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# Carbon Distribution and Multi-criteria Decision Analysis of Flexible Waste Biomass Smouldering Processing Technologies

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## Abstract

Waste biomass treatment is a globally urgent matter which highly relates to environmental quality and human health. Here, a flexible suite of smouldering-based waste biomass processing technologies is developed and four processing strategies: (a) full smouldering, (b) partial smouldering, (c) full smouldering with a flame, and (d) partial smouldering with a flame, are proposed. The gaseous, liquid, and solid products of each strategy are quantified under various airflow rates. Then, a multi-criteria analysis in terms of environmental impact, carbon sequestration, waste removal efficiency, and by-product value is performed. The results show that full smouldering achieves the highest removal efficiency but generates significant greenhouse and toxic gases. Partial smouldering effectively generates stable biochar, sequesters over 30% carbon, and therefore reduces the greenhouse gases to the atmosphere. By applying a self-sustained flame, the toxic gases are significantly reduced to clean smouldering emissions. Finally, the process of partial smouldering with a flame is recommended to process the waste biomass that can sequester more carbon as biochar, minimize carbon emissions and mitigate the pollution. And the process of full smouldering with a flame is preferred to maximally reduce the waste volume with minimum environmental impact. This work enriches strategies for carbon sequestration and environmentally friendly waste biomass processing technologies.

**Keywords:** waste biomass removal; smouldering combustion; carbon sequestration; biochar; greenhouse gases

## 1. Introduction

With the growth of population and economy, the quantity of municipal solid waste (MSW) generated has increased significantly over the last decades, which endangers the environment and contributes to climate change. Waste biomass is a significant component of MSW, which typically includes kitchen waste, wood waste, grass clippings, paper waste, etc. At present, landfilling, biological conversion (composting, anaerobic digestion, fermentation), and thermochemical treatment (pyrolysis, liquefaction, gasification, combustion) are the main ways of biowaste disposal (Basu, 2010; Lohri et al., 2017). Often, landfilling and incineration are still very common in many countries as they are the easiest and cheapest ways (Assamoi and Lawryshyn, 2012). For example, Australia deposited nearly 50% of the core organic solid waste in landfill in 2020 (Department of Agriculture, 2020). Landfilling releases large amount of greenhouse gases (GHG) (IPCC, 2006), of which $CH_4$ (a more potent GHG than $CO_2$) can reach almost 60 % and accounts for 10-15 % of the global anthropogenic $CH_4$ emissions (Gómez-Sanabria et al., 2022). Combustion (or burning/incineration) is most effective in the volume reduction of waste and can produce substantial heat for energy recovery (Lu et al., 2017). However, it releases large quantities of $CO_2$, particulate matters (PM), and air pollutants (especially the dioxins) (Lin et al., 2018; Ravindra et al., 2019). According to EEA report (European Environment Agency, 2015), 150 Tg/yr of $CO_2$ eq were emitted from MSW incineration in 2015 Therefore, nowadays, effective and environmentally friendly treatment facilities to cope with the large quantities of biowaste are still in absence, posing a globally big challenge to mankind.

Smouldering is an emerging method for organic solid waste removal, especially for those with high moisture content, like the biowaste. Smouldering is slow, low-temperature and flameless burning of porous fuels (Ohlemiller, 1991; Rein, 2014) which often occurs on the burning of charring materials, such as wood (Tissari et al., 2008), coal (Wu et al., 2015), and peat (Huang and Rein, 2016a). Different from flaming combustion, smouldering occurs when oxygen directly attack the hot surface of reactive porous media (Ohlemiller, 1991; Rein, 2014). Generally, the characteristic temperature (500-800°C) and heat of combustion (~10 MJ/kg) of smouldering are smaller than those of a flame (Rein, 2014), and it can be easily initiated by a relatively small amount of energy and sustain the process in extreme conditions such as poor oxygen supply (~12 % $O_2$) and large fuel moisture content (MC) (>100%) (Huang and Rein, 2016a). The overall smouldering combustion process can be approximated as two lumped chemical pathways, namely pyrolysis and char oxidation, as described in Eq. (1-2) (Huang and Rein, 2016b). Pyrolysis generates flammable pyrolyzates and char,

$$\text{Fuel (s)} + \text{Heat} \rightarrow \text{Pyrolyzates (g)} + \text{Char (s)} \qquad \text{[pyrolysis] (1)}$$

where both can be further oxidized. The heterogeneous char oxidation leads to smouldering as

$$\text{Char (s)} + O_2 \rightarrow \text{Heat} + CO_2 + H_2O + \text{other gases} + \text{Ash (s)} \qquad \text{[smouldering] (2a)}$$

The homogeneous gas-phase oxidation of pyrolyzates results in a flame as

$$\text{Pyrolyzates (g)} + O_2 \xrightarrow{\text{flame}} \text{Heat} + CO_2 + H_2O + \text{other gases} \qquad \text{[flaming] (2b)}$$

Over the past few decades, many existing investigations and practices have revealed the ability of smouldering to remove organic wastes like bioliquid, faeces (Fabris et al., 2017; Yermán et al., 2015), wastewater sludges (Feng et al., 2021), and food wastes (Song et al., 2023, 2022). Such a method demonstrates many attractive advantages, like a safer operation process, minimising pre-treatments (e.g., drying and grinding) (Rashwan et al., 2016; Yermán et al., 2015), the applicability for wastes with high moisture contents (MC), and minimum extra energy input.

In addition, smouldering also shows strong potential as a green waste-to-energy technology, which has been applied to produce biochar (Rashwan et al., 2021; Rein, 2008) and bio-oil (Feng et al., 2022; Yerman et al., 2017). This is because a smouldering front contains pyrolysis reaction (Eq. (1)), and pyrolysis is an endothermic process that has been widely used to convert biomass feedstock into biochar (solid), bio-oil (liquid), and bio-gas (Babu, 2008; Demirbas and Arin, 2002; Wang et al., 2010). Biochar has potential usage in a wide field and is suitable for carbon sequestration due to its resistance to chemical and biological decomposition. Converting biomass to biochar avoids the complete return of greenhouse gases (GHG) to the atmosphere, compared to natural decay or burning processes (Lee et al., 2020; Woolf et al., 2010). However, the traditional pyrolysis technology usually requires additional energy to keep the high temperature (typically >300℃). In comparison, pyrolysis in smouldering process can be sustained by the heat generated from the weak oxidation of original fuel, thus requires no external energy or fuel gas. This feature makes smouldering a more energy-efficient way to transform waste to energy than pyrolysis and suitable for on-site applications.

However, as an incomplete burning process, smouldering tends to release many toxic emissions, like CO, $CH_4$, $NO_x$, volatile organic compounds (VOCs) and particulate matters (PM) (Hu et al., 2018; Raza et al., 2023), posing severe threats to human health and the environment. This limits its further promotion and application in waste management. Considering there are still substantial flammable hydrocarbons and CO in smouldering emissions, we intend to use a flame to clean the smouldering emissions by converting most of the flammable and pollutant species into $H_2O$ and $CO_2$. A flame (or flare) is a common method of disposal of unwanted waste gases in oil and gas industries (Akeredolu and Sonibare, 1998). Generally, it is often used to burn off the C1-C6 hydrocarbons and hydrogen sulfide from the upstream and downstream of oil industry (Gai et al., 2020). Besides, it has also been utilized to remove the landfill gases and biogases (mainly $CH_4$) in landfill management (Environmental Protection Agency, 1997). Sometimes, auxiliary fuel must be introduced to support the flame if the waste gas does not meet the minimum heating value (Environmental Protection Agency, 1995). More importantly, it has been reported that if operated properly, the combustion efficiency of the flare could reach 99%, eliminating most of the flammable waste gas (Gai et al., 2020; Pohl et al., 1986).

