

Comparison of Particulate Size Distributions from Advanced and Conventional Combustion - Part I: CDC, HCCI, and RCCI

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ABSTRACT

Comparison of particulate size distribution measurements from different combustion strategies was conducted with a four-stroke single-cylinder diesel engine. Measurements were performed at four different load-speed points with matched combustion phasing. Particle size distributions were measured using a scanning mobility particle sizer (SMPS). To study the influence of volatile particles, measurements were performed with and without a volatile particle remover (thermodenuder) at low and high dilution ratios. The use of a single testing platform enables quantitative comparison between combustion strategies since background sources of particulate are held constant. A large number of volatile particles were present under low dilution ratio sample conditions for most of the operating conditions. To avoid the impact of volatile particles, comparisons were made based on the high dilution ratio measurements with the thermodenuder. As anticipated, CDC had the highest particle number emissions for all operating conditions for particle sizes greater than the 23 nm PMP cutoff. Somewhat surprisingly, the RCCI data show significantly higher particle numbers than the HCCI data for all operating conditions. The higher RCCI particle number emissions indicate that the direct-injection of fuel and non-uniformity of the in-cylinder fuel distribution may contribute to particulate generated from RCCI. Both the RCCI and HCCI results show bimodal behavior with a large number of sub-23 nm particles present, even when high-dilution is used in conjunction with the thermodenuder. Some of these particles may be volatile particles that the thermodenuder is unable to eliminate. The nature of these particles needs to be further investigated in future work.

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INTRODUCTION

Particulate Matter (PM) formation during combustion in internal combustion (IC) engines is of major concern due to its adverse effects on the environment and human health. Additional concerns arise from the impact of PM on the formation of combustion chamber deposits and the influence of PM on engine wear [1]. Recent research on the human health impacts of PM reveals that exposure to ultrafine particles results in increased occurrence of cardiovascular events and reduction in lung function [2, 3]. In fact, diesel PM was recently classified as "carcinogenic to humans" and gasoline PM was classified as "possibly carcinogenic to humans" [4]. Due to the increasing evidence of the impacts of ultrafine particulate on human health, stricter PM regulations have been developed. The European Union established the Particle Measurement Program (PMP) to measure ultrafine particles emitted by engines in a regulatory framework, and number-based engine exhaust particle regulations have been established in Europe [5].

The particulate size distribution (PSD) of conventional diesel engine exhaust has been investigated extensively [1, 6, 7]. A bimodal distribution shape, (shown in [Figure 1](#)), has been repeatedly measured and is well accepted. Particles are divided into two categories: nucleation mode ($3\text{ nm} < D < 50\text{ nm}$) and accumulation mode ($50\text{ nm} < D < 500\text{ nm}$) particles. Typically in diesel engine exhaust, nuclei mode particles account for only a small part of total particle mass ($< 20\%$), but may contain more than 90% of the particle number [1]. Nucleation mode particles are often formed during the dilution process or as the exhaust cools and, therefore, can be highly sensitive to the dilution conditions [8]. Nucleation mode particles are usually composed of volatile organic and sulfur compounds, and potentially ash derived from the oil [9]. Accumulation mode particles are the source for the majority of the carbonaceous PM emissions. Accumulation mode particles also serve as a sink for volatile materials and ash in the exhaust when they exist in large numbers [10].

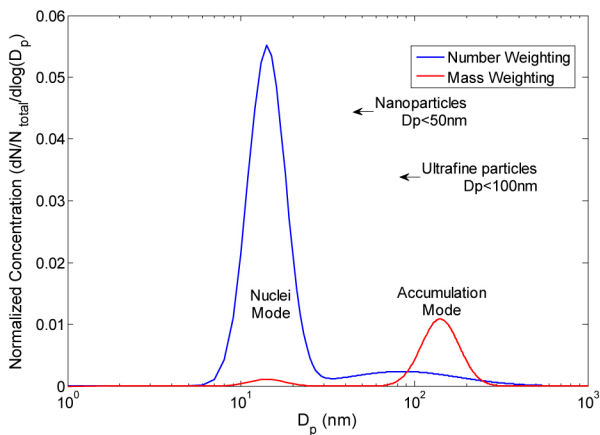


Figure 1. Typical engine exhaust size distribution both mass and number weightings (derived from [1]).

