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Effects of FAME biodiesel and HVORD on emissions from an older-technology diesel engine

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Abstract

The results of laboratory evaluations were used to compare the potential of two alternative, biomass-derived fuels as a control strategy to reduce the exposure of underground miners to aerosols and gases emitted by diesel-powered equipment. The effects of fatty acid methyl ester (FAME) biodiesel and hydrotreated vegetable oil renewable diesel (HVORD) on criteria aerosol and gaseous emissions from an older-technology, naturally aspirated, mechanically controlled engine equipped with a diesel oxidation catalytic converter were compared with those of widely used petroleum-derived, ultralow-sulfur diesels (ULSDs). The emissions were characterized for four selected steady-state conditions. When fueled with FAME biodiesel and HVORD, the engine emitted less aerosols by total particulate mass, total carbon mass, elemental carbon mass and total number than when it was fueled with ULSDs. Compared with ULSDs, FAME biodiesel and HVORD produced aerosols that were characterized by single modal distributions, smaller count median diameters, and lower total and peak concentrations. For the majority of test cases, FAME biodiesel and HVORD favorably affected nitric oxide (NO) and adversely affected nitrogen dioxide (NO₂) generation. Therefore, the use of these alternative fuels appears to be a viable tool for the underground mining industry to address the issues related to emissions from diesel engines, and to transition toward more universal solutions provided by advanced engines with integrated exhaust after treatment technologies.

Introduction

Diesel engines have been the workhorses of modern society for industrial and transportation enterprises. Diesel engines of all generations are extensively used in a variety of light-, medium- and heavy-duty applications in underground mining operations around the world. Fuels obtained from various sources and with wide-ranging chemistries have been combusted in diesel engines. However, the bulk of the currently used fuel can still be traced to nonrenewable petroleum sources. The petroleum-derived diesel fuels are primarily made up of saturated acyclic hydrocarbons (paraffins or alkanes), aromatic hydrocarbons (arenes or

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aryl hydrocarbon), and unsaturated hydrocarbons with double bonds (olefins and alkenes). Improvements in the quality of petroleum-based diesel fuels was identified in federal regulations as an important piece in resolving the puzzle of diesel engine emissions (Environmental Protection Agency, 2001, 2004). The reduction of U.S. diesel fuel sulfur content below 15 ppm had the effect of lowering sulfate emissions and enabling the development and implementation of catalyzed emissions control technology across a wide spectrum of applications.

The efforts to reduce dependency on petroleum products, reduce emissions of greenhouse gases and improve the quality of fuels have resulted in increased production of renewable biofuels such as fatty acid methyl ester (FAME) biodiesel, hydrotreated vegetable oil renewable diesel (HVORD), and biomass-to-liquid (BLT) diesel. In addition, biofuels when used in high-concentration blends are perceived as a viable control strategy to reduce emissions from a variety of diesel-powered fleets (Durbin et al., 2007) as well as exposure of workers to diesel aerosols (Bugarski et al., 2010; Bugarski et al., 2014; Bugarski, Hummer and Vander-slice, 2015).

FAME biodiesel fuels are obtained from various plant and algae oils and from animal fats through the process of transesterification (Graboski and McCormick, 1998; Wahlen et al., 2013). The chemical and physical properties of FAME fuels are highly dependent on feedstock: for example, FAME biodiesel produced from soybean oil is primarily made up of unsaturated oleic and linoleic fatty acid while the biodiesel produced from palm oil is primarily made up of saturated palmitic and unsaturated oleic fatty acid. FAME biodiesels contain on average between 10 and 11 percent oxygen. HVORD fuels are made from vegetable and algae oils and animal fats through hydrogenation and isomerization processes (Huber, O'Connor and Corma, 2007; Aatola et al., 2008; Smagala et al., 2013). By chemical composition, HVORD consists of mixtures of paraffinic and isoparaffinic hydrocarbons and is virtually free of aromatic hydrocarbons, metals, sulfur, nitrogen and oxygen-containing compounds. HVORD was recognized to contribute less to the life cycle emissions of greenhouse gases than FAME biodiesel (Sunde, Brekke and Solberg, 2011; Yano et al., 2015), and as such is perceived as the second generation of biofuels. The fuels produced from biomass using Fischer-Tropsch processes, also known as BLT fuels, are perceived as the future of diesel fuels.

