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Particulate number emissions during cold-start with diesel and biofuels: A special focus on particle size distribution

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Abstract

The share of biofuels in the transportation sector is increasing. Previous studies revealed that the use of biofuels decreases the size of particles (which is linked to an increase in particulate toxicity). Current emission regulations do not consider small particles (sub-23 nm); however, there is a focus in future emissions regulations on small particles. These and the fact that within cold-start emissions are higher than during the warmed-up operation highlight the importance of a research that studies particulate matter emissions during cold-start. This research investigates the influence of biofuel on PN and PM concentration, size distribution, median diameter and cumulative share at different size ranges (including sub-23 nm and nucleation mode) during cold-start and warm-up operations using diesel and 10, 15 and 20% mixture (coconut biofuel blended with diesel). During cold-start, between 19 to 29% of total PN and less than 0.8% of total PM were related to the nucleation mode (sub-50 nm). Out of that, the share of sub-23 nm was up to 9% for PN while less than 0.02% for PM. By using biofuel, PN increased between 27 to 57% at cold-start; while, the increase was between 4 to 19% during hot-operation. The median diameter also decreased at cold-start and the nucleation mode particles (including sub-23 nm particles) significantly increased. This is an important observation because using biofuel can have a more adverse impact within cold-start period which is inevitable in most vehicles' daily driving schedules.

Keywords: Particle size distribution; PN; sub-23 nm; biofuel; cold-start.

Word count: 7338

Abbreviations	
NOx	Nitrogen oxides
CO	Carbon Monoxide
HC	Hydrocarbon
PM	Particle mass
PN	Particle number
BSFC	Brake Specific Fuel Consumption
CMD	Count median diameter
ECU	Engine control unit
EU	European Union
VOCs	Volatile organic compounds

Highlights

1. This study investigated engine-out particle emissions and size distribution
2. Cold-start, hot-start, and two intermediate warm-up phases were studied
3. Nucleation mode particles increased as the engine warmed up
4. During cold-start, using biofuel decreased the size of particles
5. Sub-23 nm fraction contributed up to 0.02% and 9% to cumulative PM and PN

1. Introduction

The transportation sector has been undergoing a significant transformation with the utilisation of different strategies and technologies to reduce harmful emissions [1]. One of these strategies is related to fuel. The adverse effect of using fossil fuels in the transportation sector has created significant interest in renewable alternatives, such as biofuels. There have been incentives in place to increase the share of such renewable alternatives. For example, EC Directive 2003/30 was issued to increase the share of biofuel to 5.75% by 2010 and Directive 2009/28/EC targeted a share increase to 10% by 2020. One of the reasons for increasing the share of renewable fuel in the transportation sector can be the advantages it has on engine performance and exhaust emissions [2-4].

In terms of engine performance parameters, it has been reported that using biofuels (such as jatropha biodiesel) can improve the thermal efficiency of diesel engines when compared to diesel fuel; however, it depends on the type of biofuel [5-7]. Friction parameters were also reported to be lower with biodiesel owing to their better lubricity [8, 9]. However, fuel consumption parameters such as brake specific fuel consumption (BSFC) were reported to be higher and the engine power was reported to be lower with biofuel derived blends [10, 11]. The higher viscosity, higher density and lower calorific values of biofuels were reported to be primary reasons. In terms of emissions, lower hydrocarbon (HC) and carbon monoxide (CO) emissions were reported to be advantages of using biofuels [12-14]. However, it has been frequently reported that nitrogen oxides (NOx) emissions increase with biofuels (some articles claimed otherwise) [15-17]. Maybe the most highlighted advantage of using biofuel is the decrease in particulate matter emissions; however, some articles reported a different observation [18, 19]. It has been frequently reported that particulate matter emissions decrease significantly due to the oxygen content of the biofuel [18, 20, 21].