Our previous research established a novel combustion method for biowaste disposal by using a self-sustained flame (without additional fuels or heat) to purify the emissions from biowaste smouldering under an appropriate air supply (Chen et al., 2022a, 2022c, 2022b). After being ignited from the top surface of the fuel bed, a two-stage smouldering process was observed under an upward airflow, as illustrated in Fig. 1. The 1st stage is opposed smouldering (the front spreads downward), leaving abundant biochar. The 2nd stage is forward smouldering (the front spreads upwards), which burns all the char and leaves little ash. More importantly, it was found that the flame could only be piloted and sustained in the 1st stage because of the intensive pyrolysis in this stage.

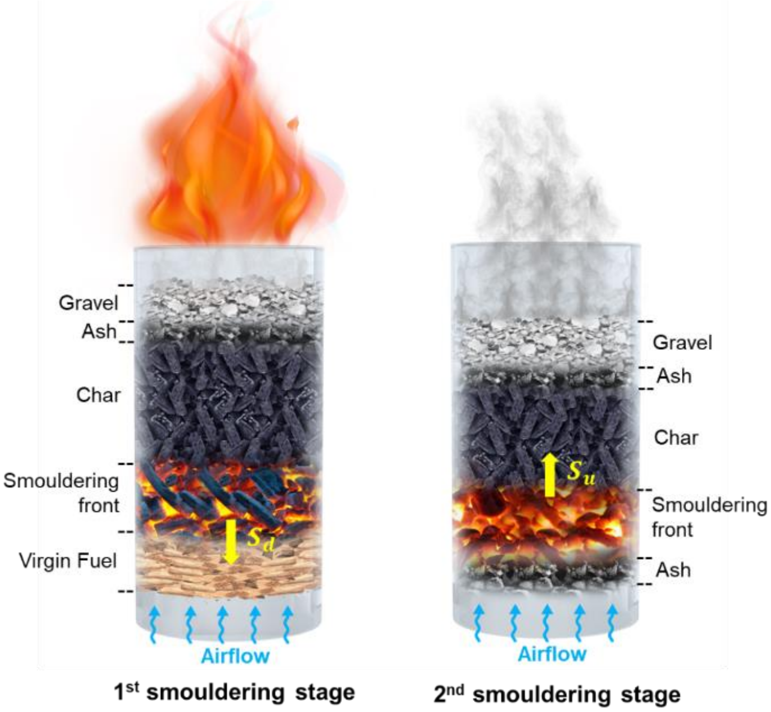


**Fig. 1.** The two-stage smouldering process with a flame sustained by the emissions from the 1st stage opposed smouldering.

Therefore, based on the two-stage smouldering process, this paper further develops a flexible suite of smouldering waste biomass processing technologies and proposes four different processing strategies. Each strategy is conducted with the wood waste under various airflow rates (6-18 mm/s). The carbon balance of each process is analysed after quantifying their gaseous, liquid, and solid products. A multi-criteria decision analysis based on PROMETHEE-GAIA algorithm is made in terms of their environmental impact, carbon sequestration, waste removal efficiency, and product value.

## 2. Materials and Methods

### 2.1 Wood waste sample

Wood (or yard) waste is a common waste biomass all over the world. For example, Hong Kong annually produces about 40,000 tonnes of wood wastes that are directly sent to landfills, bringing a huge burden to the limited land resources, and generating large amounts of GHG. Same as our previous work (Chen et al., 2022a, 2022b), the wood waste was chosen in this experiment (Fig. 2), provided by a local company (ECO-Greentech Ltd.). The particle size of the wood chips ranges from 20 mm to 30 mm, with an average of 25 mm. The dry bulk density, solid density, and porosity were measured to be 210 ± 10 kg/m$^3$, 600 ± 20 kg/m$^3$, and 0.65, respectively. The element analysis of wood waste sample shows 47.7, 6.3, 45.4, and 0.5 % mass fractions for C, H, O, and N, respectively. And the proximate analysis shows 78.4, 17.0, and 4.6 % mass fraction for volatile, fixed carbon, and ash, respectively.

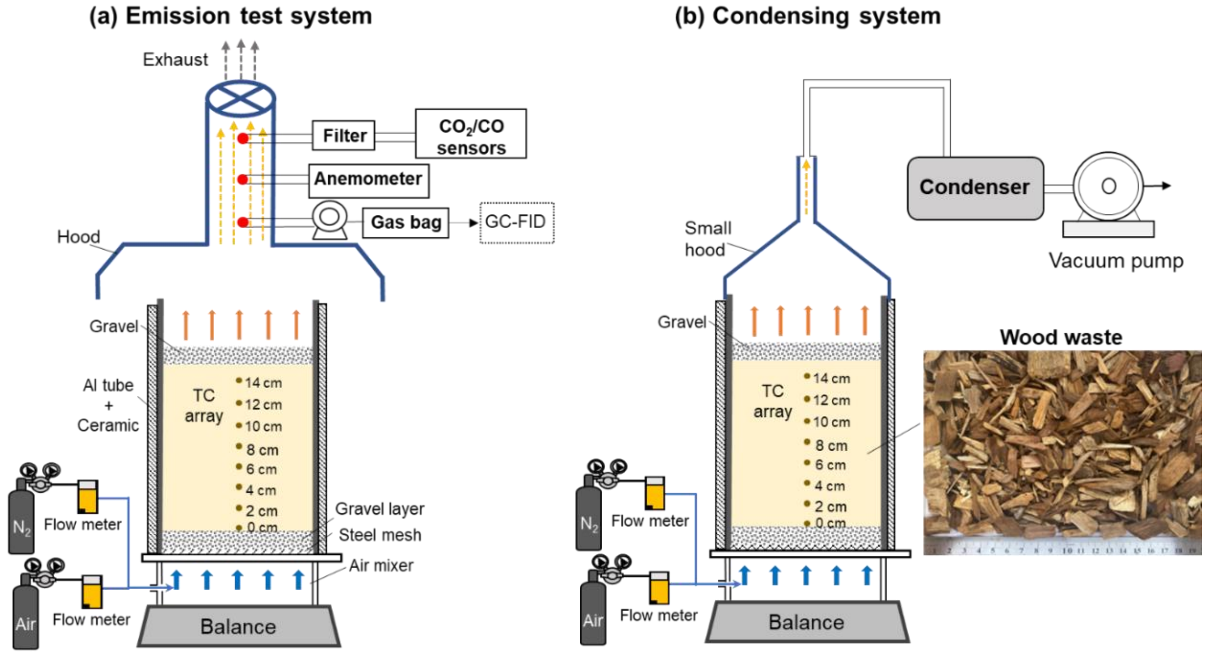


**Fig. 2.** Experimental setup for (a) emission test and (b) direct condensing.

Before the test, the raw wood chips were thoroughly dried in an oven at 90 ℃ for 48 h, and their moisture contents were measured to be <8% on a dry basis when reaching a new equilibrium with ambient moisture. Thermal analysis for the wood samples was conducted with a PerkinElmer STA 6000, and the results are same as our previous work (Chen et al., 2022a).