Diesel particulate matter (DPM) has been shown to have adverse health outcomes on the pulmonary system, cardiovascular system and brain (International Agency for Research on Cancer, 2012; Mills et al., 2005; Power et al., 2011; Lung et al., 2014). Long-term exposure to DPM in the confined spaces of occupational settings, such as underground mines and trucking depots, was linked to an increase in lung cancer risk (Attfield et al., 2012; Silverman et al., 2012; Garshick et al., 2012). Mounting concern about those adverse health outcomes resulted in extensive efforts to reduce exposures of the general population and workers to DPM, nitrogen oxides and other pollutants emitted by diesel engines. Because of their favorable effects on DPM and some gaseous emissions, biofuels — primarily FAME biodiesels — were for some time used as high-biodiesel-concentration blends or neat as a strategy to reduce diesel emissions or the exposure of workers to those pollutants (Bugarski et al., 2012). The fact that diesel engines, when fueled with FAME biodiesels in place of

ULSD, emit less total DPM by mass (Yuan et al., 2007; Sappok and Wong, 2008; Gerlofs-Nijland et al., 2013) was extensively exploited to reduce concentrations of diesel aerosols and gases in underground mines (Bugarski et al., 2010; Bugarski et al., 2014). In addition, FAME biodiesels were found to have favorable effects on carbon monoxide and hydrocarbon emission reductions (Schönborn et al., 2009; Hoekman and Robins, 2012; Bugarski et al., 2014). It is universally accepted that those reductions in particulate, carbon monoxide and hydrocarbon emissions are primarily the result of the presence of fuel-bound oxygen in FAME biodiesel fuels (Schönborn et al., 2009).

Despite the advantages of FAME biodiesel fuels, several potential drawbacks for their use as a control strategy have been identified. A number of studies linked the use of FAME biodiesel in place of petroleum-derived ULSD fuels with a small increase in nitrogen oxide ($\text{NO}_x = \text{NO} + \text{NO}_2$) emissions (Bittle, Knight and Jacobs, 2010; Hoekman and Robins, 2012; Muller, Boehman and Martin, 2014). Muller, Boehman and Martin (2014) showed that the effects of soy FAME biodiesel on increased NO_x emissions are the result of a number of coupled synergistic and antagonistic mechanisms, including those that produce higher local and average in-cylinder temperatures, advance combustion events, and changes in fuel and jet structure. The effects of FAME bio-diesel on NO_x emissions appear to depend on a number of parameters, including engine technology, certification level (Durbin et al., 2007; Hoekman and Robins, 2012), and engine operating conditions (Muller, Boehman and Martin, 2014). The aerosols emitted by engines operated on FAME biodiesel fuels were found to be characterized by smaller median diameters than the corresponding size distributions observed for the ULSD (Bugarski et al., 2010; Bugarski et al., 2014). The formation of higher number concentrations in nucleation mode aerosols relative to ULSD were reported by Schönborn et al. (2009) for several types of FAME bio-diesels. Schönborn et al. (2009) found that concentrations of nucleation mode aerosols were highest for the long-chained, fully saturated FAMES. It is important to note that the formation of nucleation mode aerosols is strongly affected by dilution and environmental conditions, and that increase in nucleation particles was not observed in the studies conducted in underground environments where soybean oil-derived FAME biodiesels were used (Bugarski et al., 2010; Bugarski et al., 2014).

Aerosols produced by diesel engines combusting FAME biodiesels in place of petroleum-derived diesel fuels might have higher pulmonary (Shvedova et al., 2013; Yanamala et al., 2013; Fukagawa et al., 2013) and reproductive (Kisin et al., 2015) toxicity. Several studies linked the increase in oxidative stress related to the use of FAME biodiesel fuels to a larger presence of oxygenated organic species in FAME aerosols than in petroleum-derived aerosols (Javala et al., 2012; Stevanovic et al., 2013). Also, Kooter et al. (2011) and Gerlofs-Nijland et al. (2013) found that particulates emitted by diesel engines fueled with neat and blended FAME biodiesel have similar oxidative potential but much higher cytotoxicity than particulates generated by the same engines fueled with petroleum diesel.