Particulate matter emissions can adversely affect our environment and are identified as a global risk factor as these emissions have been reported to be associated with cardiorespiratory health problems [22-24]. It was shown that prolonged exposure to particulate matter emissions is associated with an increase in free-radicals which adversely impact health [25-27]. Particulate matter emissions can be evaluated from two inter-correlated perspectives. The first aspect is the particulate mass (PM) which reports the mass of particles. However, this measurement might not be able to provide sufficient information when it comes to the health hazards from particles, as those very small particles which are harmful have a lower contribution to PM [28]. However, the second perspective, particulate number (PN), is more informative and has gained a lot of attention, therefore it became mandated in the recent emissions regulations [29]. For passenger cars, reporting PN emissions became mandated from the Euro 5 emissions regulation [29], while there was no regulation on PN in Euro 1-4 [29]. The PN, which is the count of individual particles can include small particles even those with nearly zero weight and contribution to PM.

Using biofuel has been reported to have different effects on PN emissions, some reported higher PN with biofuels and some reported lower [18, 30]. However, most of the reports in the literature showed that using biofuel decreases the size of particles [31, 32]. This is important as it has been reported that a decrease in particle size is associated with an increase in toxicity [33]. However, in the most recent emission regulations such as Euro 5 and 6 and WLTP (worldwide harmonised light vehicles test procedure), there is a guideline for PN measurement—PMP (particle measuring method)—which does not consider sub-23 nm particles [34-36]. PMP does specify the count efficiency of 50% (D_{50}) at 23 nm and 90% (D_{90}) at 41 nm, and it has been reported in the literature that decreasing D50 from 23 nm to smaller sizes such as 10 nm can significantly increase the PN emissions from vehicles [37, 38]. For example, Leach et al. [38] reported that decreasing D_{50} and D_{90} to 10 nm and 23 nm leads to 36% higher PN. This and the increasing share of biofuel in the transportation sector highlight the importance of studying PN emissions in more detail such as looking into the size of particles.

PN emissions from an engine depend on different factors such as operating conditions [39]. For a high portion of vehicles, the cold-start operation is a norm which occurs mostly in the morning and afternoon when people start their vehicle after some hours of engine-off and drive between home and work. It has been reported that a significant number of trips between home and work start and finish during the cold-start period [40]. For example, a study of more than one thousand trips showed that more than 30% of the trips started and finished within the cold-start period [41]. Regulation (EU Directive 2012/46/EU) considers the cold-start period from engine start—after 12 hours soak (or 6 hours forced cooled)—either for the first 5 minutes or during the time that the temperature of engine coolant increases to 70°C. Within this period, the engine temperature is sub-optimal affecting the combustion process [42]. Consequently, engine emissions and performance are different in comparison with hot-operation [43-45]. For example, fuel consumption and friction power were reported to be higher when the engine is cold [46]. This was reported to be because of the high viscosity of the engine oil at lower temperatures, which consequently leads to higher friction, therefore more fuel needs to be burnt to maintain power [40, 46]. Fuel evaporation and atomisation are also impacted by the lower temperature of the engine and fuel during cold-start which also impacts emission and performance parameters [18].

It has been reported that during cold-start, emissions are higher than when the engine is fully warmed up [47, 48]. For example, a study used a custom driving cycle and showed that during the cold-start period of the cycle, PN emissions with biofuel increased significantly compared to the hot-operation period [47]. Another study showed that around one-third of the emitted PM emissions were related to the first 12% of the total distance (Phase 1) of the LA92 Unified Driving Cycle [49]. It also reported that compared to Phase 3 of that cycle, in which the engine was fully warmed up, during Phase 1, which was cold-start, the PM emissions were 7.5 times higher. Some studies in the literature investigated other emissions such as CO, CO_2 and NOx [40, 50, 51]. However, there are a few studies that investigated PN emissions and size distribution during cold-start in detail, when compared to other emissions [52]. Also, most of the cold-start experimental studies in the literature used driving cycles such as NEDC (New European Driving Cycle) or WLTC (worldwide harmonised light vehicles test cycle) which has abrupt load/speed changes. The results of such studies were shown with the averaged value. However, it is essential to study the influence of transient engine temperature while the engine is warming up on exhaust emissions such as PN. It will be seen in this current study that cold-start and engine warm-up have different stages and therefore different impacts on exhaust emissions.

