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Application of self-sustaining smouldering combustion for the destruction of wastewater biosolids



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ABSTRACT

Managing biosolids, the major by-product from wastewater treatment plants (WWTPs), persists as a widespread challenge that often constitutes the majority of WWTP operating costs. Self-sustained smouldering combustion is a new approach for organic waste treatment, in which the waste – the combustion fuel – is destroyed in an energy efficient manner after mixing it with sand. Smouldering has never been applied to biosolids. Column experiments, using biosolids obtained from a WWTP, were employed to identify if, and under what conditions, smouldering could be used for treating biosolids. The parameter space in which smouldering was self-sustaining was mapped as a function of key system metrics: (1) sand/biosolids mass fraction, (2) biosolids moisture content, and (3) forced air flux. It was found that a self-sustaining reaction is achievable using biosolids with water content as high as 80% (with a biosolids lower heating value greater than 1.6 kJ/g). Moreover, results suggest that operator-controlled air flux can assist in keeping the reaction self-sustaining in response to fluctuations in biosolids properties. This proof-of-concept demonstrates the potential for smouldering as a new energy efficient biosolids disposal method for very wet (i.e., minimally processed) biosolids that may offer WWTPs significant operating cost savings. This study emphasizes smouldering's usefulness as a novel waste management technique.

1. Introduction

Wastewater treatment plants (WWTPs) treat sewage via various chemical, physical, and biological processes to remove harmful constituents and mitigate risk to the community and surrounding environment (Metcalf and Eddy, 2003). Municipal WWTPs are energy intensive operations which, combined with drinking water services, account for 3–4% of all energy consumption in the United States and 30–40% of total energy consumed by municipalities, costing \$4 billion/year (U.S. EPA, 2014). Furthermore, as much of North America's WWTP infrastructure approaches the end of its design life, an estimated \$298 billion is required in the United

Abbreviations: BSS, borderline self-sustaining; CI, confidence interval (%); MC, moisture content (%); HHV, higher heating value (kJ/g); HHV $_d^b$, dry biosolids higher heating value (kJ/g); LHV $_e^b$, biosolids lower heating value (kJ/g); LHV $_e^b$, effective system lower heating value (kJ/g); k_y , heat of vaporization of water (kJ/g); NSS, nonself-sustaining; S/B, sand/biosolids mass ratio (g/g); S/F, sand/faeces mass ratio (g/g); SS, self-sustaining; STAR, Self-sustaining Treatment for Active Remediation; STARX, Self-sustaining Treatment for Active Remediation applied ex-situ; TC, thermocouple; WWTP, wastewater treatment plant.

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States (and \$39 billion in Canada) to expand and upgrade WWTP infrastructure (ASCE, 2013; Félio et al., 2012). The major byproduct from WWTPs is biosolids and approximately 50% of WWTPs capital and operating costs are dedicated to processing biosolids, making it the most expensive component of the WWTP process (Khiari et al., 2004).

Biosolids are defined as the separated solids from WWTPs that undergo additional treatment for beneficial end use (U.S. EPA. 1994). These separated solids, largely organic, are first settled out from the liquid stream either before treatment (primary sludge) or after biological processing (waste activated sludge). The resulting sludge contains 88.00-99.75% moisture content (wet mass basis) (Droste, 1997; Metcalf and Eddy, 2003). This sludge undergoes various processing steps (e.g., dewatering, thickening, conditioning) to reduce its volume and improve aesthetic qualities for easier management, or undergoes stabilization to permit safe land application (Droste, 1997). The major disposal (or end use) methods for biosolids in Canada include incineration, land application for agricultural purposes, and landfilling (Apedaile, 2001). All of these methods are expensive in that they require high energy input, many person-hours, and/or large transportation distances (Bellur et al., 2009; Wang et al., 2008; Werther and Ogada,

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1999). In addition, land application is controversial and subject to restrictions and uncertain risks stemming from contaminants of emerging concern (Bolong et al., 2009; Giger et al., 2003; Hale et al., 2001; Ternes et al., 2004; U.S. EPA, 1995; Venkatesan and Halden, 2014). In general, managing biosolids is a major challenge for WWTPs and there is a strong need to provide novel alternatives (Tyagi and Lo, 2013).

This work explores, for the first time, the possibility of using smouldering combustion as a new method for biosolids management. STARx (Self-sustaining Treatment for Active Remediation applied ex-situ) refers to the commercial technology that uses smouldering combustion to destroy organic wastes; to date it has been shown to be promising for a variety of organic industrial liquid wastes including coal tar, crude oil, and mixed hydrocarbons (Pironi et al., 2011; Switzer et al., 2009, 2014). STAR applied insitu has completed numerous pilot tests in North America and Europe (Scholes et al., 2015) and is now being applied to a coal tar contaminated site in the United States in its first full scale application. STARx has undergone numerous pilot tests for treating mixed oil waste (Switzer et al., 2014) and crude oil sludge but has yet to be applied at the full scale. As explained below, smouldering has the potential to treat organic wastes in an energy efficient and cost-effective manner.

1.1. Smouldering combustion

Smouldering combustion is a flameless, heterogeneous (i.e., fuel and oxidant in different phases) oxidation reaction limited by the rate at which oxygen diffuses into the surface of a solid or liquid fuel (Ohlemiller, 1985; Switzer et al., 2009; Rein, 2016). Smouldering is self-sustaining when, after a short and localized energy input for ignition, the reaction propagates using only the heat produced by the fuel's oxidation (Switzer et al., 2009). Self-sustained smouldering requires a porous material, which provides a high surface area for reaction and adequate permeability for air flow (Drysdale, 2011). The majority of smouldering research has been performed in the context of fire prevention, and thus has focused on solid fuels such as polyurethane foam or stored biomass under natural air flow (e.g., He and Behrendt, 2009; Palmer, 1957; Quintiere et al., 1982; Rein et al., 2006). STARx accelerates the reaction by using forced air flow and, taking advantage of the buoyant hot combustion gases, utilizing upwards forward smouldering. In this configuration, the reaction propagation and oxidizer flow are both in the upward direction against gravity (Torero and Fernandez-Pello, 1996).

Upwards forward smouldering promotes efficient heat transfer ahead to unburned fuel, which extends the fuel's limits with respect to quenching (i.e., the suppression of chemical processes driving combustion) relative to those for flaming combustion (Hadden and Rein, 2011; Howell et al., 1996; Ohlemiller, 1985; Yermán et al., 2015). This means that smouldering is much less susceptible to extinction than flaming and can achieve a self-sustaining reaction using fuels with very low effective calorific values and/or significant moisture content (Hadden and Rein, 2011; Yermán et al., 2015).