## 2.2 Experimental setup

The wood waste was burnt in an open-top cylindrical reactor with a depth of 20 cm and an internal diameter of 14 cm (Fig. 2). More detailed information about the burner could be found in our previous work (Chen et al., 2022c). For each test, a wood waste sample with a controlled mass of 410 ± 5 g was fed into the reactor at a constant height of 15 cm. An array of eight K-type thermocouples were inserted into the fuel along the axis from 0 cm (bottom) to 14 cm (top) with an interval of 2 cm to monitor the temperature and trace the position of the smouldering front. A lighter was used to initiate the smouldering of the wood waste and ignite the smouldering emissions after the smouldering ignition. A forced airflow was supplied from the bottom end of the reactor and the airflow rate was controlled by a flow meter. A fume hood was located directly above the reactor to collect and transport all the emissions.

### 2.3 Processing strategies and experimental procedure

As shown in Fig. 3, four processing strategies are proposed and can be achieved by ceasing the smouldering process at different stages.

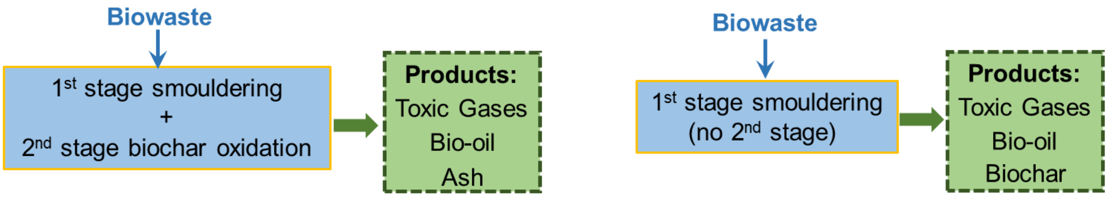
(a) Full smouldering (F-SM) includes the 1st-stage opposed propagation (strong pyrolysis and weak char oxidation) and the 2nd-stage forward propagation (strong char oxidation).

(b) Partial smouldering (P-SM) includes the 1st-stage opposed propagation (strong pyrolysis and weak char oxidation) only, after which smouldering is quenched with biochar left.

(c) Full smouldering plus flaming (F-SM+FL) upgrades the F-SM by applying a self-sustained flame to purity the smouldering emissions during the 1st stage. No bio-oil is generated from the F-SM+FL.

(d) Partial smouldering plus flaming (P-SM+FL) also upgrades the P-SM by applying a self-sustained flame to purity the smouldering emissions. No bio-oil is generated, but biochar is left.

(a) Full Smouldering (F-SM) (b) Partial Smouldering (P-SM)



(c) Full Smoldering + Flaming (F-SM+FL) (d) Partial Smouldering + Flaming (P-SM+FL)

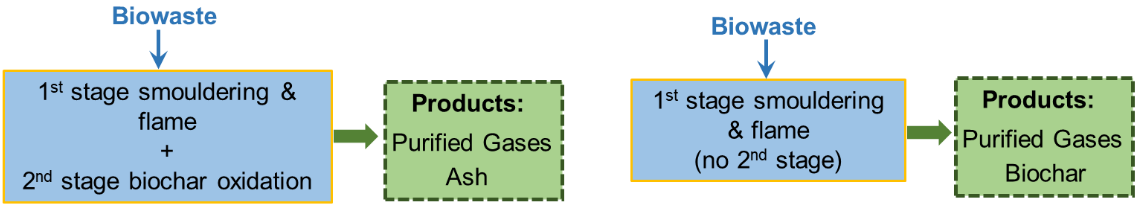


**Fig. 3.** Different strategies for the waste biomass smouldering process.

Initially, a lighter was used to initiate the smouldering combustion from the top surface of the fuel bed, and the ignition time was fixed for three minutes for each experiment. After successful smouldering ignition, a forced airflow was supplied from the bottom of the reactor, and the airflow rate was controlled by a flow meter. To control the airflow rate to smouldering front, we used airflow velocities ($u$) from 6 mm/s to 18 mm/s, where the emissions from wood waste smouldering could be ignited as a self-sustained flame, as verified in the previous experiments (Chen et al., 2022c).

To achieve different processing strategies, experimental procedure differs from each other. For the full-smouldering (F-SM), smouldering was sustained until the wood waste was completely converted into a small amount of ash after two stages. For the partial-smouldering (P-SM), smouldering was ceased after the 1st stage (when the last TC (0 cm) reached its first peak, or when the flame disappeared, and the smoke became clearly less) by supplying sufficient $N_2$ from the bottom. The whole process lasted until all TCs dropped to the room temperature, leaving abundant biochar in the reactor. Furthermore, for the processes with a flame (F-SM+FL and P-SM+FL), the smouldering emissions were ignited using the lighter in the 1st stage. For the processes without a flame (F-SM and P-SM), the smouldering emissions from 1st stage were condensed via the condenser shown in Fig. 2b.

All products (gas emissions, condensed liquid, and solid residual) from each experiment were further analyzed. And for each case, at least two repeating tests were carried out to ensure the repeatability of the experiments.

## 2.4 Product analysis

### *2.4.1 Gas products*

To quantify the emissions from different smouldering-driven processing strategies, a gas collection and measurement system was built, as shown in Fig. 2a. The emissions were entirely collected using a fume extraction hood located above the reactor. Three measuring points were designed in the centreline of the hood duct. The flow rate in the test point was measured by an anemometer (Testo 405i), which was constant at 0.038 ± 0.002 $m^3/s$ during the tests. Two major carbon-containing gas species were measured using portable gas analysers after the particulate matter was removed via a quartz filter. Specifically, carbon dioxide ($CO_2$) was measured by SKY6000-M2 Gas Analyzer and carbon monoxide (CO) was measured by Testo 300. Meanwhile, the emissions were also collected using a 10-L gas bag every 10 minutes to quantify the hydrocarbons ($C_xH_y$(g)) using Gas Chromatography Plus Flame Ionization Detection (GC-FID).

### *2.4.2 Liquid products*

As a pyrolysis-dominant process, significant liquid (called bio-oil) including the condensable organic compounds (tar) and water is generated from the 1st downward-propagation stage (Neves et al., 2011). Thus, a condensation system was built to quantify the liquid product from the 1st stage, as shown in Fig. 2b. The pyrolysis emission from the opposed propagation was collected via a hood entirely covering the top outlet of the burner. Then, the vapour was passed through a long pipe and finally condensed in the ice-cooled trappers. The mass of the bio-oil produced was obtained by weighing the trappers.

### *2.4.3 Solid products*

After each experiment (when all TCs drop to room temperature), the total solid residue was collected, and its mass was measured via an electrical balance. To quantify the carbon content and stability of the solid residue, all solid samples were further analysed by elemental and thermal analysis. Before analysis, all samples were milled to a homogenous fine powder and dried at 90 °C for 48 h.

Elemental analysis was carried out using PerkinElmer Elemental Analyzer. Moreover, for all biochar samples, H:C and O:C molar ratios were calculated and plotted on a van Krevelen diagram to estimate their stability, where H:C is an indicator of the condensation degree and O:C reflects the oxidation degree (Santin et al., 2020).

