



Effects of oxygenated fuel blends on carbonaceous particulate composition and particle size distributions from a stationary diesel engine



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HIGHLIGHTS

- Oxygenated fuel blends influence particulate composition and size distributions.
- They reduced EC and DPM emissions with butanol being the most effective.
- They showed different effects on counts of nanoparticles and larger particles.
- Both the oxygen content and the type of oxygenates affect DPM characteristics.
- The change in thermo-physical properties affects the level of DPM emissions.

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ABSTRACT

A systematic study was conducted to evaluate and compare the effects of blending five different oxygenated compounds, diglyme (DGM), palm oil methyl ester (PME), dimethyl carbonate (DMC), diethyl adipate (DEA) and butanol (Bu) with ultralow sulfur diesel (ULSD), on engine performance, particulate mass concentrations, organic (OC) and elemental (EC) carbon fractions of the particles and particle size distributions from a single cylinder, direct injection stationary diesel engine with the engine working at a constant engine speed and at three engine loads. A small increase in the brake specific fuel consumption (BSFC) and brake thermal efficiency (BTE) was observed with the use of oxygenates blended with ULSD. All five oxygenates were found to be effective at reducing particulate mass emissions at medium and high engine loads, with butanol being the most effective and DGM being the least effective. Analysis of the relative contribution of changes in the OC and EC emissions to the reduction of particulate matter indicated that under the same oxygen content, EC made a dominant contribution to the reduction of particulate mass. The results also indicated that reduction in both particle mass and number emissions was affected not only by the oxygen content, but also by the chemical structure and thermophysical properties of oxygenates as well as engine operating conditions.

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1. Introduction

Research on sustainable and cleaner fuels continues to receive considerable attention because of the motivation to reduce dependence on fossil fuels and to mitigate environmental and health impacts associated with particulate emissions from both on-road and non-road diesel engines [1]. Numerous oxygenated compounds in the form of alcohols, ethers, and esters have been investigated as additives to conventional diesel fuel in the context of reducing soot and diesel particulate matter (DPM) emissions

[1–9]. There is a general consensus that the fuel oxygen content has a significant influence on soot formation and DPM emissions. However, there are conflicting reports on the relative impacts of different oxygenated functional groups [2–9]. For example, Miyamoto et al. [2], Choi and Reitz [3], and Cheng et al. [4] concluded that reduction in soot or particulate mass was mainly dependent on the percentage of oxygen in the fuel blend while other researchers reported that apart from the oxygen content, the chemical structure of oxygenates also influenced the amount and toxicity of DPM emissions [5–10]. Moreover, different kinds of oxygenates have different thermophysical properties, which tend to affect not only the fuel injection and combustion processes but also the DPM emissions [11,12]. In addition, even with the

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same oxygen content, the extent of soot and DPM emissions reduction with oxygenated fuel blends may also be dependent on the engine types and operating conditions [3,12]. However, no systematic studies have been conducted to make a comparative evaluation of several oxygenates with a range of thermophysical properties on DPM emissions from stationary diesel engines, such as diesel generator sets. These engines are widely used, and emit a substantial fraction of DPM on a global level because they have limited emission control measures. For example, the US Environmental Protection Agency (EPA) estimates that non-road diesel engines contribute to about 44% of the DPM emissions nationwide [13].

DPM consists mostly of carbonaceous materials (total carbon, TC), which are often classified as elemental carbon (EC) and organic carbon (OC) [14]. EC stems from fuel droplet pyrolysis in the fuel-rich zone under high pressure and temperature, while OC originates from unburned fuels, lubricating oil, and combustion byproducts [14]. The previous work reported in the literature mainly focused on studying the soot or DPM emissions [2–8], with little investigation on the variation of EC and OC in response to the types and amounts of oxygenates used [15]. Furthermore, from the environmental perspective, EC suspended in the atmosphere can effectively absorb solar radiation, thus affecting the energy redistribution and global climate [16], while some semi-volatile hydrocarbons in the OC fraction are suspected human carcinogens [17]. To improve our understanding of the environmental and health effects of oxygenates, there is a strong need to conduct a systematic investigation of changes in carbonaceous matter composition of particulate emissions when the petroleum diesel blended with oxygenates is used in diesel engines.

DPM typically consists of fine particles ($PM_{2.5}$) inclusive of a large number of ultrafine particles (UFP) and nanoparticles (NP) with their aerodynamic diameter ≤ 100 nm and 50 nm, respectively. UFP and NP have recently drawn considerable research attention due to their higher rate of pulmonary deposition, ability to travel from lung to systemic sites, higher inflammation potential, and more biological activities than those of particles with larger sizes [17]. UFP and NP dominate the number concentration profiles of DPM, although they do not make a significant contribution to the total mass concentration. In previous studies, the use of oxygenates as blending agents showed reduction in smoke opacity or DPM emissions, caused by a reduction in larger particles, while there might be an increase in sub-micron sized particles [12,18]. Thus, the increased emission of the sub-micron particles and the reduced emission of larger size particles would change particle size distributions in ambient air as well as the characteristics of the inhalation exposure [17]. Because of these reasons, the mass-based emission reduction from oxygenated fuels may not be effective in mitigating their influence on the environment and human health. However, systematic studies and a comparative evaluation of the potential impacts of different oxygenated fuels on UFP and NP emission characteristics are currently lacking.