Recently, some underground operations in the United States started fueling their diesel-powered equipment with blends of HVORD. HVORD has properties very similar to ULSD and can be used in diesel engines without any modifications. HVORD is favored over FAME biodiesel due to lower environmental impact (Yano et al., 2015). Compared with ULSD,

HVORD has favorable effects not only on particulate, carbon monoxide and total hydrocarbon emissions but also NO_x emissions (Aatola et al., 2008; Happonen et al., 2012; Kim et al., 2014; Westphal et al., 2013; Bugarski et al., 2015). Westphal et al. (2013) found that HVORD particulate extracts have lower mutagenicity than ULSD and rape-seed and jatropha FAME biodiesel particulate extracts. The particulates generated from HVORD were found to have oxidative potential lower than particulates generated from ULSD and rapeseed oil-based FAME biodiesel (Javala et al., 2012).

Switching the fuel supply from petroleum-based fuels to alternative fuels has some challenges. The issues with FAME biodiesel fuels used in high concentration blends are those operational problems associated with oxidative stability, engine oil dilution, formation of deposits in fuel injection systems, compatibility with some materials, and low-temperature operability (National Renewable Energy Laboratory, 2009). Due to technical issues with high-pressure injection systems in the new-technology diesel engines, the majority of engine manufacturers only support the use of blends with low FAME-biodiesel content. The main restriction of HVORD was found to be compatibility with some materials (Smagala et al., 2013), lubricity and cold flow properties (Lapuerta et al., 2011).

The results of direct comparison of the effects of FAME biodiesels, HVORD and petroleum diesel on emissions from turbocharged, electronically controlled engines are available from the literature (Hajbabaei et al., 2012; Westphal et al., 2013; Kim et al., 2014), but the equivalent information for mechanically controlled, naturally aspirated engines is not readily available. This information is critical to underground mining operators that still have large fleets of light- and medium-duty vehicles powered by those engines.

Methodology

The current study was conducted to directly compare the effects of corn oil-based FAME biodiesel and HVORD on the performance and the particulate and gaseous emissions of an older-technology, naturally aspirated, mechanically controlled engine equipped with a diesel oxidation catalytic converter (DOC). The results were used to assess the potential of those alternative fuels as a control strategy for reducing exposure of underground miners to diesel emissions.

The emissions were characterized for an engine operated with two similar ULSDs from the same local supplier, as baseline fuels; neat corn-based FAME biodiesel; and neat HVORD. Although the baseline fuels were from two different batches, they had similar properties (Table 1). The neat corn-based FAME biodiesel was supplied by Peter Cremer NA (Cincinnati, OH) and the neat HVORD was supplied by Neste Oil's Porvoo refinery. The results of analysis performed on the fuels by Cashman Fluids Laboratory (Sparks, NV) are summarized in Table 1, which show that HVORD had substantially higher cetane number and API gravity than the other fuels.

The layout of the sampling and measurement systems used in this study is shown in Fig. 1. The 1999 Isuzu C240 (Isuzu Motors Ltd, Tokyo, Japan) older-technology, mechanically controlled, naturally aspirated and directly injected nonroad light-duty diesel engine

conforms to U.S. EPA Tier 1 emissions standards. The engine was not adjusted to compensate for the substantial differences in physical and chemical properties between the tested fuels. In order to simulate practice in the underground mines that are using biofuels, the engine was retrofitted with a DOC from Lubrizol (New Market, Ontario, Canada). The DOC is representative of those traditionally marketed to the underground mining industry for effective control of carbon monoxide and hydrocarbon emissions. The engine was coupled to an SAJ SE150 (Pune, India) 150-kW water-cooled, eddy-current dynamometer. Testing was done for four steady-state operating conditions: (1) intermediate speed, 50 percent load (I50), (2) intermediate speed, 100 percent load (I100), (3) rated speed, 50 percent load (R50), and (4) rated speed, 100 percent load (R100). The results for at least three runs were used to calculate averages.

