

# Smouldering Fire and Emission Characteristics of Eucalyptus Litter Fuel

Houzhi Wang<sup>a,b\*</sup>, Philip J. van Eyk<sup>c</sup>, Paul R. Medwell<sup>a</sup>, Cristian H. Birzer<sup>a</sup>, Zhao F. Tian<sup>a</sup>, Malcolm Possell<sup>b,d</sup>, Xinyan Huang<sup>e,\*</sup>

<sup>a</sup>*School of Mechanical Engineering, The University of Adelaide, Adelaide, SA 5005, Australia.*

<sup>b</sup>*Bushfire and Natural Hazards CRC, Melbourne, VIC 3002, Australia.*

<sup>c</sup>*School of Chemical Engineering and Advanced Materials, The University of Adelaide, Adelaide, SA 5005, Australia.*

<sup>d</sup>*School of Life and Environmental Sciences, The University of Sydney, Sydney, NSW 2006, Australia.*

<sup>e</sup>*Department of Building Services Engineering, Hong Kong Polytechnic University, Kowloon, Hong Kong*

\*Corresponding to [houzhi.wang@adelaide.edu.au](mailto:houzhi.wang@adelaide.edu.au) (HW), [xy.huang@polyu.edu.hk](mailto:xy.huang@polyu.edu.hk) (XH)

## Abstract:

*Eucalyptus* is one of the most widespread genera around the world and a key element in recent wildfires. In a *Eucalyptus* forest, the accumulation of litter builds up a ground fuel layer that can support both flaming and smouldering wildfires. This work investigates the smouldering wildfire on leaf, bark, and twig beds (bulk density: 70-140 kg/m<sup>3</sup>) of *Eucalyptus* species. Two-stage smouldering spread processes are observed. The first-stage smouldering fire has the peak temperature of 600-700 °C and spread rate of 5-9 mm/min. The measured emission factors are 1000-1500 g/kg (CO<sub>2</sub>), 180-450 g/kg (CO), 9-16 g/kg (CH<sub>4</sub>), and 2-6 g/kg (H<sub>2</sub>), respectively. The CO/CO<sub>2</sub> ratio ranges from 0.15 (leaf) to 0.8 (bark). Laboratory experiments demonstrate that the smouldering fire spread is slower in leaf than those in bark and twig. The burning of stringy barks is less complete, compared to smooth barks. For leaf and twig beds, the influence of *Eucalyptus* species and heating value on smouldering fire is negligible. This is the first work to reveal smouldering fire behaviours on different *Eucalyptus* litter fuels and provides valuable information for understanding the effects of *Eucalyptus* species and plant parts on smouldering combustion.

**Keywords:** wildland fire; ground fuel; smouldering combustion; fire emission factor.

## 1. Introduction

Wildfire is a global issue, and the frequency of mega wildfires is increasing, due to climate change<sup>1-3</sup>. Wildfire is also one of the significant contributors to the global emission of greenhouse gases<sup>4</sup>, especially for the smouldering wildfires<sup>5-7</sup>. Furthermore, extreme wildfires may slow down the natural regeneration of vegetation, which reduces the net carbon sink. A vicious circle between climate change and wildfires is gradually forming<sup>8</sup>. Hence, it is crucial to have a better understanding of wildfires to mitigate their impacts.

There are two types of combustion processes involved in wildfires, namely, smouldering and flaming combustion<sup>9-11</sup>. Smouldering wildfire also poses a significant hazard in prescribed burning and wildfire fighting<sup>12-14</sup>, as it is difficult to predict, detect, and suppress smouldering wildfire<sup>15-17</sup>. Under windy and drying environment, smouldering wildfires can also transition to flaming fires, especially for the litter fuel layer on the ground<sup>18-21</sup>. Moreover, smouldering fire has negative impacts on ecosystems because it can kill seeds and roots<sup>22</sup>, which significantly prolong the plant restoration process<sup>15</sup>. The delay of the plant restoration process also reduces the carbon offset capability of a forest after a wildfire<sup>23</sup>.

Vegetation plays a vital role in wildfires, as it determines the size and amount of fuel available for wildfires. Much past research has studied the smouldering fire on the litter layer of radiata pine and pine needle beds<sup>12-14,20,21</sup>. *Eucalyptus* has been introduced and widely planted all around the world, because of its great environmental and economic benefits. There are approximately 20 million hectares of *Eucalyptus* forest planted on the Earth, and the majority of them were planted outside Australia<sup>24,25</sup>. Although the planting of *Eucalyptus* trees can bring many benefits to people, it also dramatically increases the risk of wildfires. For example, *Eucalyptus* was introduced to Portugal to prevent soil erosion and has since become a widely distributed species across the country. *Eucalyptus* trees were a predominant factor in the 2017 Portuguese wildfires, which were the country's worst wildfires to-date<sup>26</sup>. As *Eucalyptus* wildfires are an increasingly important issue, more research on this type of fire is urgently needed to understand the associated combustion processes better. Similar to other wildfires, smouldering is a critical combustion phenomenon in eucalyptus fires. Hence, this study focuses on smouldering combustion with an emphasis on eucalyptus.

*Eucalyptus* species can have effects on the physical and chemical properties of *Eucalyptus* litter fuels. A previous study has also shown that the leaf oils from different *Eucalyptus* species have different chemical compositions<sup>27</sup>, and the difference in chemical compositions could have effects on combustion processes. Hence, it is crucial to know whether smouldering fire behaviour would be different for different *Eucalyptus* species. In a *Eucalyptus* forest, the accumulation of fuel, such as leaves, bark, and twigs, builds up a fuel bed for fire events. The fire behaviour of these plant parts (leaf, bark, and twig) can be different due to their various physical and chemical properties. However, understanding of the smouldering fire spread and

emission for different *Eucalyptus* species and their plant parts is still lacking, and this knowledge is essential for a better understanding of *Eucalyptus* wildfires.

To investigate the smouldering fire of different *Eucalyptus* species and plant parts, it is necessary to characterise different fuel samples as the fuel properties have critical effects on the combustion process. The fuel properties include both physical and chemical properties. It is known that the chemical composition of leaves is qualitatively different from that of barks or twigs<sup>28</sup>. However, there is a lack of quantitative analysis of the chemical composition of leaf, bark, and twig<sup>29</sup>; not to mention the chemical properties that are relevant to combustion. The chemical properties in this study will be focused on proximate and ultimate analyses. The physical properties of the fuel particles include their shape and size, which affects air permeability. Air permeability has significant effects on the combustion of a fuel bed. Hence, the physical and chemical characterisations of the different plant parts from different *Eucalyptus* species are essential to understand the combustion process of these typical fuels.

The research first quantified of the physicochemical properties of the different plant parts from different *Eucalyptus* species. Then, laboratory experiments were conducted to investigate the smouldering fire behaviours of litter layer from different *Eucalyptus* species, specifically, smooth-bark and stringy-bark *Eucalyptus* trees. Finally, the influence of the litter layer with different *Eucalyptus* parts on smouldering fire and emission characteristics was investigated.