Advanced combustion strategies such as homogenous charge compression Ignition (HCCI) and dual-fuel reactivity controlled compression ignition (RCCI) have been widely investigated [11, 12, 13, 14, 15, 16]. These combustion strategies utilize dilution to achieve relatively low flame temperatures resulting in reduced formation of nitrogen oxides (NO_x) and take advantage of premixing of the air-fuel mixture to suppress soot emissions (in this paper we use PM to signify both the volatile fraction and non-volatile fraction of the exhaust aerosol and soot is reserved to refer specifically to non-volatile carbonaceous component of the PM).

Several studies have focused on detailed measurements of particulate characteristics for gasoline fueled HCCI and gasoline-diesel fueled RCCI. Kaiser et al. [17] demonstrated the influence of air-fuel (A/F) ratio on the particle size distribution (PSD) for gasoline fueled HCCI operation. They observed bimodal distributions similar to those for conventional diesel combustion (CDC) with the relative magnitudes of the two modes varying with A/F ratio. Diesel HCCI combustion PSD has also been studied by Agarwal et al. [18]. Their work also reveals the PSD dependence on A/F ratio, as well as a correlation between increasing number of accumulation particles and higher benzene soluble organic fraction (BSOF) of particulate. Prikhodko et al. [19] compared particulate size distributions from RCCI combustion with conventional diesel and diesel PPCI combustion. The RCCI results showed lower particle concentrations compared to the other two combustion strategies, and the particulate size distribution for RCCI was highly dominated by nucleation-mode particles. Additionally, the filter-collected mass samples revealed some evidence for the presence of the semi-volatile particles in RCCI exhaust. Kolodziej et al. [20] operated RCCI with both fuels direct-injected into the cylinder. The data show a bimodal PSD for RCCI that is sensitive to the gasoline SOI, as well as, the gasoline/diesel ratio. Additionally, the particle concentration for RCCI was observed to be lower than for LTC diesel combustion.

It is difficult to compare the results of particulate size distributions between works from different investigators, even if they focus on the same topic, due to the fact that measurements of particle size distributions are very sensitive to the sampling/dilution process. Measurements by Abdul-Khalek et al. [8] indicate that diesel particle number shows strong sensitivity to dilution conditions such as dilution air temperature, dilution ratio, and residence time. Luders et al. [21] also demonstrated that inadequately adjusted dilution parameters, including dilution ratio and dilution air temperature, can impact ultrafine particle formation. Thermodynamics and fluid mechanics of mixing during dilution are very complicated and vary widely between dilution setups, further impacting the comparison of measurements between different experimental setups.

Other background factors such as engine oil, ambient air purity, and the use or lack of use of volatile particle removers may also affect the measured PSD [22, 23]. Although data on the PSDs for HCCI, RCCI, and CDC are available in the literature, a comparison between these strategies in the same engine under similar dilution conditions is lacking. The previous studies of HCCI and RCCI particulates generally did not utilize volatile particle removers, so the extent to which the PM distributions were influenced by volatile particles that may be removed by oxidation catalysts on a vehicle was uncertain. To eliminate the influences of varying background sources on particle number measurements, the most direct and simplest method is to use a common engine platform and dilution system for a series of experiments.

The objective of this study is to carry out PSD measurements in a single base experimental platform. Measurements of conventional diesel combustion, RCCI, and HCCI were performed at four different load-speed points with matched combustion phasing for comparison. The sensitivity to dilution conditions was studied for each operating point, and the influence of applying a volatile particle remover (a thermodenuder) after the dilution process was studied. The comparison in the same engine with similar dilution conditions and with a volatile particle remover provides a clearer picture of the difference among combustion strategies with regards to the characteristics of their PM number distributions at multiple operating points.

