Fuel Properties and Emissions of Soybean Oil Esters as Diesel Fuel

David Y.Z. Chang[a], Jon H. Van Gerpen[a,*] Inmok Lee[b], Lawrence A. Johnson[b],
Earl G. Hammond[b], and Stephen J. Marley[c]

[a]Department of Mechanical Engineering, [b]Center for Crops Utilization Research and Department of Food Science and Human Nutrition, and [c]Department of Agriculture and Biosystems Engineering, Iowa State University, Ames, Iowa 50011

ABSTRACT: The effects of using blends of methyl and isopropyl esters of soybean oil with No. 2 diesel fuel were studied at several steady-state operating conditions in a four-cylinder turbocharged diesel engine. Fuel blends that contained 20, 50, and 70% methyl soyate and 20 and 50% isopropyl soyate were tested. Fuel properties, such as cetane number, also were investigated. Both methyl and isopropyl esters provided significant reductions in particulate emissions compared with No. 2 diesel fuel. A blend of 50% methyl ester and 50% No. 2 diesel fuel provided a reduction of 37% in the carbon portion of the particulates and 25% in the total particulates. The 50% blend of isopropyl ester and 50% No. 2 diesel fuel gave a 55% reduction in carbon and a 28% reduction in total particulate emissions. Emissions of carbon monoxide and unburned hydrocarbons also were reduced significantly. Oxides of nitrogen increased by 12%.

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KEY WORDS: Biodiesel, diesel engine, engine emissions, engine performance, fuel properties, isopropyl ester, methyl soyate, particulate emissions, winterization.

Diesel engines are widely used as power sources for medium- and heavy-duty applications because of their lower fuel consumption and emissions of carbon monoxide (CO) and unburned hydrocarbon (HC) compared with gasoline-fueled engines. Alternate fuels for diesel engines have become increasingly important due to decreasing petroleum reserves and the environmental consequences of exhaust gases from petroleum-fueled engines. A number of studies have shown that vegetable oils hold promise as alternative fuels for diesel engines (1,2). However, the high viscosity, low volatility, and poor cold flow properties of vegetable oils, which result in severe engine deposits, injector coking and piston ring sticking, have prevented vegetable oils from being used directly in diesel engines (3–7). One way to improve the fuel properties of vegetable oils is transesterifying the oils with alcohols to form monoesters. The monoesters of vegetable oils have fuel properties that are much closer to No. 2 diesel fuel than the oils (2,8,9) and provide similar engine performance with attractive emission levels. The esters contain 10 to 11% oxygen, by weight, which may encourage more complete combustion than petroleum-based fuels in engines. Such monoesters also have high cetane numbers and contain no sulfur and no aromatics. These characteristics of monoesters also reduce the CO, HC, and particulates in exhaust gas, compared with No. 2 diesel fuel. Schumacher *et al.* (10) tested neat methyl esters of soybean oil (methyl soyate) as a fuel in a 5.9-L direct-injection turbocharged Cummins diesel engine, installed in a Dodge pickup, and found that the engine power with neat methyl soyate was 5% less than that of No. 2 diesel fuel. The CO emissions were about the same, and HC and particulate emissions were reduced by 48 and 20%, respectively. Oxides of nitrogen (NO_x) emissions were increased by 13%. These results confirmed earlier work by Clark *et al.* (11). Alfuso *et al.* (12) reported similar results with methyl ester of rapeseed oil as a fuel.

One serious limitation to the use of monoesters of vegetable oils is their tendency to crystallize at low temperatures. Methyl and ethyl esters of soybean oil will crystallize and separate from diesel fuel at temperatures often experienced in winter time operation. One solution to this problem may be the use of branched-chain esters, such as isopropyl esters. The isopropyl esters of soybean oil crystallize 7–11°C lower than the corresponding methyl esters (13). Another method to improve the cold flow properties of vegetable oil esters is to remove high-melting saturated esters by inducing crystallization with cooling, a process known as winterization.