This research aims to study the influence of fuel and transient engine temperature at different stages of engine warm-up (including cold-start) on particulate matter emissions from different aspects including PN, PN and PM size distribution and median diameter, the share of particles at different sizes, nucleation mode particles and sub-23 nm particles. The increasing share of biofuels in the market, with the inherent decrease in particle size and the associated increase in toxicity, increased emissions during cold-start coupled to future emissions regulations on small particles, and the fact that the current emission regulations do not consider sub-23 nm particles, all highlight the importance of this study. By using and comparing different biofuel blending ratios (10, 15 and 20%), this study can be helpful to engine researchers when it comes to nucleation mode and sub-23 nm particles the upcoming emissions regulations.

2. Methodology

The engine used in this study was a Cummins ISBe220 diesel engine (designed and manufactured by an American company) which is an in-line 6-cylinder, turbocharged, common-rail engine, as shown in Table 1. The maximum torque and power with this engine are 820 Nm (at 1500 rpm) and 162 kW (at 2500 rpm) and the engine was coupled to an in-house hydraulic dynamometer (electronically-controlled). The experiments were done at QUT Biofuel Engine Research Facility (BERF) in Brisbane, Australia.

Table 1 Engine specifications

Model	**Cummins ISBe220 31**
Aspiration	Turbocharged
Fuel injection	Common rail
Cylinders	6 in-line
Capacity	5.9 L
Bore × stroke	102 × 120 (mm)
Maximum torque	820 @ 1500 (Nm @ rpm)
Maximum power	162 @ 2500 (kW @ rpm)
Compression ratio	17.3:1

Figure 1 shows a schematic diagram of the experiment setup. Engine and dynamometer data were collected with Dynolog software. The in-cylinder data such as crank angle and injection signals were collected using a Kistler (6053CC60) transducer, Kistler type 2614 sensor and DT9832 A-to-D convertor, which all were connected to an in-house National Instruments

LabView program [53, 54]. The accuracy of the measuring instruments are shown in Appendix (Table A1).

To better evaluate the pure effect of fuel properties and also the engine temperature, this study evaluated the engine-out emissions instead of tailpipe emissions (which are sampled before any after-treatment system). This way the emissions do not depend on the after-treatment systems performance/type. Therefore, the fundamental study will not be limited and results can give more information about the real engine dependent emission [55]. In order to measure particulate matter emissions (such as particle number, mass, size distribution), this study used a TSI scanning mobility particle sizer (SMPS) consisting of a TSI 3071A classifier (which preselects the particles in different sizes) and a TSI 3782 CPC (which grows the particles to make them detectable for the optic). SMPS is designed and manufactured by TSI which is an American company. SMPS has a size resolution capability of 128 channels per decade, which results in 192 channels in total. The exhaust gas (which had a temperature of ~350 °C) was directed to the SMPS, but after being diluted (~20 times) with ambient air (~23 °C) which was passed through a HEPA filter in a constant volume dilution system (CVS). The CVS setup was followed by the European legislation (Commission Regulation (EU) 2017/2400). To calculate the dilution ratio, a CAI-600 and a SABLE CA-10 CO_2 analysers (designed and manufactured by American companies) were used before and after the dilution system measuring the CO_2 emissions.

The exhaust particles are made of solid particles, volatiles and liquid droplets. Usually, the PMP method uses a volatile particle remover (VPR) system including three stages of hot dilution (PND_1), heated evaporative tube (ET) and cold dilution (PND_2) to minimise the effect of volatiles and liquid droplets ensuring that the particle counter measures solid particles

(ECE/TRANS/WP.29/GRPE/2016/3 amended by GRPE-72-09-Rev.2) [56]. Regarding the difficulty of sub-23 nm measurement with current instruments in the market [36], it is worth mentioning that SMPS is capable to measure particles above 10 nm including solid particles and volatiles.