A fuel measurement system supplied by Max Machinery Inc. was used to measure mass-based fuel consumption. The aerosol sampling and measurements were conducted in DOC-out exhaust diluted approximately 30 times ($DR = 30$) in FPS4000 partial dilution system supplied by Dekati (Tampere, Finland). This dilution rate is typical of that of the diesel engines operated in underground mines in the United States. In the dilution system, the exhaust was diluted in two stages: the primary dilution ($DR \sim 1.7$) occurred in the perforated disk diluter, and the secondary dilution ($DR \sim 17$) occurred in the ejector diluter. The residence chamber was inserted between those two stages to allow for potential formation of nucleation aerosols. The effects of the fuels on mass concentrations of aerosols emitted by the test engine were assessed using the results of the gravimetric and carbon analysis performed on triplicate filter samples of DOC-out exhaust collected from the dilution system using custom-designed sampling systems. The carbon analysis on DPM samples was performed using the thermal optical transmittance-evolve gas analysis (TOT-EGA) known as NIOSH Method 5040, from the U.S. National Institute for Occupational Safety and Health (NIOSH, 1999). Number concentrations and size distributions of aerosols in diluted exhaust were measured using a TSI 3936 scanning mobility particle sizer spectrometer (TSI Inc., Shoreview, MN). The effects of the fuels on concentrations of NO and NO₂ in the DOC-out exhaust were determined using the results of measurements performed in undiluted exhaust with a Gaset DX-4000 Fourier transform infrared (FTIR) gas analyzer (Gaset Technologies, Helsinki, Finland).

Results and discussion

Effects of FAME biodiesel and HVORD on fuel consumption

The inherent energy content, typically expressed in terms of heating value per mass of the fuel, is traditionally considered to be the primary property affecting fuel consumption. In the case of fuels used in this study, the heating values of FAME biodiesel and HVORD were approximately 10 percent lower and 1 percent higher, respectively, than that of the corresponding ULSDs (Table 1). However, the direct mass-based measurements of the fuel consumption showed that on average, the test engine consumed more of both biofuels than respective ULSDs in all test cases (Fig. 2). In the case of the I50, I100 and R50 tests, the relative increases in the mass of fuel consumed were higher for FAME biodiesel than for HVORD. For R100, the changes in mass-based fuel consumption were quite similar for the

FAME biodiesel and HVORD cases. These results indicate that, due to substantial differences in the specific gravities of the evaluated fuels, one should also consider the energy capacity expressed per volume of fuel when assessing fuel consumption (Lapuerta et al., 2011). The findings of this study relative to fuel consumption are in general agreement with the findings for FAME (Graboski and McCormick, 1998; Wahlen et al., 2013; Kim et al., 2014) and HVORD fuels (Kim et al., 2014).

Effects of FAME biodiesel and HVORD on DPM emissions

The effects of the fuels on the mass concentration of DPM were assessed using the averaged results of gravimetric analysis and TOT-EGA performed on the DOC-out samples. Compared with the corresponding ULSDs, both FAME biodiesel and HVORD contributed substantially less to total mass concentrations of DPM emitted by the test engine (Fig. 3a). For I50, R50 and R100 conditions, the differences in reductions in total mass concentration (TMC) between those two fuels were within the margin of experimental error. For I100 conditions, the reductions in total mass concentrations were more substantial for HVORD than for FAME biodiesel. Figures 3b and 3c show that both FAME biodiesel and HVORD reduced mass concentrations of total carbon (TC) and elemental carbon (EC). For I50, R50 and R100 conditions, the reductions in TC and EC were more substantial for FAME biodiesel than for HVORD. Only in I100 conditions were the reductions in TC and EC more substantial for HVORD. The marginal reductions in TC and EC emissions for I100 conditions when the engine was fueled with FAME biodiesel could be attributed to poor combustion of that fuel at peak torque conditions.

The effects of the fuels on the size distributions of aerosols were examined using the results of selected measurements performed in diluted exhaust. The statistical parameters for the observed size distributions are summarized in Table 2. The concentrations were normalized to a dilution ratio of 30 ($DR = 30$). For both tested fuels, aerosols emitted by the test engine were distributed in single accumulation mode (Table 2). For all test conditions, use of FAME bio-diesel and HVORD resulted in size distributions that were characterized by smaller count median diameters (CMDs) and lower total number concentrations (TNCs) of aerosols compared with the corresponding ULSD tests.

Compared with ULSD, both FAME biodiesel and HVORD reduced the average TNCs of aerosols in the exhaust of the tested engine (Fig. 4). For I50, I100 and R50 conditions, the observed differences in reductions in TNC for FAME biodiesel and HVORD were within the margin of experimental error. For R100 conditions, the reductions were more substantial for FAME biodiesel.