The objectives of this investigation were to compare the fuel properties of methyl, isopropyl, and winterized methyl esters of soybean oil with those of commercial No. 2 diesel fuels and to compare the exhaust emission levels of fuel blends of 20, 50, and 70% esters with No. 2 diesel fuels.

EXPERIMENTAL PROCEDURES

Test fuels. Five fuels were selected as the base fuels for this study. Two commercial diesel fuels were used as baseline fuels: a high-sulfur No. 2 diesel fuel and a low-sulfur No. 2 diesel fuel. Methyl esters of soybean oil were purchased from Interchem Environmental, Inc. (Overland Park, KS), and iso-

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*To whom correspondence should be addressed at 2022 Black Engineering Building, Department of Mechanical Engineering, Iowa State University, Ames, IA 50011.

propyl esters and winterized methyl esters of soybean oil were prepared in the facilities of the Center for Crops Utilization Research at Iowa State University (Ames, Iowa). The fuel blends tested are shown in Table 1. The key fuel properties investigated in this study were cetane number and fuel composition. The cetane number was determined by Phoenix Chemical Laboratory (Chicago, IL), and the composition of the monoesters of soybean oil by the Food Science and Human Nutrition Department at Iowa State University.

Test engine. The engine used in this study was a John Deere model 4276T four-cylinder, four-stroke, turbocharged diesel engine with a bore of 106.5 mm, a stroke of 127.0 mm, a displacement of 4.53 L, and a compression ratio of 16.8:1. The combustion system of the engine was a bowl-in-piston, direct-injection, medium-swirl type. It was connected to a 150 HP General Electric (Schenectady, NY) model TLC 2544 direct current dynamometer.

Test procedure. The engine test procedures described below were largely dictated by the small amounts of fuel available for testing. There were four fuel groups, and each of them included No. 2 diesel fuel for comparison. Fuels in the first group were tested under two steady-state engine operating conditions, 20 and 100% of full load at 1400 rpm, and the other three fuel groups were tested only at the 100% condition.

To determine the gaseous emissions, a portion of the engine exhaust gas was passed through a desiccant dryer for measurement of CO and CO_2 by infrared analyzers, and O_2 by a polarigraphic oxygen monitor. Another portion of the engine exhaust gas was directly drawn from the exhaust pipe through a 450 K heated sampling line to determine the concentrations of unburned HC by a heated flame-ionization detector, and NO_x by a chemiluminescence analyzer.

Total particulate emissions were measured with a full-flow dilution tunnel. The diluted sample passed through two 110-mm Pallflex T60A20 (Putnam, CT) glass fiber filters, which were weighed before the engine test, and again 24 h after they were loaded and returned to a temperature- and humidity-controlled chamber. The total particulate matter (TPM) mass was the net weight gain of the filters. The soluble organic fraction (SOF) of the particulate was determined by Soxhlet extraction of the loaded filters with methylene chloride. Data presented in this paper are means of three duplicate tests. All data were analyzed statistically by means of least significant difference (LSD) at $P < 0.05$.

TABLE 1
Tested Fuels

Group	Fuels	Engine test conditions
	HSD2[a]	20%, 100% load @ 1400 rpm
1	20% MS[b] + HSD2	20%, 100% load @ 1400 rpm
	50% MS + HSD2	20%, 100% load @ 1400 rpm
	70% MS + HSD2	20%, 100% load @ 1400 rpm
	LSD2[c]	100% load @ 1400 rpm
2	20% IP[d] + LSD2	100% load @ 1400 rpm
	50% IP + LSD2	100% load @ 1400 rpm
	LSD2	100% load @ 1400 rpm
3	20% MS + LSD2	100% load @ 1400 rpm
	50% MS + LSD2	100% load @ 1400 rpm
	LSD2	100% load @ 1400 rpm
4	20% WMS[e] + LSD2	100% load @ 1400 rpm
	50% WMS + LSD2	100% load @ 1400 rpm

[a] High-sulfur No. 2 diesel fuel.
[b] Methyl esters of soybean oil.
[c] Low-sulfur No. 2 diesel fuel.
[d] Isopropyl esters of soybean oil.
[e] Winterized methyl esters of soybean oil.

