radius, then the centre of the reactor – and that curvature increased with time (i.e., increased spread of those arrival times). This dynamic behaviour corresponds to dynamic propagation velocities near the wall, which were initially fast but then decelerated to equal mean centreline values (see Supplementary Materials, Fig. S.5).

Figure 4 plots the peak temperatures from all GAC experiments (DRUM and LAB) versus non-dimensional reactor radius ($r/r_o$). Peak smouldering temperatures were observed to decrease from the centreline towards the wall; this matches observations from virtually every other non-adiabatic smouldering study (e.g., [25, 27, 48, 73, 78, 84]). The small error bars in Fig. 4 reveals the peak temperatures at each radius were relatively steady (as shown for DRUM R2 in Fig. 3 and further examined in the Supplementary Materials, Fig. S.6). Zanoni et al., [19] suggests that the majority of heat losses in applied smouldering systems emanate from the thick cooling zone [19, 25-27]. In other words, the reaction zone is nearly adiabatic in many applied smouldering systems. Therefore, it is hypothesized here that the radial peak temperature gradient observed in the reaction zone (Fig. 4) results from radial heat losses from the cooling zone. This hypothesis is further explored below.
3.2. Quantitative analysis of non-uniform air flux in experiments

Equation (8) was applied to the temperature data in each experiment to map the non-uniform air flux as a function of time at four radial locations in the reactor. DRUM R2 is shown here as a typical example. Figure 5 reveals that the air flux was approximately uniform across the radius at the beginning and end of the experiment (when the temperature was most uniform) and was non-uniform across the radius in between those times. The air flux non-uniformity increased throughout smouldering propagation as temperature differences between the walls and centre increased. This non-uniformity continued to increase for some time after smouldering was complete, since the walls cool much faster than the centre, leading to continued growth in radial temperature gradients. The maximum radial temperature gradients caused the air flux vectors to diverge in the cooling zone from the reactor centre to the wall, thereby decreasing the effective axial air flux near the centre of the reactor by approximately 50% and increasing the effective axial air flux near the reactor wall by approximately 50%.

The evolution of non-uniform air flux observed in Fig. 5 aligns with the evolution of the smouldering front curvature observed in Fig. 3. Together the data sets indicate that a flat front early in propagation coincided with more uniform flux, and front curvature increased as the extent of air flux non-uniformity increased throughout propagation. This suggests a link between non-uniform air flux and the smouldering front curvature (as hypothesized in Fig. 1(b)).

The same analysis in Fig. 5 was performed on all robust GAC DRUM experiments and are presented in the Supplementary Materials, Fig. S.2. In summary, the results from DRUM R0 and R1 (in Figs. S.2(a) and S.2(b), respectively) were similar to the DRUM R2
results in Fig. 5; i.e., non-uniform air flux evolved similarly among all DRUM GAC experiments.

3.3. Cooling as a function of radius under non-uniform air flux: Experiments and analytical modelling

Equations (18-20) were used to model the axial cooling profiles at multiple radial locations and these predictions were compared with experimental results. This comparison was completed in two ways: \( (I) \) assuming the air flux was constant with radius and equal to the injected air flux along the centreline, i.e., \( \dot{m}_{g,x}(r_i = 0, t) = \dot{m}_{g,\text{inj}} \) and \( (II) \) assuming the air flux varied across the radius and time, using the \( \dot{m}_{g,x}(r_i, t) \) values provided in Fig. 5. Though the values from Fig. 5 represent the axial air flux across the entire reactor length, they were used to approximate the time-averaged axial flux just across the cooling zone. Therefore, while this analysis does not fully resolve the dynamics within the cooling zone, it aims to highlight the key trends driving cooling. Both modelling attempts \( I \) and \( II \) used the parameters from Tables 1 and 2, including the constant ambient temperature \( T_{\text{amb}} \) injected into the plenum to sustain propagation and the peak temperatures from Fig. 4 as the boundary and initial conditions at multiple radial locations in Eqs. (12) and (13), respectively. Modelling attempt \( II \) approximates the effects of heat losses on cooling by assuming they manifest predominantly in the radial peak temperature distribution and non-uniform air flux, as heat losses are not explicitly modelled in Eqs. (18-20).

Figure 6 compares the modelled cooling behaviour and experimental results along the centreline in DRUM R2 at three times, following modelling attempt \( I \). The figure reveals that, although the cooling profile is well-predicted initially, the error between the modelled
and measured temperatures increases with time. The model predicts faster cooling than was observed. This observation suggests that the key assumption in modelling attempt I, that the air flux along the centreline was constant and equal to the injected air flux, is incorrect.