Moreover, thermal analysis of biochar was conducted with a PerkinElmer STA 6000 Simultaneous Thermal Analyzer in an air atmosphere at a heating rate of 15 K/min. The recalcitrance index $R_{50}$, another important parameter to evaluate the char stability, was calculated from the TG results as (Harvey et al., 2012):

$$R_{50} = T_{50,char}/T_{50,graphite} \tag{3}$$

where $T_{50,char}$ is the temperature value at which 50% of the mass of the char sample is lost, and $T_{50,graphite}$ is the temperature value at which 50% of the total mass of graphite is lost, which is given as 823 °C (Harvey et al., 2012). The recalcitrance/carbon sequestration potential of the studied material can be classified as Class A: $R_{50}<0.50$; Class B: $0.50 \le R_{50}<0.70$; Class C: $R_{50} \ge 0.70$, where higher $R_{50}$ represents better carbon sequestration potential (Harvey et al., 2012).

## 2.5 Carbon balance calculations

The carbon balance of each strategy calculated in this study includes the total carbon in permanent gaseous emissions, solid residues, and liquid products (tar). For each smouldering-driven processing strategy, 410 g of wood waste with the carbon content ($X_{C,F}$) of 47.7 % is provided. Thus, the total carbon of the fuel ($m_{C,F}$) is:

$$m_{C,F} = m_F \cdot X_{C,F} = 410 \times 47.7\% = 195.6 \text{ g} \tag{4}$$

The carbon from the gaseous emissions ($m_{C,G}$) is defined as a sum of the carbon content in the major gas species as

$$m_{C,G} = \sum_j m_j \cdot X_{C,j} \tag{5}$$

where $m_j$ is the mass of gas species *j* (g), $X_{C,j}$ is the carbon molar mass fraction in species *j* (%), and subscript *j* indicates the major carbon-containing gases, e.g., $CO_2$, CO, $CH_4$, $C_2H_4$, and $C_2H_6$. The gas mass can be calculated as

$$m_j = \int_0^t \rho_j [j](t) \dot{V} \times 10^{-3} \tag{6}$$

where $\rho_j$ is the density of species *j* calculated based on the assumptions of the ideal gas law (kg/m$^3$), $[j](t)$ is the real-time concentration of the species *j* (ppm) (subtracting species' background concentration), $\dot{V}$ is the volume flow rate in the duct (m$^3$/min), and *t* is the total processing time (min). A representative concentration evolution of the gas species is shown in Fig. S3 in Supplemental files.

The carbon from the solid residue ($m_{C,S}$) is calculated as

$$m_{C,S} = m_S \cdot X_{C,S} \tag{7}$$

where $m_S$ is the mass of the solid residue (g), which is directly measured via a balance after the smouldering process, $X_{C,S}$ is the mass fraction of carbon content of the burnt residue (%), which is determined via the elemental analysis.

The carbon inside the liquid product tar ($m_{C,T}$) is calculated as

$$m_{C,T} = m_T \cdot X_{C,T} = (m_L - m_W) \cdot X_{C,T} \tag{8}$$

where $m_T$ is the mass of the tar (g), $m_L$ is the mass of the condensed liquid (g) and $m_W$ is the mass of the condensed water (g), $X_{C,T}$ is the carbon mass fraction of the tar (%). In this paper, the average value of $m_W$ is about 30 % $m_L$ (Xiu and Shahbazi, 2012). $X_{C,T}$ is estimated from its relationship with $X_{C,F}$ according to previous research (Neves et al., 2011), where $X_{C,T}$=1.14$X_{C,F}$. As a result, $X_{C,T}$ is calculated as 54.4 % here.

### 2.6 PROMETHEE-GAIA algorithm

Preference Ranking Organization Method for Enrichment Evaluations and Geometrical Analysis for Interactive Aid (PROMETHEE-GAIA is) a kind of multicriteria decision analysis (MCDA) algorithm and has been proved to be a useful approach in environmental applications (Behzadian et al., 2010; Surawski et al., 2013). It facilitates a rational decision-making process, which is achieved by virtue of a decision vector that directs the decision maker toward “preferred” solutions (Brans and Mareschal, 1994). A description on the principle of PROMETHEE-GAIA algorithm is provided in the Supplemental files.

This study applies the PROMETHEE-GAIA algorithm to the proposed four processing strategies under five different airflow velocities (totally 20 conditions) to decide the most preferred condition (alternative) for achieving different goals by considering seven criteria. A full listing of the abbreviations used for alternatives and criteria, along with information on how each criterion is treated by the PROMETHEE-GAIA algorithm is provided in Table S3-S6.

## 3. Results and Discussion

### 3.1 Smouldering temperature

Fig. 4 exhibits the smouldering temperatures of the full smouldering process and the partial smouldering process at a representative airflow velocity of 14 mm/s. It should be noted that the flame and the smouldering are separated by a gravel layer, so the flame above will not affect the smouldering temperatures. As expected, there are two separate characteristic peaks in Fig. 4a which correspond to the two-stage propagation of full smouldering process. The peak temperature of the 1st stage ($T_{\max,1}$) is generally lower than that of the 2nd stage ($T_{\max,2}$) because the endothermic fuel pyrolysis dominates in the 1st stage while the exothermic char oxidation dominates the 2nd stage. Comparatively, for the partial smouldering process, there is only one peak in Fig. 4b of which the value is the same as $T_{\max,1}$ in Fig. 4a. Once the bottom thermocouple (0 cm) reached its maximum, the air supply was replaced by the pure nitrogen ($N_2$). Afterwards, the smouldering temperature starts to decline to room temperature under the cooling effect of the supplied $N_2$. The summary of $T_{\max1}$ and $T_{\max2}$ under various airflow velocities is shown in Fig. S4. In general, $T_{\max2}$ is larger than $T_{\max1}$, and both $T_{\max1}$ and $T_{\max2}$ increase with the increased airflow velocity because of stronger char oxidation.

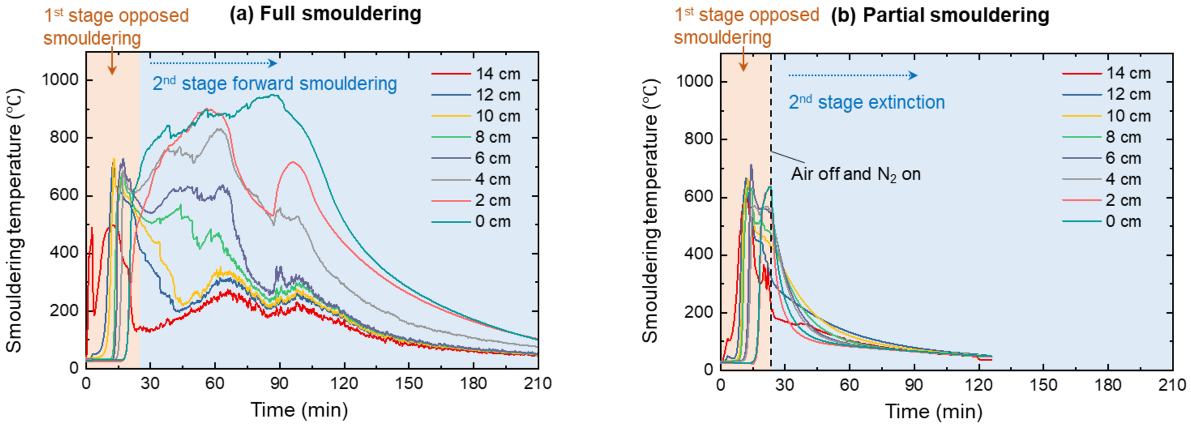


**Fig. 4.** Smouldering temperature evolution of (a) full smouldering process and (b) partial smouldering process at *u*=14 mm/s.