RESULTS AND DISCUSSION

Fuel properties. Table 2 shows the composition of the neat esters. The greater molecular weight, number of double bonds, lack of aromatic compounds, and presence of oxygen in the esters suggest that they will respond differently in the engine than diesel fuel. While the methyl and isopropyl esters contained 14.6 and 14.8% saturates, respectively, the winterized methyl esters had only 5.2%. The lower amount of saturates in the winterized methyl esters was responsible for its better cold flow properties than nonwinterized methyl esters.

Because soybean oil esters do not contain any aromatics, they decreased the concentration of aromatic compounds in the fuel blends. Aromatics are known to contribute to particulate formation, and reducing their concentration in the fuel should reduce particulate emissions. Table 3 shows the fuel properties of the base fuels.

The cetane numbers of the fuel blends are shown in Figure 1. The cetane numbers of all fuel blends were improved by increasing the concentration of soybean oil esters, which means that engines would experience a decrease in ignition delay. Pure methyl esters had the highest cetane number, 59.1 (Fig. 1). The cetane number of the isopropyl esters was smaller than that for the methyl esters, possibly because of the carbon chain branching of the isopropyl esters.

Table 3 shows the oxygen content of the neat esters to be between 10 and 11%, by weight. The proportion of oxygen in the fuel blends increased with increasing concentration of esters. The presence of oxygen in the fuel may have contributed to the observed reduction in solid carbon emissions and other products of incomplete combustion, such as CO and HC.

The heating values of the methyl, isopropyl, and winterized methyl esters were 12.6, 12.2, and 12.1% lower, respectively than that of No. 2 diesel fuel. The lower heating value

TABLE 2
Compositions of Esters of Soybean Oil (by weight)

Fatty ester	Methyl (MS)	Isopropyl (IP)	Winterized methyl (WMS)
Palmitate 16:0	10.7	11.0	4.1
Stearate 18:0	3.9	3.8	1.1
Oleate 18:1	22.1	20.4	22.4
Linoleate 18:2	55.1	55.9	62.2
Linolenate 18:3	8.3	9.0	10.2
Saturated	14.6	14.8	5.2

TABLE 3
Base Fuel Properties

Fuel	HSD2	LSD2	100% MS	100% IP	100% WMS
Cetane number	46.3	47.4	59.0	52.6	51.9
AMW[a]	198.0	195.0	291.6	320.6	293.8
API gravity @ 60°F	34.9	33.5	30.4	28.9	28.1
Carbon, % (wt)	86.46	86.41	78.16	78.48	77.99
Hydrogen, % (wt)	13.32	12.97	12.06	12.21	11.39
Oxygen, % (wt)	—	—	10.99	10.00	10.90
C/H ratio	6.49	6.66	6.48	6.45	6.57
Sulfur, % (wt)	0.240	0.045	0.020	0.021	0.012
HHV[b], MJ/kg	45.36	45.33	39.72	39.98	39.83
LHV[c], MJ/kg	42.53	42.58	37.17	37.39	37.41
Aromatics, % (vol/vol)	43.7	39.2	0.0	0.0	0.0
Distillation (ASTM D86)					
Initial boiling point, K	455	452	—	—	—
50% recovery, K	538	535	—	—	—
90% recovery, K	589	588	—	—	—
End point, K	615	618	—	—	—

[a] Average molecular weight.
[b] Higher heating value.
[c] Lower heating value (calculated based on SAE J1498 MAY90).

of the isopropyl esters was slightly greater than that of the methyl esters (0.6%).

Engine performance. The brake-specific fuel consumption (BSFC) and brake-fuel-conversion efficiency of the fuels are shown in Figures 2 and 3, respectively. The term brake-specific is used to designate quantities that have been normalized by dividing by the engine's power; thus, the BSFC is equal to the fuel flow rate divided by the engine's power. Engine performance of all fuel blends was similar to No. 2 diesel fuel. All blends showed a nearly linear increase in BSFC with increasing fraction of esters. The greatest fuel consumption increase was 9.3% for the fuel with 70% methyl esters. Figure 3 shows that the brake-fuel-conversion efficiency is the same for all fuels tested, indicating that the increase in BSFC can be attributed entirely to the lower energy content of the esters. The esters converted their chemical energy to mechanical work with the same efficiency as diesel fuel.