Several studies have explored the self-sustained smouldering of peat in the context of forest fires, which is relevant in this context because peat, like biosolids, exhibits high moisture and inorganic contents (Rein, 2013). Frandsen (1987) detailed the impact that moisture and inorganic contents had on self-sustaining smouldering of peat moss between 0–1 moisture/organic ratio and 0–5 inorganic/organic ratio. It was found that the moisture/organic ratio limit that permitted self-sustaining smouldering linearly declined with increasing inorganic/organic ratio. Using an ignition protocol roughly equivalent to the heating from a flaming stump, 100 W for 30 min, Rein et al. (2008) found the critical moisture content for

smouldering ignition to be 55 ± 2% (wet mass basis). Huang et al. (2015) and Huang and Rein (2015) developed a numerical model to explore the effects that the moisture and inorganic contents have on self-sustaining smouldering within peat. The latter study revealed that in a peat sample with little inorganic content a self-sustaining smoulder is possible with moisture content as high as 72% (wet mass basis) with a critical moisture content for ignition of 54% (wet mass basis). Prat et al. (2014) found a dramatic drop in smouldering propagation velocity above 20% moisture content (wet mass basis).

1.2. Application of smouldering for waste management

Intentional smouldering for mass destruction was first developed for the in-situ remediation of soil contaminated by organic industrial liquid wastes (Pironi et al., 2011, 2009; Salman et al., 2015; Switzer et al., 2009) and is commercially available as the STAR technology (Self-sustaining Treatment for Active Remediation). In this case, the fuel (i.e., contaminant) occupies a fraction of the pore space of an inert porous medium (i.e., soil). STARx extends this concept to intentionally mixing liquid wastes in above ground applications, which may be recently produced by industrial operations or were historically disposed in lagoons, with sand to form a smoulderable mixture. In addition to providing increased surface area for reaction and permeability for air (oxidant) flow, the sand promotes the efficient storage, transfer, and recycling of the released reaction energy (Switzer et al., 2014). Smouldering of organic liquids in sand typically achieves peak temperatures between 500 and 800 °C for many minutes in one location resulting in upwards of 99% consumption of fuel, effectively producing clean, sterile sand that can be reused (Switzer et al., 2009).

In examining the sensitivity of a smouldering reaction, of interest is the effect of a variable on peak temperatures and reaction propagation rates in the self-sustaining regime, and the boundary between self-sustaining and non-self-sustaining reactions. Pironi et al. (2011) studied the influence of the fraction of pore space occupied by water on the smouldering of coal tar in sand at the laboratory scale. Though increasing water content reduced the peak temperature and propagation velocity, a self-sustaining reaction was achieved in all cases from 0% to 75% water-filled porosity and 25% coal tar-filled porosity. This demonstrated the ability of an exothermic smouldering reaction to propagate itself and, with the excess energy generated, to sustain a water boiling/evaporation front ahead of the reaction. The propagation rate of the reaction (and thus the waste destruction rate) was shown to be linearly dependent on air injection rate across the range of air flow rates examined, which is expected for an oxygen-limited reaction (Dosanjh et al., 1987; Pironi et al., 2009; Schult et al., 1996). Smouldering was also shown to be sensitive to sand grain size and initial contaminant concentration; the reaction was non-self-sustaining for sands with average grain sizes greater than 10 mm and initial coal tar concentration lower than 25,000 mg/kg. These specific numbers are expected to be a function of experimental scale, with larger grain sizes and lower initial concentrations likely to be selfsustaining at larger scales (Switzer et al., 2014). This is because self-sustainability depends on a positive energy balance (heat generation minus heat losses) and increased scale means less relative heat loss to the external boundary due to lower surface area/volume ratio (Switzer et al., 2014). At a given scale, a boundary between self-sustaining and non-self-sustaining smouldering behavior depends on complex interactions between variables that affect heat generation (e.g., fuel energy content, oxygen supply), heat retention (e.g., sand and fuel heat capacities) and heat loss (e.g., volatile compounds, moisture content) and thus needs to be determined experimentally (Torero and Fernandez-Pello, 1996).

Smouldering research on materials similar to biosolids is largely limited to understanding the self-heating and fire hazards associated with their storage (e.g., (Chirag et al., 2011; Della Zassa et al., 2013; He et al., 2014)). The exception is a study on smouldering faeces for international development purposes by Yermán et al. (2015). That work explored the self-sustainability of upwards forward smouldering using surrogate-human faeces and dog faeces mixed with sand as a function of faeces moisture content, sand/faeces mass ratio, contaminant pack height, and airflow rate. This study showed that faeces with moisture content as high as 70% (wet mass basis) could achieve a self-sustaining smoulder in a 98 cm tall and 16 cm diameter fixed bed using 108 g/min airflow and a sand/faeces mass ratio of 4.25. The cool porous medium ahead of the reaction provides a condensation zone, which can be problematic for very wet fuels (Ohlemiller, 1985). Indeed, accumulated condensation ahead of the smouldering zone, resulting from evaporated/boiled water and water produced during oxidation, was shown to limit the pack height for a self-sustaining reaction as faeces moisture content increased (Yermán et al., 2015).

These findings suggest that self-sustaining smouldering may provide an attractive biosolids destruction option for WWTPs, as it may minimize energy input and biosolids processing (e.g., reducing the need for dewatering). Thirty experiments were completed to (i) demonstrate, for the first time, the potential for WWTP biosolids to be smouldered, (ii) map the parameter space (i.e., the biosolids moisture content and sand/biosolids mass ratio) in which biosolids will permit self-sustained smouldering, and (iii) understand the sensitivity of the reaction to sand/biosolids mixture energy content and air flux, a key operator parameter. This work aims to provide basic design information for considering smouldering combustion as an alternative biosolids disposal option.

2. Experimental procedure

Biosolids were obtained from Greenway Pollution Control Centre (Greenway) in London, Ontario, Canada. Greenway utilizes a typical wastewater treatment path, where primary clarification initially removes settleable solids (primary sludge). The waste activated sludge is settled out in secondary clarification following aerobic digestion, and is thickened in either dissolved air flotation units or rotating drum thickeners. The combined primary and waste activated sludge is dewatered via centrifugation with polymer addition, and the sampling point for all biosolids used in this study was located after this final processing step.

Biosolids initial moisture content (MC) and ash content, measured by EPA Method 1684 (U.S. EPA, 2001), and biosolids dry higher heating value (HHV_d^b) $\left(\frac{|\mathbf{y}|}{g}\right)$, measured using a bomb

Table 1 Biosolids Key Properties.