### 3.2 Carbon distribution

Different processing strategies will cause a considerable distinction in the carbon distribution in different by-products (gas, liquid, and solid), thus leading to different environmental impacts. The carbon balance of each strategy is calculated according to the method presented above. As a result, the by-products yields and corresponding carbon distributions are illustrated in Table S1.

Fig. 5 shows the mass fraction of carbon ($Y_C$, %) in the gas, liquid, and solid products from different strategies. Here, $Y_{\Delta C}$ (carbon difference) for all the tests are lower than 20%, which is acceptable for carbon balance research (Dufour et al., 2009). As exhibited in Fig. 5a, after the full smouldering process (F-SM), about 70 % of the carbon is distributed into the gaseous product, with about 20% into the liquid product and less than 5% into the solid residue (ash). This means that most of the carbon is released into the atmosphere after F-SM, which can be a detriment in view of its effect of accelerating global warming. Further, if a flame is applied (Fig. 5c), the carbon in gas will increase to over 90% because tar is mostly consumed by the flame, and its carbon distribution is insensitive to the airflow velocity within the tested range.

Regarding the partial smouldering, Fig. 5(b, d) show that over 30% carbon is fixed in the solid residue (biochar), and this carbon fraction decreases with the increased airflow velocity. In contrast, the carbon in the gaseous products from partial smouldering increases with the airflow velocity. Specifically, as the airflow velocity increases from 6 mm/s to 8 mm/s, the carbon fraction in gas from P-SM and P-SM+FL increases from 16 % and 21 % to 34 % and 49 %, respectively. This is because a larger airflow velocity strengthens the oxidation process existing in the 1st stage, and more heat is released to decompose more organic material into volatile matters through pyrolysis. In addition, only processes without flame can generate liquid (mainly water and condensable organic compounds), and the carbon fraction in liquid increases with the airflow velocity.

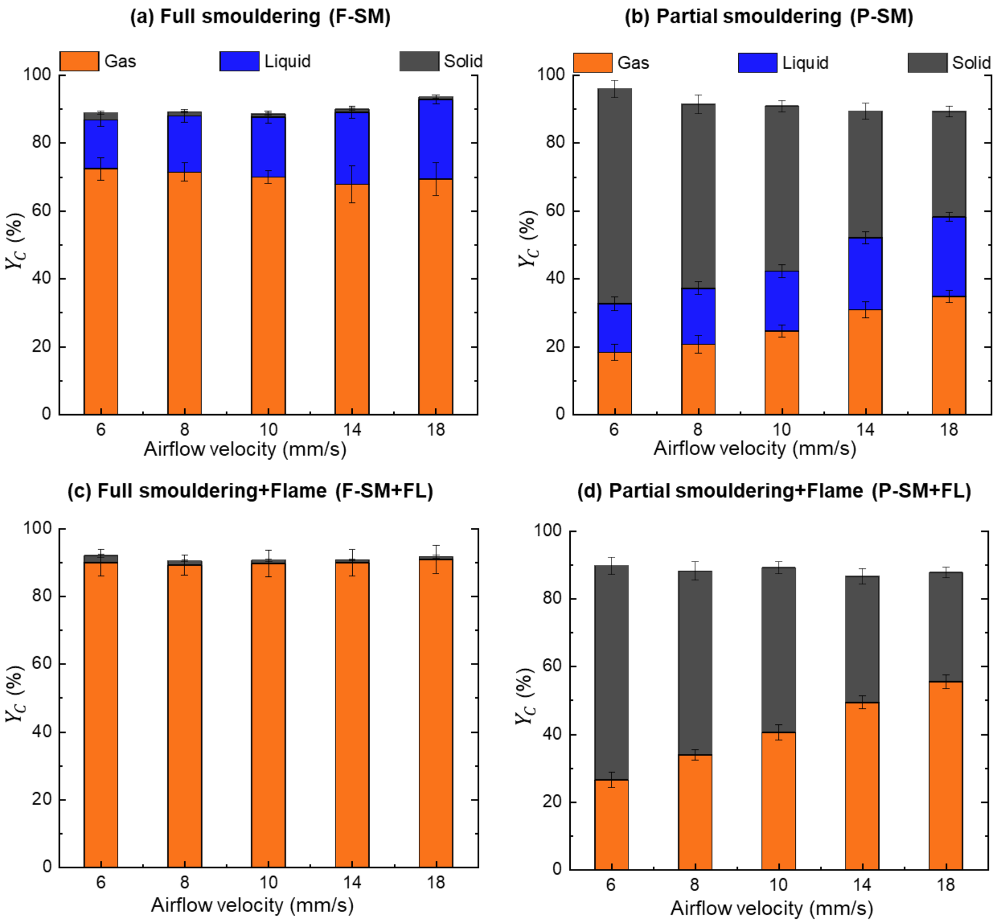


**Fig. 5.** Carbon fraction in gas, liquid, and solid products under various airflow velocities for four different strategies, (a) full smouldering (F-SM), (b) partial smouldering (P-SM), (c) full smouldering plus flame (F-SM+FL), and (d) partial smouldering plus flame (P-SM+FL).

In a short summary, such a thermochemical conversion process has significant effects on carbon partitioning in the products. Full smouldering will release much more carbon into the atmosphere, while partial smouldering can effectively generate biochar and achieve carbon sequestration. Therefore, by regulating the processing strategies and controlling the supplied airflow, different objectives can be achieved. Besides the distribution of carbon in different forms of products, the properties of the products are also very important as their characteristics determine their environmental impacts and their potential usage (Lehmann et al., 2021). Therefore, a detailed discussion of the properties of the products from each processing strategy is given in the following sections with a focus on the gaseous and solid products.

### 3.3 Gas-phase emissions characteristics

In this work, five major carbon-containing gas species are measured, that is, carbon dioxide ($CO_2$), carbon monoxide (CO), methane ($CH_4$), ethylene ($C_2H_4$), and ethane ($C_2H_6$). Among these gases, $CO_2$ and $CH_4$ are well-known greenhouse gases (GHG), which contribute significantly to global warming

(Wang et al., 2014). CO is the second main compound emitted from any combustion process of biomass, which is toxic and may cause poisoning deaths in humans (Manisalidis et al., 2020). In terms of the gaseous hydrocarbons ($C_xH_y$(g)), the mass of $CH_4$ is the largest, while the masses of $C_2H_4$ and $C_2H_6$ are much less and similar.