Emissions. The brake-specific CO and HC emissions for the fuel blends are plotted in Figures 4 and 5, respectively. The CO emissions of all fuel blends were significantly ($P < 0.05$) lower than for No. 2 diesel fuel. Blends with 50% methyl esters had the greatest reduction of CO emissions, which was 25.3%. All ester blends lowered HC emissions except one. The maximum reduction in HC emissions was 29.0% for 50% isopropyl esters, and 50% methyl esters had a 7.4% reduction in HC emissions. However, 20% methyl esters blended with low-sulfur diesel gave a 2.8% increase in HC emissions, compared with No. 2 diesel fuel. This increase was not statistically significant.

Emissions are sensitive to environmental conditions that cannot be fully controlled. Most of the engine tests were run on different days when the ambient air temperature, pressure,

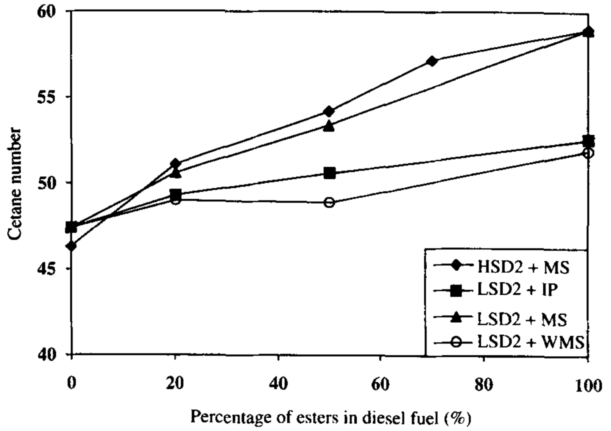

FIG. 1. Cetane number of fuels. HSD2 = high-sulfur diesel #2, LSD^2 = low-sulfur diesel #2, MS = methyl soyate, WMS = winterized methyl soyate, IP = isopropyl soy esters.

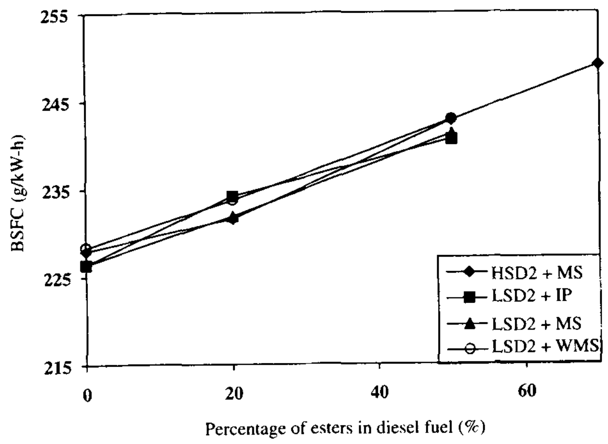

FIG. 2. Brake-specific fuel consumption. For abbreviations see Figure 1.

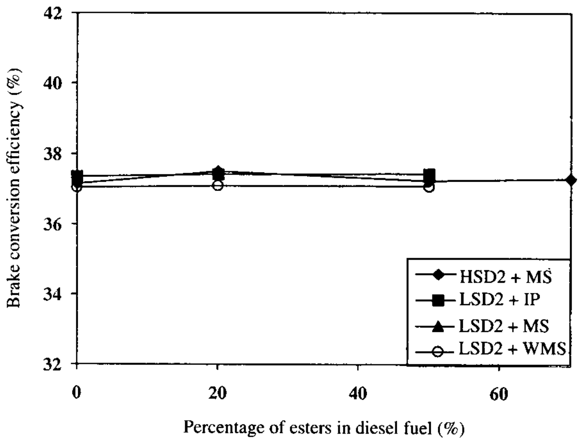

FIG. 3. Brake conversion efficiency. For abbreviations see Figure 1.