## **2. Methods**

### **2.1. Characteristics of Litter fuel**

*Eucalyptus* species can be divided into two types: smooth-bark and stringy-bark<sup>30</sup>. In the present study, four *Eucalyptus* species are investigated, namely, two smooth barks and two stringybarks. These four species were collected from Black Hill Conservation Park, South Australia. For each species, three plant parts (bark, leaves, and twigs) were collected. Among the four species and three plant parts, a total of 12 different fuels were considered. All the leaf samples were freshly collected from ten to fifteen mature trees for each *Eucalyptus* species in the same area, and only the adult leaves were later selected for the experiment. Similarly, all the twig samples were trimmed and collected from ten to fifteen mature trees for each *Eucalyptus* species in the same area. All the bark samples were collected from the lower trunk of ten to fifteen mature trees for each species in the same area. One of the focuses of the current study is to investigate the effects of three plant parts (leaf, twig and bark) on smouldering combustion. Natural litter is a very complex fuel with various physical and chemical properties, as it is hard to control the factor of natural decomposition in the litter layer<sup>31</sup>. Hence, instead of collecting and separating the litter layer, the three plants were collected separately to reduce the variability between samples. For the leaf samples, the fresh adult leaves were picked directly from trees for consistency. For the twig samples, twigs were cut

from trees. The bark samples were collected from the trunk. To reduce variability between samples, all the fuel samples were milled, and sieved ( $1 \text{ mm} < d < 2 \text{ mm}$ , where  $d$  is the sieve opening size) and then dried in an oven at  $105^\circ\text{C}$  for 24 hours, consistent with the drying method presented in a previous study<sup>32</sup>.

All the fuel samples were characterised based on their physical and chemical properties. Chemical characterisation of the fuels was conducted using thermogravimetric and ultimate analyses, and air permeability was used to determine the physical properties of the milled fuel. The details about the fuel properties can be found in Supplementary Information, Section A. The milled and sieved fuel sample was loaded into the air permeability testing rig. For more details about the rig, please refer to<sup>33</sup>. The pressure drop across the fuel sample was measured under different inlet airflow velocities. Then, the air permeability and Forchheimer coefficient were calculated using the Forchheimer equation (Equation A1 in Supplementary Information).

## 2.2. Experimental setup

The experiment was designed to investigate the smouldering spread and emission characteristics of the different *Eucalyptus* species and the plant parts on combustion. The combustion was conducted using the same testing rig as in previous studies<sup>13,14</sup>. The experimental apparatus (Figure 1) consists of six main components: an infrared heat lamp, a smouldering reactor, thermocouples, a gas scrubber, and a gas analyser. The reactor has a square cross-sectional area of  $14.4 \text{ cm}^2$  ( $3.8 \text{ cm} \times 3.8 \text{ cm}$ ) and a height of 12.0 cm. There are two main reasons for choosing the current reactor. First, a small area enables a more uniformly distributed heat flux profile. Second, the heat loss from the reactor walls was calculated using the model presented in<sup>13</sup>. The heat loss to the reactor walls is approximately 13% of the heat generated by exothermic reactions. This amount of heat loss will reduce the temperatures near the walls, but is sufficiently low to consider the reactor close to one-dimensional.

The infrared heat lamp is used to heat the fuel sample in the smouldering reactor. Five thermocouples installed in the reactor measure the temperature: one above the fuel bed (the freeboard, FB) and four embedded in the fuel (TC1–4), which has a height interval of 1.0 cm. The smouldering reactor has one oxidizer inlet at the bottom of the reactor, and one output connected to a gas analyser (VARIOPlus, MRU Instrument Inc., Neckarsulm, Germany). A gas scrubber, described in the Supporting Information of<sup>14</sup>, is installed between the smouldering reactor and the gas analyser to remove heavy hydrocarbons and tars, in order to avoid contamination within the gas analyser. The gas analyser measures the dry-basis volumetric concentration of five gases, namely,  $\text{O}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{H}_2$ , and  $\text{CH}_4$ . The sampling rate of the gas analyser is 0.75 L/min, and the accuracies of  $\text{O}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{H}_2$ , and  $\text{CH}_4$  measurement are 0.1 %, 0.1 %, 0.3 %, 0.2 % and 0.2 %, respectively.

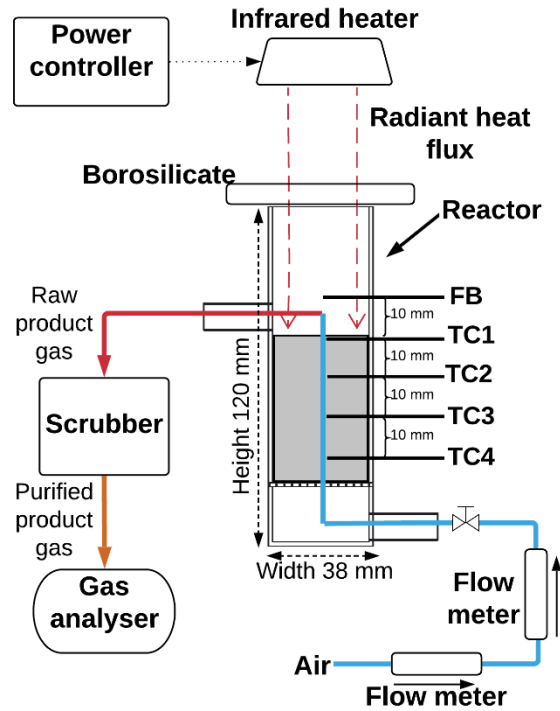


Figure 1. Schematic diagram of the experimental apparatus

The combustion of fuel samples was initiated by the radiant heat flux generated by an infrared heat lamp. The level of the radiant heat flux was controlled by power. The radiant heat flux level was set at  $40 \text{ kW}\cdot\text{m}^{-2}$ , which simulated the radiant heating from nearby flame or hot smoke plume<sup>14</sup>. The combustion experiment in this study focused on smouldering combustion, a slow form of combustion which shows no flame<sup>34</sup>. Hence, the input oxidizer flow velocity was set to  $15 \text{ mm}\cdot\text{s}^{-1}$ , as smouldering combustion generally requires a lower airflow velocity than for flaming combustion<sup>13</sup>. The oxidizer used in this study is the air.

### 2.3. Test procedures

The combustion experiment was conducted for the 12 fuel samples. As the fuel samples have various densities, the height (or volume) of the testing samples was kept constant as 60 mm, in order to reduce variability between fuel samples. Another reason that the fuel bed height was set at 60 mm is to ensure that the bottom four thermocouples (TC1-4) are buried inside the fuel bed and the freeboard thermocouple (FB) is above the top of the fuel bed at the beginning of the combustion experiment. The mass of the sample used in each run was 6 g for the *E. baxteri* and *E. obliqua* bark samples (i.e., the density of  $69 \text{ kg}\cdot\text{m}^{-3}$ ), and 12 g for the other fuel samples (i.e., the density of  $138 \text{ kg}\cdot\text{m}^{-3}$ ). Fuel was loaded into the reactor without compaction. The reason that the mass of the *E. baxteri* and *E. obliqua* bark samples was less than that of the other fuel samples, because the bark of the stringybark *Eucalyptus* species (*E. baxteri* and *E. obliqua*) is fibrous and less dense than the other fuel samples.