	Initial moisture content% (g_{water}/g_{total})	Ash content% (g_{ash}/g_{dry})	Dry higher heating value (kJ/g_{dry})		
Measured on Greenway samples	72.2, 79.9	18.6, 22.8	17.2, 18.1		
Literature comparison	70.0-90.0 ^a	17.8, 27.5 ^b	23.0–29.0° 20.0–23.0°		

^a Biosolids moisture content range following dewatering via centrifugation (Metcalf and Eddy, 2003).

calorimeter, are summarized in Table 1. The presented values represent the minimum and maximum average properties measured from four batches (19–120 L) gathered across 10 months. Each value in the first row of Table 1 is an average of at least three measurements from a batch that were all within 10% of the average. Note that the ash content represents (dry solids – volatile solids). Table 1 further demonstrates that the measured values compare well against values from literature. However, since the measured HHV_d^b range was slightly lower than typical, this suggests that the employed biosolids provided a conservative sample with respect to biosolids energy content (Metcalf and Eddy, 2003; WEF, 1988).

The biosolids were dried to <1% MC and stored in sealed 20 L pails in a 5 °C cold room, which successfully preserved the biosolids between the time of collection and experimentation. When preparing an experiment, the stored biosolids were manually crushed to a particle diameter near 1 cm and mechanically mixed (KSM7581MS, KitchenAid) with water to reach the desired biosolids MC. The biosolids re-wetted to the initial MC exhibited some qualitative differences from the virgin biosolids, such as less absorbed (i.e., interstitial, bound) water (Vesilind and Martel, 1990). This is likely due to irreversible changes in the biosolids particles' structures upon drying and rewetting (Vesilind and Martel, 1990). Several smouldering experiments were conducted with virgin biosolids to confirm that, despite any qualitative differences in the appearance of the fuel, the rewetted biosolids provided indistinguishable quantitative smouldering data as detailed in Section 3.2.

Column smouldering experiments followed established experimental procedures (Pironi et al., 2011; Switzer et al., 2009; Yermán et al., 2015), which are briefly reviewed here. Fig. 1 illustrates the experimental setup and data collection equipment. A 60 cm tall and 15 cm diameter stainless steel column rested upon a base component that housed a coiled resistive heater (450 W, 120 V, Watlow Ltd.) and an air injection manifold. Sixteen thermocouples (Type K, Omega Ltd.) inserted horizontally into the center of the column at 3.5 cm intervals along the column height were employed to track temperatures. The heater was connected to a 120 V AC, single-phase variable power supply (STACO Energy Products) and the entire column was wrapped in insulation (5 cm thick mineral wool, McMaster Carr) as is typical in smouldering studies to reduce external heat loss associated with the scale of the experimental system (Switzer et al., 2009). The apparatus rested on a mass balance (KCC150, Metler Toledo) to provide real-time mass loss. The emissions from a select number of experiments (presented in Rashwan (2015) and noted in Table 2) were analyzed for volume fractions of carbon monoxide, carbon dioxide, and oxygen using a gas analyzer (MGA3000C, ADC). The mass balance, thermocouples, and gas analyzer were connected to a data logger (Multifunction Switch/Measure Unit 34980A, Agilent Technologies) and personal computer, which logged all readings every two seconds.

Clean sand was packed into the apparatus base until just covering the air diffuser and heater. Coarse sand (Number 12, Bell & Mackenzie Co., mean grain diameter = 0.88 mm, coefficient of uniformity = 1.6) and biosolids, mechanically mixed in three batches, were packed in the column above the base to a height of 40 cm in three equal lifts. This was topped with 15 cm of clean sand. All experiments were subjected to a uniform ignition procedure. This started with a preheating phase in which the heater was turned on, with no airflow, until the first thermocouple (2 cm above the heating element) reached 200 °C. Air injection was then initiated, achieved by a mass flow controller (FMA5400/5500 Series, Omega Ltd.) connected to laboratory compressed air at ambient temperature. This introduction of oxygen initiated smouldering that caused

^b Ash content range from two samples of raw sewage sludge (Cui et al., 2006).

^c Primary (high range) and activated sludge (low range) higher heating value ranges (Metcalf and Eddy, 2003; WEF, 1988).

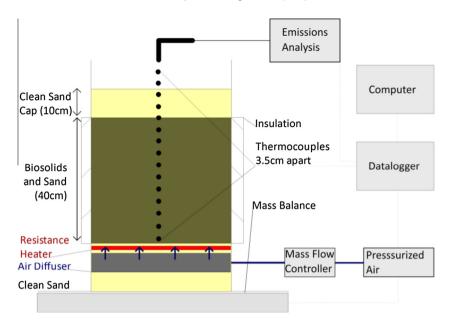


Fig. 1. Illustration of the experimental set up.

a spike in temperature at the first thermocouple, at which time the heater was turned off. Subsequent combustion was sustained by the energy released by the biosolids during upwards forward smouldering. Excluding heater inefficiency, the resistive heater provided a constant heat flux of approximately 408 W (3.4 A \times 120 V). The length of the preheating phase varied between 1 and 4 h depending on the biosolids MC and sand/biosolids mass ratio (S/B). The air remained on until the reaction extinguished and the entire column cooled to ambient temperatures.

summarizes the twenty-three experiments (Experiments No. 0-23, excluding 10) performed to map the self-sustaining smouldering parameter space using biosolids with initial MCs ranging from 74% to 85% and S/Bs from 1.5 to 23 g/g. The focus was on the most economical condition for biosolids treatment, where the least processed (i.e., the wettest) biosolids can be destroyed at the maximum organic destruction rate (i.e., using the least amount of sand). All experiments were classified as self-sustaining (SS), non-self-sustaining (NSS), or borderline self-sustaining (BSS) (further defined in Section 3.1). These experiments also served to quantify the changes in key combustion metrics (peak temperature, front propagation velocity, and mass loss rate) within this parameter space. These experiments all used the same sand and an injected air flux of 3.3 cm/s (volume of air at ambient temperature per unit total cross-sectional area of column per time, i.e., Darcy flux). Seven additional experiments (Experiments No. 10 and 24-29) were performed to examine the sensitivity of the process to air fluxes between 1.6 and 8.1 cm/s for biosolids at both 73% and 79% MC. In all cases, the reported combustion metrics were measured from the smouldering front passing through the middle 50% of the contaminant pack, between thermocouples (TCs) 3 and 9, as this region was unaffected by boundary effects (further discussed in Section 3.1). The smouldering front propagation velocities were calculated by averaging all local velocities (i.e., distance between successive thermocouples divided by time of arrival of the front), following the method of Pironi et al. (2009). Moving averages (heuristically chosen) of 2.7 and 1.7 min was applied to the mass loss data and the calculated mass loss rate, respectively, to reduce minor random noise in the mass balance data.