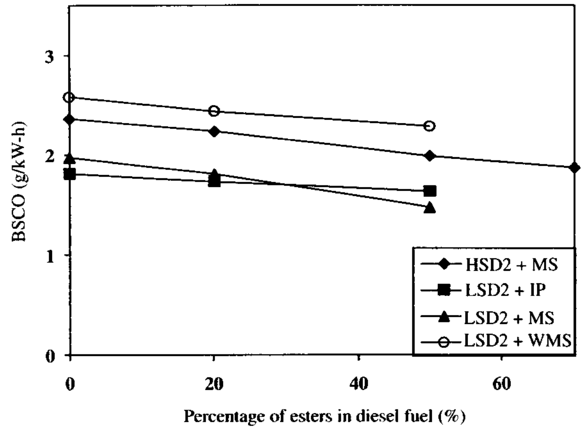

FIG. 4. Brake-specific CO emissions. See Figure 1 for abbreviations.

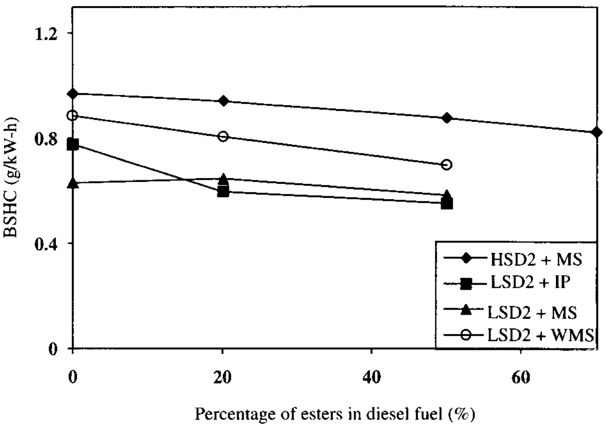

FIG. 5. Brake-specific HC emissions. See Figure 1 for designations.

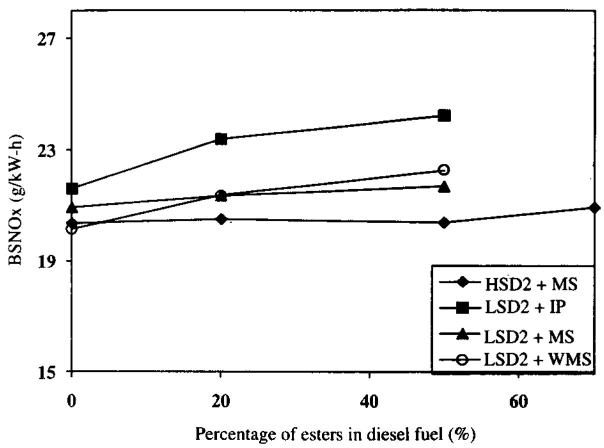

FIG. 6. Brake-specific NO_x emissions. See Figure 1 for abbreviations.

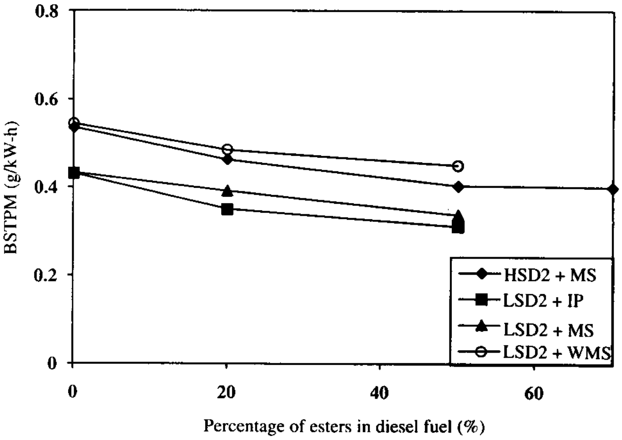

FIG. 7. Brake-specific TPM emissions. See Figure 1 for abbreviations.