Figure 7 compares the modelled cooling behaviour at various radial positions with the experimental results from DRUM R2 at three times, following modelling attempt II. This figure reveals that, by accounting for $\hat{m}_{g,x}(r_i, t)$, this model predicts the observations much better than model I. Both the model and the data exhibit cooling to be slowest in the centre and fastest near the reactor wall. As predicted by Eq. (17), the rate of cooling was proportional to the air flux. This direct proportionality inhibited cooling along the centreline, so that it proceeded approximately 40% slower than expected during propagation. Therefore, the radial differences in cooling are caused by non-uniform air flux in the cooling zone.

Furthermore, by comparing the position of the cooling front at various radial positions and times, the cooling front shape across the reactor can be observed. Figure 7 includes vertical dotted lines highlighting the approximate cooling front position (halfway between the peak and ambient temperatures) across the radius at each time. Figure 7 demonstrates that the cooling front position at the half-radius and wall were ahead of the cooling front in the centreline. Therefore, the cooling front was concave and became increasingly curved over time. The cooling front curvature aligns with the smouldering front curvature (Fig. 3). This is strong evidence that both curvatures were driven by non-uniform air flux.

It is noted that the model and experimental comparisons are best made along the centreline (Figs. 7(a-c)). This is because the centreline was best instrumented with TCs
and the centreline cooling front shape was least influenced by radial heat losses (aside from the indirect consequences from non-uniform air flux and the radial peak temperature distribution) [25, 68, 70]. The same trends were observed in DRUM R1 in the Supplementary Materials, Fig. S.7. It is the fact that the centreline should be predicted most accurately by the model that most clearly distinguishes model I from model II and emphasizes the important role played by non-uniform air flux here.

3.4. The heat of smouldering under non-uniform air flux: Experiments and Modelling

Equation (23) was used to estimate the effective heat of smouldering at various radial positions in all GAC experiments (i.e., LAB R1, R2 and DRUM R0, R1, and R2). These approximated values were normalized to the observed heat of smouldering, calculated from the experimental CO and CO$_2$ emissions (i.e., a bulk value specific to each experiment from Eq. (25) and detailed in [48]). The analysis used relevant values estimated from Tables 1 and 2, the radial peak temperature profiles from Fig. 4 (which were all far from initial effects), each experiment's mean centreline $v_{o, o}$, and the constant $m_{g, inj}'$ specific to each experiment.

Though smouldering is generally a dynamic process, robust smouldering in applied systems can reach a nearly steady-state condition away from initial and end effects. This is observed when key smouldering metrics are nearly constant over time (e.g., propagation velocities, peak temperatures, and CO/CO$_2$ fractions [48, 49, 73, 74]). Therefore, the values used to solve Eq. (23) represent this nearly steady-state condition, when the temperatures and CO/CO$_2$ fractions were nearly constant, and the $v_{o, o}$ at all
radial locations approached the mean centreline $v_{oxid}$. Additional details on the
implementation can be found in [48] and in the Supplementary Materials, Figs. S.5 and
S.6.

Figure 8 plots the non-dimensional effective heat of smouldering values at various radial
locations in all GAC experiments. A value of 1 on the y-axis indicates that the effective
heat of smouldering at a radial location is equal to the average heat of smouldering
observed from the experiments. A value of 1.3 on the y-axis indicates the maximum
$\Delta H_{oxid}$ of the embedded fuel assuming complete one-dimensional, adiabatic combustion
in Eq. (23) (i.e., if all C oxidized to CO$_2$). Like the radial peak temperatures in Fig. 4, the
effective heat of smouldering values diminished from the centre to the reactor wall.
Furthermore, Fig. 8 shows that axial flow differences from non-uniform air flux amplified
the differences in values across the radius (as identified from the labels and light-coloured
markers) but did not significantly affect the main trends.

The lower effective heat of smouldering values near the reactor wall may be due to a
combination of factors: (i) changes in reaction chemistry could favour more incomplete
oxidation towards the wall because of lower heating rates (Fig. 3), lower peak
temperatures (Fig. 4), and lower oxygen residence times because of higher air velocities
(i.e., all driving non-uniform reactions, Fig. 1(a)); (ii) a heat loss correction may be needed
in Eq. (23), especially in calculations near the reactor wall that were most affected by
radial losses, e.g., as done by Bar-Ilan et al., [31, 44]. However, these two factors do not
explain the high $\Delta H_{oxid, eff}(r_i)$ estimates toward the reactor centre: nearly all values at the
centreline in Fig. 8 were greater than the maximum $\Delta H_{oxid}/\Delta H_{oxid,obs} = 1.3$. These high
\( \Delta H_{\text{oxid,eff}}(r_i) \) values show a super-adiabatic effect, where the reactor centre was heated beyond the maximum \( \Delta H_{\text{oxid}} \) of the embedded fuel.