The energy content of the biosolids is a function of MC. The biosolids lower heating value (LHV^b) $\left(\frac{k!}{g}\right)$ was estimated (Boundy et al., 2011):

$$LHV^{b} = \left[\left(1 - \frac{MC}{100} \right) \cdot HHV_{d}^{b} \right] - \left[\frac{MC}{100} \cdot L_{\nu} \right] \tag{1}$$

where MC(%) is the biosolids MC and $L_{\nu}\left(\frac{|\mathbf{y}|}{g}\right)$ is the latent heat of vaporization of water, 2.447 kJ/g at 25 °C. The equation demonstrates that as the MC of the fuel increases, the energy content linearly decreases. The *LHV* calculation assumes: (1) complete combustion (i.e., full oxidation of carbon compounds to carbon dioxide and water vapor) (Washburn, 1933), and (2) none of the energy lost during water evaporation/boiling is recovered (Boundy et al., 2011). Neither of these assumptions is true for smouldering; some incomplete combustion is expected (Ohlemiller, 1985; Yermán et al., 2015) and some heat recovery from steam is achieved. Therefore, these values are qualitative, presented to quantify the relative change in energy content as biosolids MC changes.

The energy content of a unit mass of sand and biosolids mixture depends on the S/B in addition to the MC of the biosolids. This is quantified by defining an effective system lower heating value (LHV_s^s) :

$$LHV_e^s = \frac{LHV^b}{\left[1 + \frac{S}{B}\right]} \tag{2}$$

The equation reveals that as the amount of fuel dilution by sand increases, the effective energy content of the mixture linearly decreases. Note that LHV^b and LHV^s_e are bulk metrics considering each batch column experiment as a whole. However, it is acknowledged that individual smouldering column experiments are dynamic systems in which local properties such as MC, S/B, and reaction and condensation zone thicknesses vary in space and in time. Nevertheless, LHV^b and LHV^s_e are considered suitable metrics for the purpose of categorizing the differences in the bulk, static energy balance between batches.

Table 2 Input parameters and summarized results for all experiments.

No.	Input parameters							Results				
	Biosolids moisture content (%)		Biosolids lower heating value ¹ (kJ/g)	Sand/biosolids (g/g)		Effective system lower heating value ² (kJ/g)	Pack height	Darcy air flux (cm/s)	Self sustaining ?	Average front velocity ³ (cm/min)	Average peak temp. ³ (°C)	Average mass loss rate ³ (g/min)
	Initial	After preheating		Initial	After preheating		(cm)			±1.9%	±0.75%	±0.31%
0 ^a	74	73	3.0	4.5	4.7	0.52	40.5	3.3	SS	0.41	529	17
l ^b	74	-	2.8	4.5	_	0.51	34.5	3.3	SS	0.47	555	_
2	80	80	1.6	3.5	3.5	0.35	40.0	3.3	BSS	0.19	456	12
3	85	80	1.6	2.6	3.5	0.36	40.0	3.3	SS	0.21	481	13
4	74	74	2.9	4.5	4.6	0.51	40.0	3.3	SS	0.38	548	18
5	80	79	1.8	4.9	5.1	0.29	40.0	3.3	SS	0.27	470	10
6	85	83	0.9	5.2	5.8	0.13	40.0	3.3	NSS	=	_	=
7	74	72	3.2	23	25	0.12	40.0	3.3	NSS	=	_	=
8	74	72	3.2	9.9	11	0.28	40.0	3.3	SS	0.34	467	7
9 ^a	80	80	1.6	2.6	2.6	0.44	37.0	3.3	NSS	=	_	=
10 ^a	80	80	1.6	2.6	2.6	0.44	37.0	8.1	NSS	=	_	=
11	80	80	1.6	2.6	2.6	0.44	40.0	3.3	NSS	-	_	=.
12	85	84	0.8	6.0	6.5	0.11	38.5	3.3	NSS	-	_	-
13	85	82	1.1	6.5	7.7	0.13	37.5	3.3	NSS	-	_	-
14	85	83	0.9	7.5	8.3	0.10	36.0	3.3	NSS	-	_	-
15	74	72	3.2	2.0	2.2	1.00	40.5	3.3	SS	0.38	612	23
16	74	71	3.5	1.5	1.7	1.30	39.0	3.3	SS	0.25	657	25
17	83	80	1.6	3.5	4.1	0.31	35.0	3.3	SS	0.20	456	9
18	83	82	1.1	3.0	3.1	0.27	35.0	3.3	NSS	_	_	_
19	77	75	2.6	2.0	2.2	0.82	40.5	3.3	SS	0.19	500	15
20	77	77	2.2	1.5	1.5	0.87	39.0	3.3	NSS	_	_	_
21	83	83	1.0	4.0	4.0	0.19	35.5	3.3	NSS	_	_	_
22	77	77	2.2	2.5	2.5	0.62	37.5	3.3	BSS	0.16	462	12
23	80	76	2.4	3.5	4.2	0.46	40.0	3.3	SS	0.27	499	12
24 ^c	74	73	3.0	4.5	4.7	0.53	41.5	6.5	SS	0.62	569	25
25°	74	72	3.1	4.5	4.8	0.54	42.5	1.6	SS	0.23	481	11
26 ^c	83	78	1.9	3.5	4.5	0.35	38.0	6.5	SS	0.45	495	17
27 ^c	83	79	1.7	3.5	4.3	0.33	38.0	1.6	SS	0.18	391	6
28 ^c	83	80	1.5	3.5	4.0	0.29	36.0	3.3	SS	0.28	482	12
29 ^c	83	78	1.9	3.5	4.5	0.35	37.5	3.3	SS	0.26	441	10

¹ Calculated with Eq. (1). ² Calculated with Eq. (2).

a Virgin biosolids were used for comparative purposes.
b No mass loss data is available, the amount of lost water from preheating is assumed negligible.
c Experiments were analyzed for carbon monoxide, carbon dioxide, and oxygen volume fractions.

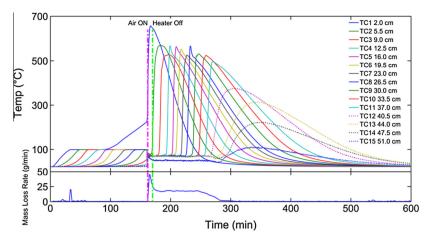


Fig. 2. Temperature and mass loss rate for Experiment No. 4, a self-sustaining smouldering experiment with 74% moisture content re-wetted biosolids in a fixed bed at a 4.6 g/g sand/biosolids mass ratio. The solid lines represent thermocouples within the contaminant pack.