Before each experiment, the predetermined amount of the pre-dried fuel sample was loaded into the reactor. The input oxidizer flow velocity was set, and an aluminium shutter was placed above the reactor to initially isolate the fuel sample from the radiant heat flux. After the heat flux levels were adjusted to the predetermined value, temperature, product gas concentration, and mass change recordings commenced. Then, the aluminium shutter was removed, so that the fuel bed was directly exposed to the radiation. The heat lamp was kept on for 900 seconds. The completion of the combustion process was determined based on the measurements of temperature and gas analysis. The combustion experiment has been repeated three times for each of the 12 fuel samples.

### 3. Results and Discussion

#### 3.1. Fuel characterisation

All the fuel characterisation analyses have been repeated three times for each of the 12 fuel samples. The chemical characteristics of the fuel samples were determined based on the results of proximate and ultimate analyses. **Table 1** shows the average percentages of volatile matter, fixed carbon, and ash. Further details can be found in Table A1 of Supplementary Information. Fixed carbon is the solid combustible residue (char) that remains after volatile matter distils off. While, volatile matter represents the non-water gases formed from a fuel sample during heating in an oxygen-free environment. Clearly, most of the twig and leaf samples have similarities in the contents of volatile matter, fixed carbon, and ash; while for bark samples, the composition is more diverse. Theoretically, smouldering combustion would occur preferentially in the twig samples, as they have a higher content of fixed carbon. This is because, in self-sustained smouldering combustion, the majority of heat is from the oxidation of carbon<sup>35</sup>.

**Table 1.** Proximate analysis results of the 12 fuel types investigated (wt%, dry basis).

|                        | <i>E. camaldulensis</i> |      |      | <i>E. fasciculosa</i> |      |      | <i>E. baxteri</i> |      |      | <i>E. obliqua</i> |      |      |
|------------------------|-------------------------|------|------|-----------------------|------|------|-------------------|------|------|-------------------|------|------|
|                        | Bark                    | Leaf | Twig | Bark                  | Leaf | Twig | Bark              | Leaf | Twig | Bark              | Leaf | Twig |
| <b>Volatile Matter</b> | 73.0                    | 77.2 | 69.9 | 76.0                  | 72.0 | 70.6 | 81.2              | 76.8 | 75.7 | 73.8              | 76.7 | 70.6 |
| <b>Fixed carbon</b>    | 24.4                    | 18.0 | 25.3 | 19.8                  | 24.4 | 24.9 | 18.3              | 19.4 | 20.0 | 25.0              | 20.3 | 25.8 |
| <b>Ash</b>             | 2.6                     | 4.8  | 4.8  | 4.2                   | 3.6  | 4.5  | 0.5               | 3.8  | 4.3  | 1.2               | 3.0  | 3.6  |

Most of the leaf samples have a higher content of volatile matter (72.0-77.2 %), a low content of fixed carbon (18-24.4 %). Hence, the leaf samples produce more volatile matter. Similar results were also found for *Pinus Sylvestris* pine<sup>29</sup>. The volatile matter is essential to produce flammable pyrolysis gases for flame<sup>36</sup>, so that the leaves are the most flammable components of the tree, as expected. Moreover, there is no significant correlation between the *Eucalyptus* species and the contents of volatile matter, fixed carbon and ash. However, the barks of the stringy-bark *Eucalyptuses* (*E. baxteri* and *E. obliqua*) have a lower content

of ash (0.5-1.2 %) than those (2.6-4.2 %) of the smooth-bark *Eucalyptus* (*E. camaldulensis* and *E. fasciculosa*).

Table 2 shows the results of ultimate analysis, and further details can be found in Table A1 of Supplementary Information. Clearly, leaf samples have similarities in the carbon, hydrogen, and oxygen contents. Furthermore, the carbon content of the leaf (50-51 %) samples are higher than that of the twig (41-48 %) and bark (43.5-47.5 %) samples; and the hydrogen content of the leaf (7-7.5 %) samples is also slightly higher than that of the twig (5.5-7 %) and bark (6-6.5 %) samples. The elemental composition is closely related to the heat of combustion<sup>37,38</sup>; hence, the complete combustion of the leaf samples could release more energy per unit mass of the fuel than that of the twig and bark samples. The high carbon and hydrogen contents are because the volatile matter content of the leaf samples is generally higher than that of the bark and twig samples. This can also be seen from the proximate analysis results in **Table 1**.

**Table 2.** Ultimate analysis of 12 fuel types investigated (wt%, dry basis), where oxygen is measured by difference.

|                 | <i>E. camaldulensis</i> |      |      | <i>E. fasciculosa</i> |      |      | <i>E. baxteri</i> |      |      | <i>E. obliqua</i> |      |      |
|-----------------|-------------------------|------|------|-----------------------|------|------|-------------------|------|------|-------------------|------|------|
|                 | Bark                    | Leaf | Twig | Bark                  | Leaf | Twig | Bark              | Leaf | Twig | Bark              | Leaf | Twig |
| <b>Carbon</b>   | 43.6                    | 49.3 | 41.1 | 43.6                  | 50.0 | 44.2 | 47.0              | 50.0 | 47.3 | 46.4              | 48.9 | 45.9 |
| <b>Hydrogen</b> | 6.0                     | 7.2  | 5.7  | 6.5                   | 7.1  | 6.3  | 6.5               | 7.4  | 6.8  | 6.3               | 6.8  | 6.5  |
| <b>Nitrogen</b> | 0.1                     | 1.2  | 0.4  | 0.2                   | 0.9  | 0.4  | 0.5               | 0.9  | 0.4  | 0.6               | 1.7  | 0.4  |
| <b>Oxygen</b>   | 50.3                    | 42.3 | 52.8 | 49.7                  | 42.0 | 49.1 | 46.0              | 41.7 | 45.5 | 46.7              | 42.6 | 47.2 |

According to the Kjeldahl method<sup>39</sup>, the nitrogen indicates the protein content, so that the leaf samples contain more protein. Table 2 shows that there is no significant correlation between the *Eucalyptus* species and the carbon, hydrogen, and oxygen contents of the leaf samples. However, there is a significant correlation between the *Eucalyptus* species and the carbon content of the twig and bark samples. Table 2 also shows that bark samples from the stringybark *Eucalyptus* (*E. baxteri* and *E. obliqua*) have a lower carbon content than that of the smooth-bark *Eucalyptus* (*E. camaldulensis* and *E. fasciculosa*). Thus, the carbon content can be used to distinguish the bark and twig between smooth and stringybark *Eucalyptus*.

**Table 3.** Measured higher heating value (MJ·kg<sup>-1</sup>, dry basis) of 12 litter samples.

| Heating value (MJ·kg <sup>-1</sup> ) | Bark | Leaf | Twig |
|--------------------------------------|------|------|------|
| <i>E. camaldulensis</i>              | 16.2 | 20.0 | 14.9 |
| <i>E. fasciculosa</i>                | 16.6 | 20.2 | 16.6 |
| <i>E. baxteri</i> <sup>#</sup>       | 18.0 | 20.4 | 18.5 |
| <i>E. obliqua</i> <sup>#</sup>       | 17.8 | 19.5 | 17.6 |

<sup>#</sup>E. baxteri and E. obliqua bark density is 70 kg·m<sup>-3</sup>, all other density is 140 kg·m<sup>-3</sup>.