EXPERIMENTAL SETUP

Engine Platform

A modified single-cylinder version of a GM 1.9 L four-stroke light-duty diesel engine with a bore of 82 mm and stroke of 90.4 mm was used in this study. The stock piston with a reentrant piston bowl was utilized. Literature has shown that PM emission can be reduced by at least an order of magnitude utilizing an optimized piston bowl geometry [24], but for the purpose of consistency the stock piston is used in this study. The engine has four valves per cylinder with a variable valve timing camshaft valve actuation arrangement. A schematic of the

engine experimental setup is shown in Figure 2. The details of the engine specifications are listed in Table 1. The intake air pressure is controlled by a series of choked flow orifices and the temperature is regulated within $\pm 2^\circ\text{C}$ with an inline air heater. The engine oil is heated to 70°C and the coolant is controlled at 80°C .

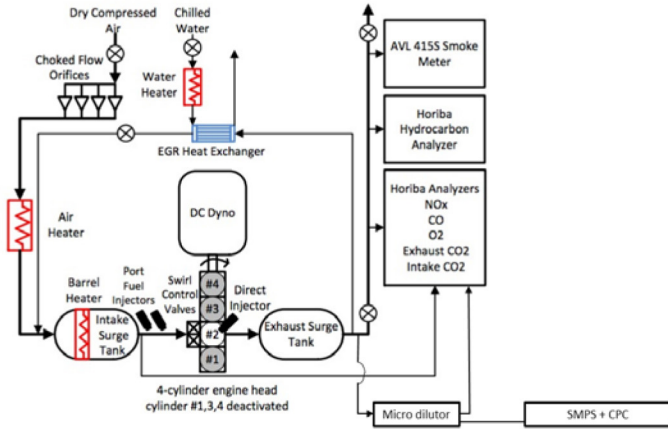


Figure 2. Schematic of the engine experimental setup.

Table 1. Engine specifications.

Base Engine	Single-cylinder version of GM 1.9L 4-cylinder DI Diesel
Bore x Stroke	82.0 mm x 90.4 mm
Connecting Rod Length	145.54 mm
Displacement	0.477 L
Compression Ratio	16.7:1
Intake Valve Opening/Closing	344°/-132° aTDC
Exhaust Valve Opening/Closing	112°/388° aTDC
Swirl Ratio	1.5
Piston Bowl Type	Re-entrant bowl
Injector	Bosch CRIN2 154 μm x 6 at 144°

Fuel System

The intake manifold is equipped with two port fuel injectors to deliver premixed fuel into the swirl-actuated intake port. The port-fuel-injection (PFI) fuel was injected at 200°bTDC during the intake stroke. A centrally mounted common-rail injector is located in the cylinder head and is used to deliver direct-injection (DI) fuel. Fuel rail pressure is controlled by Mototune software, and NI Driven software is used to control all of the injectors. The gasoline used in this study was a standard 87 AKI pump gasoline, and the diesel fuel used was an ultra-low sulfur diesel sourced from a local fuel supplier. For HCCI operation, one of the PFI injectors delivers gasoline, and the other is fed with gasoline mixed with n-heptane at 20% by volume. The addition of n-heptane helps to achieve the combustion phasing required to match the CDC, and RCCI phasing. The small portion of n-heptane additive (~10% of the total premixed fuel volume) should not have any significant influence on soot formation. Dempsey et al. [25] conducted a study comparing the performance of pump fuel vs. Primary research fuel (PRF) in the same engine platform of this research, and they found no significant difference in the smoke meter measurements for all cases.

Data Acquisition

National Instruments LabView software is utilized for system control and data acquisition. A Kistler 6125A pressure transducer together with a Kistler 4010 charge amplifier is used to measure the in-cylinder pressure. A 1440 pulse per revolution shaft encoder allows for pressure acquisition at a resolution of 0.25 crank angle degrees (CAD). Pressure data are acquired for 300 consecutive cycles at each experimental point. Horiba gaseous emissions benches are used to measure emissions including NO_x , UHCs, O_2 , CO, CO_2 , and EGR/diluted exhaust (for particulate sampling) CO_2 . All species are measured on a dry basis, except for UHCs which are measured on a wet basis. (UHCs are subsequently converted to a dry basis; all species are reported as such.) Filter smoke number (FSN) is measured with an AVL smoke meter.

