

Improving Biodiesel Blended Fuels: Overcoming the NO_x Penalty and Enhancing the Engine's Regulated Emissions Profile

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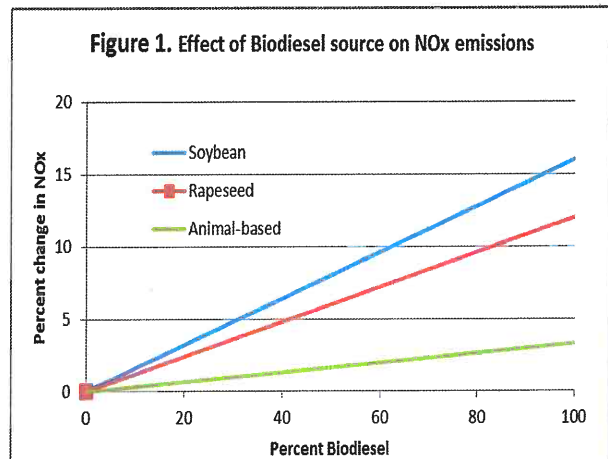
ABSTRACT

Emulsified B20 fuel containing 6.5% mass water has been shown to fully eliminate the NO_x penalty associated with diesel blends containing biodiesel. The soybean biodiesel fuel used in this study showed a 6% increase in NO_x when formulated as B20 blend in a 2004 Tier 2 Cummins QSM11C engine. This is about twice as high as the average for B20 fuels on account of the higher degree of unsaturation in the fatty acid backbone of soybean oil. In conjunction with a diesel oxidation catalyst (DOC) the emulsified fuel gave impressive reductions in HC, CO and PM with no increase in NO_x making emulsified biodiesel fuels with or without a DOC and attractive option for heavy duty applications.

INTRODUCTION

The use of biodiesel blended fuels offer the prospect of mitigating the increasing levels of CO₂ in the atmosphere emanating from combustion of fossil fuels. The performance and emissions characteristic of burning biodiesel containing fuels have been studied and reported extensively [1]-[4]. There is general agreement that increasing the amount of biodiesel in blended fuels reduces emissions, chiefly of PM, HC, and CO. However the effect on NO_x emissions is pejorative – it increases as the biodiesel content increases. McCormick et al. [1][2] concluded that this NO_x increase in both old and new engines for B20 is around 3%. In the first instance the NO_x increase was correlated with the bulk modulus of compressibility of the fuel. The higher compressibility of biodiesel it was argued results in an advance in injection timing and start of combustion. This seemed to explain the NO_x increase. However when tests were carried out in a modern engines, with common rail instead of electronic unit injectors, the NO_x increase with biodiesel fuels persisted. In these engines the compressibility mechanism is virtually negated. The authors [1] concluded that the higher bulk modulus of compressibility of biodiesel cannot be the only mechanism for NO_x increase. They observed that the bulk modulus of compressibility also correlated with

the degree of saturation of the fatty acid methyl ester (biodiesel). A US EPA report [4] has drawn attention to the dependence of NO_x increase on the source of biofuel (Figure 1). The magnitude of the NO_x increase is directly related to the increase in unsaturation of the fatty acid.



(US EPA document 420-P-01_001 [4])

Water-in-diesel emulsion fuels have been known for decades. Recent commercial developments aimed at using high water content to maximise the reduction in PM and NO_x associated with diesel emissions. Engine and fuel developments have provided alternative, effective ways of reducing these undesirable emissions. The strategy of adopting high water containing diesel emulsions also gave rise to variable THC (total hydrocarbon) and CO (carbon monoxide) emissions as well as poor performance, including unacceptable power loss, which ultimately limited the success of this fuel. However the increasing use of biodiesel in environmentally sensitive applications has prompted the development of a new generation of emulsion fuels. The water content is low, sufficient to neutralise the NO_x increase resulting from the use of the biofuel. The additive chemistry has been optimised to specifically stabilise these emulsions.