The higher heating value of fuel samples was determined using a bomb calorimeter (6400 Automatic Isoperibol Calorimeter; Parr Instrument Company, Moline IL). Table 3 presents the measured bulk density and the higher heating value of all litter samples. The initial mass of the fuel was measured before it was loaded into the combustion reactor. The bulk density of the fuel bed is calculated based on the mass of fuel per fuel bed volume. It is noticed that measurements of bulk densities for the fuel beds in the combustion reactor were much higher than the eucalyptus litter bulk densities reported in the literature<sup>40-42</sup>. Smooth-bark samples have slightly lower heating values than that of the stringy-bark samples, because the stringy-bark samples normally have a higher carbon content than the smooth-bark samples (see Table 2). Moreover, leaf samples have the highest heating values (19.5-20.4 MJ·kg<sup>-1</sup>) among three plant parts. Smooth-bark samples have slightly lower heating values than stringy-bark samples because of lower carbon content.

**Table 4** Permeability ( $k_F$ ) and coefficient ( $\beta$ ) of the pulverised and sieved fuel material of 1-2 mm particle size (The error in the table is the standard error).

| Species                 | Forchheimer permeability, $k_F$ ( $\times 10^{-9} \text{ m}^2$ ) |             |             | Forchheimer coefficient, $\beta$ (-) |            |            |
|-------------------------|--|-------------|-------------|--------------------------------------|------------|------------|
|                         | Bark   | Twig        | Leaf        | Bark                                 | Twig       | Leaf       |
| <i>E. camaldulensis</i> | 5.82 ± 0.21  | 5.87 ± 0.26 | 1.45 ± 0.07 | 3414 ± 102                           | 3749 ± 157 | 6975 ± 146 |
| <i>E. fasciculosa</i>   | 4.87 ± 0.12  | 4.68 ± 0.09 | 1.76 ± 0.09 | 3110 ± 124                           | 4231 ± 131 | 6774 ± 203 |
| <i>E. baxteri</i>       | 5.33 ± 0.19  | 5.35 ± 0.23 | 2.87 ± 0.07 | 457 ± 20                             | 4017 ± 189 | 3622 ± 76  |
| <i>E. obliqua</i>       | 5.8 ± 0.13   | 4.02 ± 0.16 | 2.08 ± 0.04 | 650 ± 18                             | 5052 ± 182 | 901 ± 36   |

The results in Table 4 show that the air permeability of the bark and twig samples is similar for all the *Eucalyptus* species, and the leaf samples have the smallest air permeability<sup>33</sup>. This is because the leaf samples create a more compact fuel bed than that of the bark and twig samples due to their flaky shape. In comparison with the plant parts, there is no significant correlation between the *Eucalyptus* species and the air permeability.

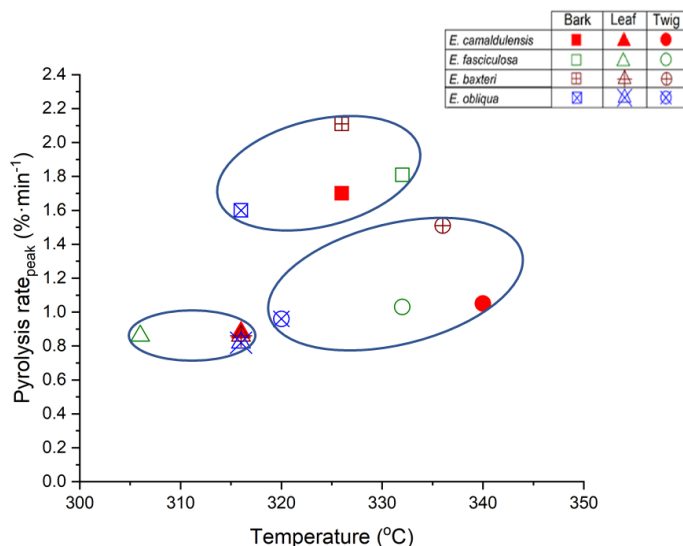
### 3.2. Thermal analysis

The pyrolysis of the fuel samples is heavily dependent on the chemical composition of the fuel samples. The standard thermogravimetric analysis (TGA) is performed under the non-oxidative ambient, and the heating rate of 2 K·min<sup>-1</sup>. The raw data of thermal analysis are shown in Figure A1 of supplemental material. Figure 2 summarizes the peak pyrolysis rate and the peak-pyrolysis temperature of the TGA data under the non-oxidative ambient, at which the peak pyrolysis rate occurred, for all litter fuel samples.

Clearly, the bark, leaf, and twig samples have different peak pyrolysis rates and peak-pyrolysis temperatures. Notably, the leaf sample has the lowest peak pyrolysis rate and reaches the peak pyrolysis rate at the lowest temperature<sup>43</sup>. The low pyrolysis rates suggest that the hemicellulose and cellulose components of the leaf are more and easier to decompose than those of the twig and bark samples. For example, cotton has more than 90% of cellulose<sup>44</sup>, which is much larger than bagasse. Thus, under the



same heating rate, the peak pyrolysis rate of cotton ( $16.1\% \cdot \text{min}^{-1}$ ) is higher than that of bagasse ( $12.0\% \cdot \text{min}^{-1}$ )<sup>45</sup>.