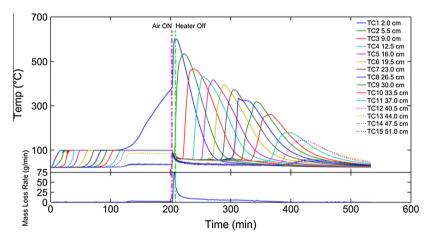


Fig. 3. Temperature and mass loss rate for Experiment No. 6, a non-self-sustaining smouldering experiment with 83% moisture content re-wetted biosolids in a fixed bed at a 5.8 g/g sand/biosolids mass ratio. The solid lines represent thermocouples within the contaminant pack.

The 'Initial' and 'After preheating' biosolids MC values provided in Table 2 quantify the loss of moisture from the reaction zone prior to ignition during the preheating phase. There were two main sources of losses: (1) evaporation/boiling out of the column top, and (2) downwards water migration into the base of the apparatus and below the heater. These quantities were estimated with the mass loss data: the mass loss observed during preheating was assumed to be water boiling, while the mass remaining after the reaction reached the top of the column (TC13) was assumed to be water remaining below the heater due to early downwards mobilization. A mass balance analysis presented in Rashwan (2015) indicates that these assumptions are robust and provide a conservative (i.e., low) estimate of the MC in the reaction zone as the front progresses. Table 2 reveals that overall the decrease in MC in the biosolids to be treated was 0-5%, with the higher values in the wettest S/B mixtures. It is noted that the mass of water below the heater could not be estimated by this method for the NSS and BSS experiments, because not all of the wet biosolids were removed from the reaction zone, so only evaporation/boiling was accounted for in those cases. In the Results and Discussion, all reported biosolids MC and S/B values are 'After preheating' values (i.e., corrected for moisture changes occurring after packing) unless specified as 'Initial'. In Table 2, every LHV^b and LHV^s is computed for 'After preheating' conditions.

3. Results and discussion

3.1. Definition of self-sustaining and non-self-sustaining experiments

Figs. 2 and 3 illustrate the temperature profile and mass loss rate over time from a SS and NSS experiment, respectively. Fig. 2 shows smouldering Experiment No. 4 (Table 2) with 74% MC biosolids mixed at 4.6 g/g S/B. Until 34 min, the data illustrates the water boiling out of the 2 cm region closest to the heater (TC1). The drying region (including both evaporation and boiling) lengthens until 90 min when the bottom 2 cm region dried and the temperature rose above 100 °C; when it reached 200 °C (162 min), the airflow was turned on. In this case, the water boiled during preheating did not leave the column but condensed in the upper, cooler regions of the column; this is inferred from the negligible rate of mass loss and the cooler temperatures in the upper regions of the column during the preheating period. The forced airflow ignited the biosolids nearest to the heater, as seen in TC1's sharp increase to 657 °C at 167 min, at which time the heater was turned off. The smouldering behavior in Fig. 2 is identified as SS due to (1) the consistent peak temperatures, here 548 °C, as the smouldering front propagates the full length of the column, and (2) the steady rate of mass loss, here 18 g/min in the central portion of the column. This represents the first ever demonstration of the selfsustaining smouldering of WWTP biosolids.

Consistent boundary effects hold for all SS smouldering column experiments. First, an atypically high peak temperature observed at TC1 (and corresponding high mass loss rate) is observed due to excess energy supplied from the heater. Second, a declining mass loss rate is observed at late time as, presumably, the smouldering front thins as it approaches the top of the contaminant pack. For these reasons, only the data between TC3 and TC9 was considered when quantifying the combustion metrics for all SS experiments. The minimal mass loss from boiling and water migration below the heater in this case means that the 'After preheating' MC is unchanged from 74% (see Table 2). The total mass loss indicated that 98% of the biosolids' volatile solids and water within the system were removed over the experiment's duration. The remaining mass is primarily trapped as condensed pyrolysate in the top sand cap, and as char remaining in a thin layer around the inside wall of the column due to boundary (external heat loss) effects. The remaining mass is not associated with remaining biosolids as the sand in the reaction zone was found to be completely clean and dry upon excavation; this is true for all of the SS smouldering experiments. The smouldering front propagation velocity was constant at 0.38 cm/min. TC12 and TC13 are in the clean sand cap and TC14 and TC15 are in the air-filled cavity above the sand; their low temperatures during the smouldering phase show the extent of energy capture and recycling by the unburned fuel. After the biosolids are destroyed, TC12 and TC13 reveal the forward propagation of the heat wave and convective cooling of the clean, hot sand by forced

In contrast, Fig. 3 illustrates a NSS reaction for Experiment No. 6 (Table 2) with 83% MC biosolids mixed at 5.8 g/g S/B. The preheating and ignition characteristics are similar to Fig. 2. The subsequent declining peak temperatures of 465-319 °C from TC3 to TC9 reveals insufficient energy released relative to the heat losses; here the heat losses are considerably increased due to almost doubling the water/dry biosolids mass ratio relative to Experiment 4 (74% MC) as discussed above. The mass loss rate correspondingly steadily declines from 9 to 5 g/min over this same region, providing additional evidence that the smouldering reaction is dying after ignition. The decrease in the initial magnitude of the mass loss rate relative to Experiment No. 4 is due to a decrease in smouldering reaction robustness and an increase in sand dilution from 4.6 g/g to 5.8 g/g. Clearly, biosolids can be smouldered in a self-sustaining manner under some conditions and not others; the approximate bounding limits within this parameter space is mapped out in Section 3.4 below.

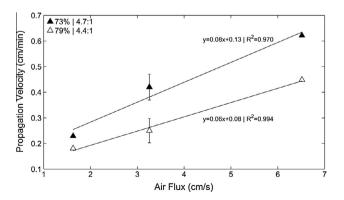


Fig. 4. Average smouldering propagation velocity as a function of air flux for 73% moisture content biosolids in a fixed bed at a 4.7 g/g sand/biosolids mass ratio (black triangles) and for 79% moisture content biosolids in a fixed bed at a 4.4 g/g sand/biosolids mass ratio (white triangles). The error bars indicate the 95% confidence intervals from three repeat experiments.