Various factors appear to contribute to the reduction in soot formation when fuels are emulsified. These include the micro-explosion mechanism [6] leading to

better dispersion of the fuel mixture, a volume expansion and accelerated flame-propagation, and accelerated soot oxidation by OH radicals formed by the dissociation of water [6]. The delay in ignition associated with the emulsified fuel would have the effect of increasing soot and HC, while reducing NO_x. Various studies [7]-[9] with emulsified biodiesel have been reported. In many of these studies the water content is quite high. Indeed the various emulsified fuels verified by CARB (California Air Resources Board), for example 'PuriNOx' emulsion fuel commercialized by the Lubrizol Corporation, opted for water contents of up to 20%_m. This high water containing fuel was also registered by the U.S. Environmental Protection Agency under its fuel registration program [10].

In this paper two engine test studies are presented, which were undertaken to investigate (1) the effect of biodiesel content in biodiesel blended fuels on regulated emissions; (2) the effect of varying the water content in emulsified B20 biodiesel blended fuels on regulated emissions; (3) the effect of a diesel oxidation catalyst in combination with emulsified biodiesel blended fuels on emissions. The first study was carried out on a 1991 12.7 litre turbocharged engine in accordance with the Texas Low Emissions Diesel (TxLED) Program [11]. A shorter second engine test evaluation [12] was undertaken on a Tier 2, 2004 engine. This has led to the development of an emulsified biodiesel blended containing 6.5%_m water and 20%_v biodiesel (B20) made with a biodiesel of soybean origin. In the second study the additional benefit of incorporating a diesel oxidation catalyst (DOC) was investigated. Many of the operational difficulties associated with high water containing emulsions are thus avoided while retaining the major benefits associated with both emulsions and biodiesel, with and without a diesel oxidation catalyst.

EXPERIMENTAL DETAILS

FIRST ENGINE TEST STUDY

This engine test study was conducted at an independent commercial laboratory in Texas.

Engine

Throughout this study a 1991 12.7 Detroit Diesel Series 60 engine (serial No. 06R0038671) was used. This engine had an inline, six-cylinder configuration rated for 365hp at 1800rpm. It was turbocharged and used a laboratory water-to-air heat exchanger for a charge air intercooler. Table 1 lists the engine specifications and features.

Table 1. Engine Specifications and Features

Engine Parameter	Comment
Engine Serial Number	06R0038671
Make	Detroit Diesel
Engine displacement and configuration	12.7L; I-6
Emission Family	MDD12.7FZAK
Rated Power	365 at 1800rpm
Electronic Control Module	DDEC-II
Aspiration	Turbocharged

Test Cycle.

The US Federal Test Procedure (FTP) was used in this work. The EPA transient cycle under the FTP is described by means of the percent of maximum torque and percent of rated speed for each one second interval over a cycle of 1199 seconds. In order to generate the transient cycle the engine's full load torque curve is obtained from an engine speed below curb idle speed to maximum no-load engine speed. Data from this torque map are used with specified speed and load percentages to form a transient cycle. Only hot starts were used, in triplicates, for this study. Hot starts involved running the engine over a "prep" cycle. It is then stopped and allowed to stand for 20 minutes after which the hot-start EPA transient cycle was begun with engine cranking. All the test cycles were within the tolerances set by the Code of Federal Regulations (CFR).

The exhaust gases were routed to a full constant volume sampler (CVS) that utilised a positive displacement pump (PDP). Total flow in the tunnel was maintained at a nominal flow rate of about 2000 SCFM. Sample zone particulate, heated NO_x, heated hydrocarbons THC, CO, CO₂ measurements were connected to the main tunnel. Probes for background gas measurements were connected downstream of the dilution air filter pack, but upstream of the mixing section. The dilution system was equipped with pressure and temperature sensors at various locations in order to obtain all necessary information required by the 40 CFR, Part 86, Subpart N.

Fuels.

The reference and untreated candidate fuels were both ultra-low sulfur diesels (<15ppm S) which meet fuel specifications under TCEQ Chapter 114, Subchapter H. The biodiesel obtained commercially met the specification ASTM D6751. The biodiesel blended fuels were prepared using the base diesel and the B100 biodiesel. Emulsions of varying water content were prepared by APT using a pilot scale blender. The final emulsions were characterised for stability and water content was measured using the Karl Fisher method. The proprietary additive treat rate was fixed for all emulsion fuels, irrespective of the water content. Table 2 summarises the fuel analyses obtained.