This study reports PN and PM, and also PN and PM size distribution. Regarding the PN and PM size distribution, SMPS measures the number of concentration in a given channel (dN) and divides it by the geometric width of the size of channel, and reports the normalised number concentration ($dN/dlogD_p$), where D_p is the geometric midpoint of the particle size channel. The conversion of PN to PM was done through the Aerosol Instrument Manager Software for SMPS Spectrometer using the formula $dM = dN \cdot (\pi/6)D_p^3 \rho$, where ρ is density, and reporting the normalised mass concentration using the formula $dM/dlogD_p = dN/dlogD_p \cdot (\pi/6) D_p^3 \rho$ [57]. In general, based on the size distribution, the measured ultrafine particles can be classified into two modes; nucleation mode and accumulation mode [28]. The nucleation mode particles are defined as particles with diameters less than 50 nm, and the size of accumulation mode particles is between 50-500 nanometres, and these definitions are used in this study to better interpret the data [28].

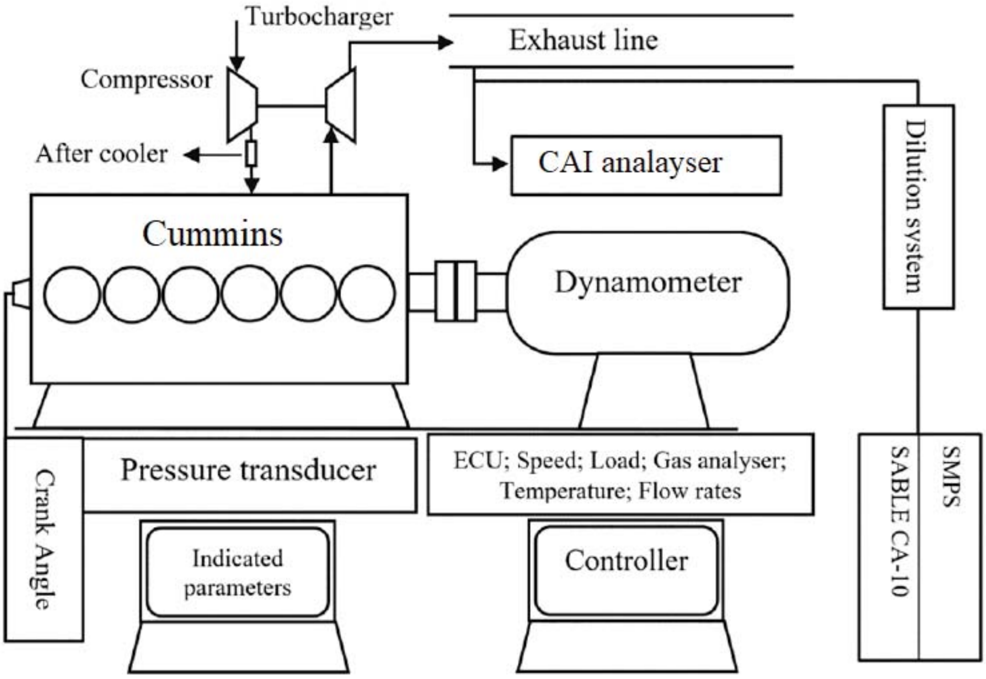

Figure 1 Test setup schematic diagram

Given that this study intended to investigate particulate matter emissions based on the current and future emissions regulation requirements, the selection of the fuels was conducted in a way to cover past, current and future biofuel blending ratio in the market. Therefore, it used diesel, and then made three diesel/biofuel blends of 10, 15 and 20% (by volume) denoted as D90C10, D85C15 and D80C20 using coconut oil biofuel (this study is a part of a project which investigates the potential of using different biofuels in diesel engines in Marshal Islands which have a high resource of coconut). In the tested fuel names, D stands of diesel and the digits after that show the volume of diesel in the blend. And C stands for coconut biofuel and the two digits after C shows the blending ratio. These fuel blends will be evaluated against diesel (D100).

