



# Particulate matter emission from bio-oil incomplete combustion under conditions relevant to stationary applications



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## HIGHLIGHTS

- PSDs of PM<sub>10</sub> and its key inorganic elements from bio-oil incomplete combustion are reported.
- Fine chars in bio-oil play important roles in PM<sub>10</sub> emission during bio-oil incomplete combustion.
- Atmosphere has significant influence on PM<sub>10</sub> emission during bio-oil incomplete combustion.
- Incomplete combustion in O<sub>2</sub> leads to higher PM<sub>1</sub> yield but lower PM<sub>1–10</sub> yield than that in air.

## ARTICLE INFO

### Article history:

Received 21 August 2015  
Received in revised form 16 December 2015  
Accepted 17 December 2015  
Available online 28 December 2015

### Keywords:

Bio-oil  
Filtrated bio-oil  
Incomplete combustion  
PM<sub>10</sub>  
Carbonaceous material

## ABSTRACT

A raw bio-oil from biomass fast pyrolysis and a filtrated bio-oil, which was prepared from the raw bio-oil via syringe filtration to remove fine char particles, were atomized via an air-assist nozzle set and then combusted in a laboratory-scale drop-tube furnace (DTF) at 1400 °C under incomplete combustion conditions. Both air and oxygen (O<sub>2</sub>) atmospheres were considered to produce PM with aerodynamic diameter of <10 μm (PM<sub>10</sub>). Regardless of experimental conditions, it was found that the particle size distributions (PSDs) of PM<sub>10</sub> follow a bimodal distribution. Under such incomplete combustion conditions, the PM<sub>10</sub> samples apparently contain substantial amounts of carbonaceous material. Whereas the PSDs of Na, K, Cl and S (in the form of SO<sub>4</sub><sup>2-</sup>) exhibit a unimodal distribution, those of Mg and Ca in PM<sub>10</sub> are dependent on combustion atmosphere, i.e. a unimodal distribution for air combustion and a bimodal distribution for O<sub>2</sub> combustion. The results show that under incomplete combustion conditions, the fine char particles in the raw bio-oil play significant roles in the emission of PM<sub>10</sub> as well as Mg and Ca in PM<sub>10</sub>. The removal of the fine char particles in the raw bio-oil leads to considerable reductions in the mass of PM with aerodynamic diameters of 0.1–10 μm as well as that of Mg and Ca in the PM with a size range of 0.372–10 μm from the filtrated bio-oil combustion, compared to those from the raw bio-oil combustion. Combustion atmospheres also have significant effects on the emission and chemical composition of PM<sub>10</sub> due to incomplete combustion. Switching combustion atmosphere from air to O<sub>2</sub> increases the PM<sub>1</sub> yield by ~74.2% due to the increased yields of Na, K, Mg, Ca, SO<sub>4</sub><sup>2-</sup> and PO<sub>4</sub><sup>3-</sup> in PM<sub>1</sub>, but decreases the PM<sub>1–10</sub> yield by ~27.2%, apparently as a result of improved burnout and thereby decreased amounts of unburned carbonaceous material in PM<sub>1–10</sub>.

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## 1. Introduction

While it can be upgraded/refined for producing liquid transport fuels, bio-oil produced from biomass fast pyrolysis is also a suitable fuel for stationary combustion applications [1–4]. Bio-oil combustion offers several advantages over direct biomass combustion. For

instance, bio-oil has a high volumetric energy density suitable for transport [5] and after biomass pyrolysis the majority of key ash-forming inorganic species are retained in biochar [6]. Therefore, bio-oil combustion may potentially mitigate ash-related issues including ash deposition [7], corrosion [8], and fine particulate matter (PM) emission [9] that are often encountered during biomass combustion.

