

The Impact of Biodiesel on Particle Number, Size and Mass Emissions from a Euro4 Diesel Vehicle

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ABSTRACT

New European emissions legislation (Euro5) specifies a limit for Particle Number (PN) emissions and therefore drives measurement of PN during vehicle development and homologation. Concurrently, the use of biofuel is increasing in the marketplace, and Euro5 specifies that reference fuel must contain a bio-derived portion.

Work was carried out to test the effect of fuels containing different levels of Fatty Acid Methyl Ester (FAME) on particle number, size, mass and composition. Measurements were conducted with a Cambustion Differential Mobility Spectrometer (DMS) to time-resolve sub-micron particles (5-1000nm), and a Horiba Solid Particle Counting System (SPCS) providing PN data from a Euro5-compliant measurement system. To ensure the findings are relevant to the modern automotive business, testing was carried out on a Euro4 compliant passenger car fitted with a high-pressure common-rail diesel engine and using standard homologation procedures.

It was found that using FAME decreased total PN emissions, by 16% over the Type I drive cycle (NEDC) for a 30% biodiesel (B30) compared to mineral fuel (B0). FAME also decreased accumulation mode PN and carbonaceous mass, by 20-30% for B30 versus B0, with a consequent reduction in Diesel Particulate Filter loading rate. A 25% increase in the nucleation mode PN was found when using B30 versus B0, and the higher molecular-weight organic mass fraction also increased. Increases in nitrogen oxides when using FAME-containing fuels were also confirmed.

INTRODUCTION

Introduction of a Particle Number (PN) limit in Euro5 emissions legislation, and the associated necessity for PN measurement, is driven by two things. Firstly, there are concerns over the resolution of the existing gravimetric technique for particulate mass measurement (PM), against a backdrop of ever-lower limits. Anticipating a point where the viability of this technique is questionable, confidence needs to be gained in another particle measurement approach. Secondly, ongoing medical research indicates that particle mass may not be the most relevant metric, with various studies instead implicating particle size, number and surface area [1 and references contained therein].

Concurrently, concerns over global warming continue to drive the use of biodiesel through the belief that less carbon dioxide is produced on a 'source-to-wheels' basis than with fossil fuels. The biodiesel being used is Fatty Acid Methyl Ester (FAME) and this is blended with mineral-diesel. Incorporating FAME extends the size of the diesel fuel pool in the EU, and reduces reliance on fossil-fuel producing regions. Furthermore, fuel retailers and vehicle operators are free to blend and use FAME at levels above that specified by EN590 (7% v/v) because FAME content is not legally controlled as with 'environmental parameters' such as the amount of Sulphur or Poly-Aromatic Hydrocarbons (PAH) in the fuel.

This situation of increased biodiesel use, with elevated FAME levels, against a background of type approval for particle number, raises some questions for the automotive business. Particle generation in the engine is fundamentally linked to the combustion process of a fuel-air mixture, so it may be surmised that changes in chemical composition of the

fuel due to FAME blending will affect particle formation and/or oxidation during the engine operating cycle. Therefore, one might expect differing particle emission levels to be seen when comparing mineral diesel with biodiesel. Many studies in the literature show that FAME has an impact on engine-out particulate mass [2, 3], but there are fewer studies into tailpipe particle number from light-duty applications over the homologation test. Although there is a relationship between particle mass and number, this is dependent on the particle size distribution, so it is important to understand the effects of FAME content on size distribution to quantify the PN-PM relationship and predict any impact on aftertreatment performance.

The particle sizes of interest are in the nano-metre (nm) range, and it is well established in the literature that particles can be separated into three size ranges, known as the nucleation mode, the accumulation mode, and the coarse mode [4,5,6,7]. The nucleation mode contains the smallest particles (3nm to 50nm [4, 5]), and is formed mainly through the nucleation of material from the gas-phase during exhaust cooling, although it can also be solid material such as ash. The accumulation mode consists of agglomerated solid particles formed from soot precursors, plus the extraction of material from the gas-phase [1], and has a typical size range of 30nm to 500nm [4]. The coarse mode (>500nm) does not come from combustion. The accumulation mode contributes most of the particle mass, and therefore this has been the area receiving the bulk of automotive research. Particle formation and composition is complex, but two key particulate contributors are the carbonaceous fraction and organic fraction. The carbonaceous fraction arises from pyrolysis of fuel in locally fuel-rich areas during combustion, or pyrolysis of oil escaping into the combustion chamber. The organic fraction can come either directly from unburnt fuel/oil, or through incomplete pyrolysis of fuel/oil.