To characterize the environmental impacts of the emissions from different strategies and various airflow velocities, we quantify four widely used parameters in environmental research, that is, the $CO/CO_2$ mass ratio, $CH_4/CO_2$ mass ratio, $C_xH_y$ (g)/$CO_2$ mass ratio, and equivalent GHG (Fig. 6). Fig. 6a shows that the $CO/CO_2$ ratio decreases with the airflow velocity for all processes. The reason is that the combustion is more complete under larger airflow (sufficient oxygen supply). In addition, it is observed that the $CO/CO_2$ ratios of processes with flames (F-SM+FL and P-SM+FL) are all lower than 0.1, which is significantly smaller than those without flame. This proves that the flame could effectively reduce the toxic CO in smouldering emissions, agreeing well with previous work (Chen et al., 2022c).

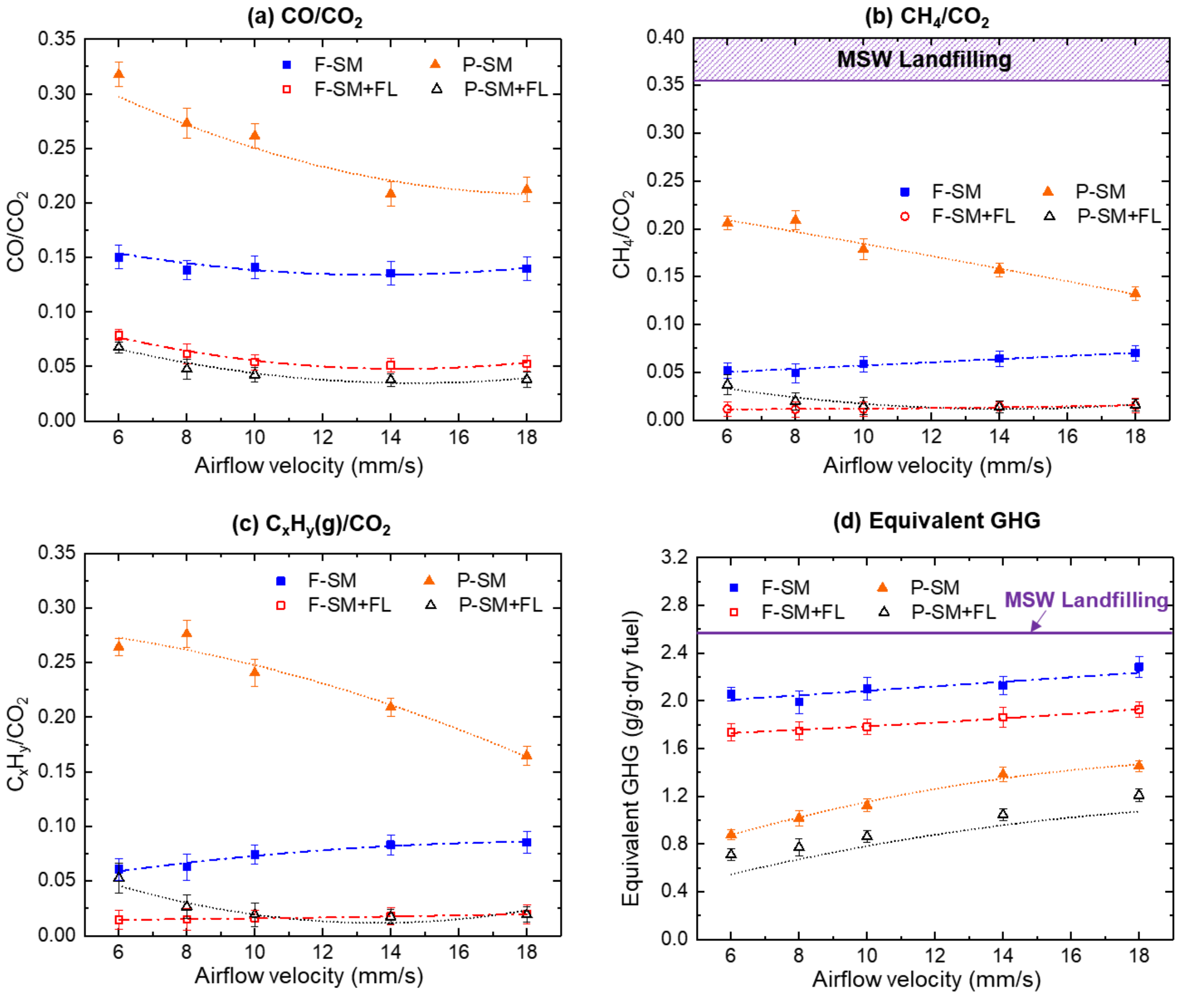


**Fig. 6.** (a) $CO/CO_2$ ratio, (b) $CH_4/CO_2$ ratio, (c) $C_xH_y$(g)/$CO_2$ ratio, and (d) equivalent GHG of different processing strategies under various airflow velocities, where the shadow area indicates the emission parameters of treating MSW via landfill in literature.

Fig. 6(b, c) show $CH_4/CO_2$ ratio and $C_xH_y$ (g)/$CO_2$ ratio respectively, which are very close as $CH_4$ accounts for the majority. For the gases emitted from landfilling of MSW, $CH_4$ accounts for a primary component with the $CH_4/CO_2$ mass ratio usually larger than 0.36 (0.36-0.80) (Davis et al., 2022; Hilger and Humer, 2003; Krause et al., 2016; Valencia et al., 2009). In comparison, the $CH_4/CO_2$ ratios of all the four smouldering processes are significantly smaller than that of the MSW landfilling (see Fig. 6b).

Global warming caused by GHG emissions has attracted worldwide attention. Each GHG has a different global warming potential (GWP) and persists for different lengths of time in the atmosphere (Buss et al., 2022). Equivalent GHG is a parameter that converts all greenhouse gas emissions into $CO_2$ equivalents so they can be correctly compared in terms of their greenhouse impacts (Lee et al., 2020). In this work, the GHG from each process is simplified as a mixture of $CO_2$ and $CH_4$ (the amount $N_2O$ is very small and is ignored here), and the equivalent GHG ($m_{GHG}$, g/g· dry fuel) is estimated as

$$m_{GHG} = \frac{GWP_{CO_2} \cdot m_{CO_2} + GWP_{CH_4} \cdot m_{CH_4}}{GWP_{CO_2} \cdot m_F} \tag{9}$$

where *GWP* refers the 100-year global warming potential of the species. $CO_2$ is taken as the reference gas and given a 100-year *GWP* of 1, and the 100-year $GWP_{CH_4}$ is given as 25 here (IPCC, 2006). $m_{CO_2}$ and $m_{CH_4}$ are the total mass of $CO_2$ and $CH_4$ produced, respectively. $m_F$ = 410 g is the mass of fuel.