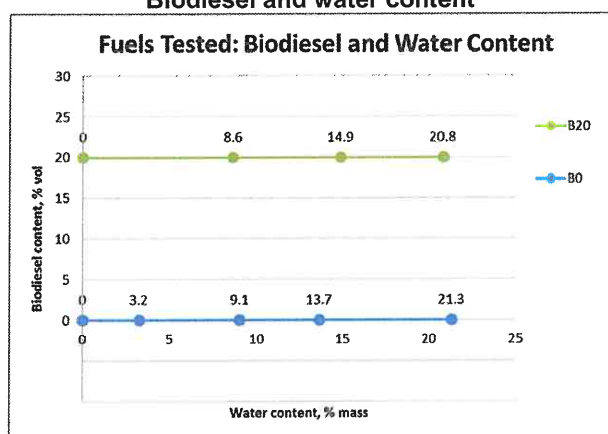
Nine test runs are reported here (each one in triplicate). These include a range of biodiesel and water contents in the fuels. Conventionally a 20% biodiesel in diesel blend is referred to as B20, to indicate the volume of the fatty acid methyl ester (FAME) or neat biodiesel (20% vol.) and volume of diesel making up the balance (80% vol.). However in the case of water-in-diesel emulsion fuels the convention is to refer to the *mass* of water in the fuel. So a 13%_m diesel emulsion fuel contains 13g of water in 100g of fuel. This equates to about 11.2% water on a volumetric basis. Table 2 below shows the volume content of biodiesel and both the volume and mass content of water in the emulsified fuels.

Table 2: Composition of Diesel, Biodiesel and emulsified fuels Tested.

Test No.	Code	Biodiesel %v. FAME	Water % vol.	Water % mass
1	B0-1	0	0	0
2	B0-2	0	2.7	3.2
3	B0-3	0	7.7	9.1
4	B0-4	0	11.7	13.7
5	B0-5	0	18.5	21.3
6	B20-1	20	0	0
7	B20-2	20	7.4	8.6
8	B20-3	20	12.9	14.9
9	B20-4	20	18.2	20.8

Note: FAME – fatty acid methyl ester (B99 material)

Figure 2: Fuels Tested Biodiesel and water content



SECOND ENGINE TEST STUDY

This study was conducted by Olson-EcoLogic Engine Testing Laboratories, California.

Engine

A Tier 2 Model year 2004 Cummins QSM 11C engine was chosen for this work. The QSM 11 C is rated at 330hp at 2100 rpm. The EPA and ARB standards for this engine are 4.9g per bhp-hr for NO_x +NMHC and 0.15g per bhp-hr. Its emissions were shown to comply with the standards.

Table 3 Engine Specifications and Features

Engine Parameter	Comment
Engine Serial number	60420004
Make	Cummins
Model	QSM 11C
Engine Displacement	10.8 litre
Emission Family	4CEXL0661AAC
Rated Power	330 hp
Aspiration	Turbocharged

Emissions Measurements

Dilute exhaust gases from the dilution tunnel are continuously collected and routed to the gaseous analyzers for analysis by corrected volume and for final

calculation of corrected mass concentrations using temperature, barometric pressure and humidity. All engine test related variables are automatically integrated from the second by second raw dilution data and automatically corrected in accordance with the applicable 40 CFR Part 89 for dilution ratio, temperature, humidity and mass and automatically calculated by the computer program to provide the second-by-second integrated final results in g/bhp-hr and g/kWh. Simultaneously and continuously dilute exhaust samples are routed to the AVL particulate sampler for capture of secondary diluted samples over the test cycle on a pre-weighed paper filter media and weighed again to determine the mass concentration of PM. All PM filter preparation and subsequent weighing is done in accordance with 40 CFR Part 86.

The computer software program captures and integrates (when appropriate) all raw data continuously over the test cycle recording and collecting the data every second over the 1200 second test. These measured and recorded data, after correction, become the basis for the final mass per brake horsepower-hour data that are provided in the summary reports. In addition to correction of the raw data for temperature, barometric pressure and humidity the data are corrected for any hydrocarbons and carbon monoxide present in the dilution air introduced through the dilution tunnel. This is done by continuously collecting a dilution air sample over the test cycle in a bag for analysis of the background dilution air at the end of the engine test cycle. The measured dilution air bag concentrations of selected gases are subtracted from the continuously integrated dilute exhaust gas samples to provide the corrected exhaust gas values.