3.2. Comparison between rewetted biosolids and virgin biosolids and repeatability

Comparing re-wetted to virgin biosolids experiments indicates that the employed method of re-wetting biosolids does not significantly affect the propagation of a SS reaction, as all of the combustion metrics differed less than 6% (see Experiments No. 0, 1, and 4 in Table 2). However, when comparing conditions that result in NSS reactions, re-wetted biosolids led to more rapid decay of the reaction than did virgin biosolids (data presented in Rashwan (2015)). This may result from the increased downward migration of water toward the smouldering front; recall that re-wetted biosolids had a much higher fraction of free (i.e., mobile) water. Therefore, using re-wetted biosolids in place of virgin biosolids provides meaningful results that are nearly identical to those obtained from virgin biosolids, though re-wetted biosolids likely provide a conservative (i.e., lower MC) estimate of the smouldering bounding limits.

To assess the experimental variability amongst SS experiments, three repeat experiments were completed far from the bounding limits (73% MC biosolids at 4.7 g/g S/B) and near the bounding limits (79% MC biosolids at 4.4 g/g S/B). These experiments provided a mean smouldering propagation rate of $0.42\pm12\%$ (95% CI) and $0.25\pm19\%$ cm/min, respectively, and a mean peak temperature of $544\pm2.8\%$ and $460\pm5.1\%$ °C, respectively. These low CI values on the mean peak temperature are typical of all SS experiments in this work, underscoring that the peak temperatures are very consistent for self-sustained biosolids smouldering and that the mean is representative of the experiment as a whole. These experiments further demonstrate good repeatability and suggest that changes observed between experiments outside of these uncertainties are the result of intentionally varied experimental parameters.

3.3. Smouldering propagation velocity sensitivity to air flux

The smouldering combustion's sensitivity to varying air flux was explored as it can be considered a key operator control parameter for the mass destruction rate and will aid in full scale system design. Fig. 4 presents the smouldering propagation velocity as a function of air flux far from the bounding limits (73% MC biosolids, 4.7 g/g S/B) and near the bounding limits (79% MC biosolids, 4.4 g/g S/B). The smouldering front velocity in the 73% MC experiments increased from 0.23 to 0.62 cm/min (2.7 times increase) resulting from increasing the air flux from 1.6 to 6.5 cm/s (4.1 times increase). Over the same increase in air flux, the 79% MC experiments exhibited slower front velocities from 0.18 to 0.45 cm/min (2.5 times increase).

A linear relationship was also observed for smouldering of contaminated soil over a similar range of air fluxes at a comparable scale (Pironi et al., 2009). This suggests that the reaction is oxygen limited, which is typical for smouldering, over the employed air flux range (Dosanjh et al., 1987; Schult et al., 1996).

3.4. Mapping the self-sustaining smouldering parameter space

Mapping the parameter space in which biosolids facilitate a SS smouldering reaction is important for determining the maximum destruction rate possible and minimum amount of sludge predrying necessary. This information is essential in gauging the economic potential of smouldering biosolids. In Fig. 5, the vertical axis plots biosolids MC while the horizontal axis considers S/B, as these represent two key system metrics to be optimized. The parameter space map distinguishes between SS (green, square symbols) and NSS (red, triangle symbols) experiments. Beige indicates the parameter space that encompasses self-sustaining conditions while white indicates non-self-sustaining conditions; the region

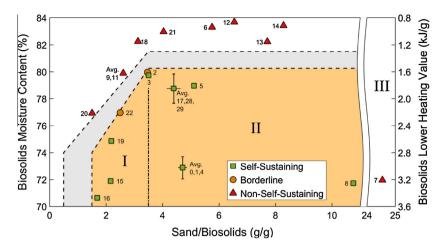


Fig. 5. Parameter space illustrating the biosolids moisture content, biosolids lower heating value, and sand/biosolids mass ratio (S/B) combinations that facilitate self-sustained smouldering at a Darcy air flux of 3.3 cm/s. The error bars denote 95% confidence intervals obtained from three repeat experiments. All relevant experiments are numbered (see Table 2) and the three S/B regions, I, II, and III, are further discussed in the text. The beige region encompasses self-sustaining conditions, the white region is non-self-sustaining conditions, and the gray region is the area of uncertainty between the two.

of uncertainty between these two, shaded gray, identifies the bounding limits for self-sustained smouldering at the column scale.

Fig. 5 reveals, first, that biosolids with a MC above 70% and up to 80% can be successfully smouldered in a SS manner. It is noted that even incineration of biosolids was determined to infeasible above a moisture content of 70% (Roy et al., 2011). Therefore, this is a novel, important result, the consequences of which are further discussed in subsequent sections. Second, it is apparent that adding sand facilitates that ability to smoulder biosolids in a self-sustaining manner. Fig. 5 reveals that SS smouldering is possible over a wide range of S/B ratios and that the chosen S/B can influence the biosolids MC that can be smouldered.

To aid analysis and discussion, the parameter space is divided into three regions. In Region I, where there is minimal dilution of the fuel with sand (S/B < 3.5 g/g), the results indicate that a linear increase in biosolids MC can be smouldered with increasing S/B. Three possible reasons for this are: (1) higher S/B means less biosolids filling the sand pore space, which increases the surface area available for smouldering combustion; (2) increased permeability to air, which decreases the pressure drop across the column and decreases air pore velocities thus increasing the oxygen residence time through the reaction zone; and (3) increased effective heat capacity (sand plus biosolids) in the drying zone, which promotes heat retention and mitigates heat losses ahead of the smouldering front. These hypotheses are not tested in this work and remain open questions for future study.

In Region II, where fuel dilution by sand is intermediate (3.5 g/g < S/B < 11 g/g), a MC limit of approximately 80% biosolids MC is observed above which SS smouldering is not possible regardless of further increases in the amount of sand (Fig. 5). The LHV^b – which is constant for a given MC and independent of S/B (see Eq. (1); see right vertical axis of Fig. 5) – appears to mark the SS/NSS divide in this region. Greater than 80% MC corresponds to an LHV^b less than 1.6 kJ/g; for comparison purposes, the LHV of wood with 10–60% MC is 17–8.4 kJ/g, respectively, and for charcoal with 1–10% MC is 32–25 kJ/g, respectively (Quaak et al., 1999). It is expected that the heat losses in the system, primarily due to the large fraction of water when biosolids MC > 80%, exceeds the energy produced during smouldering. However, Fig. 5 reveals that SS smouldering is possible over a large range in this region for MC < 80%, with S/B as high as 11 g/g.