It is hypothesized that this super-adiabatic heating was due to radial heat transfer towards the centre from outer radial locations. Centre preheating from the walls occurred because the smouldering front exhibited concave curvature (Figs. 1(b) and 3). Simulations from Zanoni et al., [33] illustrate that matching weak smouldering experiments requires super-adiabatic heat of combustion values. Simulations from Lutsenko [66] suggest a local radial velocity component into the reactor centre may have governed this extra heat transfer. Ultimately, this super-adiabatic effect towards the centreline is an indirect consequence of heat losses from the trailing cooling zone causing non-uniform air flux.

To provide extra confidence in the interpretations above, the results from Fig. 8 were averaged over each experiments’ reactor radius using Eq. (24). Figure 9 shows the \( \Delta H_{\text{oxid,avg}} \) (area-weighted average) from each experiment normalized to the \( \Delta H_{\text{oxid,obs}} \) from experimental observations [48]. The close agreement between \( \Delta H_{\text{oxid,avg}} \) and \( \Delta H_{\text{oxid,obs}} \) shows that that smouldering propagation here is well-described assuming the one-dimensional, steady approximation with Eqs. (23-25). Moreover, Fig. 9 further substantiates that minimal energy was lost from the reaction zone and instead was redistributed into the centre of the reactor, as Eqs. (23-25) assume propagation is adiabatic. Therefore, though significant energy was lost from the experiments as heat losses (approximately 35 ± 3% and 14 ± 5% of the energy inputted for ignition and released from smouldering in the LAB and DRUM experiments, respectively [48]) these losses drew primarily from the thick cooling zone not from the thin reaction zone.
As argued by Bar-Ilan et al., [44], if significant energy was lost from the reaction zone, the $\Delta H_{\text{oxid,avg}}$ values in Fig. 9 would be systematically lower than $\Delta H_{\text{oxid,obs}}$. Instead, most $\Delta H_{\text{oxid,avg}}$ estimates in Fig. 9 are higher than $\Delta H_{\text{oxid,obs}}$. This overprediction may be due to compounded errors, which are approximated in the error bars in Fig. 9, or an incomplete description of the local energy balance. For example, the influence of ignition was included by Torero and Fernandez-Pello [62] and other multidimensional effects (e.g., air channelling) were also neglected. Overall, the $\Delta H_{\text{oxid,avg}}$ and $\Delta H_{\text{oxid,obs}}$ estimates are sufficiently close (e.g., like the propagation velocity matching in [34, 62, 63, 85]) to validate that smouldering propagation is well characterized by Eqs. (23-25).
4. Conclusions and Implications

Overall, heat losses have a complicated impact on smouldering propagation. It is proposed that the smouldering front shape results from the competition between non-uniform reactions (leading to a convex shape) and non-uniform air flux (leading to a concave shape). In this work, heat losses led to lower peak temperatures towards the wall and a smouldering front that curved towards the reactor wall in the direction of air flow (concave).

Like the smouldering front, the trailing cooling front was shown to be similarly curved (concave). The curvatures of both fronts reflected the evolving differences in resistance to air flow across the reactor radius. These thermophysical-induced air resistance differences led to non-uniform air flux. Non-uniform air flux caused the highest air mass flux near the wall (~50% increase), which was comparatively cooler because of heat losses in the cooling zone. At the same time, axial convection was reduced towards the reactor centre, so centreline cooling proceeded approximately 40% slower than expected during propagation. Moreover, it was shown that the curved smouldering front further cools peak temperatures near the reactor wall and provides super-adiabatic heating towards the centre of the reactor (due to radial heat transfer inwards). This is another, previously unreported consequence of non-uniform air flux.

Altogether, the multiple and integrated analyses used here describe a variety of heat loss effects on smouldering propagation with important implications across a broad spectrum of smouldering conditions. However, heat losses play a complicated role in smouldering systems and are responsible for multiple feedback loops that are difficult to decouple. Especially regarding non-uniform air flux. Non-uniform air flux is a flow redistribution
problem that would benefit from an expanded analysis to resolve the changing air flow field throughout smouldering propagation. Therefore, it would be worthwhile to further investigate these effects with a validated, multi-dimensional numerical model to account for non-linearities and heterogeneities, similar to [25, 66]. Here, unique experimental evidence and targeted analyses are provided to help guide this future work on better understanding the consequences of heat losses in smouldering systems and exploring interventions to overcome, or make use of, those consequences.
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6. References