Engine and related test variables, including automatically calculated values, are recorded second-by-second at all times during testing. They include the following:

Work, horsepower	Relative Humidity, %
Speed, RPM	CO ₂ , %
Torque, lb-ft	CO, ppm
Throttle Setting, lb-ft	NO _x , ppm
Dyno set point, RPM	NO, ppm
Throttle output, volts	THC, ppm
Dyno output, volts	CH ₄ , ppm
Oil Temperature, °F	Total Flow, SCMM
Coolant Temperature, °F	Total Inlet, mm Hg
Exhaust Temperature, °F	Total Temperature, C
Charge Air Temperature, °F	Dilute Flow, SCMM
Oil Pressure, PSIG	Dilute Inlet, mm Hg
Boost Pressure, PSIG	Dilute Temperature, C
Exhaust Back Pressure, in H ₂ O	Dilute Throat, mm Hg
Air Inlet Temperature, °F	Time, Hours
Manifold Vacuum, in H ₂ O	Time, Minutes
Barometric Pressure, in Hg	Time, Seconds
Inlet Air Flow, SCFM	Additional variables can also be recorded

Table 4: Emission Testing Equipment

Pollutant	Instrument	Instrument Description
CO	Horiba AIA-23	NDIR
CO ₂	Horiba AIA-23	NDIR
CH ₄	CAI Model 600	HFID
HC	CAI Model 300	HFID
NOx	CAI Model 400	HCLD
NO	CAI Model 400	HCLD
PM	AVL PM Sampler	1ary tunnel dilution followed by 2ary dilution/gravimetric
Dynamometer	Baldor controlled 450 HP	Full Electric

Notes:

CO/CO₂ – Non dispersive infra-red spectroscopic method.
 CH₄/HC – both measured using a flame ionisation detector. The CAI Model 600 was used in the Methane only mode.
 NOx/NO – measured by chemiluminescent gas analyser. The detector measures NO concentrations. In the NOx mode the NO₂ is converted to NO before passing through the detector.

Test Cycle

The engine is tested according to the Non Road Transient cycle (NRTC), an engine dynamometer transient driving schedule of total duration of 1200 seconds.

Fuels

The baseline diesel fuel is commercial California ultra-low sulfur diesel fuel. The B100 Biodiesel was prepared by Community Fuels in Stockton, California from 100% soybean biodiesel feed stock. The B20 blend was prepared by Ramos Oil in West Sacramento.

The emulsified fuel, EmB20 used in this study was found to contain 6.55% mass water (Karl Fisher method). The fuel compositions and characteristics are shown in Table 5. The stable emulsion was prepared using an APT commercial blender and additive.

Table 5: Composition of EmB20

Fuel	Density (19°C)	% mass	% vol
B20	0.842	93.45	94.43
Water		6.55	5.57
EmB20	0.855	100.00	100.00

The fuels were engine tested in the sequence shown in Table 6. All fuels were conducted in duplicate, totaling eight tests.

Table 6: Engine Testing Plan

Fuel	Test No.
Diesel	A1
(B0)	A2
B20	B1
	B2
EmB20	C1
	C2
EmB20DOC	D1
	D2

RESULTS AND DISCUSSION

The overall conclusions from both of the studies presented here are summarized in Table 7. The effect on emissions of emulsified water was to decrease PM and NOx with both diesel and biodiesel blends, however an increase in THC was recorded; the effect of increasing the biodiesel content (FAME) in the diesel blend was to reduce THC and PM but to increase NOx. Diesel oxidation catalyst (DOC) was neutral to NOx but reduced THC, CO and PM. The combination of emulsified B20 (EmB20) and DOC was to reduce all four emissions. The actual emissions obtained from the two studies are shown in Table 8, 9, 14 and 15.

Table 7: Effect of Water and FAME in diesel and DOC on Regulated emissions

	<i>Changes relative to Diesel</i>			
	Water	FAME	DOC	EmB20+DOC
THC	+	-	-	-
CO	-	-	-	-
NOx	-	+	0	-
PM	-	-	-	-

Key: Green (-) indicated a reduction in emissions; Red (+) indicates an increase; Blue (0) indicates no change.

1. First Study – Regulated Emissions

The engine performance was evaluated before commencing the tests with the experimental fuels using the appropriate reference fuel. The engine demonstrated the expected emissions, (in g/bhp-hr, 0.062 THC; 2.99 CO; 4.41 NOx; 0.243 PM and BSFC of 0.384lb/hp-hr).