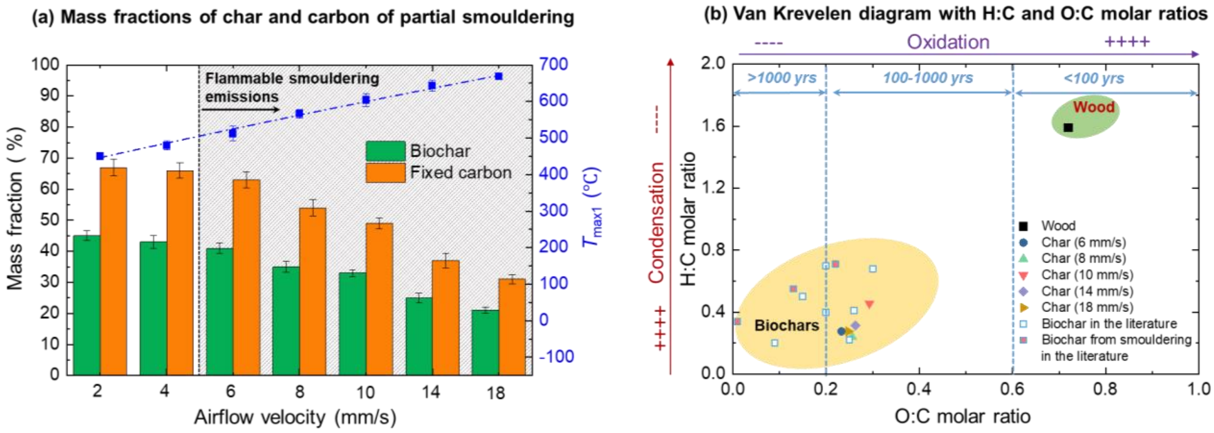
The calculated results of the equivalent GHG ($m_{GHG}$) are shown and compared with that from MSW landfilling (Lou and Nair, 2009) in Fig. 6d. Various theoretical and experimental investigations have suggested that a large variation of GHG emitted from 1 ton of waste landfilling (Ayalon et al., 2000; Humer and Lechner, 1999; Themelis and Ulloa, 2007). For comparison, we take an average 1.28 ton $CO_{2\text{-e}}$/ ton of waste generated from landfills (Kumar and Sharma, 2014; Lou and Nair, 2009) and assume that the moisture content of the waste is 50%, then 2.56 ton $CO_{2\text{-e}}$/ ton dry waste is obtained. As shown in Fig. 6d, the equivalent GHG ($m_{GHG}$) emitted from the proposed waste processing strategies is basically lower than that of waste landfilling, especially for the partial smouldering processes. Moreover, $m_{GHG}$ increases as the airflow velocity increases for all processing strategies. For example, $m_{GHG}$ of F-SM increases from 2 to 2.3 g/(g dry fuel) as the airflow velocity increases from 6 mm/s to 18 mm/s. Besides, $m_{GHG}$ from partial smouldering plus flame (P-SM+FL) is the smallest, followed by that from P-SM, F-SM+FL and F-SM. Specifically, $m_{GHG}$ from P-SM+FL is about 30 % smaller than that from P-SM, 55 % smaller than that from F-SM+FL, 60 % smaller than that from F-SM, and 67 % smaller than that from landfilling.

Therefore, on the one hand, the process of partial smouldering plus self-sustained flame can effectively reduce the global warming effect and toxicity degree of emissions. On the other hand, the supplied airflow velocity is much better to be regulated at a moderate degree (~6-10 mm/s), where not only the emissions are cleaner with lower $CO/CO_2$ and $CH_4/CO_2$ ratios, but also generating less greenhouse effect with a lower $m_{GHG}$.

### 3.4 Char yield and stability

The partial smouldering process can generate abundant biochar, which is a solid product of biomass pyrolysis and is a promising concept for climate change mitigation and adaptation (Woolf et al., 2010). In this section, the char yield and its stability from each process are investigated. Note that the char produced from processes P-SM and P-SM+FL are the same because their pyrolysis conditions are same under the same airflow supply.

Fig. 7a shows the char yield as well as carbon mass fraction fixed in char of the partial smouldering process under various airflow velocities. Two smaller airflow velocities (2 and 4 mm/s) were also measured here to show the carbon sequestration potential of the proposed partial smouldering process under smaller airflow velocities. The blue symbols in Fig. 7a indicate the average peak smouldering temperature of the 1st smouldering stage.


(a) Mass fractions of char and carbon of partial smouldering

(b) Van Krevelen diagram with H:C and O:C molar ratios

(c) TGA results for char samples

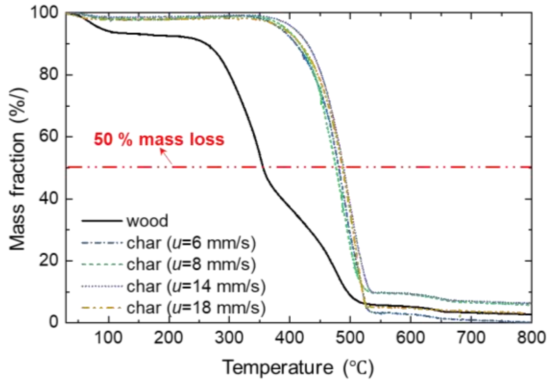


**Fig. 7.** (a) Mass fractions of char and carbon from the partial smouldering process with average $T_{max1}$ marked, where the shadow region indicates the smouldering emissions is flammable enough to sustain a flame, (b) Van Krevelen diagram with H:C and O:C molar ratios, where the half-lives range in the X-axis is defined according to Spokas (Spokas, 2010), and (c) TGA results for char samples under various airflow velocities.

As suggested, both the char yield and fixed carbon fraction decrease as the airflow velocity is increased. Specifically, when the airflow velocity decreases from 18 mm/s to 2 mm/s, the char yield increases from 21 % to 45 % and the fixed carbon fraction increases from 31 % to 66 %. This is because more organic material inside the fuel will be decomposed into volatile material and released to the atmosphere under higher temperatures ($T_{\max1}$) (Tripathi et al., 2016), leaving less char and fixing less carbon. Moreover, prior research (Wyn et al., 2020) has shown that under the internal airflow velocities of 8 mm/s and 2 mm/, the yields of charcoal from forward smouldering are 40 % and 53 %, respectively, which are slightly larger than the yields obtained in this work under the same airflow velocities. This is because the charcoal yields reported in the literature only account for the fuel in close proximity to the airflow supply, while this work calculates the biochar yield for the whole fuel bed.

Char stability is an essential property which demonstrates the longevity of stored carbon and therefore establishes an effective means for carbon abatement (Crombie et al., 2013). In this work, the stability of the produced biochar is assessed by two methods: O:C and H:C molar ratios (Santin et al., 2020), and recalcitrance index $R_{50}$ (Harvey et al., 2012). The O:C and H:C ratios are plotted in Fig. 7b (elemental analysis results are shown in Table S2). For comparison, the H:C and O:C molar ratios of biochar from smouldering and other thermal conversions in literature are also summarized in Fig. 7b (Crombie et al., 2013; Kambo and Dutta, 2015; Santin et al., 2020). The finding is that the char samples produced from partial smouldering have similar condensation ($0.2<\text{H:C}<0.5$) and oxidation ($0.2<\text{O:C}<0.3$) degrees. In comparison, the charcoal from forward smouldering under the airflow velocity of 8 mm/s in the literature has H:C and O:C ratio ranging from 0.01 to 0.23, and 0.35 to 0.71 respectively (Wyn et al., 2020). The relatively low O:C ratio indicates that the biochar obtained in this work can be preserved in the environment over long timescales with half-lives of over 100 years. $R_{50}$ of wood and biochar are calculated from their TG curves (Fig. 7c) according to Eq. (3). It is found that $R_{50}$ of the char from partial smouldering is similar under all airflow velocities, which is about 0.6, larger than that of the virgin wood (~0.4). According to the classification (Harvey et al., 2012), the char produced belongs to Class B ($0.5 \leq R_{50} < 0.7$), which has intermediate carbon sequestration potentials. In this way, we prove that the proposed partial smouldering is a good biowaste disposal method for long-term carbon sequestration from both char yield and char stability.

