



## Size distributions of PM, carbons and PAHs emitted from a generator using blended fuels containing water

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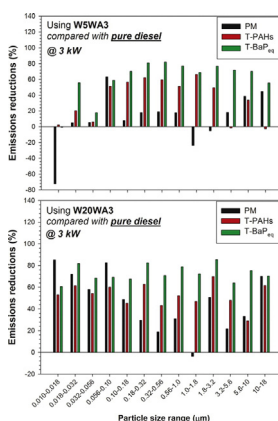
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### HIGHLIGHTS

- Size distributions of diesel particles using water-containing fuels were studied.
- Biodiesel containing 3% WA reduces PM, carbon, and PAHs in 0.010–18  $\mu\text{m}$  size ranges.
- These reductions of emissions of submicron particles exceeded 85%.
- Biodiesel blended with recycled waste solvents may be used as an alternative fuel.
- Water plays a significant role on reducing biodiesel engine pollutant emission.

### GRAPHICAL ABSTRACT



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### ABSTRACT

This investigation studied the size distributions of particulate matter (PM), particulate carbon, and polycyclic aromatic hydrocarbons (PAHs) that are emitted from a generator that is fueled by diesel that is blended with waste-edible-oil-biodiesel and water-containing acetone. PM samples were collected using a micro-orifice uniform deposit impactor (MOUDI) and a Nano-MOUDI (with aerodynamic diameters of 0.01–18  $\mu\text{m}$ ). The results reveal that waste-edible biodiesel blended with water-containing acetone (W5WA3 or W20WA3) at a load of 3 kW emitted lower  $\Sigma\text{PM}$ ,  $\Sigma\text{PM-EC}$ ,  $\Sigma\text{PM-OC}$ ,  $\Sigma\text{T-PAHs}$  or  $\Sigma\text{T-BaP}_{\text{eq}}$  concentrations than did D100, in all 13 particle size ranges, and these reductions of emissions of submicron particles exceeded 85%. Furthermore, W20WA3 emitted significantly lower concentrations of Total-PAHs and Total-BaP<sub>eq</sub> in four nano/ultrafine particle size ranges. Therefore, water-containing acetone biodiesel fuels can be utilized as alternatives to petroleum diesel as fuel to reduce the dangers to human health that are posed by emissions from diesel engines.

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## 1. Introduction

Many studies of particulate matter (PM) have shown that mobile mission sources are the main sources of PM<sub>2.5</sub> (Lin et al., 2005, 2008a). Diesel engines are used in many buses, heavy trucks, tools and machinery, vessels, and generators owing to their high fuel efficiency, high output power, and energy-saving performance (Schinder, 1992). Diesel particulate matter (DPMs) and gaseous pollutants (such as SO<sub>x</sub>, NO<sub>x</sub>, CO, CO<sub>2</sub>, etc.) that are emitted by combusting traditional petrochemical diesel in diesel engines severely pollute the environment. DPM contains carbon particles (~80%, mainly elemental carbon (EC) and organic carbon (OC)), ash, the soluble organic fraction (SOF), sulfides, and other hazardous organic hydrocarbons (HCs) (such as polycyclic aromatic hydrocarbons, PAHs) (Durbin et al., 2000). Research has demonstrated that the aerodynamic diameters of diesel exhaust particles (DEPs) are in the range 0.05–1 µm (Kittelson, 1998; Jacobson and Seinfeld, 2004; Canesi et al., 2008; Tsai et al., 2012). PMs with diameters of less than 2.5 µm can easily enter the human respiratory system and reach the alveolar-interstitial region, severely harming human health (Brook et al., 2010; Maté et al., 2010). In June 2012, the International Agency for Research on Cancer (IARC, WHO) officially recognized diesel engine exhaust as a human carcinogen (Group 1), which has been identified in many countries as having adverse health effects.

In general, DPMs can be divided by aerodynamic diameter into coarse particles (2.5 µm < D<sub>p</sub> < 10 µm), fine particles (D<sub>p</sub> < 2.5 µm), ultrafine particles (0.01 µm < D<sub>p</sub> < 0.1 µm), and nanoparticles (0.01 µm < D<sub>p</sub> < 0.056 µm). Most of the mass of these particles is associated with the accumulation mode between 0.05 and 1 µm (Kittelson et al., 2004). Fine particles typically have higher surface areas than coarse particles, so most PAHs are absorbed into fine particles and nanoparticles (Kahandawala et al., 2004; Westerdahl et al., 2005). Additionally, although the total masses of coarse particles and fine particles are equal, the former have a lower PAH toxicity (Donaldson et al., 1998; Obersdorster, 2001). Fine particles can enter the human body by inhalation, being deposited in the respiratory track, promoting respiratory track disorders (Harrison and Yin, 2000; USEPA, 2002; HEI, 2002). Epidemiological research has shown that exposure to large amounts of DPM increases the rates of incidence of respiratory track disorders and associated mortality (Davis et al., 2006), and so is harmful to human health (Reynolds and Richards, 2001). Therefore, the DPM size distribution and the PAH content in differently sized PMs must be studied to clarify their potential effects on human health.