The 9 test fuels (shown in Table 8a) and engine configurations were then tested, in triplicate. The averaged results of the regulated emissions are shown in Table 8b and 9.

Table 8a: Fuel Formulations and Test Runs

Test No.	Code	Biodiesel %v. FAME	Water % vol.	Water % mass
1	B0-1	0	0	0
2	B0-2	0	2.7	3.2
3	B0-3	0	7.7	9.1
4	B0-4	0	11.7	13.7
5	B0-5	0	18.5	21.3
6	B20-1	20	0	0
7	B20-2	20	7.4	8.6
8	B20-3	20	12.9	14.9
9	B20-4	20	18.2	20.8

Table 8b: Emissions Regulated Results

Test No.	THC	CO	NOx	PM
	g / bhp-hr			
1	0.08	3.14	4.9	0.241
2	0.09	2.75	4.63	0.195
3	0.12	2.19	4.34	0.123
4	0.13	2.07	4.25	0.101
5	0.3	2.66	4.02	0.086

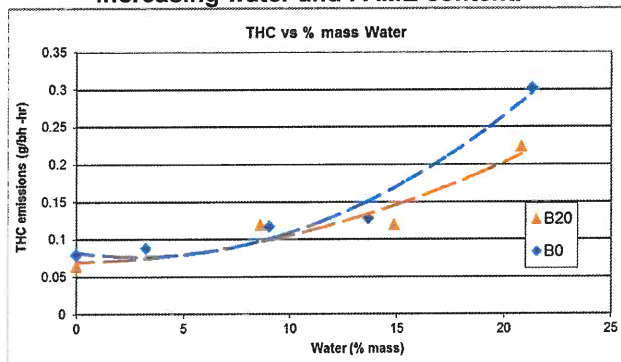
Test No.	THC	CO g / bhp-hr	NOx	PM
6	0.06	2.77	4.92	0.198
7	0.12	2.24	4.43	0.118
8	0.12	1.84	4.27	0.077
9	0.22	2.34	3.99	0.075

The results in Table 9 are shown graphically in Figures 3, 4, 5, and 6.

Table 9: Regulated Emissions, % change

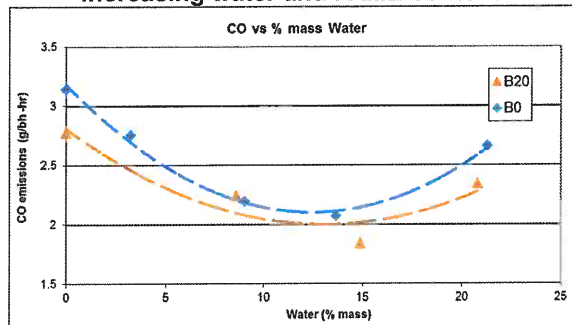
Test No.	THC	CO	NOx	PM
1	0	0	0	0
2	12.8	-12.4	-5.5	-19.1
3	50.0	-30.3	-11.4	-49.0
4	64.1	-34.1	-13.3	-58.1
5	287.2	-15.3	-18.0	-64.3
6	0	0	0	0
7	85.9	-19.1	-10.0	-40.4
8	85.9	-33.6	-13.2	-61.1
9	248.4	-15.5	-18.9	-62.1

Figure 3: Variation in THC emissions with increasing water and FAME content.



The intercept on the y-axis is a measure of the decrease in THC emissions for biodiesel fuels of increasing biodiesel content without water. There is a suggestion that the rate of increase in THC accelerates as the water content rises above 5% water.

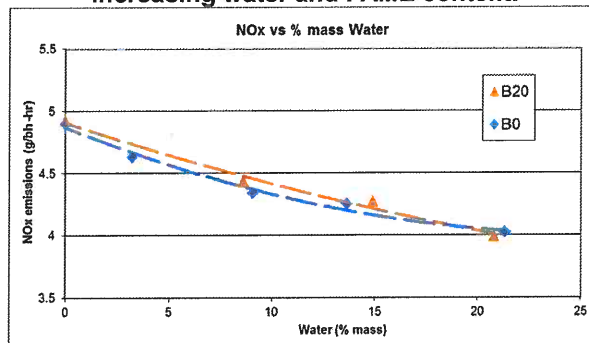
Figure 4: Variation in CO emissions with increasing water and FAME content.