and humidity were different. Also, due to limited diesel fuel storage capacity, some of the tests were run with different baseline diesel fuels. However, within a fuel group, the same diesel fuel was always used. Tests of fuels in a specific group (Table 1) were run sequentially, i.e., tests of one fuel group were completed before a second fuel group was tested. This was necessary to obtain consistent and reproducible results within the three repetitions of each test. In Figures 4 through 9, environmental variations account for the differences shown among the LSD2 fuels at 0% esters in the fuel. At 0% esters, emissions for the three LSD2 fuel groups should be the same, but often they were not. Thus, Figures 4 through 9 accurately show the effects on emissions of increasing fuel ester content, but they should not be interpreted to reflect differences among the fuel groups.

The emissions of NO_x from all fuel blends were higher than for No. 2 diesel fuel, as shown in Figure 6. The NO_x emissions of 20 and 50% isopropyl and winterized methyl esters blended with No. 2 diesel fuel showed significant increases; the 50% isopropyl ester blend had 12.1% higher NO_x emissions, which was the greatest increase among the fuels tested. The blends with methyl esters had the lowest increase in NO_x emissions, which was below 4.0%.

The total particulate matter emissions are shown in Figure 7. All fuel blends had significantly improved particulate emis-

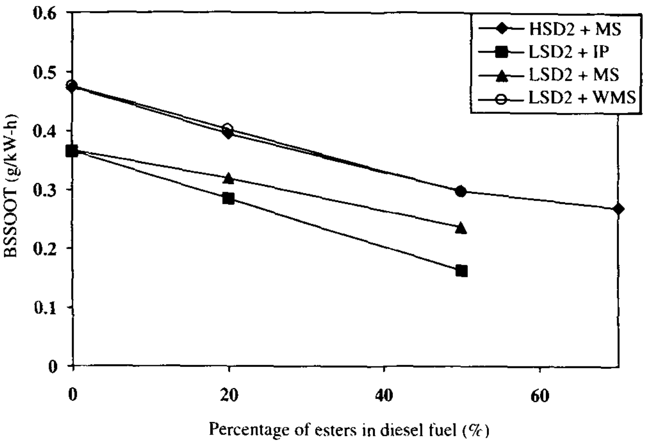

FIG. 8. Brake-specific soot emissions. See Figure 1 for abbreviations.

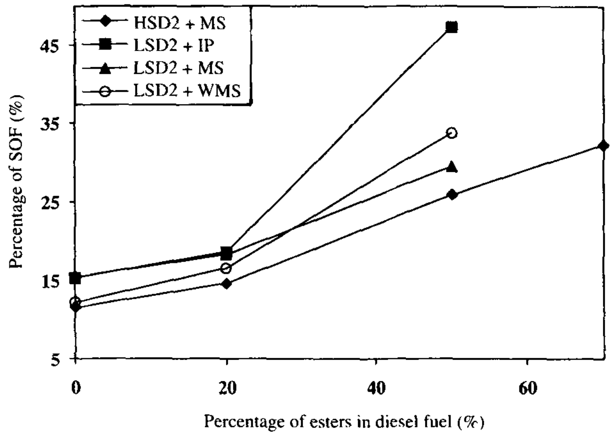

FIG. 9. Soluble organic fraction of particulate. See Figure 1 for abbreviations.

sions, compared to the No. 2 diesel fuels. All 50% ester blends decreased particulate emissions at least by 17.4%, and 20% ester blends reduced particulate emissions at least by 9.6%. Isopropyl ester blends (20 and 50%) had the best effects on particulate reduction. The largest reduction of particulate emissions was 28.0%, which was produced by the 50% isopropyl esters with low-sulfur No. 2 diesel.

TPM consists of an SOF and a solid fraction, which contains carbon, sulfates, and other inorganic material (soot). To determine the proportion of each fraction in the particulates, all primary filters were Soxhlet-extracted with methylene chloride for 60 to 70 cycles, (about 4 h). The secondary filter mass was generally too small to obtain an accurate measurement of the soluble organic fraction. The soot emissions that are shown in Figure 8 had the same trends as the TPM emissions. The soot emission levels of all fuel blends were significantly decreased compared with No. 2 diesel fuel. Among the tested fuels, isopropyl esters had the lowest soot emission. The largest reduction of soot was achieved with 50% isopropyl esters, which was 55.3% lower than low-sulfur No. 2 diesel fuel, and the 20% isopropyl esters reduced soot by 22.0%.