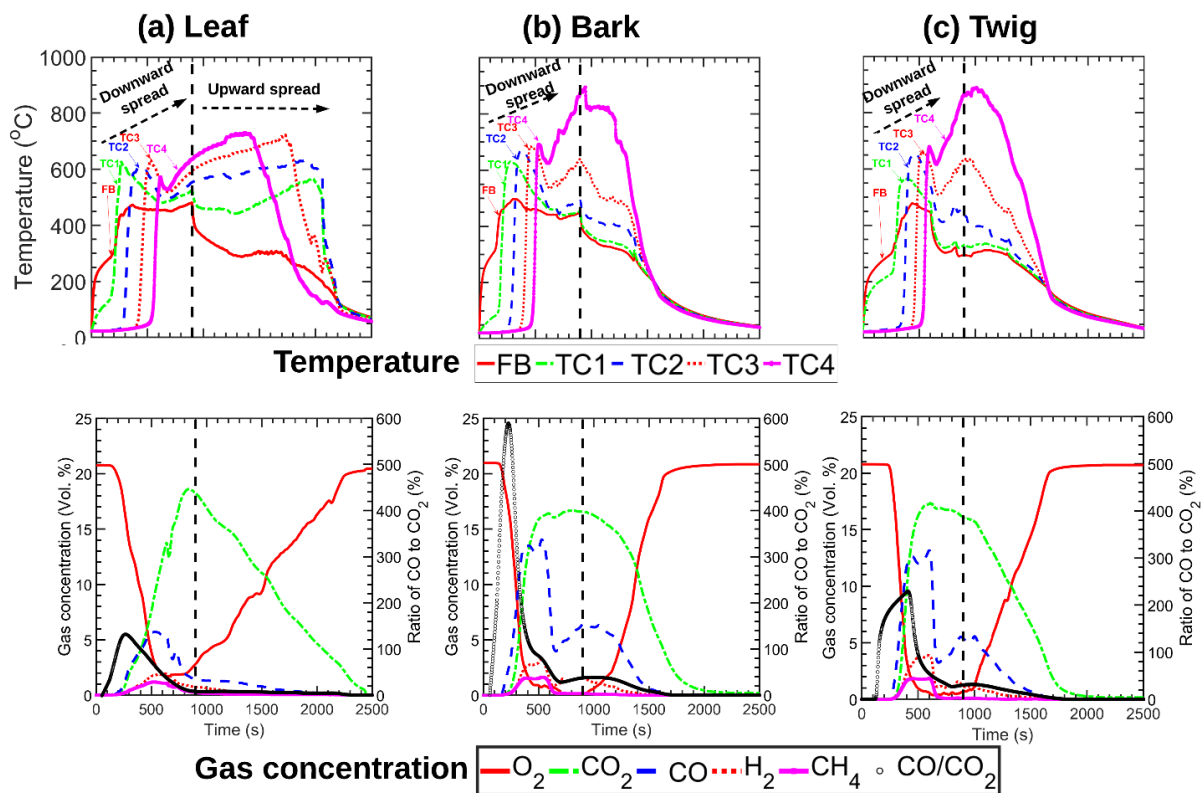
**Figure 2.** Plot of average peak pyrolysis rate value and the temperature at which the peak pyrolysis rate occurred in TGA test, where the heating rate is  $2\text{ K} \cdot \text{min}^{-1}$  under the non-oxidative ambient.

For bark and twig samples, there are connections between the *Eucalyptus* species and the peak pyrolysis rates and the peak-pyrolysis temperature. For instance, the bark and twig samples of *E. baxteri* have higher peak pyrolysis rates compared to the other *Eucalyptus* species, while the bark and twig samples of *E. obliqua* have the lowest peak-pyrolysis temperature. For leaf samples, there is no significant correlation between the *Eucalyptus* species and the pyrolysis rates. In general, the pyrolysis characteristics of the same plant parts are similar, as seen in Figure 2. In each plant part, the *Eucalyptus* species that have the lowest peak-pyrolysis temperature, e.g. *E. fasciculosa* for leaf, and *E. obliqua* for bark and twig, generally have a lower content of volatile matter and a higher content of fixed carbon. For bark and twig, samples with the highest peak pyrolysis rate have the highest volatile matter among all the *Eucalyptus* species. This result shows that for the same plant part, the samples that have a low content of volatile matter and a high content of fixed carbon, leads to a low peak-pyrolysis temperature. The bark and twig samples with a higher content of volatile matter lead to a higher peak pyrolysis rate. Although leaf samples generally have a high content of volatile matter, they have the lowest peak pyrolysis rate, probably because of its high content of oils that evaporate at low temperatures ( $105\text{-}250\text{ }^\circ\text{C}$ ).

### 3.3. Smouldering fire behaviours

Figure 3. Transient temperature and emission gas concentration from the litter bed of (a) leaf, (b) bark, and (c) twig of *E. camaldulensis*, where the heat flux level is  $40\text{ kW} \cdot \text{m}^{-2}$ , the heating time is 900 s

(lamp off indicated by vertical dashed line), and the flow velocity is  $15.0 \text{ mm}\cdot\text{s}^{-1}$ . A moving average method (Span=0.1) is used to smooth the ratio of CO and CO<sub>2</sub> data. shows the transient temperature and emission gas concentrations of the various plant parts of *E. camaldulensis*. These curves are similar between different plant species, so not all of them are shown here. The freeboard (FB) thermocouple gives the temperature inside the litter bed. The vertical dashed line at 900 s represents the time when the infrared heat lamp was turned off. The smouldering combustion process can be divided into two stages: (1) downward (opposed) smouldering spread, and (2) the upward (concurrent) smouldering burning. In the Stage I spread, char oxidation was firstly initiated by the external radiation and propagated downward through the fuel bed. In this phase, char oxidation provides the heat needed for the first peak temperature and the initiation of the pyrolysis zone that is below the smouldering combustion zone. CO and other volatile matters are not combusted, as O<sub>2</sub> is sufficiently depleted above the combustion zone. After the Stage I spread, most of the litter fuels are pyrolyzed and charred, and there is sufficient O<sub>2</sub> left over for the oxidation of CO. This can be seen from the low CO emission during the upward spread.



**Figure 3.** Transient temperature and emission gas concentration from the litter bed of (a) leaf, (b) bark, and (c) twig of *E. camaldulensis*, where the heat flux level is  $40 \text{ kW}\cdot\text{m}^{-2}$ , the heating time is 900 s (lamp off indicated by vertical dashed line), and the flow velocity is  $15.0 \text{ mm}\cdot\text{s}^{-1}$ . A moving average method (Span=0.1) is used to smooth the ratio of CO and CO<sub>2</sub> data.

A similar two-stage opposed-to-concurrent smouldering process has been observed for peat<sup>16</sup> and biochar<sup>46</sup>. Note that the existence of external radiation does not affect the overall two-stage smouldering, but primarily acts as the ignition source<sup>14</sup>. It is evident from the temperature profiles which continues to increase after the lamp is switched off. The high CO/CO<sub>2</sub> emission ratios at the beginning could attribute to the nondispersive infrared sensors' response time. The concentration of the gases from the reactor was measured and recorded every 2 seconds. The concentration of CO and CO<sub>2</sub> in the flue gas increases dramatically when the fuel bed is ignited. The CO and CO<sub>2</sub> measurements will increase to the actual concentrations over a period of time.

The temperature evolution and 1<sup>st</sup> peak temperature (about 650 °C) of *Eucalyptus* species are similar to radiata pine of previous work<sup>13,14</sup>. The 2<sup>nd</sup> temperature peak is caused by the initiation of the char combustion<sup>14</sup>. The existence of external radiation also increases the char oxidation and the 2<sup>nd</sup> peak temperature, so that without radiation, the leaf has a much lower 2<sup>nd</sup> peak temperature. Moreover, the bottom temperature (TC4) is much higher than the 1<sup>st</sup> peak and higher locations (TC1-3), because the bottom has the greatest oxygen supply. For leaf beds, not all oxygen is consumed during the 2<sup>nd</sup> stage. Thus, the oxidation is also intense in higher locations, showing a peak temperature similar to the bottom. On the other hand, for bark and twig samples, the oxygen concentration between 500 s and 1100 s is near zero, indicating the oxygen supply decreases from bottom to top. Thus, the 2<sup>nd</sup> peak temperature decreases as the height is increased.