Region III is characterized by high fuel dilution by sand (S/ B > 11 g/g). The transition between Regions II and III was explored

with only two experiments – Experiments 7 and 8 (Table 2) – since it is far from the optimal region for commercial treatment of biosolids. For 72% MC biosolids, it was found that increasing the S/B from 11 to 25 g/g shifted from SS to NSS smouldering, corresponding to a drop in LHV_e^s from 0.28 to 0.12 kJ/g, respectively. The high S/B limit is likely governed by a minimum system energy content value in this range, which is a function of the heat losses at a given scale of experimentation (Switzer et al., 2014; Yermán et al., 2015). It is important to note that the LHV_e^s and LHV^b , which correspond to critical thresholds of SS smouldering in Regions II and III, are not alone able to predict the SS/NNS divide in Region I, as neither account for the complexity of interrelated factors discussed above that cause the reaction to decay when the dilution by sand is minimal.

Two BSS experiments (borderline SS; orange, circle symbols) are included on the SS boundary in Region I. These experiments exhibited SS smouldering through >65% of the contaminant pack, but abruptly extinguished leaving un-burnt, wet biosolids above the corresponding height. It is suspected that these BSS experiments were sensitive to excessive condensed moisture accumulating ahead of the smouldering front; similar causes of extinction at late time were observed for smouldering of wet faeces in tall columns (Yermán et al., 2015). This means the lower part of the column was in the SS region of the parameter space but the MC conditions were changing in the upper part of the column causing the local system to leave the SS region, leading the reaction to decay. It is interesting to note the close proximity of SS and BSS Experiments Nos. 3 and 2, respectively. This suggests that the boundary defining the limit of SS behavior is likely sensitive to small amounts of experimental variability at this scale (e.g., column packing, water redistribution, preheating time, etc.).

A potential financially optimal condition for operating a biosolids smouldering system is observed at the top of the dashed black line marking the distinction between Regions I and II in Fig. 5: biosolids with a MC of 80% within a S/B of the mixture of 3.5 g/g (i.e., in the vicinity of Experiment No. 3). Minimizing the amount of sand results in less fuel dilution and thus maximizes the rate of biosolids destruction. Maximizing MC is significant because this reduces requirements for (i) extensive processing (e.g., dewatering, thickening, pre-drying) or (ii) supplemental fuel for disposal in incinerators, and it is these costs that often dominate the disposal operating cost (Murakami et al., 2009; Werther and Ogada, 1999).

It is noted that this financially optimal location in Fig. 5, as well as the region boundaries identified in the figure, are not absolute.

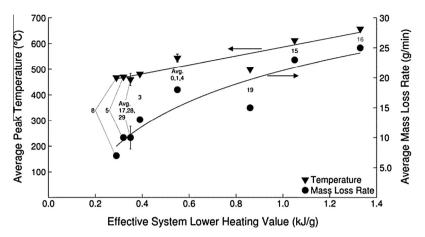


Fig. 6. The average peak temperatures (triangles) and average mass loss rate (circles) from self-sustaining experiments versus effective system lower heating value. All experiments employed a Darcy air flux of 3.3 cm/s. The error bars indicate the 95% confidence intervals from three repeat experiments and all experiments are numbered (see Table 2) for further discussion in the text.

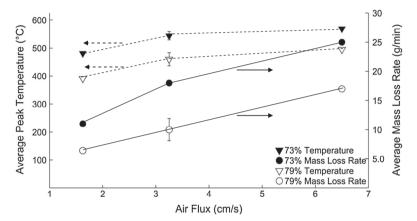


Fig. 7. The average peak temperatures (triangles) and average mass loss rate (circles) from self-sustaining experiments far from the bounding limits (73% MC biosolids, 4.7 g/g S/B) (solid) and near the bounding limits (79% MC biosolids, 4.4 g/g S/B) (open) resulting from increasing Darcy air flux. The error bars indicate the 95% confidence intervals from three repeat experiments.

These limits are specific to the system variables employed, specifically: the scale of experiment, the sand grain size and distribution, the airflow injection rate, and the biosolids properties. It is expected that the limits would shift in response to changing system variables. For example, it is known that larger scale systems are more energy efficient because the surface area-to-volume ratio of the reactor is reduced, which reduces heat losses (Switzer et al., 2014). Thus a larger smouldering system may shift the boundary between Region I and II upwards and to the left, since reduced heat losses would allow wetter biosolids to smoulder for the same energy content. The most representative process limits and optimal system conditions should be determined at the pilot scale for a given application (biosolids and sand). Furthermore, it is noted that the boundary between Regions I and II represents both a theoretical and a practical limit on smouldering, since as MC is increased the energy balance is shifted further toward extinction and the boundary identifies where this balance tips. However, the boundary between Regions II and III is only a practical limit and not a theoretical limit; theoretically, there is no limit on how much sand dilution can occur since the energy dilution could be compensated for by other factors (e.g., increased airflow rate). Despite all these qualifications, we expect that the optimal location and boundary limits identified in Fig. 5 are (a) conservative, because of the previously discussed experimental assumptions and that this small scale exhibits maximum heat losses (Switzer et al., 2014), and (b) broadly representative of those expected for smouldering of biosolids. Overall, these findings provide an important starting place to support further research and up-scaling, and reflects the important benefits smouldering combustion can provide due to its energy efficiency.

3.5. Smouldering robustness among self-sustaining experiments

While Section 3.4 separated all experiments into SS, BSS, and NSS cases, here we discuss the robustness of the smouldering reaction amongst the SS experiments. This is explored by comparing the mass loss rate and average peak temperatures as a function of both LHV_e^s and air flux in Figs. 6 and 7, respectively. These two parameters are of practical importance as they can be monitored in a WWTP and be altered to control the smouldering process.

The SS reactions that are most robust, or farthest from the bounding limits, are expected to exhibit the highest average peak temperatures and highest mass loss rates (Yermán et al., 2015).

Recall that LHV_e^s is a measure of the energy content of the fuel/sand mixture and depends on both biosolids MC and S/B. Fig. 6 shows that increasing the LHV_e^s , by decreasing biosolids MC and/or decreasing S/B, linearly increased the average peak temperatures (y = 175x + 412, $R^2 = 0.850$) and logarithmically increased mass loss rates (y = 10.2ln(x) + 21.2, $R^2 = 0.871$) for SS experiments. This is due to excess energy driving hotter reactions that, in turn, support faster reaction kinetics. These trends are clearest amongst the SS experiments that lie furthest from the low S/B bounding limits, as defined in Fig. 5 and Section 3.4 above (i.e.,

Experiments No. 0, 1, 4, 8, 15, and 16). The remaining SS experiments (i.e., Experiments No. 3, 5, 17, 19, 28, and 29) are near the low S/B bounding limits (i.e., near the black dashed lines from Region I and II in Fig. 5) and exhibit some scatter around the trends in Fig. 6. Overall, it is observed that within the SS set of experiments, there is considerable variability in smouldering metrics. Nearly a fourfold increase in biosolids mass destruction rate is possible, all other operational factors being equal, by maximizing the LHVs of the sand/biosolids mixture.