The aim of this study was to characterize and compare the effects of five oxygenates (diglyme (DGM), palm oil methyl ester (PME), dimethyl carbonate (DMC), diethyl adipate (DEA) and butanol (Bu)) with a range of thermophysical properties on carbonaceous particulate emissions from a stationary diesel engine. To this end, the oxygenates were blended with ultralow sulfur diesel (ULSD) fuel at 2 wt.% and 4 wt.% oxygen. The influence of oxygen content in the blends, the specific types of oxygenates, and the engine operation parameters on carbonaceous particulate composition, particle number and size distributions were investigated. The outcome of the study may offer insights into the effect of oxygenated fuel blends on diesel particulate emissions, and prove to be useful for controlling and regulating DPM emissions from non-road diesel engines.

2. Experimental section

2.1. Test engine and fuels

A schematic of the experimental system employed in this study is shown in [Supplementary Fig. S1](#). Experiments were carried out on a single cylinder, naturally aspirated, four-stroke, direct injection diesel engine (L70AE, Yanmar Corporation) connected to a 4.5 kW generator. The diesel engine has a displacement of 296 mL with bore and stroke of 78 mm and 62 mm, a fixed speed of 3000 rpm (revolutions per min). The main specifications of the engine are shown in [Supplementary Table S1](#). In this study, ultra-low sulfur diesel fuel with less than 10-ppm (parts per million) by weight of sulfur was used as the baseline fuel. Five oxygenates, namely, PME as biodiesel, DMC, DGM, DEA and Bu which represent different chemical groups, namely ester, ethers and alcohols were used as oxygenated fuels. The PME used in this study was obtained from a palm oil-based biodiesel plant in Malaysia operated by Vance Bioenergy, and the fuel properties were provided by the biodiesel supplier. The other four oxygenate compounds were purchased from Sigma-Aldrich. The major properties of the tested fuel are listed in [Supplementary Table S2](#). The properties of each fuel blend can be estimated on the basis of properties of the baseline fuel and the oxygenated compounds using the method recommend by Wang et al. [19], with the results shown in [Supplementary Table S3](#). Each fuel blend having the same oxygen concentrations of 2% and 4% by weight was used for this study. The same amounts of oxygen in fuel blends have been used in many other studies, and also proved to be effective in reducing DPM emissions significantly without the need for engine modification as documented in many previous studies [20–23].

2.2. Particulate sampling and testing

A two-stage Dekati mini-diluter (DI-2000, Dekati Ltd.) was used for diluting the engine exhaust gas for DPM sampling. The diluter provides primary dilution in the range of 8:1–6:1, depending on the engine operating conditions, while the secondary dilution system provides a further dilution of 8:1. The actual dilution ratio was evaluated based on measured CO_2 concentrations in the raw exhaust, in the background air and in the diluted exhaust. The CO_2 concentration was measured with a non-dispersive infrared analyzer (MRU Vario Plus, Germany, $\pm 0.5\%$ accuracy). This measurement was done for every test, and all data presented in this article have been dilution-corrected to represent engine-out condition.

DPM emissions from the first-stage diluter were collected onto 47 mm Teflon filters (Pall Life Sciences, Ann Arbor, MI) and pre-combusted (650 °C for 12 h) 47 mm quartz fiber filters (Whatman, USA), by using two Mini-Vol particulate samplers (Air metrics Ltd.; 5 L min^{-1} flow rate), respectively. Particles collected on the Teflon filters were used for gravimetric analysis, while those collected on quartz fiber filters were processed for subsequent OC/EC analysis. Before and after sampling, the filters were allowed to equilibrate in a humidity-controlled cabinet at 25 °C and 30% relative humidity (RH), and weighed using a microbalance (Sartorius MC5, accuracy of $\pm 1 \mu g$) for quantifying total particulate mass emissions. After being weighed, the filters were kept in glass petri dishes and stored under refrigeration at -20 °C for the subsequent analysis. A thermal/optical carbon aerosol analyzer (Sunset Labs, Forest Grove, Oregon, USA) was used to quantify EC and OC emissions according to the NIOSH 5040 reference method [24]. The number concentration and size distribution of particulate emissions in the secondary dilution stage were measured by a Fast Mobility Particle Sizer (FMPS, Model 3091, TSI Incorporated, USA) in the size

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