The percentage of SOF in the particulates is shown in Figure 9. The SOF increased with increasing proportion of esters in the fuel blends, and the 50% isopropyl ester blend had the highest SOF, which was 47.3%. The isopropyl esters had the highest SOF among the 20% fuel blends, which was 18.6%. The SOF level for 50% isopropyl esters was 209.1% higher than that of No. 2 diesel fuel. The SOF increases of methyl esters were the lowest for the same-percentage fuel blends.

Only the fuel blends of methyl esters with high-sulfur diesel fuel were tested at the light-load engine condition. The emission data that were collected at this light-load condition (Table 4) revealed some different characteristics from the tests at full load. All fuel blends revealed a significant decrease of unburned hydrocarbon emissions. The more methyl esters in the blends, the greater the reduction of HC emissions. CO emissions were also reduced by the methyl ester blends.

Though NO_x emissions of the 50 and 70% methyl ester blends showed 3 and 8% increases, compared with No. 2 diesel fuel, these were within the range of experimental variation. The results for particulate emissions from the fuel blends at 20% of full load were contrary to those observed at the full-load conditions. As shown in Table 4, the particulate emissions increased with increasing percentage of methyl ester. The 20, 50, and 70% blends had 33.3, 68.3, and 113.5% higher particulate emissions, respectively, than No. 2 diesel fuel. The increase of total particulate emissions was caused by a large increase in the soluble organic portion of the particulate. At light load, even with the diesel fuel, over 80% of the particulate mass was soluble organic material. When the concentration of methyl esters reached 70%, the percentage of SOF was 95.2%. Thus, all of the increase in particulates can be attributed to SOF. The soot emissions seemed to be irregular, but this probably is due to the uncertainty in the determination of SOF because the soot is determined by subtraction. When SOF is high, the amount of soot may be so small that slight errors in the SOF percentage can cause large percentage errors in the soot fraction.

Monoesters of soybean oil have very different chemical structures from petroleum-based diesel fuels. No. 2 diesel fuel typically contains 30–35% aromatic hydrocarbons and 65–70% paraffins, with a trace of olefins. It contains a large number of straight-chain molecules in the C_{10}–C_{16} range. However, the es-

TABLE 4
Emissions of Methyl Ester Blends with Diesel @ 1400 rpm and 20% of Full Load

Fuel	Diesel	20% MS	50% MS	70% MS
CO[a]	8.73	8.46	8.04	8.32
HC[a]	6.06	5.18	4.51	4.05
NO_x[a]	14.5	14.2	15.0	15.6
PM[a]	1.26	1.68	2.12	2.69
SOOT[a]	0.19	0.25	0.38	0.13
SOF[a]	1.07	1.43	1.74	2.56

[a] Unit in g/kW-h.

ters of soybean oil contain primarily C_{16} and C_{18} fatty acyl groups with from 0 to 3 double bonds per molecule.

Cetane number depends primarily on fuel structure (straight-chain vs. branched-chain), molecular weight, volatility, and the number of double bonds (14,15). The longer the straight carbon chains and the lower the concentration of aromatic content in the fuels, the higher the cetane number. The methyl and isopropyl esters of soybean oil have longer carbon chains than the hydrocarbon in diesel fuel, and no aromatic content.

Because esters of soybean oil contain little sulfur, they diluted the sulfur concentration of the blends when they were blended with diesel fuel. The effect of fuel sulfur content on the diesel particulate emissions has been studied widely by many researchers (16,17). Low-sulfur diesel fuel can significantly reduce the particulate emissions by lowering the amount of sulfate and its bound water. The more esters in the fuel blends, the lower the sulfur concentration. The lack of aromatic content of the esters also contributed to the lower aromatic content in the fuel blends, which is known to reduce particulate emissions and improve combustion (16,17).