Some previous research has covered the topic on PN emissions from engines operating on biofuel. Zhang *et al.* [8] suggested the total absence of nucleation mode particles with 100% biodiesel (B100), as well as varying relationships between biodiesel content and the engine operating point, at the sub 400nm size. Li *et al.* [9] showed lower PN levels with biodiesel in the nano-metre range at two different loads, and that specific reductions compared to mineral diesel are dependant on whether the sample is taken before or after the oxidation catalyst. Some researchers have found that biodiesel increases the number of smaller particles when compared to mineral diesel [10,11,12,13,14,15,17]. Schönborn *et al.* conducted tests on a single-cylinder engine operating in the steady-state, and found that biodiesel molecules with longer fatty acid chain lengths tend to produce larger numbers of small particles (5-30nm diameter) than biodiesel molecules with shorter fatty acid chains [17]. Schönborn also found that the degree of saturation of fatty acid alcohol molecules affects particle emissions. Czerwinski

et al. [12] showed that the particle size distribution is moved to smaller sizes with both crude rapeseed oil and Rapeseed Methyl Ester (RME), and the nuclei mode is increased. Krahel *et al.* [13] states that particle numbers below 30nm decrease with fossil fuels but exhibit a 10-fold increase with methyl-ester fuels derived from various sources, and Trapel *et al.* [14] found that with increasing FAME content, the particle size distribution moves towards smaller particles, although the total particle number decreases drastically. There is other evidence of increases in the smaller particles [10], Bertola *et al.* [11] found a large amount of ultra-fine particles (<20nm) in the exhaust during operation with RME, and Krahel *et al.* [15] showed that RME leads to more particles in the range of 10nm to 40nm compared to mineral diesel and less particles for the larger diameters.

The variety of results obtained demonstrates the considerable complexity involved when trying to establish the effect of biodiesel on particle number. A significant portion of the studies are on heavy-duty applications or steady-state testing, leaving many questions unanswered for the light duty application over the homologation test, particularly for modern technology such as common rail fuel injection equipment and vehicles fitted with particle aftertreatment. Therefore, the main objective of this work is to quantify any change in total particle number and size distribution due to FAME content, over type-approval emission tests with an engine/vehicle representative of current technology.

TEST FUELS

The baseline fuel was a mineral diesel meeting the European diesel fuel standard EN 590 and the legislative fuel specification CEC RF-06-03 for European exhaust emissions testing [16]. An analysis of the fuel properties can be found in [Table 1](#), from where it can be seen the baseline fuel did not contain any FAME or ester based additives such as lubricity improvers.

Biodiesel blends were formulated by blending the baseline mineral diesel fuel described above with an EN 14214 compliant FAME, a.k.a. biodiesel fuel. As discussed in the introduction, FAME is a renewable blending component that is widely blended with diesel fuel in many European markets, and in North America. Currently, EN 590 permits a maximum FAME content of 7% v/v, although the European Commission has issued a mandate for this to be increased to 10% v/v in the next revision of the standard. As the 7% limit in EN 590 is not legally binding, fuel retailers are free to operate with blend levels above the limit specified in EN 590, and this is often the case for operators of captive fleets. In North America, the ASTM have released the D7467 standard for diesel fuel containing between 6 and 20% v/v FAME, and such fuels are becoming widely available. In the experiments described here, FAME was blended volumetrically with the mineral diesel baseline fuel at concentrations of 5% v/v (B5),

Table 1. Properties of mineral diesel and FAME used in this work.