In Fig. 7, an increase in smouldering robustness as a function of air flux is observed for the two SS cases, one far from the bounding limits (Experiments No. 0, 1,4, 24, 25; Table 2) and one near the bounding limits (Experiments No. 17, 26, 27, 28, 29; Table 2). For the 73% MC experiments (far from the bounding limits), increasing the air flux from 1.6 to 6.5 cm/s increased the average mass loss rate and average peak temperature from 11 g/min and 481 °C to 25 g/min and 569 °C, respectively. For the 79% MC experiments (near the bounding limits), the same increase in air flux produced lower mass loss rates and average peak temperatures from 6.4 g/ min and 391 °C to 17 g/min and 495 °C, respectively. The increase in robustness with air flux, especially the logarithmic increase in temperature, is largely due to the increase in the oxygen concentration in the bulk pore space in the reaction zone and associated increase in oxygen flux diffusing into the smouldering fuel surface (Dosanjh et al., 1987; Leach et al., 2000; Yermán et al., 2015). For both cases, the emissions' carbon monoxide/carbon dioxide volume ratio was found to decrease with increasing air flux, indicating more complete combustion and providing further evidence of increased robustness (for details see Rashwan, 2015).

This raises the question of to what degree the SS/NSS distinctions identified in Fig. 5 are a function of the fixed air flux of 3.3 cm/s used in those experiments. Increasing the air flux from 3.3 to 8.1 cm/s did not affect the NSS result in Experiments Nos. 9 and 10 (80% biosolids MC and 2.6 g/g S/B, Table 2). As these experiments were quite close to the low S/B bounding limits in Fig. 5, it is concluded that – at least at this scale of experimentation - increasing the air flux will likely not overcome the heat losses in many NSS smouldering cases limiting the ability to significantly stretch the SS parameter space toward the left (i.e., further into the more economical S/B region). Conversely, there is a minimum air flux required for SS smouldering, which, from the SS results in Fig. 7, is lower than 1.6 cm/s for mixtures either near or far from the bounding limits identified in Fig. 5. For comparison, Pironi et al. (2011) found SS smouldering achievable in coal tar and sand using an air flux at least 0.5 cm/s and Yermán et al. (2015) found the minimum air flux was a function of sand/faeces mass ratio (S/F). where a SS reaction was achieved with 0.74 cm/s when the S/F was at least 3.75 g/g. The minimum air flux has not been explored in this study as it represents an uneconomical condition (i.e., minimum mass destruction rate from Fig. 7).

This gradation in SS smouldering robustness has important implications when considering the applicability of smouldering as a practical process for WWTPs. Generally, biosolids experience temporal variability in properties due to plant conditions, seasonality, weather conditions, etc. (King and Painter, 1986). This variability is expected to shift the sand/biosolids mixture properties (i.e., LHV^b and LHV^s_e) with respect to the bounding limits. Regular monitoring of biosolids key properties (e.g., Table 1) would assist in (a) ensuring the system is in the SS region where it is sufficiently robust and can withstand variations, and (b) balancing proximity to the bounding limits, where the system is most economical (minimum sand, maximum destruction rate). Manipulating the S/B or the amount of biosolids pre-drying may be cumbersome in response to property changes. Manipulating the air flux may be the most straightforward tool for increasing the smouldering's

robustness near the bounding limits. However, the limits of this approach need to be fully understood as there may eventually be diminishing returns or an upper limit on the benefit that can be achieved from further increasing the air flow (Sato and Sega, 1985). It is noted that the column scale is the most challenging for testing smouldering, since the surface area to volume ratio – and thus heat losses – are maximized (Switzer et al., 2014). This means the results presented are likely highly conservative relative to a full scale application. In other words, the SS/NSS boundary identified in Fig. 5 may represent robust smouldering conditions at full scale application where heat losses are minimized. For these reasons, optimization of the system operation and boundaries of the parameter space for treatment at a WWTP should be done at the pilot or full field scale.

4. Summary and conclusions

Experimental results indicate that biosolids with moisture content as high as 80%, with a lower heating value greater than 1.6 kJ/ kg, may be successfully smouldered in a self-sustaining manner. This is significant as WWTP disposal methods often require expensive dewatering, thickening, and conditioning processes to reduce the biosolids moisture content significantly below this value prior to disposal. The biosolids need to be mixed within a fixed sand bed, where sand/biosolids mass ratios from 1.7 to 11 g/g lead to selfsustaining smouldering, even with effective energy contents of the sand/biosolids mixtures as low as 0.28 kJ/g. With a sand/biosolids mass ratio between 1.7 and 3.5 g/g, increasing biosolids moisture content from 73% to 80% was possible by linearly increasing the amount of diluting sand. Above a sand/biosolids ratio of 3.5 g/g, the upper biosolids moisture content limit for smouldering (80%) was independent of the amount of sand added. The exception was an upper limit on the amount of sand that can be added, with smouldering dying when the effective energy content of the sand/fuel mixture was diluted to 0.12 kJ/g at a sand/biosolids mass ratio of 25 g/g. These limits are specific to the employed experimental system but are expected to be conservative and broadly representative.

The robustness of the self-sustaining smouldering reaction, as identified by peak temperatures and mass destruction rates, is lowest near the identified bounding limits. However, increasing the effective energy content of the sand/biosolids mixtures (e.g., by lowering biosolids moisture content, by decreasing the sand/biosolids ratio) and/or increasing the applied air flux both improve smouldering robustness. Increasing air flux also proportionally increased the smouldering front propagation velocity. As air flux can be easily varied by the system operator, this presents a means for controlling the mass destruction rate.

This work suggests that smouldering destruction presents significant potential as a new, low energy, on-site alternative disposal method for biosolids. As it appears to be able to (i) reduce biosolids pre-treatment (particularly energy intensive drying) and (ii) minimize biosolids disposal costs, it may provide WWTP operating cost savings. This is a new, beneficial environmental application of smouldering in addition to destroying liquid industrial waste and faeces treatment for sanitation as previously reported. However, further study is required before it can be fully exploited or its cost/benefit properly evaluated. For example, exploration into the characteristics of the resulting ash, emissions analysis and treatment, and examining how smouldering is affected by scale of the reactor would be beneficial. Additional research into energy recovery (e.g., from the emissions, from the hot sand) may provide additional value. Most of these issues would be best addressed in the implementation of a pilot scale smouldering reactor operated on site at a WWTP.

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