Table 2 shows the tested fuel properties and Table 3 shows the chemical composition of fuels analysed by a GC/MS instrument (Trace 1310 Gas chromatograph, model ISQ, single quadrupole MS). Diesel (D100) contained aromatic compounds (benzene and its derivates, naphthalene, xylene, phthalan, mesitylene) and aliphatic compounds (mainly alkanes with 7-13 carbons, low concentrations of limonene). D90C10, D85C15, and D80C20 also contained cycloalkanes, cyclohexane and cyclooctane. With D100, the aromatic content was higher compared to other fuels. Compared to neat diesel, fuels with a lower blending ratio do not change the fuel properties significantly. For example, the density of the tested fuels, D100, D90C10, D85C15 and D80C20 were 0.84, 0.843, 0.845 and 0.846 g/cc. Also, the lower heating value of the tested fuels, D100, D90C10, D85C15 and D80C20 were 41.77, 41.31, 41.09 and 40.86 MJ/kg, respectively. However, even small changes in fuel properties can affect engine performance and emissions. For example, the higher density and lower calorific value of biofuel blends can negatively impact engine power and also fuel consumption parameters such as BSFC [8]. The fuel oxygen content of biofuel is another property that distinguishes biofuel from diesel (which has no oxygen content). It has been frequently reported that fuel oxygen content is the primary driver for decreased PM emissions with biofuels [18, 19, 58].

Table 2 Fuel properties

	D100	C100	D90C10	D85C15	D80C20
Density at 15 ºC (g/cc)	0.84	0.87	0.84	0.84	0.85
Kinematic viscosity at 40 °C (mm^2/s)	2.64	4.82	2.86	2.97	3.08
Cetane number	53.30	58.60	53.83	54.10	54.36
Lower heating value (MJ/kg)	41.77	37.20	41.31	41.08	40.86
Higher heating value (MJ/kg)	44.79	39.90	44.30	44.06	43.81

Table 3 Fuel analysis using GC/MS (Model ISQ, single quadrupole MS, Trace 1310 Gas chromatograph).

	Area %				
	aromatic	aliphatic	cyclic hydrocarbons	hydrazide	oxygenated hydrocarbons
D90C10	0.0879-0.169	0.0866-0.367	0.119-1.12	1.09-2.62	0.192-0.305
D80C20	0.0433-0.0968	0.0398-0.741	0.0394-0.222		0.0416-0.076
D100	1.43-5.66	1-12.24			

The fuels used in this study were tested under constant load (25%) and speed (1500 rpm). The rationale for using a constant load and speed was to facilitate the fundamental investigation into the influence of fuel type & engine temperature under cold-start and during engine warm-up. Most of the studies on cold-start in the literature used a drive cycle such as NEDC or WLTC and compared the first part of the cycle with the rest and made the conclusion about cold-start contribution. However, these driving cycles consist of frequent load and speed changes which add more variables to the analysis, complicating and limiting the investigation about the pure influence of fuel type and engine temperature on emissions within cold-start period. Therefore, this experimental investigation used a constant speed and engine load to limit the number of influential factors that can potentially aid a better judgment on the effects of engine temperature and fuel properties.

Experiments were done every morning with at least 12-hours of engine-off period at the ambient temperature on consecutive days in an engine laboratory. Before starting each test, coolant temperature and lubricating oil temperature were checked to be the same as ambient temperature, as per the regulation (EU Directive 2012/46/EU). Given that the engine room had an air-conditioning system, the ambient temperature during the test stayed constant (23±5 °C). For each test, the engine was started and ran under a quarter load (25%) at the speed of 1500 rpm for more than 30 min to stabilise. The statistical analysis of the test repeatability is shown in Appendix.