The CO emissions decrease as the biodiesel content increases (intercept on the y-axis). Initially the

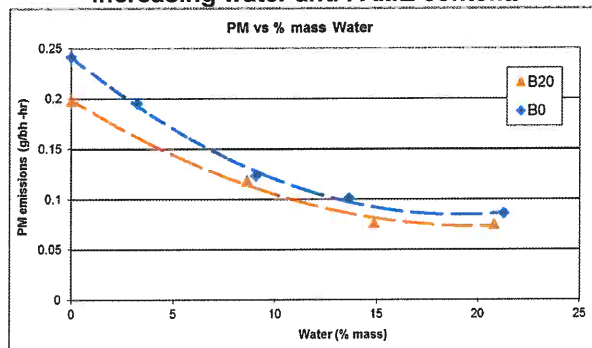
inclusion of water enhances the CO reductions. At around 12% water the trend appears to reverse.

Figure 5: Variation in NOx emissions with increasing water and FAME content.



The small increase in NOx due to the inclusion of biodiesel is indicated by the intercept of the y-axis. The slope of graphs, the rate of decrease in NOx emissions with increasing water content, diminishes as the water content increases. The measured NOx emission for B20 observed in this study is slightly higher but consistent with the EPA models. As expected the NOx emission of B20 is greater than the NOx emission of diesel and water emulsion reduces the NOx emission of both of the fuels.

Figure 6: Variation in PM emissions with increasing water and FAME content.



The intercept on the y-axis illustrates the difference between diesel and biodiesel (B20) PM emission. As noted for NOx, the rate of reduction of PM as the water content increases diminishes for higher water contents.

The decrease in PM and the increase in THC, which has been reported elsewhere [6] is pursued further in reports to be published. It suggests that emulsified fuels increase the VOC (volatile organic compounds) portion of the Total Particulate Matter (TPM), while reducing the soot content. Diesel Oxidation Catalysts are particularly effective at reducing the VOC.

The convergence in PM and NOx emissions for the various fuels with increasing water content and the reduction in the slopes of the individual curves implies diminishing reductions in these emissions as the water content is progressively increased. This is illustrated graphically (Figures 7 and 8) by calculating the % reductions for a percent of water between each two successive measurements for each fuel. In both cases the maximum gains are obtained at low water contents. The first 5% water would appear to decrease NOx

by ~1.2 to 1.7% per %m water, and PM by 5 to 6% per %m water. Above 10% water the improvements in emissions (PM and NOx) for every % increase in water content are considerably smaller.

Figure 7: NOx reductions per %m water as a function of water content

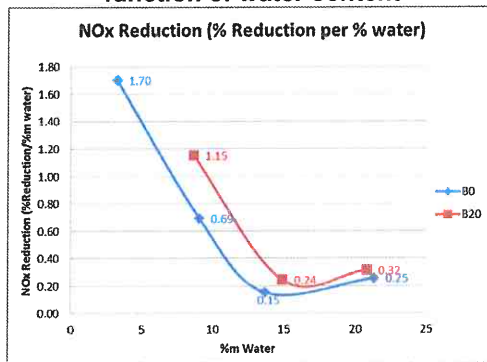
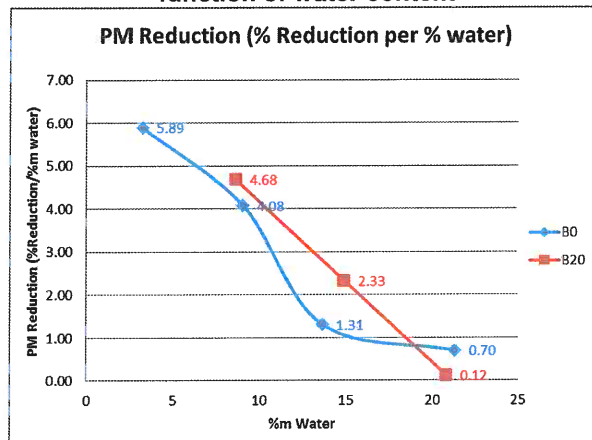


Figure 8: PM reductions per %m water as a function of water content



2. Second Study – Regulated Emissions

The second engine test study was conducted with a 6.5% water-in-B20 fuel (referred to here as EmB20), with and without a DOC fitted. The ability of the DOC in reducing the THC is of particular interest (see Table 10 and 11).