For the leaf sample, the 1<sup>st</sup> peak temperature (~635 °C) in the Stage-I downward spread is slightly lower than that of the bark sample (~650 °C), mainly because of the particle shape and fuel bed structures. The results in Table 4 show that the air permeability of the leaf samples is different from the bark and twig samples due to the particle shape; and the results also show that the air permeability of the bark and twig samples are similar. Hence, this could explain the similarity between the bark and twig samples. It is also noticed that the self-sustained smouldering combustion for the leaf sample lasted for about 1300 s, which is much longer than that of the bark sample (~700 s). As the mass of the fuels loaded into the reactor was the same for the bark and leaf samples; this indicates that the reaction rate of smouldering combustion is slower in the leaf sample than that in the bark sample. Figure 3a shows that the emission of CO in the self-sustained smouldering stage of the leaf sample is much lower compared with the bark sample. As shown in Tables 1 and 2, the leaf samples have a high content of volatile matter and elemental carbon; but a low content of fixed carbon. This means that a large amount of the elemental carbon is released through the volatile matter of the leaf sample. The low content of fixed carbon suppresses smouldering combustion; hence; the emission of CO was quite low in the self-sustained smouldering combustion stage. Furthermore, the leaf samples have a relatively higher content of volatile matter and lower content of fixed carbon, so

the heat release from the oxidation of char (or fixed carbon) is smaller, leading to a lower temperature than that of the bark and twig samples.

Figure 3. Transient temperature and emission gas concentration from the litter bed of (a) leaf, (b) bark, and (c) twig of *E. camaldulensis*, where the heat flux level is  $40 \text{ kW}\cdot\text{m}^{-2}$ , the heating time is 900 s (lamp off indicated by vertical dashed line), and the flow velocity is  $15.0 \text{ mm}\cdot\text{s}^{-1}$ . A moving average method (Span=0.1) is used to smooth the ratio of CO and CO<sub>2</sub> data. also compares the evolution of emission gases of CO, CO<sub>2</sub>, H<sub>2</sub>, CH<sub>4</sub> and the ratio of CO to CO<sub>2</sub> for different parts of *E. camaldulensis*. Despite the overall similarity with radiata pine in previous work<sup>13</sup>, the concentration of CO during the 2<sup>nd</sup>-stage char oxidation (600–1600 s) for the bark of *E. camaldulensis* is higher than that of the *P. radiata*. The stage of smouldering has a major impact on the emission gas composition. The value of CO/CO<sub>2</sub> ratio is much higher in the 1<sup>st</sup>-stage smouldering fire spread than that of 2<sup>nd</sup>-stage smouldering burning. Particularly, when oxygen consumption is small, the pyrolysis of litter is dominant, and CO is the major carbon emission. As the oxidation reaction becomes strong, the depletion of O<sub>2</sub> rapidly increases, the emission of CO<sub>2</sub> exceeds that of CO. Comparatively, the elimination of the radiative heating only has a secondary effect on the emission of CO and CO<sub>2</sub> from smouldering litter.

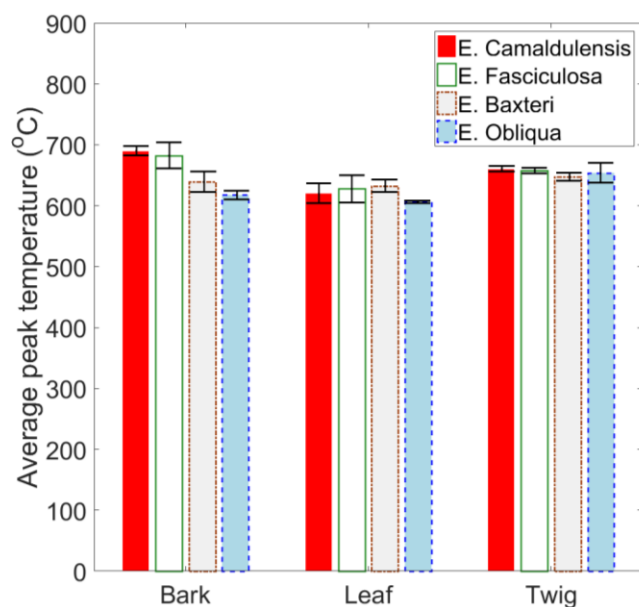
The results in Figure 3 show the temporal and spatial temperature profiles of the twig of *E. camaldulensis*. The temperature profiles of the twig sample are similar to that of the bark sample. The results in Table 1. Proximate analysis results of the 12 fuel types investigated (wt%, dry basis). show that the content of fixed carbon is similar for the bark and twig sample of *E. camaldulensis*. Hence, it implies that there is a positive correlation between the content of fixed carbon and the temperature of the combustion process. Figure 3 shows the temporal gas concentration profiles of the twig of *E. camaldulensis*. The temporal gas concentration profiles of the twig sample are also similar to that of the bark sample. Hence, there is no significant difference between the combustion of the bark and twig samples of *E. camaldulensis*. Figure 3 also shows that the leaf of *E. camaldulensis* is burnt differently from the bark and the twig samples; and the combustion of the bark and twig of *E. camaldulensis* is similar based on the temperature and gas concentrations measurements. This could be because the particle shape has significant effects on the combustion process.

### 3.4. Smouldering temperature and spread rate on litter layer

Figure 4 compares the average peak temperature measurements for the different Eucalyptus species and their plant parts. The average peak temperatures were calculated based on the average peak temperatures of TC1–4. In general, the peak smouldering temperature of all fuel samples ranges from 600 to 710 °C, which is comparable to duff, pine needles<sup>13,14</sup>, and peat<sup>47</sup>. For the bark samples of the different *Eucalyptus* species, the peak temperatures of the smooth-bark *Eucalyptus* (*E. camaldulensis* and *E. fasciculosa*) are

higher than those of the stringybark *Eucalyptus* (*E. baxteri* and *E. obliqua*). This may be because the bark of the stringybark *Eucalyptus* is fibrous, which decreases the bulk density of the fuel bed (Table 3). A lower bulk density leads to lower peak temperature.

For the leaf samples of the different *Eucalyptus* species, there is no significant correlation between the peak temperature and the *Eucalyptus* species. The peak temperatures of leaf samples are about 60 °C lower than those of bark samples of the smooth-bark *Eucalyptus*, and close to the bark samples of the stringybark *Eucalyptus*. The lower peak temperatures of the leaf samples may be because of the shape of the leaf particles. The leaf particles are flaky, and its surface to volume ratio is large; which results in high convective heat loss. Same for the twig samples, no significant correlation can be seen between the peak temperature and the *Eucalyptus* species.

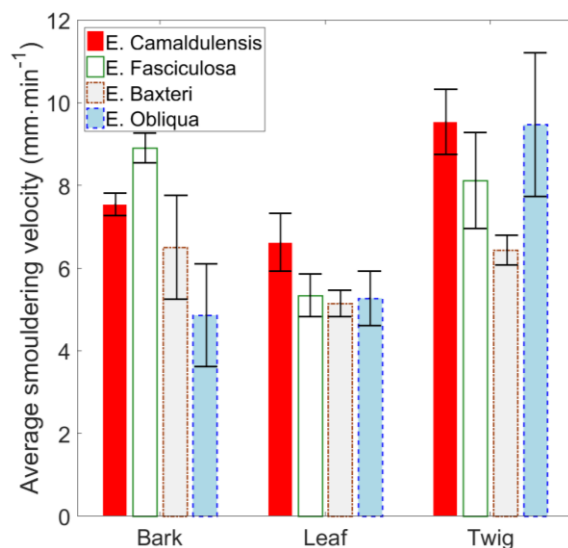


**Figure 4** The average peak temperature measurements of smouldering combustion. Error bars represent the standard error of the peak temperature measurements of the thermocouples (TC1–4) between multiple repeated experiments for each fuel sample.