These observations on the effects of FAME biodiesel and HVORD on aerosol emissions are in general agreement with the findings of a number of studies conducted using turbocharged, electronically controlled engines operated on neat FAME biodiesel (Yuan et al., 2007; Sappok and Wong, 2008) and HVORD (Aatola et al., 2008; Kim et al., 2014; Westphal et al., 2013).

Effects of FAME biodiesel and HVORD on emissions of nitrogen oxides

The effects of FAME and HVORD on averaged NO emissions are shown in Fig. 5a. For a majority of the cases, using the alternative fuels in place of ULSD resulted in lower NO concentrations in the exhaust. In general, the NO₂ levels in the DOC-out exhaust were relatively low for the I50 and R50 conditions and slightly elevated for the I100 and R100 conditions (Fig. 5b). This difference can be explained by the effects of exhaust temperature on catalyst activity, and oxidation of NO to NO₂. The effect of FAME biodiesel on averaged NO₂ concentrations was favorable in the case of I50 conditions and adverse in the case of R100 conditions. For the other two test cases, the NO₂ concentrations were similar to those observed for ULSD. For all test conditions when the engine was fueled with HVORD, the DOC-out NO₂ concentrations were substantially higher than for the corresponding cases when the engine was fueled with ULSD.

With the exception of the R50 tests, emissions of nitrogen oxides (NO_X = NO + NO₂) were found to be lower when the engine was fueled with both FAME and HVORD than with ULSDs. This finding is in disagreement with the slight increase in NO_X emissions previously reported when FAME biodiesel was used in turbocharged, electronically controlled engines in place of ULSDs (Bittle et al., 2010; Hoekman and Robins, 2012; Muller et al., 2014) and in general agreement with reductions in NO_X emissions previously observed when HVORD was used in similar engines in place of ULSDs (Aatola et al., 2008; Kim et al., 2014; Westphal et al., 2013).

Conclusion

This study shows that FAME biodiesel and HVORD both had favorable effects on DPM, TC and EC emissions from an older, mechanically controlled, naturally aspirated engine. The magnitude of reductions in total mass concentrations of DPM, TC and EC in the exhaust were found to be comparable for FAME biodiesel and HVORD. Combustion of these alternative fuels in place of ULSD also produced aerosols with smaller median diameters and in lower number concentrations. However, the combustion of all of these fuels with drastically different chemical compositions produced emissions with different chemical compositions and toxicities (Javala et al., 2012; Shvedova et al., 2013; Yanamala et al., 2013; Fukagawa et al., 2013; Westphal et al., 2013; Kisin et al., 2015).

Use of these alternative fuels appears to be a viable tool for the underground mining industry to address the issues related to emissions from older- and newer-technology diesel engines and transition toward more universal solutions provided by advanced engines with integrated exhaust after-treatment technologies (Bugarski et al., 2012; Scheepers and Vermeulen, 2012). The benefits of using biofuels as a DPM emissions control strategy would be relatively limited in the case of diesel engines equipped with diesel particulate filters. More research on the toxicology outcomes is warranted before a wide implementation of these biofuels, particularly HVORD, occurs.