Regulated emissions with and without DOC

The data obtained are summarised in the tables and figures below:

Table 10: Average of Two runs: Emissions (g/bhp-hr)

Fuel		HC	CO	NOx	NO	CO ₂	NMHC	PM
B0	A	0.249	1.238	4.9	4.726	627	0.247	0.12
B20	B	0.214	1.241	5.18	4.996	620	0.212	0.097
EmB20	C	0.236	1.231	4.93	4.743	608	0.234	0.085
EmB20DOC	D	0.112	0.2895	4.91	4.108	600	0.11	0.073

Table 11: Emissions (% relative to B0)

Fuel		HC	CO	NOx	NO	CO ₂	NMHC	PM
B0	A	100	100	100	100	100	100	100
B20	B	85.9	100.2	106	105.7	99	85.8	80.4
EmB20	C	94.8	99.4	101	100.4	97	94.9	70.8
EmB20DOC	D	45.1	23.4	100	86.9	96	44.4	60.4

Figure 9: Second Study- Regulated Emissions (g/bhp-hr)

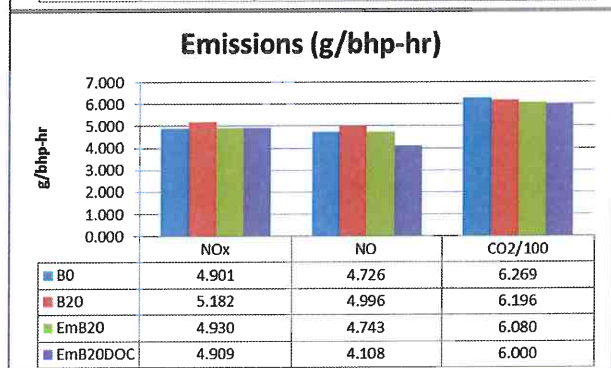
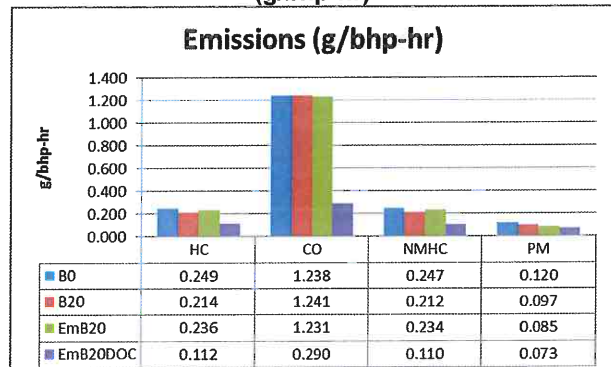


Figure 10: % Change in Emissions relative to B0 (diesel), relative to B20 and relative to EmB20 respectively.

