Effects of a diesel oxidation catalyst on gaseous pollutants and fine particles from an engine operating on diesel and biodiesel

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Abstract The effects of a diesel oxidation catalytic (DOC) converter on diesel engine emissions were investigated on a diesel bench at various loads for two steady-state speeds using diesel fuel and B20. The DOC was very effective in hydrocarbon (HC) and CO oxidation. Approximately 90%–95% reduction in CO and 36%–70% reduction in HC were realized using the DOC. Special attention was focused on the effects of the DOC on elemental carbon (EC) and organic carbon (OC) fractions in fine particles (PM_{2.5}) emitted from the diesel engine. The carbonaceous compositions of PM_{2.5} were analyzed by the method of thermal/optical reflectance (TOR). The results showed that total carbon (TC), OC and EC emissions for PM_{2.5} from diesel fuel were generally reduced by the DOC. For diesel fuel, TC emissions decreased 22%–32% after the DOC depending on operating modes. The decrease in TC was attributed to 35%–97% decrease in OC and 3%–65% decrease in EC emissions. At low load, a significant increase in the OC/EC ratio of PM_{2.5} was observed after the DOC. The effect of the DOC on the carbonaceous compositions in PM_{2.5} from B20 showed different trends compared to diesel fuel. At low load, a slight increase in EC emissions and a significant decrease in OC/EC ratio of PM_{2.5} after DOC were observed for B20.

Keywords diesel oxidation catalyst (DOC), diesel particulate matters, elemental carbon (EC), organic carbon (OC), biodiesel

1 Introduction

Particulate matter (PM) emitted from a diesel engine consists mostly of fine particles (PM_{2.5}) composed of carbonaceous materials, small amounts of sulfate, nitrate, trace elements, water, and unidentified compounds [1,2]. Carbonaceous materials contributed most to the mass of diesel PM, which can be typically divided into two classes: elemental carbon (EC) and organic carbon (OC). EC is traditionally used as a marker for human exposure to diesel exhaust and OC provides a significant amount of the mutagenic and carcinogenic chemicals in the air. The emission rates of EC and OC from a diesel engine and the ratios of EC to OC can be influenced by many factors including engine load, vehicle type, fuel composition and exhaust aftertreatment [3–5].

Diesel oxidation catalysts (DOC) have long been used to reduce hydrocarbons and CO emissions from a diesel engine. Research has shown that DOC could affect PM emissions in three ways: soot oxidation, soluble organic fraction (SOF) removal, and sulfate formation [6,7]. PM mass emissions may be increased by a DOC through significant sulfate formation when the fuel sulfur content is high [6]. The impact of the DOC on PM mass emission is a combination effect of SOF reduction and sulfate formation. In recent years, with the fuel sulfur content decreasing, the particle mass emission reduction has been achieved mainly by lowering the SOF of the particles with slight sulfate formation [8].

The vegetable oil ester-based biodiesel has long been used as fuel for diesel engines. Many studies of biodiesel fuel use in diesel engines have been performed and some of them have been well reviewed [9,10]. Biodiesel has properties similar to those of traditional fossil diesel fuel and it may be used directly in a diesel engine or blended with diesel fuel. Studies clearly indicate that the use of

biodiesel may potentially reduce society's dependence on petroleum diesel fuel and improve air quality. Substantial reduction in particulate emissions may be obtained through the addition of biodiesel to diesel fuel. B20 (a mixture of 20% biodiesel and 80% petroleum diesel) is the most popular biodiesel fuel blend being studied [11–13].

The purpose of this study was to examine the effect of a DOC converter on the carbonaceous compositions (OC and EC fractions) of $PM_{2.5}$ from a diesel engine. The $PM_{2.5}$ in the exhaust before and after the DOC was sampled at steady-state operating conditions and analyzed by thermal/optical reflectance (TOR). Additionally, a blend of 20% biodiesel with diesel fuel (B20) was prepared and examined in the same manner. The results from the diesel fuel and B20 were compared and discussed.