Extensive research has been conducted on bio-oil evaporation and combustion behaviour via thermogravimetric analysers [10], fibre-suspended droplet tests [11], and entrained flow reactors [12,13]. It was proposed that bio-oil combustion consists of four major stages [12,13], including (i) quiescent combustion of light

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volatiles, (ii) droplet swelling and distortion, followed by microexplosion, which usually results in the breakup of droplet to various degrees, (iii) droplet coalescence, and (iv) its subsequent combustion. Recent studies on the combustion of bio-oil or bio-oil/ethanol blends [1,4,14–16] on pilot- or commercial-scale combustion systems led to better understanding on ash deposition and emissions of pollutant gases (e.g.,  $\text{NO}_x$ ,  $\text{SO}_2$  and HCl). While scattered data were reported on the PM produced from the combustion of bio-oil/ethanol blends [14–16], there is no systematic study on the emission of PM with aerodynamic diameter of  $<10\ \mu\text{m}$  ( $\text{PM}_{10}$ ) which is an important environmental concern for fuel utilisation [17–20]. Especially, bio-oil has some undesired fuel properties such as high viscosity, high water content and high coking propensity [12,14,21–24], leading to achieving complete combustion being challenging. In addition, particle size distributions (PSDs) of key inorganic elements in  $\text{PM}_{10}$  are also seldom reported, which is of great importance to understanding the transformation of these inorganic species during bio-oil combustion. Most importantly, bio-oil generally contains substantial amounts of fine char particles of micron and, to a lesser extent, submicron size ranges [25], which accelerate microexplosion during bio-oil combustion [12]. However, the contribution of these fine char particles in  $\text{PM}_{10}$  emission remains unclear due to the lack of comparison study on the combustion of both char-removed bio-oil and its corresponding raw bio-oil. Such knowledge is important in developing strategies for PM reduction during bio-oil combustion.

Therefore, this study aims to conduct a set of systematic research on the emission of  $\text{PM}_{10}$  from the incomplete combustion of both a raw bio-oil and its filtrated bio-oil at  $1400\ ^\circ\text{C}$  in air and oxygen ( $\text{O}_2$ ) atmospheres. A laboratory-scale drop tube furnace (DTF) was employed for the experiments relevant to stationary combustion applications. The produced  $\text{PM}_{10}$  samples were collected and characterised to understand its emission behaviour. The complete PSDs of  $\text{PM}_{10}$  and its key inorganic species were reported. It is noted that this study focuses specially on  $\text{PM}_{10}$  emission during bio-oil incomplete combustion.

## 2. Experimental section

### 2.1. Bio-oil and its atomization

A bio-oil sample (denoted as “raw bio-oil”, produced at  $500\ ^\circ\text{C}$  via a fluidised bed reactor using pine wood as feedstock) was supplied by a company who has chosen to remain anonymous. The raw bio-oil was filtered using a  $0.45\ \mu\text{m}$  polyvinylidene difluoride (PVDF) syringe filter to remove the suspended fine char particles and prepare a filtrated bio-oil sample. The bio-oil samples were stored in fridge prior to experiments and further analyses. The bio-oil samples were injected into a DTF system for combustion via atomization. The bio-oil atomization was achieved by an air-assist nozzle set (model: VLA-3, VLT-1 and VLB, Paasche Airbrush) which was installed into the feeding system of the DTF. Briefly, bio-oil was injected into an inner tube via a syringe pump (model: KDS LEGATO 210) and a 20 ml stainless steel syringe, and atomised at the end of the inner tube which was connected with the air-assist nozzle and then sprayed out. The bio-oil was injected into the furnace at a volumetric flow rate of  $\sim 1.6\ \text{mL}/\text{min}$ , with the assistance of atomization gas (either air or  $\text{O}_2$ , flow rate:  $10\ \text{L}/\text{min}$ ). In each experiment,  $\sim 20\ \text{mL}$  bio-oil was injected into the DTF.