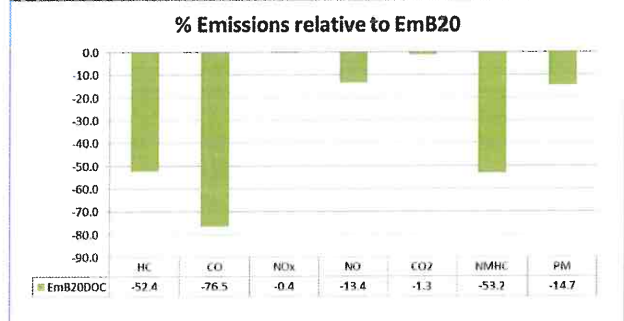
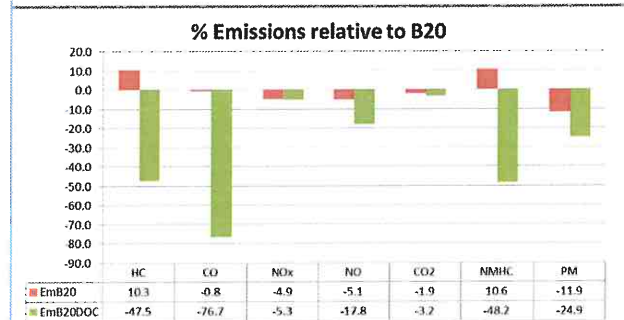
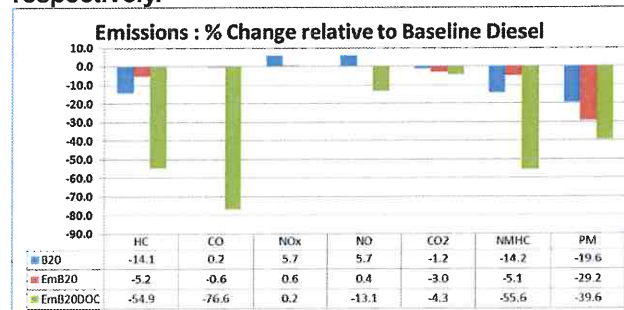
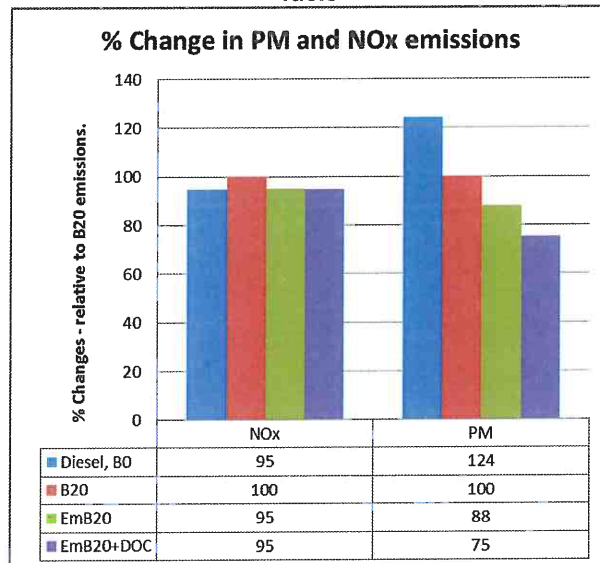


Figure 11: PM and NOx emissions with various test fuels



- In Figure 9 and Tables 10 and 11 the NO and NOx increases for B20 relative to diesel are ~ 6%, slightly higher but consistent with the EPD data [4] (see Figure 1) for soybean B20.
- Comparison of the NO and NOx values in Figure 9 indicate that around 95% of the NOx is present as NO. The NO content is down to around 80% when the Diesel Oxidation Catalyst is fitted. The DOC oxidises NO to NO₂.
- Figures 9, 10 and 11 show that the 6% water in B20 emulsion fuel, EmB20, is able to reduce the NO and NOx emissions back to the baseline level obtained with diesel.
- Table 11 (and Figure 11) illustrate the stepwise reduction in PM emissions when the fuel is changed from diesel (100%) to B20 (80%) to EmB20 (70%) and to EmB20+DOC (60%).
- In this study the increase in HC, NMHC and CO with the emulsified fuel relative to the B20 fuel is marginally lower than the emissions seen with diesel.
- As expected the DOC is able to more than half the emissions of NC, CO and NMHC (Figure 9, Table 11).

CONCLUSION

The detailed study enabled some general conclusions to be drawn about the effect of water, biodiesel content and DOC on engine emissions. The combination of a DOC, 6% water in B20 emulsion offers a very exciting combination with significant reductions in THC, CO, PM and NOx.

1. The rate of hydrocarbon emissions increase for emulsion fuels is greatest when water content is above 10%*m*.
2. The CO emissions were shown to decrease up to 10% water content.
3. The reduction in PM and NOx with increasing water content is not linear. The maximum reduction per %*m* water is obtained at the lower water contents (below 10%*m*).

4. The DOC unit has a significant effect on THC, CO and PM. The combined effect of DOC and biodiesel is cumulative.
5. DOC and emulsified fuel appear to have a synergistic effect – the DOC is better suited for reducing THC and CO, while emulsion fuel are better at reducing soot content.
6. Soybean biodiesel in a B20 blend causes an increase of 6% in NOx emissions and a 20% decrease in PM emissions relative to diesel in a 2004 tier 2 Cummins QSM11C engine.
7. A stable emulsion containing 6.5% mass water in B20 fuel reduced the NOx emissions to the same level obtained with diesel. The PM emissions were reduced further. It is interesting to note that all the emissions obtained with the emulsified B20 fuel (EmB20) alone were lower than those obtained with diesel.
8. A diesel oxidation catalyst fitted to the exhaust system in combination with the emulsified biodiesel fuel reduced HC and CO by more than 50%, PM by 40% and showed no increase in NOx relative to baseline diesel.

ACKNOWLEDGEMENTS

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