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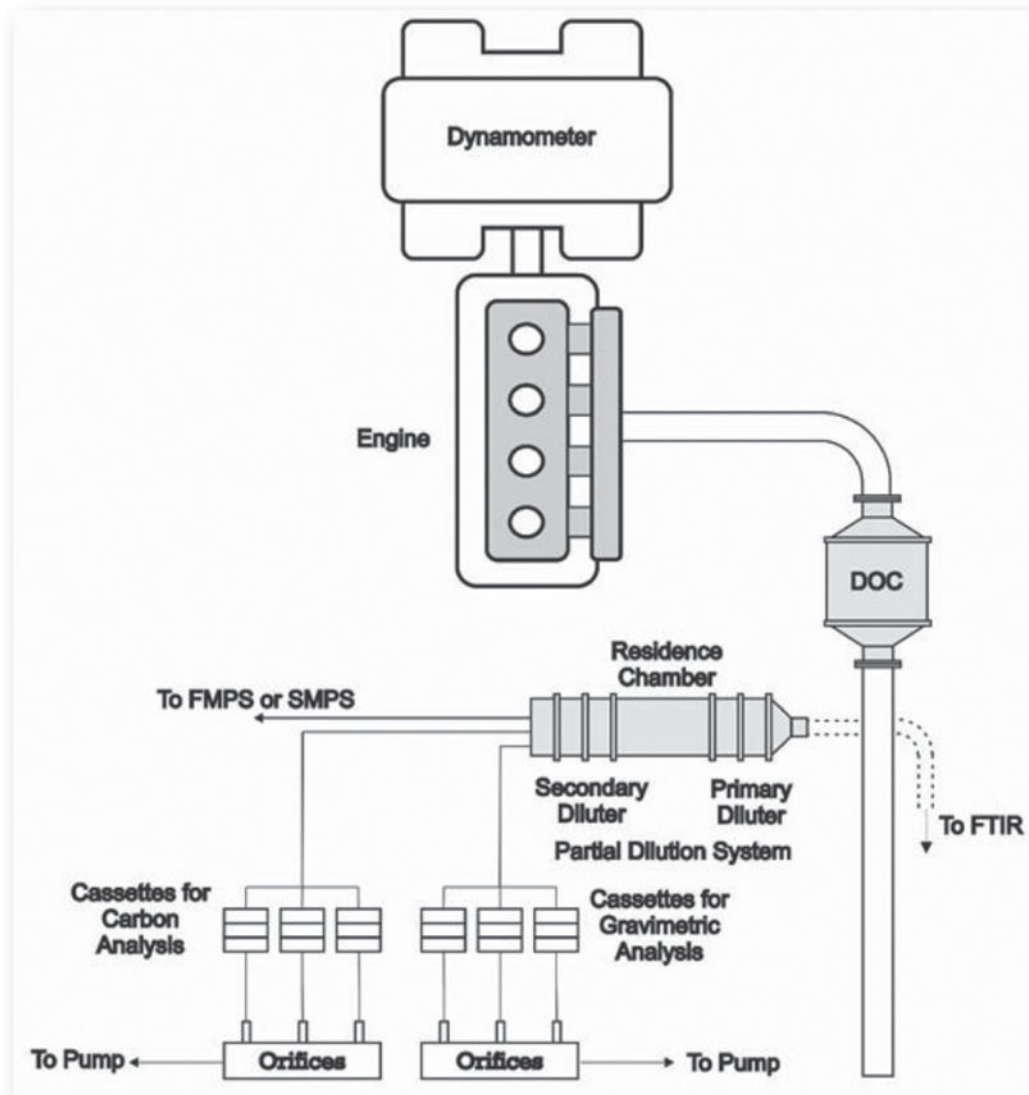


Figure 1.
Experimental layout.

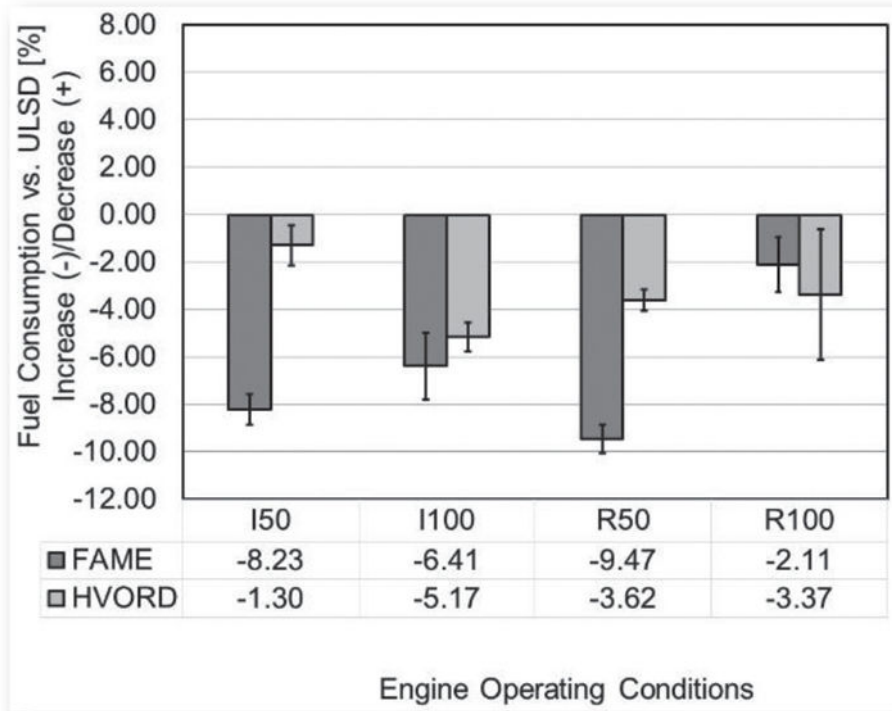


Figure 2.
Changes in fuel consumption with respect to ULSD.