### 2.2. Combustion and PM collection

For both the raw and filtrated bio-oil samples, a set of combustion experiments were conducted at  $1400\ ^\circ\text{C}$  using a DTF system

which is detailed elsewhere [26], with a modified feeding probe that includes the bio-oil atomization nozzle. The feeding probe was protected using a stream of protection gas (either air or  $\text{O}_2$ , depending on combustion atmosphere,  $0.2\ \text{L}/\text{min}$ ) and cooling water to maintain the temperature at the nozzle's outlet below  $70\ ^\circ\text{C}$  to prevent bio-oil coking [27]. The flow rate of total combustion gas, including atomization gas and protection gas, is thus  $10.2\ \text{L}/\text{min}$ . To achieve different levels of burnout, combustion experiments were carried out under both air and  $\text{O}_2$  conditions, with the values of  $\lambda$  (i.e., the ratio of the actual air/fuel ratio to the stoichiometric air/fuel ratio) being  $\sim 1.8$  and  $\sim 8.6$ , respectively. The residence time of the fuel in the isothermal zone of the DTF was estimated as  $\sim 0.9\ \text{s}$ . Before each experiment, the reactor tube of the DTF was thoroughly washed with water to remove impurities (if any) in its inner wall. Under the experimental conditions, complete combustion of the bio-oils was not achieved, as demonstrated by the fact that the collected  $\text{PM}_{10}$  samples are black. The produced PM samples were collected using a combination of a cyclone and a Dekati Low Pressure Impactor (DLPI), following a procedure detailed elsewhere [26]. The temperature of flue gas at the outlet of sampling probe was kept at  $115\ ^\circ\text{C}$  to avoid the condensation of acidic gases (e.g.,  $\text{SO}_3$  and HCl). Experiments were conducted at least in duplicate to ensure repeatability and to produce PM samples for different analyses.

### 2.3. Sample analysis

The water contents of the two bio-oils were analysed using Karl-Fischer titration, following ASTM E203-96 [28]. The titrator (model: Mettler V30) was calibrated using the reagents (CombiTitrant 5 Keto) recommended by a VTT publication [27]. To determine the solids content in the raw bio-oil, the bio-oil was firstly diluted with acetone at a mass ratio of 1:2 and then filtrated with a  $0.45\ \mu\text{m}$  PVDF syringe filter. The mass of the filtrated solids was measured to determine the solid contents of the raw bio-oil. The ash contents of the raw and filtrated bio-oils were determined via ashing the samples in a muffle furnace at  $600\ ^\circ\text{C}$ , following a specifically-designed temperature-time program used in a previous study [29]. Ultimate analysis of the raw and filtrated bio-oils was carried out using a CHN elemental analyser (model: PerkinElmer 2400 series II). The contents of alkali and alkaline earth metallic (AAEM) species (mainly Na, K, Mg and Ca) in the two bio-oils were determined using a method recently developed [29]. Cl and S in the two bio-oils were quantified via an improved Eschka method detailed elsewhere [30]. The quantification of P was conducted via a method that can be found elsewhere [31]. The data on the fuel properties of the raw and filtrated bio-oils are presented in Table 1. The ash content of the raw bio-oil is  $\sim 0.07\ \text{wt}\%$  (dry basis), which is slightly higher than that of most bio-oils reported in previous studies [16,22]. Ca is the most abundant element in the raw bio-oil, followed by K, Mg, and Na in a decreasing order. Filtration results in only slight changes in the contents of water, ash, C, H, N and AAEM species in the filtrated bio-oil, compared to those in the raw bio-oil. This is reasonable because the total solid content of the raw bio-oil is only  $\sim 0.17\ \text{wt}\%$  (as-received basis).

The mass of  $\text{PM}_{10}$  was measured via a Mettler MX5 microbalance (accuracy:  $0.001\ \text{mg}$ ). The AAEM species, water-soluble chloride ( $\text{Cl}^-$ ), phosphate ( $\text{PO}_4^{3-}$ ), sulfate ( $\text{SO}_4^{2-}$ ) in the PM samples were quantified following methods detailed elsewhere [32]. Selected PM samples were also characterised using Neon scanning electron microscopy (SEM) instrument equipped with an energy dispersive X-ray spectrometer (EDS) for morphology and chemical composition analysis.

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