2 Experiment

2.1 Engine, fuel, catalysts, and test modes

A 2.77 L, four-cylinder, direct injection/turbocharged intercooled diesel engine meeting Euro II emission standards was used in this study (Aaling 4JB1 series diesel engine, Foton, Beijing, China). It was connected to an electric (AC) dynamometer (HT 350, SCHENCK, Germany). The compression ratio of the engine was 18.2:1. Engine rated power was $80\text{ kW}/3600\text{ r}\cdot\text{min}^{-1}$ and maximum torque was $245\text{ N}\cdot\text{m}/1400\text{ r}\cdot\text{min}^{-1}$.

The diesel fuel is a commercial fuel with a fuel sulfur content below $50 \times 10^{-6} w$. Biodiesel is methyl soyate made from soybean oil and methanol; more details about the biodiesel can be found in our previous study [14]. A blend of 20% biodiesel with diesel fuel (B20) was prepared for testing.

The DOC coated with precious metals was provided by Kunming Institute of Precious Metals (Kunming, China) with volume of 2.5 L. However, more details of the DOC, such as precious metal content, washcoat, catalyzed volume were not provided by the supplier. The converter was placed about 1.5 m downstream of the engine. Engine out emissions and emissions after the DOC were measured at two steady speeds with loads of 25%, 50%, and 75% at 2125 and 2300 $\text{r}\cdot\text{min}^{-1}$, corresponding to modes 7, 5, 6, and modes 9, 3, 4 of the European Stationary Cycle (ESC). The exhaust gas temperatures of test modes were about 275°C, 349°C, and 410°C for 25%, 50%, and 75% load at 2125 $\text{r}\cdot\text{min}^{-1}$, respectively, and 294°C, 367°C, and 464°C for 25%, 50%, and 75% load at 2690 $\text{r}\cdot\text{min}^{-1}$, which were measured before the inlet of DOC converter (temperature sensor was 15 cm upstream of the converter).

NO_x , HC, and CO emissions were measured by a gaseous pollutant detector (AMA4000, AVL, Austria). A flame ionization detector (FID) was used to measure THC; non-dispersive infrared analyzer was used to detect CO and CO_2 ; and paramagnetic O_2 analyzer was used for O_2

measurement. The relative standard deviations of the analyzer are less than 1% for NO_x , 5% for CO, 3% for THC, and 0.2% for CO_2 . At each operating condition for regulated gaseous measurements, the sampling values were averaged over a duration of 5 min.

2.2 PM sampling and composition analysis

The particle samples were collected by a MSP Model 130 High-Flow Impactor from MSP Corp (USA). The High-Flow Impactor was installed downstream of a dilution tunnel from Dekati Ltd. (Finland). After dilution, the temperature of the diluted gas was maintained at about 40°C. Based on CO_2 concentrations in exhaust gas and diluted gas, the dilution ratio was calculated as in our previous work [14]. The dilution ratio for particle collection was controlled at 8 during the test. Fine particles ($PM_{2.5}$) typically dominate among the diesel particles. Therefore, two stages were selected for particle sampling according to their diameters: 2.5–10 μm and less than 2.5 μm ($PM_{2.5}$). Particles at the different engine operation conditions were collected onto quartz filters (Pall #2500 QAT2U, PALL, USA) installed in the High-Flow Impactor. To reduce blank carbon levels, the quartz filters were calcined at 450°C for 4 h before used. At each operating condition, the particle sampling time was maintained for 10–20 min at a steady flow of $100\text{ L}\cdot\text{min}^{-1}$. Particle sampling was repeated twice at each operating condition. The schematic diagram of dilution and the particle sampling system is showed in Fig. 1.

Fig. 1 Schematic diagram of dilution and sampling system

OC and EC in PM were measured by a Thermal/Optical Carbon Analyzer (DRI Model 2001, Atmoslytic Inc., Calabasas, CA), using the IMPROVE (Interagency Monitoring of Protected Visual Environments) protocol. A 0.495 cm^2 sample punch from the quartz fiber filter is heated first in a pure helium environment at four steps of 120°C (OC1), 250°C (OC2), 450°C (OC3), 550°C (OC4), whereupon an oxidative atmosphere containing 2% O_2 in helium was applied at temperatures of 550°C (EC1), 700°C (EC2), and 800°C (EC3). The total carbon (TC) emissions were calculated as the sum of OC and EC.

Inorganic water-soluble ions (SO_4^{2-} , NO_3^- , F^- , Cl^- , NO_2^- , NH_4^+ , Na^+ , K^+ , Ca^{2+} , and Mg^{2+}) in particles collected on filters were analyzed by Ion Chromatography (Dionex 600, DIONEX, USA) after the filters were first extracted by ultrapure water.