Fig. 1. Conceptual model of the relative impacts of heat losses on the curvature of the smouldering front. (a) convex smouldering front (non-uniform reactions in the reaction zone dominate) versus (b) concave smouldering front (non-uniform air flux in the cooling zone dominates). The key zones are labelled for discussion: (i) Inert Heating Zone, (ii) Reaction Zone, and (iii) Cooling Zone. Common temperatures and pressures at the boundaries associated with constant injected air mass flux experiments are highlighted.
Fig. 2. Conceptual model of the effect of non-uniform temperature driving air thermophysical variations that lead to non-uniform air flux. The resistance to flow in the axial direction is treated in series and the resistance in the radial direction is treated in parallel. The concept of dividing the reactor into a series of concentric cylinders is illustrated.
Fig. 3. Temperature histories from DRUM R2 plotted until each thermocouple’s (TC’s) peak temperature to highlight the smouldering front behaviour.
Fig. 4. Median peak temperatures from all robust GAC experiments across the normalized reactor radius. All peak temperatures were measured after initial ignition effects until the end of propagation, which captured the smouldering front travelling 0.485 to 0.800 m and 0.120 to 0.540 m in the DRUM and LAB experiments, respectively. The peak temperatures were all relatively steady during this period (see an expanded discussion in the Supplementary Materials, Fig. S.6). The error bars note the maximum and minimum observations from each experiment.
Fig. 5. Non-uniform air flux estimates smoothed over time using the temperatures from DRUM R2 (see raw results and validation to measured pressures in the Supplementary Materials, Figs. S.2 and S.3, respectively).
Fig. 6. Experimental data from the centreline showing axial cooling behind the smouldering front from DRUM R2 compared to the modelled sand and air temperatures from Eqs. (18-20). The frames show cooling at (a) 3600 s, (b) 7200 s, and (c) 9840 s from fixing the air flux after turning off the heater. The axial air flux through the centreline is assumed uniform and constant throughout time \( \dot{m}_{g,x}(\zeta, t) = \dot{m}_{g,inj} \). The plenum air temperature measured 0.05 m below the fuel bed was used as the 0 m temperature in all frames (see the experimental layout in the Supplementary Materials, Fig. S.1).
Fig. 7. Experimental temperature data showing cooling in DRUM R2 compared to the modelled temperatures from Eqs. (18-20). The 0 m values were from the air plenum 0.05 m below the fuel bed (Fig. S.1). The frames show cooling at: (a, d, g) 3600 s, (b, e, h) 7200 s, and (c, f, i) 9840 s from fixing the air flux after turning off the heater along: (a-c) the reactor centreline, (d-f) 0.135 m from the centre, and (g-i) 0.227 m from the centre. The local air mass flux \( \dot{m}_{g,x}(r_i, t) \) is compared to the injected air mass flux \( \dot{m}_{g,\text{inj}}'' \) for reference and the dashed lines note the approximate cooling front positions.
Fig. 8. The $\Delta H_{\text{oxid,eff}}(r)$ calculated at every radial position from Eq. (23) using the $T_{\text{peak}}(r_i)$ profiles in Fig. 4 and the values from Tables 1 and 2 normalized to the $\Delta H_{\text{oxid,obs}} = 24.9 \text{ MJ kg}^{-1}$ (observed experimentally from the reaction stoichiometry in [48]). The $\Delta H_{\text{oxid,eff}}(r_i)$ from DRUM R0, R1, and R2 are also calculated after 9840 s of cooling using the $\dot{m}_{g,x}(r_i, t = 9840 \text{ s})$ results from Figs. 5 and S.2. The extremes of reaction completeness are highlighted for comparison in the maximum and minimum $\Delta H_{\text{oxid}}$ values assuming either no CO was produced or only CO was produced, respectively. The $\dot{m}_{g,x}(r)$ and $\Delta H_{\text{oxid,eff}}(r)$ relationship indicate the direct implications of non-uniform air flux on Eq. (23). The error bars note conservative estimates of experimental error and are only included on the DRUM R1 for clarity.
Fig. 9. The $\Delta H_{\text{oxid,avg}}$ from averaging all estimated $\Delta H_{\text{oxid,eff}}(r_t)$ values from each experiment in Fig. 8 using Eq. (25) and normalized to the $\Delta H_{\text{oxid,obs}} = 24.9$ MJ kg$^{-1}$ (observed experimentally from the reaction stoichiometry in [48]). The extremes of reaction completeness are highlighted for comparison in the maximum and minimum $\Delta H_{\text{oxid}}$ values assuming either no CO was produced and only CO was produced, respectively. The filled and open markers indicate the $\Delta H_{\text{oxid,avg}}$ values estimated using the constant $\dot{m}_{g,\text{inj}}''$ and $\dot{m}_{g,x}(r, t = 9840$ s) values specific to each experiment, respectively. The error bars note conservative estimates of experimental error [48].