Figure 5 shows the 1<sup>st</sup>-stage downward smouldering spread rates for the different *Eucalyptus* species and their plant parts. It has been demonstrated that the fire spread rate can be quantified using thermocouples<sup>48</sup>. The propagation velocity was calculated from the time between the arrival of the smoulder front (1<sup>st</sup> peak temperature) at TC1 and TC4 and the known distance between the thermocouples. The average smouldering propagation velocity varies from approximately 5 to 9 mm·min<sup>-1</sup>. For the bark samples of the different *Eucalyptus* species, the average smouldering spread rate of the smooth-bark *Eucalyptus* (*E. camaldulensis* and *E. fasciculosa*) are faster than those of the stringybark *Eucalyptus* (*E. baxteri* and *E. obliqua*). As

observed from Figure 4, the bark of the smooth-bark *Eucalyptus* has higher peak temperatures than those of the stringybark *Eucalyptus*.

For the leaf samples of the different *Eucalyptus* species, there is no significant correlation between the average smouldering propagation velocity and the *Eucalyptus* species. Overall, the average smouldering propagation velocity of the leaf samples are the lowest compared to the bark and twig samples. This finding is consistent with the finding in Figure 3 and 4, where the temperature and gas concentrations results show that for the same experimental conditions, the combustion of the leaf samples lasted longer than that of the bark and twig samples. The longer combustion time reveals that the combustion of the leaf samples is slower than the bark and twig samples. The leaf samples have the highest content of volatile matter amongst the three plant parts (Table 1). However, the results in Figure 2 suggest more volatile matter and less fixed carbon for most leaf samples. Thus, for leaf samples, more heat is need for pyrolysis while less heat is released from the char oxidation, which reduces the smoldering temperature and spread rate.



**Figure 5** The average spread rate in the 1<sup>st</sup>-stage downward (opposed) smouldering. Error bars represent the standard error of the smouldering velocity between multiple repeated experiments for each fuel samples.

In this study, the focus is on smouldering combustion, so conditions were set to initiate smouldering combustion. Under these conditions, the majority of the volatile matter is not oxidised, which can be seen from the temperature and gas concentrations (Figure 3 and 4) profiles. If the strong oxidisation of volatile matter existed, it would lead to a high temperature or even a flame to further oxidize H<sub>2</sub> and CH<sub>4</sub>. In the Stage-1 spread, the majority of the gaseous emissions is from the pyrolysis of the original fuel sample. It can also be seen in Figure 3 that the gaseous emission peaks all occur in the Stage-1 spread. The oxidation of the volatiles does not occur in the Stage 1 spread due to the lack of oxygen. In the Stage-2 spread, as fuel has been converted into char through thermal degradation, CO and CO<sub>2</sub> are the main product gases in this

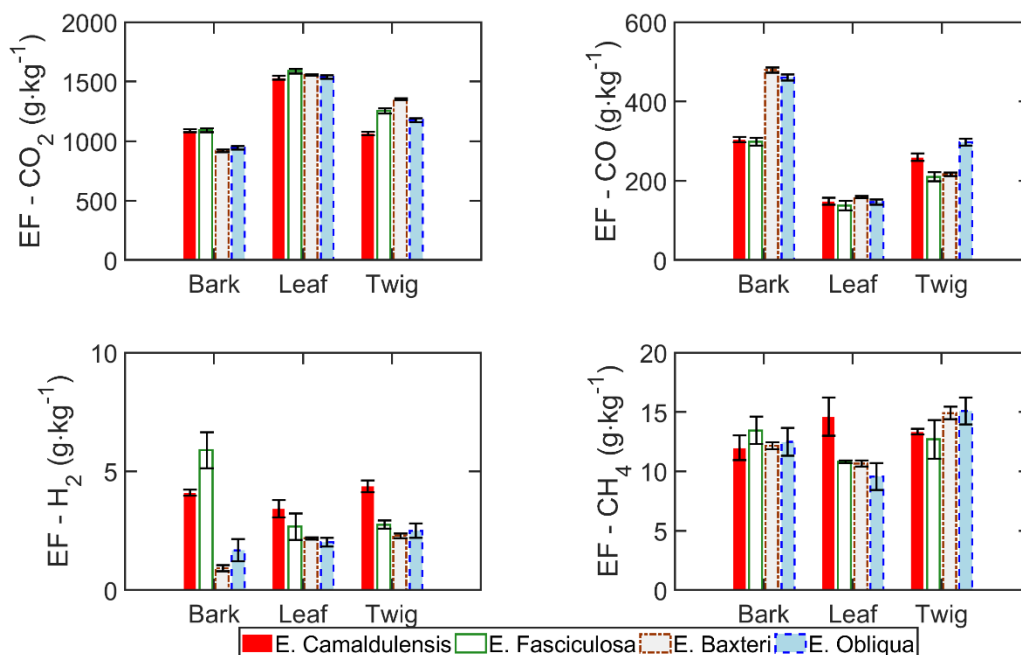
stage. The particle shapes also have effects on the heat transfer, which eventually slows down the combustion process. For the twig samples of the different *Eucalyptus* species, there is also no significant correlation between the average smouldering propagation velocity and the *Eucalyptus* species.

### 3.5. Emission characteristics

For the low-heating rate (<1 K/s) pyrolysis process driven by the char oxidation, low-molecular gases (e.g., H<sub>2</sub>O, CO<sub>2</sub>, CO, H<sub>2</sub> and CH<sub>4</sub>) are the major gaseous products, and comparatively, the production of large-molecular liquid particles (or tar) are small [1–3]. By assuming that major carbon-containing emissions include CO<sub>2</sub>, CO and CH<sub>4</sub>, the emission factor (EF)<sup>49</sup> can be calculated as

$$EF_{b,i} = F_c \left( \frac{1000 \text{ g}}{1 \text{ kg}} \right) \left( \frac{MM_i}{12} \right) \left( \frac{C_i}{C_T} \right) \quad (1)$$

where  $F_c$  is the fuel carbon content (% , on the dry mass basis),  $MM_i$  is the molar mass of species  $i$ ,  $C_i$  is the number of moles of species  $i$ , and  $C_T$  is the total mole number of carbon emitted through CO<sub>2</sub>, CO and CH<sub>4</sub>. It is worth noting that for the pyrolysis gas products, the CH<sub>4</sub>, CO<sub>2</sub>, and CO are still the major gas compositions as found for many biomasses<sup>50-52</sup>. The emission factor is based on the conservation of carbon element, so that it is valid for different chemical (combustion) and physical (phase-change) processes. Ideally, it is also possible to use the conservation of oxygen element. However, it is difficult to identify if the water comes from the chemical reactions or the drying of biomass, and some water vapor may condense and stay in the upper unburnt fuel. Thus, the principle of carbon element conservation is used to quantify the emission factor.