In the current study, the particles were collected on quartz filters through a one stage dilution system without preheating; the water in exhaust gas could be easily adsorbed by filters and might have influenced the accuracy of the particle weight measurement. Because of this, the $\text{PM}_{2.5}$ mass emissions in this paper were calculated from the carbon analysis results. The carbonaceous mass (CM) was calculated by

$$\text{CM} = \text{EC} + 1.2 \times \text{OC}, \quad (1)$$

where the 1.2 accounts for hydrogen and oxygen present in the OC; EC and OC refer to the mass of EC and OC collected [5]. In this paper, the PM mass was calculated as the addition of CM and the mass of water-soluble ions. Although some other trace elements and ions may also be attributed to the PM mass, the addition of CM and inorganic water-soluble ions was estimated to comprise most of the PM mass.

3 Results and discussion

3.1 Regulated gaseous and $\text{PM}_{2.5}$ brake-specific emissions

The brake specific emissions of NO_x , HC, CO, and $\text{PM}_{2.5}$ from the engine out exhaust and after the DOC converter when using diesel fuel are showed in Fig. 2. For a given speed, the engine out NO_x , HC, and CO emissions decreased with an increase in engine load. The DOC is very effective in CO and HC reduction. The CO reductions at all test modes were higher than 90%: 90%–96% at $2125 \text{ r} \cdot \text{min}^{-1}$ and 95%–97% at $2690 \text{ r} \cdot \text{min}^{-1}$. The HC emissions were reduced by the DOC from 43% to 70% at $2125 \text{ r} \cdot \text{min}^{-1}$ and from 36% to 68% at $2690 \text{ r} \cdot \text{min}^{-1}$. The DOC had very little effect on NO_x emissions; these emissions were slightly decreased with the maximum reduction of 7% at the engine operating mode of 75% load/ $2125 \text{ r} \cdot \text{min}^{-1}$. The NO_2 emissions were not be measured in the current study. It is reported that DOC has the potential to increase NO_2 emissions [15]. In this study, the $\text{PM}_{2.5}$ contributed at least 90% to the PM_{10} emissions. $\text{PM}_{2.5}$ emissions were observed to decrease at all test modes following passage through the DOC. The reduction rates of $\text{PM}_{2.5}$ emissions were 20%–39% depending on the operating modes. The DOC has a high efficiency for HC reduction while it showed limited reduction of $\text{PM}_{2.5}$. This can be explained by the composition of the $\text{PM}_{2.5}$. The condensable volatile hydrocarbons may be oxidized by DOC before forming OC, while the soot produced during combustion was

difficult to oxidize. The DOC reduces the $\text{PM}_{2.5}$ emissions mainly by reducing the OC of the particles.

The reductions of NO_x , CO, and HC emissions by the DOC when the diesel engine was fueled by B20 were similar to the results from diesel fuel and are not shown here.

3.2 Carbonaceous compositions in engine out $\text{PM}_{2.5}$

The EC and OC in $\text{PM}_{2.5}$ from diesel fuel before and after the DOC were analyzed. The results of the EC, OC and TC fractions in $\text{PM}_{2.5}$ were presented as brake specific emissions and illustrated in Fig. 3. For a given engine speed, the EC emissions increased with increasing engine load, that is to say, the EC emissions rose with increasing engine power output. This is understandable since EC consists of soot which is formed mostly during combustion. Higher engine power consumed more fuel and more EC emissions were observed. However, the OC emissions decreased with the increasing load at a given speed. This suggests that volatile hydrocarbons condensed or adsorbed on soot easier at low load than at high load due to the lower temperature associated with low load [16]. The maximum OC emissions were observed at low load (25% here) and contributed to approximately 90% of TC. The change of TC emissions with engine load was the combination of OC and EC emissions. At $2690 \text{ r} \cdot \text{min}^{-1}$, highest TC emissions were found at 25% load.