Particulate Sampling System

Particulate was sampled from the exhaust after the exhaust surge tank and diluted using a two stage micro-diluter (Dekati FPS-4000). A heated probe extracts a sample from the exhaust flow that is sent to a heated perforated tube primary diluter, and subsequently undergoes a room temperature secondary dilution with an ejector diluter. Exhaust dilution is used to achieve an appropriate particulate inlet concentration for the scanning mobility particle sizer (SMPS) used to measure the PSDs. A schematic of the micro-diluter system is shown in Figure 3.

The SMPS system consists of a long differential mobility analyzer (DMA, TSI model 3081) controlled by an electrostatic classifier (EC, TSI model 3080). An x-ray based neutralizer (TSI model 3088) is used to establish the bipolar charge distribution on the particles. The particle sample sized by the DMA is measured using a condensation particle counter (CPC, TSI model 3010). The system has the capability of resolving particle mobility diameters from 7 nm to 300 nm (the scan range used in the current study). Before entering the SMPS, the sample aerosol goes through an inertial impactor with a 600 nm cut-point diameter to remove large particles. The Aerosol Instrument Manager (AIM) software (TSI) is used to acquire and process the particulate size distribution (PSD) data. Both the correction for particle diffusion losses in the SMPS and the multiple charge correction were applied to the current data set.

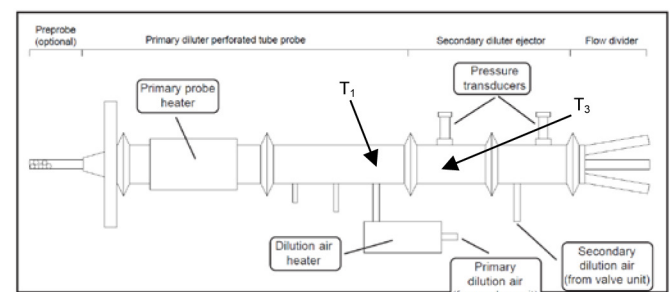


Figure 3. Schematic of the micro-diluter control unit and micro-diluter cross-section [26]

The dilution process is carried out in two phases. The first (primary) dilution is conducted in a perforated probe located near the tip of the diluter. The primary dilution air is heated to prevent condensation of volatile species. Thermocouple T1 (labeled in Fig 3) is used to monitor the temperature of the primary dilution air. The second phase is an ejector-type diluter, located downstream from the primary dilution. The ejector diluter acts as a pump, sucking an amount of a diluted sample from the primary dilution and further diluting the sample from the primary dilution with room temperature air. The dilution air is filter with a HEPA filtration system prior to being used to dilute the sample and contains negligible particles in the size range for the current measurements. The sample temperature following the primary dilution, before the ejector diluter, is also recorded and referred to as T3. When repeating experiments at the same operating conditions both T1 and T3 were matched as closely as possible, since the sample dilution process is very sensitive to the temperature profiles within the micro-diluter. The flow rates of the primary dilution air and the secondary dilution air are controlled by solenoid ON/OFF valves in the control unit for the micro-diluter. The dilution ratio (DR) is determined by CO₂ measurements in both the diluted and undiluted exhaust samples:

$$DR = \frac{[CO_2]_{\text{diluted}} - [CO_2]_{\text{background}}}{[CO_2]_{\text{undiluted}} - [CO_2]_{\text{background}}}$$

In this study we use two sets of dilution air valve settings to achieve a low dilution ratio (LDR, ~ 18:1) and a high dilution ratio (HDR, > 50:1).

Absolute uncertainties for the SMPS number concentration measurements are difficult to quantify, however, comparison with measurements from an Engine Exhaust Particle Sizer show generally good agreement in total number magnitude, within better than a factor of two. The mobility diameter accuracy has been verified at one diameter to be within the uncertainty associated with the diameter bin size of the SMPS, which is diameter dependent. We focus on repeatability of the measurements as the most pertinent measure of the relative uncertainty to determine the significance of trends. An example of this is shown in Figure 4 where the 95% confidence intervals are seen for CDC mode 2 for all dilution cases. Generally, the confidence intervals are tight relative to the mean, especially for mobility diameters greater than 40 nm. More variability can be present at small diameters with confidence bands spanning a factor of two to four in concentration. It should be noted that the HDR data show noticeably less variability at all diameters (less than a factor of two) and that the data used for comparisons throughout the paper are HDR data. These confidence bands are typical results for most of the data.