The use of biodiesel as an alternative fuel in diesel generators is attracting increasing attention. Numerous studies have demonstrated that biodiesel can improve engine combustion efficiency and reduce the emissions of air pollutants (such as PMs, CO, HC, and PAHs) (Lin et al., 2006, 2008b; Tsai et al., 2011b; Chang et al., 2014). The addition of organic solvents (such as ethanol, butanol and acetone) in petrochemical diesel reportedly could improve the combustion reaction in a manner that reduces PM emissions (Lin et al., 2010, 2012; Chang et al., 2013). Additionally, whereas biodiesel has a higher viscosity and a greater cetane index than traditional petrochemical diesel (Tsai et al., 2010, 2011a), bio-alcohols do not, so they cannot be added to petrochemical diesel in a large proportion to form an alternative fuel without requiring modifications to the engine. Therefore, bio-alcohols, as well as biodiesel, are added to petrochemical diesel to adjust the viscosity and cetane index of mixed fuels.

According to our earlier works (Tsai et al., 2014a, 2014b), the characteristics of total suspended particulate matter that was emitted from a small generator whose fuel was pure diesel that was mixed with 1–3% water-containing acetone (WA) or pure acetone (A), 1% isopropyl alcohol (IPA), and 1–70% waste-edible-oil-biodiesel were investigated. The results thus obtained indicated that the concentrations of diesel exhaust emissions (total PM, PM-carbons, PAHs, BaP<sub>eq</sub>, and NO<sub>x</sub>) decrease as the added percentage of water-containing acetone or pure acetone increase from 1% to 3%, regardless of the percentage of biodiesel. Therefore, 3% of

water-containing acetone or pure acetone is a reasonable addition ratio for the biodiesels used in this study to reduce emissions of gaseous and particulate pollutants from the diesel-generator.

To study further the influence of adding water-containing acetone to diesel/biodiesel blend on the distribution of DEP sizes, PM emissions and particle-bound carbon (EC and OC) size distributions from a diesel engine generator that was fuelled with different biodiesels were investigated; these biodiesels were W5 (5% waste-edible-oil-biodiesel + 95% diesel), W5A3 (5% waste-edible-oil-biodiesel + 3% pure acetone + 1% IPA + 91% diesel), W5WA3 (5% waste-edible-oil-biodiesel + 3% water-containing acetone + 1% isopropyl alcohol + 91% diesel), W20 (20% waste-edible-oil-biodiesel + 80% diesel), W20A3 (20% waste-edible-oil-biodiesel + 3% pure acetone + 1% IPA + 76% diesel) and W20WA3 (20% waste-edible-oil-biodiesel + 3% water-containing acetone + 1% IPA + 76% diesel). In this study, 1% IPA was used as a co-solvent to stabilize the water content in diesel fuels. The results thus obtained were compared with those obtained using D100 (pure diesel) under a 3 kW load. The emitted PMs were sampled using micro-orifice uniform deposition impactors (MOUDI) and a Nano-MOUDI, so the tested range of particle sizes was from 0.010 to 18 µm.

## 2. Materials and methodology

### 2.1. Instruments and sampling methods

The size distributions of particle-bound PAHs that were emitted from the generator using different fuels were measured using a micro-orifice uniform deposit impactor (MOUDI) and a Nano-MOUDI (with 0.01–18 µm aerodynamic diameters). The flow rates of the MOUDI and Nano-MOUDI were set to 30 and 10 L/min, respectively, and they were used with 37 and 47 mm quartz filters (Pall Ltd., USA), respectively. The impactors in the MOUDIs and Nano-MOUDI separated the particulate matter into 13 size ranges (at 50% efficiency) with equivalent cut-off diameters of 0.010–0.018, 0.018–0.032, 0.032–0.056, 0.056–0.1, 0.1–0.18, 0.18–0.32, 0.32–0.56, 0.56–1.0, 1.0–1.8, 1.8–3.2, 3.2–5.6, 5.6–10, and 10–18 µm.

The premium diesel fuel was obtained from the Chinese Petroleum Corporation, Taiwan. The waste-edible-oil-biodiesel was manufactured by the Taiwan NJC Corp, and the acetone was obtained from Merck Ltd. (Taiwan). Various blended fuels were tested at the stable energy output (110 V/60 Hz, 1800 rpm) of the generator under a 3 kW load. For each combination of parameters, the experiments were conducted in triplicate (each sampling time = 30 min).

Prior to use, the quartz fiber filters were heated for 2.5 h at 900 °C to reduce of their carbon blanks. This process minimized the background concentration of carbon in the quartz fiber matrix, which could affect the analysis. The treated paper filters were stored in a dry chamber at a temperature of 25 ± 3 °C and a humidity of 45 ± 5% for 24 h before and after sampling. A seven-digit precision microbalance (model UMX2; Mettler Toledo Inc., Switzerland) (with a precision of 0.1 µg) was used to weigh the paper filters before and after sampling. The filter blanks were used to correct all PM and particle-bound PAH concentrations.

### 2.2. Carbon analysis

The carbon contents (elemental carbon (EC) and total carbon (TC)) of the particles that were collected using the quartz filters were analyzed using a total organic carbon analyzer (TOC-5000A; Shimadzu Corp., Japan) that was equipped with a suspended solid measuring (SSM) instrument. To make the carbon measurements, the samples were placed in a sample boat, and were then manually pushed into a 900 °C burner that was filled with oxygen to ensure complete combustion. Following the formation of CO<sub>2</sub> and H<sub>2</sub>O, the H<sub>2</sub>O was removed using a draining device, and the CO<sub>2</sub> content was determined using a non-dispersive infrared (NDIR) gas analyzer. Finally, data were processed and calculations were made to

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