Table 1 lists the emission factors (EF) for OC, EC, and TC, expressed as g of pollutant / kg of fuel consumed. The change of EC EF with engine loads at two steady speeds was similar to that in the case of brake specific EC emission. The EC EF increased by a factor of more than 5 from 75% load to 25% load at two engine speeds. However, the variation of the OC EF as a function of engine load was less than 2% at the tests loads of $2125 \text{ r} \cdot \text{min}^{-1}$ and 50%, 75% load for $2690 \text{ r} \cdot \text{min}^{-1}$. The EC EF was reduced from 8 to 50% by the DOC while the OC EF was lowered from 18% to 50%, depending on test modes.

By TOR measurements, the OC1 to OC4, Opy fractions of OC and EC1 to EC3 fractions of EC can be obtained. In this study, EC3 was not observed for all samples. The contribution of Opy to OC was less than 1% in most cases. The percentage of carbon, excluding the EC3 and Opy is shown in Fig. 4 with the ratios of OC to EC of $\text{PM}_{2.5}$ at various operating modes. It was found that the OC contributed 63%–90% to TC. OC1 and EC2 were the most abundant compositions in OC and EC. OC1 as a percentage of OC in this study ranged from 66% to 72% at $2125 \text{ r} \cdot \text{min}^{-1}$ and from 65%–67% at $2690 \text{ r} \cdot \text{min}^{-1}$. At $2125 \text{ r} \cdot \text{min}^{-1}$, about 62%–78% EC was attributable to EC2, whereas approximately 32% to 78% EC was composed of EC2 at loads of $2690 \text{ r} \cdot \text{min}^{-1}$.

The ratios of OC to EC also reflect the variation of OC

Fig. 2 Effects of DOC on (a) NO_x , (b) HC, (c) CO, and (d) $\text{PM}_{2.5}$ emissions at various test modes (speed/load): diesel fuel

Fig. 3 Emissions of (a) TC, (b) OC, (c) EC fractions in $\text{PM}_{2.5}$ from engine out and after the DOC at various test modes (speed /load): diesel fuel

and EC emission depending on operating condition. From the values included in Fig. 4, the OC/EC of engine out $\text{PM}_{2.5}$ decreased with increasing load at a given speed. The OC/EC varied from 9.5 to 0.9 and from 5.7 to 1.4 at 2125 and 2690 $\text{r} \cdot \text{min}^{-1}$, respectively. Shi et al. investigated the particle carbon composition at 25%, 50%, and 100% load at two constant engine speeds on a turbocharged DI engine [3]. In that study, the OC and EC in PM were

determined by measuring the emissions of CO_2 produced from samples burnt in pure oxygen within a programmable furnace. The percentage of EC in PM increased with rising load while the percentage of OC in PM decreased. The OC/EC ratio was reported as 0.47 to 2.3 and decreased with the increasing power output. Similar behavior of OC/EC was reported by Ålander et al. [4], who showed that the OC/EC of particles from a DI engine decreased with increasing

Table 1 Effect of DOC on fuel based emission factor (EF) of TC, EC, and OC in PM_{2.5}, diesel fuel

speed (r·min ⁻¹) /load%	EC/(mg·g ⁻¹)		OC/(mg·g ⁻¹)		TC/(mg·g ⁻¹)	
	diesel	diesel + DOC	diesel	diesel + DOC	diesel	diesel + DOC
2125/25	0.016	0.007	0.15	0.108	0.165	0.115
2125/50	0.05	0.037	0.148	0.118	0.198	0.155
2125/75	0.084	0.082	0.144	0.073	0.229	0.155
2690/25	0.032	0.029	0.306	0.228	0.337	0.258
2690/50	0.135	0.12	0.23	0.162	0.365	0.282
2690/75	0.206	0.134	0.221	0.182	0.427	0.317

Fig. 4 Effects of DOC on the percentages of OC (OC1–OC4) and EC (EC1–EC2) fractions in TC and OC/EC ratios: (a) diesel; (b) diesel; (c) B20

engine power. Although it is difficult to compare the results of OC and EC measured by different methods, the variation of OC/EC with engine load was consistent. Shi et al. suggested that dilution condition had a greater influence on the OC fraction due to condensation of semivolatile organic compounds [3]. The OC fraction in PM mostly consists of condensed volatile organic compounds which are formed during the cooling of soot. At low load, the exhaust temperature was lower compared to that at high load, thus the OC fraction in PM was higher at low load. Shah et al. also reported that the OC fraction in TC was higher at idle than at creep: 0.80 OC/EC at idle and 21.80 at creep [5].