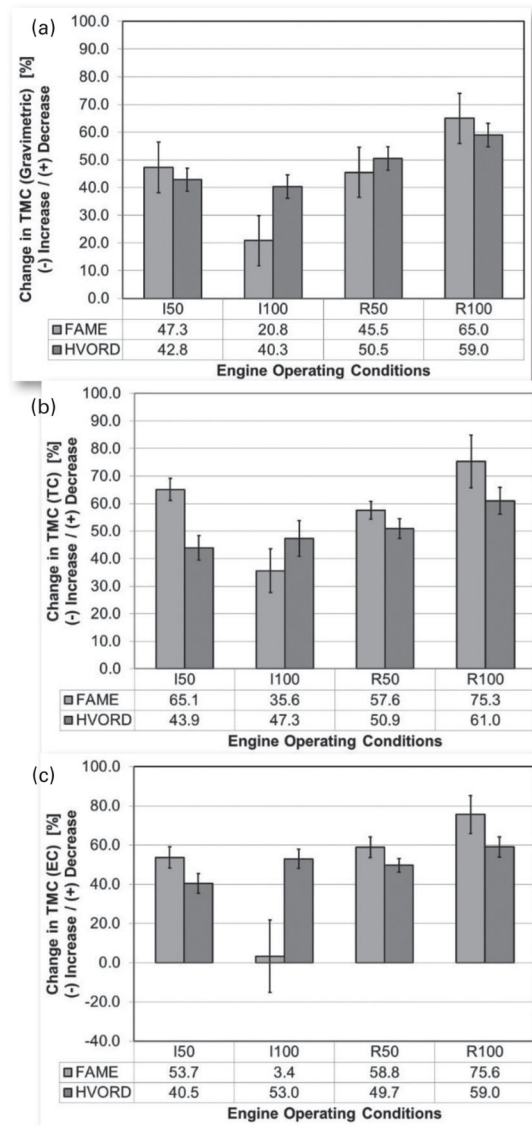


Figure 3. Effects of the tested fuels on the total mass concentrations (TMC) of aerosols as determined by (a) gravimetric analysis, (b) carbon analysis as total carbon (TC) and (c) carbon analysis as elemental carbon (EC).

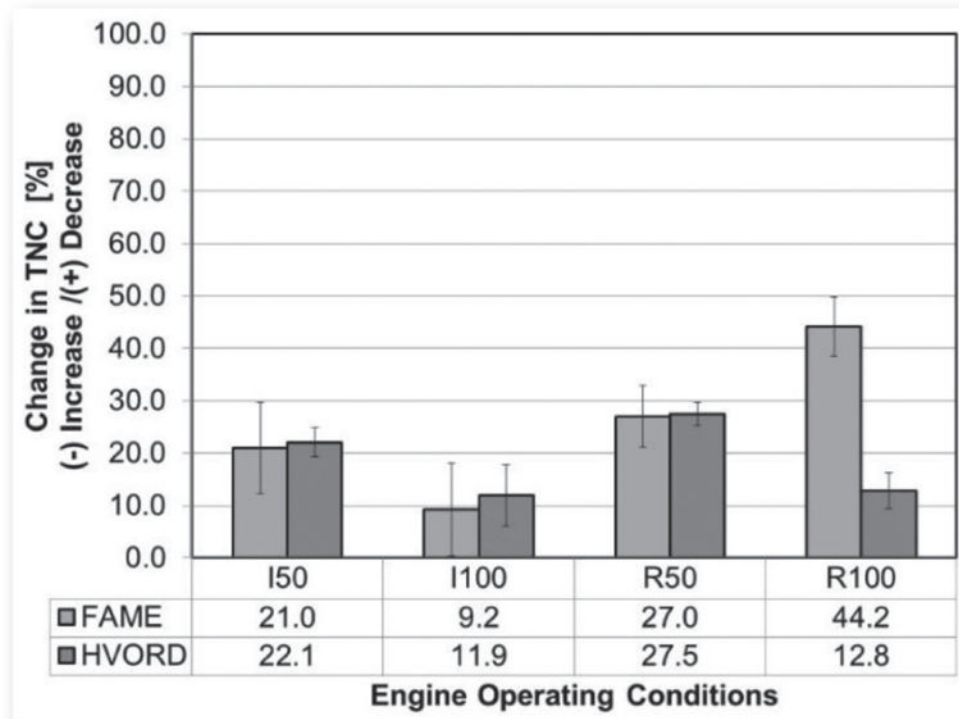


Figure 4.
Effects of the tested fuels on the total number concentrations (TNC) of aerosols.