Property (Test method)	Mineral diesel (B0)	FAME (B100)
Cetane number (D613)	53.2	52.2
Sulphur content (mg/kg) (IP490)	< 1	1.4
FAME content (% v/v) (EN14103)	0	98.8
Molar H/C (Calc.)	1.85	1.85
Molar O/C (Calc.)	< 0.0003	0.104
C (% m/m) (D5921)	86.62	77.24
H (% m/m) (D5921)	13.38	11.94
O (% m/m) (Elemental Analysis)	< 0.04	10.7
H/C (Calc.)	0.1545	0.1546
Net calorific value (MJ/kg) (IP12)	43.00	37.39
Methanol content (% v/v) (EN14110)	-	0.04
Mono-glyceride (% m/m) (EN14105)	-	0.43
Di-glyceride (% m/m) (EN14105)	-	0.09
Tri-glyceride (% m/m) (EN14105)	-	0.05
Total glycerol (% m/m) (EN14105)	-	0.14
Free glycerol (% m/m)	-	0.01

10% v/v (B10) and 30% v/v (B30). Blend levels were verified by an in-house gas chromatogram procedure [18]. This test reported FAME concentrations of 4.27% v/v for the B5, 10.24% v/v for the B10 and 31.16% v/v for the B30.

<table 1 here>

Table 1 shows that the FAME contains 10.7% m/m oxygen. This oxygen arises due to the two oxygen atoms contained in the ester functional group. Although not clearly elucidated by the data in the table, a significant fraction of the (aliphatic) side-chains in the FAME molecules are likely to be mono- or even poly-unsaturated.

The physical effects of FAME blending on the finished fuel were quantified by measurement of the ASTM D86

distillation curves and D445 viscosity at 40°C. These data are presented in the results section together with a discussion of the aromatics level in the finished test fuels.

The presence of the ester group, and associated physical effects described above are expected to influence the engine emissions performance. Sulphur was not considered in detail because both the baseline mineral diesel and the FAME contained only ~ 1 mg/kg Sulphur or less.

TEST METHODOLOGY

EQUIPMENT

Testing was conducted at the Ford Dunton Technical Centre in the UK in a test cell conforming to ECE Regulation No. 83 [19] and approved by the UK Department of Transport

Vehicle Certification Agency and German Technical Inspection Association. The equipment included a CVS/Dilution Tunnel system, particulate sampling onto filters (Pallflex TX40), gaseous analysers, and chassis dynamometer. Additional equipment used included a Combustion Differential Mobility Spectrometer (DMS500), Horiba Solid Particle Counting System (SPCS), AVL Photo-Acoustic Soot Sensor (Model 483), and Perkin-Elmer Gas-Chromatograph (GC). Test variability was minimized through the use of a robot driver, plus rapid vehicle cooling between tests which promotes test stability by allowing the vehicle to remain in position on the dynamometer during repeat testing on each fuel. The test vehicle was a CD-class model with a Euro4 compliant 2000cc high-pressure common-rail diesel engine. Data from the Powertrain Control Module (PCM) were collected for every test.

METHOD

The four fuels were tested using standard emission tests from European and Federal (USA) legislation. The European tests are of main interest here, but the Federal tests consist of more transient driving patterns potentially providing different results. Each fuel was tested with five NEDC cycles [20] and two FTP75 cycles [21]. Each test was conducted from ambient temperature at 20-30degC (colloquially a 'cold test') after a rapid vehicle cool down with chilled engine coolant and multiple high-speed cooling fans. Before changing fuel, all data were checked for quality and consistency. At the start, and after each fuel change, two conditioning drives were carried out, each consisting of three NEDC Part Two cycles (Extra-Urban). For all this work the DPF was removed from the vehicle to gain insights into particle emissions from the engine and into the DPF. The vehicle PCM was flashed with a non-DPF version of the production calibration. Real-time data were collected at 10Hz and post-processed to obtain results at the vehicle tailpipe normalised by distance, as required for reporting regulated emission results. During particle sampling, the DMS was set to a 4:1 dilution which was found suitable to prevent saturation of the DMS electrometers. Furthermore, the DMS sample was taken from the dilution tunnel which was set at a CVS flowrate of 600 cubic feet per minute. The dilution-corrected concentration from the DMS was then used in conjunction with CVS volume to determine tailpipe PN emissions.