In diesel engines, the formation of particulate matter emissions depends on a number of variables such as engine operating condition and fuel properties [18]. During cold-start operation, the engine temperature and fuel properties are significantly influential factors. Figure 2 shows how the engine coolant temperature increases during cold-start operation with the tested engine for one of the tests. According to the regulation, (EU Directive 2012/46/EU), the cold-start period is defined from when the engine starts after 12 hours soak (or 6 hours forced cooled soak) at the ambient condition either for the first 5 minutes or during the time that the temperature of engine coolant increases to 70°C. However, after this period, the coolant temperature still has an increasing trend indicating the sub-optimal engine temperature (Figure 2). Also, it can be seen that there is a lag between the engine lubricating oil temperature and coolant temperature. Compared to the coolant temperature, the engine lubricating oil temperature remains sub-optimal for a longer period). The sub-optimal temperature period outside the formal cold-start boundaries can affect the engine performance and exhaust emissions [59, 60]. Figure 2 also shows that for the tested engine, even within the formal cold-start period, when the engine temperature reaches to 65°C, the start of injection is changed by the injection strategy commanded by the ECU of the tested engine, and this injection strategy change affects the exhaust emissions and engine performance parameters. It is worth mentioning that while the start of injection changed, the injection period (therefore, the injection mass) remained constant, as the injection was controlled by the ECU and, the injection period was independent of the start of injection. Also, there was no modification to the engine ECU/calibration for different fuel blends. Therefore the start of injection timing stayed the same for all the tests.

As discussed, it can be seen that from the engine start until the stable operation, there are some periods with different characteristics and variables. Therefore, in order to better analyse the influence of fuel properties and engine temperature, this research divides the engine warm-up period into four phases to minimise the number of variables in each phase.

- Phase 1: Formal engine cold-start, constant start of injection, coolant and oil temperatures are less than 65°C.
- Phase 2: Start of injection is increasing, coolant and oil temperatures above 65°C and still increasing.
- Phase 3: Constant start of injection, optimal coolant temperature, sub-optimal oil temperatures.
- Phase 4: Engine hot-operation, start of injection is constant, coolant and oil temperatures are optimal.

This study used an SMPS analyser with a 2 min sampling time. The first two samples were measured within Phase 1, the third sample was measured in Phase 2, the fourth and fifth samples fell into Phase 3, and the last two samples were measured within Phase 4.

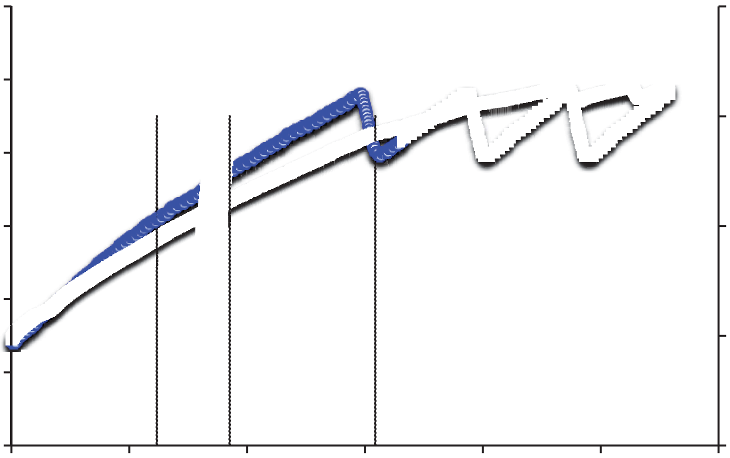

Figure 2 Engine coolant temperature, engine oil temperature and start of injection during the test with diesel

3. Result and discussion

This section analyses the PN concentration and evaluates the particle number and mass size distribution. It also investigates the median diameter and the share of PN and PM at different sizes including accumulation mode, nucleation mode and sub-23 nm particles.

Figure 3 presents the PN concentration at the four different phases. It shows that for each fuel the PN concentration in Phase 1 was lower than the other phases. Given that the engine load and speed were constant within all phases, the engine temperature can be identified as an influential factor, as shown in the literature [61]. Looking at each phase, it can be seen that the use of biofuel increased PN emissions in all of the phases. This expected result shows that fuel properties are influential, as reported before in the literature [62, 63].