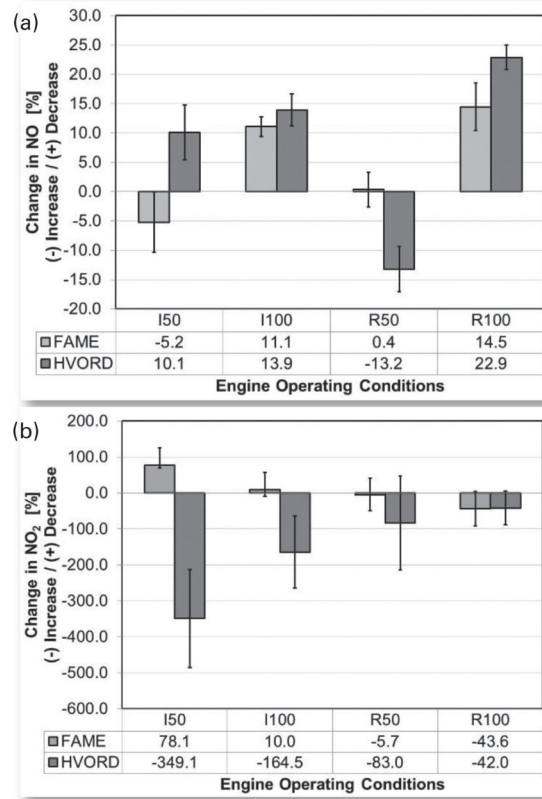


Figure 5. Effects of the tested fuels on (a) NO and (b) NO₂ concentrations.

Table 1

Properties of fuels used in this study.

Fuel properties	Test method	ULSD (FAME)	FAME	ULSD (HVORD)	HVORD
Aromatics content (vol %)	ASTM D1319	–	–	24.2	<5.0
Olefins content (vol %)	ASTM D1319	–	–	1.6	1.2
Saturates content (vol %)	ASTM D1319	–	–	74.2	>95.0
Fatty acid methyl ester content (%)	ASTM 7371	0.0	100.0	0.0	0.0
Flash point, closed cup (°C)	ASTM D93	60.5	180.0	62.5	86.6
Sulfur, by UV (ppm)	ASTM D5453	4.9	0.1	7.4	0.0
Cetane number	ASTM D613	45.3	51.2	44.5	75.2
API gravity @ 15.6 °C (°API)	ASTM D1298	35.0	28.8	36.9	49.9
Heat of combustion (MJ/kg)	ASTM D240	45.1	41.2	45.9	46.4

Table 2

Statistical parameters for size distributions of aerosols in diluted exhaust (DR = 30) observed during FAME and HVORD tests (EOC = experimental operating conditions, TNC = total number concentration; CMD = count median diameter, σ = spread).

Aerosol size distributions	EOC	FAME tests			HVORD tests		
		TNC #/cm ³	CMD nm	σ	TNC #/cm ³	CMD nm	σ
Baseline fuels	I50	1.53×10^6	58.8	1.62	1.75×10^6	57.8	1.61
	I100	1.91×10^6	65.0	1.58	2.90×10^6	68.0	1.68
	R50	2.38×10^6	53.9	1.59	2.52×10^6	53.2	1.57
	R100	2.10×10^6	55.7	1.61	2.08×10^6	59.6	1.58
	I50	1.19×10^6	44.4	1.66	1.45×10^6	50.4	1.64
Alternative fuels	I100	1.76×10^6	57.0	1.53	2.26×10^6	62.2	1.54
	R50	1.85×10^6	44.1	1.59	1.86×10^6	47.1	1.60
	R100	1.24×10^6	40.6	1.69	1.72×10^6	46.7	1.62