A blend of 20% biodiesel with diesel fuel (B20) was prepared and tested at 25%, 50%, and 75% load of 2690 r·min⁻¹. The engine out emissions of TC, OC, and EC fractions in PM_{2.5} from B20 were compared to those from the base diesel fuel in Fig. 5. Compared to diesel fuel, the engine out TC emissions from B20 was lower at the test modes with the reduction of 6%, 28%, and 50% at 25%, 50%, 75% load, respectively. Except at 25% load for

2690 r·min⁻¹, the EC and OC emissions from B20 were lower than those from diesel fuel. EC emissions were decreased by B20 at all test modes with reductions of 39%–58%. The OC/EC ratio in engine out PM_{2.5} from B20 was higher than that from diesel fuel and a significant increase in OC/EC was observed at 25% load. Most researchers reported an increase in SOF by using biodiesel or biodiesel fuel blends [10]. For instance, Bagley et al. [17] reported that the SOF of the total PM was 57% for diesel fuel and 80% for B100 and explained that it was due to a shift to higher carbon number fuel-related organic species in the exhaust that were less volatile. The increased OC/EC with the B20 is consistent with our previous study [14]. Figure 4 shows that the OC/EC in engine out PM_{2.5} also decreased with increasing load similar to the case of diesel fuel.

3.3 Effect of DOC on carbonaceous compositions in PM_{2.5}

The DOC lowered both OC and EC emissions when using diesel fuel, as shown in Fig. 3. The TC emissions were

Fig. 5 Emissions of (a) TC, (b) OC, and (c) EC fractions in $PM_{2.5}$ from engine out and after DOC using B20 and compared those with base diesel fuel

decreased from 21%–32% by DOC. The reductions of OC and EC by DOC were variable depending on the test modes. At $2125 \text{ r} \cdot \text{min}^{-1}$, the maximum reduction of OC by the DOC was observed at the 75% load, at which OC emissions were reduced approximately 50%. At $2690 \text{ r} \cdot \text{min}^{-1}$, the reduction of OC was in the range of 17%–30%. At most test modes, EC emissions were significantly decreased by the DOC with reductions in the range of 2%–53% at $2125 \text{ r} \cdot \text{min}^{-1}$ and 11%–41% at $2690 \text{ r} \cdot \text{min}^{-1}$. The EF of TC, OC and EC were also reduced by the DOC. At loads of $2125 \text{ r} \cdot \text{min}^{-1}$, the OC EF were lowered by the DOC from 20%–49% and EC EF were reduced by the DOC from 2%–53%. At loads of $2690 \text{ r} \cdot \text{min}^{-1}$, OC EF were reduced by the DOC from 25%–50% and the EC EF were reduced by 9%–35%. The effect of the DOC on the carbonaceous compositions in $PM_{2.5}$ is shown in Fig. 4. A significant decrease in the OC/EC ratio by the DOC was observed at 25% load, $2690 \text{ r} \cdot \text{min}^{-1}$, due to the high OC fraction in engine out particles at this low load.

It is reported that the primary effect of the DOC on PM is the reduction in the OC fraction (or SOF) [4,7,8,16]. Vaaraslahti et al. investigated the effect of a conventional DOC and a particle oxidation catalyst on the diesel particles [16]. They reported that the two catalytic converters significantly decreased the SOF in PM. Shah et al. examined the effect of four different control technologies on the PM emitted from two diesel fuels with different sulfur content [5]. The honeycomb-structured DOC showed a slight reduction in EC, but a significant reduction in OC emissions (56%–77%).

In the current study, the effect of the DOC on the EC fraction was much more significant than that reported by others. Tang et al. reported that for all of the fuels studied the TC mass emissions were reduced by the DOC and the EC/OC ratio was increased due to more reduction of OC than EC emissions [18]. In this study, the variation of the

OC/EC ratio after the DOC was not consistent and depended on operating conditions. For diesel fuel, the OC/EC after the DOC increased at 25% load.

The DOC showed less effect on the OC fraction in $PM_{2.5}$ from B20 at high load compared to diesel fuel. An unexpected result was observed in the case of B20 – the EC emissions increased after the DOC at 25% and 50% load. Bagley et al. found that the reduction in SOF by a DOC using pure biodiesel was slightly lower than that using diesel fuel [17].

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