### 3.5 Multi-criteria analysis

A multi-criteria decision analysis based on PROMETHEE-GAIA algorithm is conducted, which analyses three scenarios, and each scenario considers seven criteria. Specifically, the seven criteria include emission of $CO_2$, CO, $CH_4$, $C_2H_4$, $C_2H_6$, and the amount of fixed carbon and produced tar. Three scenarios indicate maximum carbon sequestration with minimum environmental impact (Scenario (a)), maximum removal efficiency with minimum environmental impact (Scenario (b)), and maximum by-product value with minimum environmental impact (Scenario (c)).The outranking result for each scenario is thoroughly listed in Fig. S5, which involves ranking the alternatives from most preferred to least preferred based on the value of their net outranking flow (Φ).

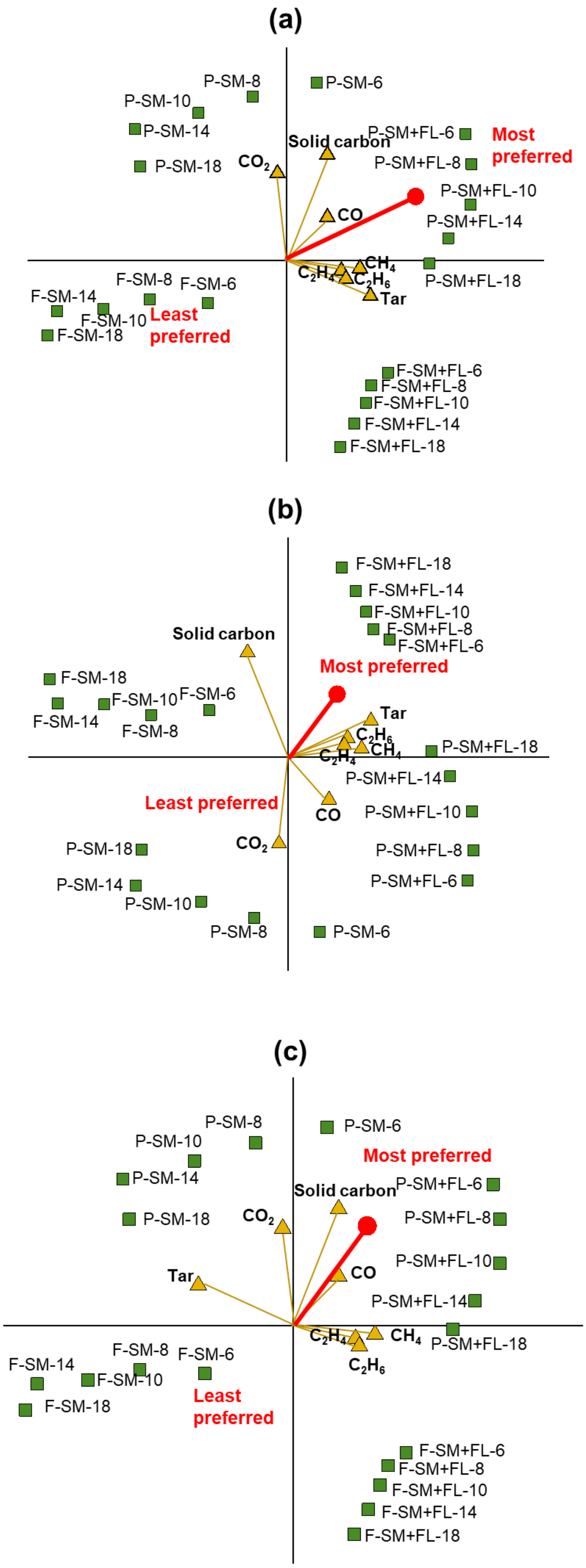


**Fig. 8.** GAIA plot of alternatives and criteria with the decision vector for different scenarios: (a) maximum carbon sequestration with minimum environmental impact, (b) maximum removal efficiency with minimum environmental impact, and (c) maximum by-product value with minimum environmental impact.

Fig. 8 shows the two-dimensional views of alternatives (green symbols) and criteria (yellow symbols with yellow lines) for a multidimensional problem obtained by Principal Component Analysis with a decision vector (red line) marked. A longer decision vector indicates greater decision-making power. In general, the length of the decision vectors for three scenarios is nearly similar, which all imply strong decision-making power. For scenario (a) (Fig. 8a), processes P-SM+FL-6, P-SM+FL-8, P-SM+FL-10, and P-SM+FL-14 are all located in close proximity to the decision vector, which means the strategy of partial smouldering plus a flame can achieve the maximum carbon sequestration and minimum environmental impact (emitted less carbon and toxic species). Moreover, a smaller airflow is preferred for this scenario.

Fig. 8b shows 5 alternatives, namely, F-SM+FL-18, F-SM+FL-14, F-SM+FL-10, F-SM+FL-8, and F-SM+FL-6, are close to the decision vector, indicating that the strategy of full smouldering plus a flame is more recommended to achieve a high removal efficiency (maximum mass reduction of the waste) with least environmental impact, and a larger airflow is preferred. For scenario (c), it is revealed by Fig. 8c that both the partial smouldering processes with and without a flame under a relatively small airflow velocity (i.e., P-SM-6, P-SM+FL-6, P-SM+FL-8, and P-SM+FL-10) are able to produce valuable by-products (either biochar or bio-oil) with minimum harm to the environment. In contrast, the full smouldering process (F-SM) located at the left-bottom side of the plot are least preferred for this scenario.

## 4. Conclusions

In summary, the proposed flexible suite of smouldering waste biomass processing technologies can be regulated with different strategies to achieve different goals. Partial smouldering process can sequester abundant stable carbon in biochar (with $0.2<H:C<0.5$, $0.2<O:C<0.3$, and $R_{50}=0.6$) and thus reduce the carbon released to the atmosphere. And a smaller airflow rate is recommended for this strategy to fix more carbon. Full smouldering process can achieve the largest waste removal efficiency, and the larger airflow rate is preferred. With a flame applied, both partial and full smouldering processes can significantly reduce the toxic emissions with lower $CO/CO_2$ and $C_xH_y$ (g)/$CO_2$ ratios compared with the processes without flame. The equivalent GHG and the $CH_4/CO_2$ ratio from the four proposed processing strategies are all smaller than those from the conventional landfilling treatment.

Finally, according to a multi-criteria analysis, the process of partial smouldering with a flame under small airflow rates is preferred to achieve the maximum carbon sequestration and by-products value with minimum environmental impact. The process of full smouldering with a flame under larger airflow rates is recommended to achieve the largest removal efficiency and the least environmental harm. Altogether, the newly developed technology shows great promise for achieving sustainable municipal solid waste management.

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## CRediT authorship contribution statement

Yuying Chen: Investigation, Methodology, Writing-original draft, Formal analysis. Shaorun Lin: Resources, Methodology, Formal analysis, Writing-review & editing. Yunzhu Qin: Resources, Formal analysis. Nicholas C. Surawski: Supervision, Writing-review & editing. Xinyan Huang: Conceptualization, Methodology, Supervision, Writing-review & editing, Funding acquisition.

## Declaration of Competing Interest

The authors declare no conflict of interest.

## Supplemental Files

Supplemental data to this work can be found in online version of the paper.

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