RESULTS AND DISCUSSION

FUEL

Figure 1 shows the effect of FAME blending on the volatility of the finished fuel. The boiling range of the FAME molecules lies within the normal diesel hydrocarbon boiling range, and so the initial and final boiling points of the fuel are unaffected. Whilst the sigmoidal shape is retained, the FAME does cause a reduction in the mid-range volatility. This can be quantified by comparison of the T50 for the B30 (298°C)

and the T50 for the mineral diesel baseline (271°C), which is 27°C lower.

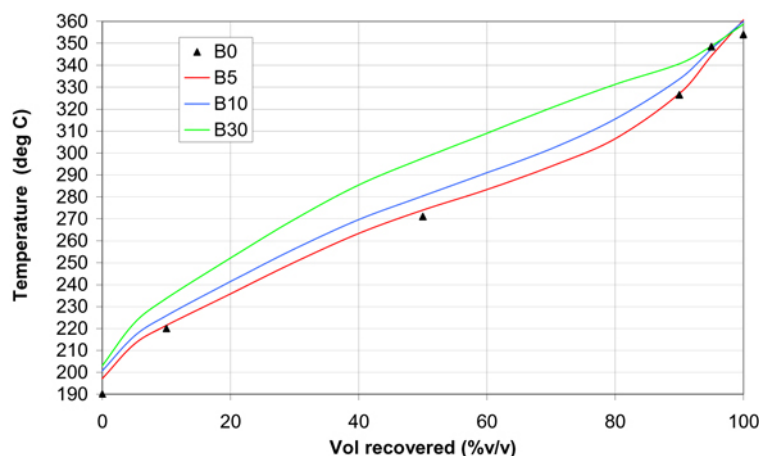


Figure 1. ASTM D86 distillation curves for the test fuels.

Figure 2 shows the effect of FAME blending on the kinematic viscosity of the finished fuel at 40°C. Consistent with the shift in distillation curve, the introduction of 30 % v/v FAME increases this viscosity from 2.5 cSt in the base fuel to 3.05 cSt.

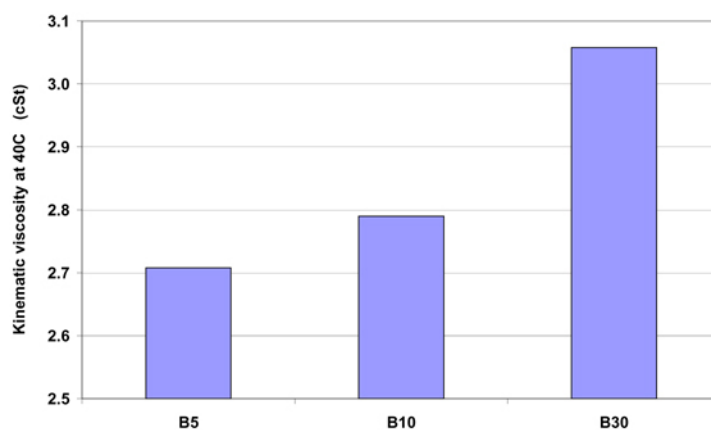


Figure 2. Kinematic viscosity at 40°C for the B5, B10 and B30 fuels, measured according to IP71/97.

In addition to the effect on the volatility (Figure 1) and viscosity (Figure 2), some authors report that FAME has a higher bulk modulus of compressibility than mineral diesel [22]. In applications with pump-line-nozzle fuel injection equipment, the timing of the fuel injection event depends on the speed of sound in the fuel, which increases in proportion to the bulk modulus. In the experiments described here, a common-rail fuel injection equipment architecture was used. The injection timing in these systems may be considered independent of the compressibility of the fuel, and so changes in injection timing due to FAME content were not considered further. Similarly, a reduction in ignition delay has been