Another significant difference between diesel fuel and the esters of soybean oil was that the esters contained more than 10% oxygen. The presence of oxygen in the esters and fuel blends lowers the local fuel/air ratio. Diesel engines always operate under overall fuel-lean conditions, but the fuel-rich core of the spray where the solid carbon is formed may behave leaner if the oxygen provided by the fuel is available for combustion.

Due to the heterogeneous nature of diesel combustion, there is a wide distribution of fuel/air ratios within the cylinder. HC emissions are attributed to (i) either fuel/air mixtures that are too lean to autoignite or to support a propagating flame, or (ii) fuel/air mixtures that are too rich to ignite. Soot formation mainly takes place in the fuel-rich zone at high temperature and high pressure, specifically within the core region of each fuel spray, and is caused by high-temperature decomposition. If fuel is partially oxygenated, it could reduce locally over-rich regions and limit primary HC and soot formation. The availability of fuel oxygen is not clear, but it is certain that the primary reason for the particulate reductions is less soot emission. Any unburned esters, due to its low volatility, will condense on the particulate filters and be measured as SOF.

The emissions of NO_x are determined by oxygen concentration, combustion temperature, and time. The availability of fuel oxygen is unclear because the esters may decarboxylate and produce CO_2 early in the combustion process. Also, because the heating value of the esters is about 12% less than for diesel fuel, the flame temperature is expected to be lower. However, the higher exhaust NO_x levels could be caused by earlier combustion timing. Because the fuel blends had higher cetane numbers than No. 2 diesel fuel (shown in Fig. 1), the engine would experience a shorter ignition delay period when it was fueled with those blends. The shorter ignition delay results in an earlier combustion timing, which means that less combustible fuel/air mixture is available before the burning starts than with longer ignition delay. The shorter ignition delay will produce lower premixed combustion temperature and pressure, which are of benefit to lowering HC emissions. But, the advance in timing tends to promote higher NO_x formation.

At part load, the combustion temperature was lower than under full-load conditions, and the fuel/air ratio was much leaner. Most light-load HC emissions come from bulk flame quenching at low temperature and from overmixed fuel and air. When HC in the exhaust gas were diluted with cold air in the dilution tunnel, more unburned HC would condense and adsorb on the surface of the soot particles and cause an increase in particulate emissions. Another possible cause of the low HC emissions is that the low-volatility methyl esters may be condensing on the emission sampling system before they reach the HC analyzer.

Based on the above discussion, the following conclusions can be stated about engine performance and emissions of methyl and isopropyl esters of soybean oil: (i) Methyl and isopropyl esters of soybean oil have higher cetane numbers than No. 2 diesel fuel, and they can be used as cetane improvers. The lower heating value of these esters is approximately 12% lower than that of No. 2 diesel fuel. (ii) The engine performance of the blends of methyl and isopropyl esters with No. 2 diesel fuel is similar to operation with No. 2 diesel fuel with equal fuel conversion efficiency, but slightly higher fuel consumption due to the lower energy content of the esters. (iii) All blends of methyl and isopropyl esters of soybean oil with No. 2 diesel fuel produce lower emissions of CO and unburned HC than the diesel fuel itself. (iv) Particulate and solid carbon emissions are significantly reduced when the diesel engine is fueled with fuel blends of methyl and isopropyl esters. Isopropyl ester blended with low-sulfur No. 2 diesel fuel at 50% gives the greatest reductions in particulate and solid carbon emissions. However, the soluble organic fraction for particulates of fuel blends increases with increasing percentage of esters in the fuel blends. (v) NO_x emissions of all ester blends are higher than that of No. 2 diesel fuel, with the maximum increase, 12%, from the blend of 50% isopropyl esters and low-sulfur No. 2 diesel fuel. (vi) At light load conditions, particulate emissions for the blends are increased due to large increases in the soluble organic portion of the particulate. The 70% methyl ester blend with high-sulfur No. 2 diesel fuel gives the highest increase in particulate emissions, 53%.

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