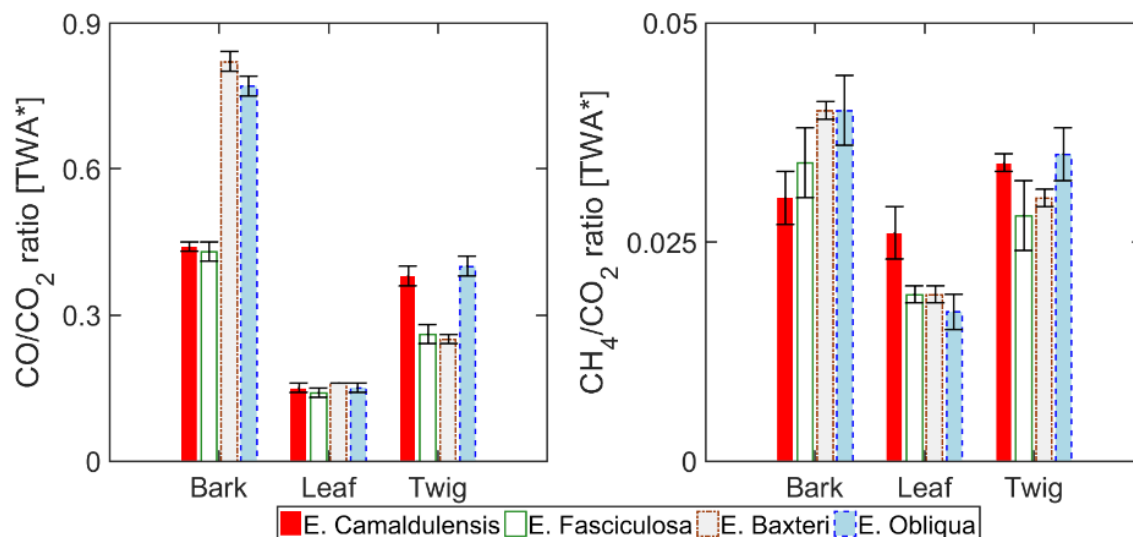


**Figure 6** The time-weighted emission factors (EFs) of (a) CO<sub>2</sub>, (b) CO, (c) H<sub>2</sub> and (d) CH<sub>4</sub> for the different *Eucalyptus* species and their plant parts. Error bars represent the standard error of the time-weighted average between multiple repeated experiments for each fuel sample

For the bark samples of the smooth-bark *Eucalyptus* (Figure 6), their CO<sub>2</sub> emission factors (1086-1092 g·kg<sup>-1</sup>) are slightly higher than the CO<sub>2</sub> emission factors of stringybark *Eucalyptus* (917-944 g·kg<sup>-1</sup>). While, the bark samples of the stringybark *Eucalyptus* have higher CO emission factors (461-479 g·kg<sup>-1</sup>) than those of the smooth-bark *Eucalyptus*. The higher yield of CO indicates that the combustion process in the bark samples of the stringybark *Eucalyptus* is more incomplete than those of the smooth-bark *Eucalyptus*. Among the three plant parts, the leaf samples have the highest CO<sub>2</sub> emission factors and the lowest CO emission factors, showing that the smouldering combustion is more complete.

From the proximate analysis (Table 1) and the cellulose, hemicellulose, and lignin analysis, it was found that there is a correlation between the content of volatile matter and the contents of cellulose/hemicellulose. The bark of *E. fasciculosa* has the highest content of hemicellulose. From the molecular formula of cellulose (C<sub>6</sub>H<sub>10</sub>O<sub>5</sub>), hemicellulose (C<sub>5</sub>H<sub>10</sub>O<sub>6</sub>) and lignin (C<sub>9</sub>H<sub>10</sub>O<sub>2</sub>), hemicellulose has the highest H/C ratio, which means for the same amount of each sample, hemicellulose releases more hydrogen than cellulose and lignin. Therefore, the fuel sample, which has a high content of hemicellulose, will produce more hydrogen. From the results of the emission factor (Figure 6), it can be seen that the bark of *E. fasciculosa* produces the most hydrogen among all the samples.





**Figure 7.** The time-weighted average (TWA) of (a) CO/CO<sub>2</sub>, and (b) CH<sub>4</sub>/CO<sub>2</sub> for the different *Eucalyptus* species and their plant parts. Error bars represent the standard error of the time-weighted average values between multiple repeated experiments for each fuel samples.

To better interpret the results of gas concentration, the ratios of time-weighted average CO and CH<sub>4</sub> to CO<sub>2</sub> are shown in Figure 7. The CO/CO<sub>2</sub> ratio gives an indication of how complete the combustion is. The high CO/CO<sub>2</sub> ratio indicates incomplete combustion. The fuel samples were combusted in the reactor which has one oxidizer inlet and the oxidizer flow was purposely limited to initiate smouldering combustion. The results in Figure 7 present that the CO/CO<sub>2</sub> is less than 0.5 for the bark of smooth barks, and larger than 0.7 for the bark of stringybarks, which is comparable to smouldering of other wildland fuels in the literature<sup>53</sup>. Hence, the combustion of the bark of stringybarks is more incomplete than that of the bark of smooth barks. This could be because the bark of stringybarks is fibrous, which has a high surface to volume ratio. The high surface to volume ratio increases the heat losses of smouldering combustion, which leads to a less complete combustion. The CO/CO<sub>2</sub> ratio of the leaf samples is the lowest among all fuel samples, which implies the combustion is more complete.

#### 4. Conclusions

This study investigated the smouldering combustion on four different *Eucalyptus* species and three plant parts and quantitatively characterised their differences. For different *Eucalyptus* bark samples, peak smouldering temperatures (~650-700 °C) of smooth bark (*E. camaldulensis* and *E. fasciculosa*) are higher than those (~610-650 °C) of stringybark (*E. baxteri* and *E. obliqua*). Also, two-stage smouldering spread processes are observed. The smouldering front also propagates faster in smooth bark (~7.2-9.3 mm·min<sup>-1</sup>) than in stringybark (~3.8-7.7 mm·min<sup>-1</sup>), and the combustion is more complete with CO/CO<sub>2</sub> of 0.5 vs. 0.8. The smouldering behaviour is insensitive to sample heating values.

The smouldering temperature of the leaf samples (~600-650 °C) is lower than twig and bark samples (660-700 °C), and its smouldering propagating velocity is also smaller (~4.6-7.2 mm·min<sup>-1</sup>). The CO/CO<sub>2</sub> ratio of leaf samples (~0.15) is much smaller, because they have a high surface to volume ratio which promotes the surface reaction. The overall smouldering combustion of the twig samples is similar to the bark samples. The emission factors of smouldering are 1000-1500 g/kg (CO<sub>2</sub>), 180-450 g/kg (CO), 9-16 g/kg (CH<sub>4</sub>), and 2-6 (H<sub>2</sub>), respectively. This is the first work to reveal smouldering fire behaviours on different Eucalyptus litter fuels, and provides valuable information for understanding the effects of Eucalyptus species and plant parts on smouldering combustion. However, more work is needed to understand how the variances in physical and chemical properties can have influences on fire behaviour.

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