A thermodenuder (TD, Dekati) is installed in one of the lines between the micro diluter and the SMPS to remove volatile particles. Measurements could be taken with or without the thermodenuder by switching the flow path with a valve. The thermodenuder operates by first vaporizing volatile compounds in an aerosol heater, which heats the sample to 250 °C. After

heating, the sample enters the absorber section, where the vaporized compounds are adsorbed by the activated charcoal filter. A particle loss correction given in the manufacturer's manual is applied to data taken with the thermodenuder during the data processing.

Operating Conditions

The engine was operated at four speed/load conditions with three combustion strategies: conventional diesel combustion, RCCI, and HCCI. The parameters for the operating conditions are given in Table 2. The four operating modes are chosen to represent a wide range of operating conditions for a light duty engine, ranging from low-speed idle conditions to relatively high-speed high-load conditions. In this study, the combustion phasing, as determined by the location of 50% energy conversion (CA50), was matched among the different combustion strategies at each speed/load condition. This provides an appropriate comparison in general. However, this will give different results than if each strategy had been individually optimized at their own CA50. The comparison of the strategies at individually optimized conditions is beyond the scope of the current work, but is a topic for future research. For mode 8 operation with RCCI, combustion became very unstable, so the CA50 was advanced to obtain an acceptable coefficient of variation (COV) of the indicated mean effective pressure (IMEP). For all combustion strategies the operating parameters (injection timing/pressure, intake temperature, EGR level, A/F ratio, etc.) are varied to achieve matched combustion phasing and to insure stable combustion (COV of IMEP <5%).

Table 2. Engine operating conditions. Variable parameters are distinguished by color (blue for CDC, green for RCCI, red for HCCI) - indicates that item is not applicable for the combustion strategy or mode.

	Unit	mode 2	mode 5	mode 7	mode 8
Speed	rpm	1490	1900	2300	2300
IMEP (gross)	bar	4.2	5.7	5.7	8
Intake Temp.	C	50/35/85	50/35/70	50/35/75	35
Intake Pressure	bar	1.15	1.3	1.3	1.88
Exhaust Pressure	bar	1.25	1.45	1.5	2.1
Injection Pressure	bar	950/500/-	950/500/-	950/500/-	950/500/-
Injection Timing	aTDC	-25/-4/-43/-	-7.5/-44/-	-10/-48/-	-9/-44/-
CA50	aTDC	4.5	4.5	4.5	7.5 (5 for RCCI)
EGR	%	-	-	-	-/48/42

Particulate Data Operating Procedure

Maintenance of the particle sampling system is conducted prior to data collection. The micro-diluter system needs to be cleaned after every 20 hours of operation to remove the soot deposits. This was important because the deposits reduce the cross sectional area that the sample flow passes through.

impacting the dilution ratio and temperatures within the dilution tunnel. A valve downstream of the flow divider on the micro-diluter system is used to control the pressure at the outlet of the micro-diluter. This insures a sample flowrate of 1.3 L/min to the SMPS. The activated charcoal filter in the TD is replaced every two days of operation with a regenerate filter. To regenerate the filter, the filter is baked in oven at 165 °C for 8 hours. During data acquisition, the time interval between acquiring each set of data was chosen to be long enough to ensure that all temperatures in the system had reached steady-state. For each operating condition, the PSD was measured using 7 scans (115 s scan-up and 30 s scan-down times). The results for the 7 scans were averaged and corrected for dilution ratio for all of the plots presented.

RESULTS AND DISCUSSION

DR/TD Investigation

Before taking the bulk of the experimental data, preliminary tests were conducted to investigate the effect of the dilution ratio, as well as sending the diluted sample through the TD, on the measured PSDs. For each operating condition and each combustion strategy the PSDs were acquired at both the LDR and HDR with and without the TD, giving four measured PSDs for each operating condition. The results for CDC Mode 2 are presented in Figure 4 as an example. All other modes show similar results, and can be found in Appendix A. In the following discussion, we refer to particles with mobility diameter smaller than 50 nm as nucleation (or nuclei) mode particles, while particles larger than 50 nm are referred to as accumulation mode particles.

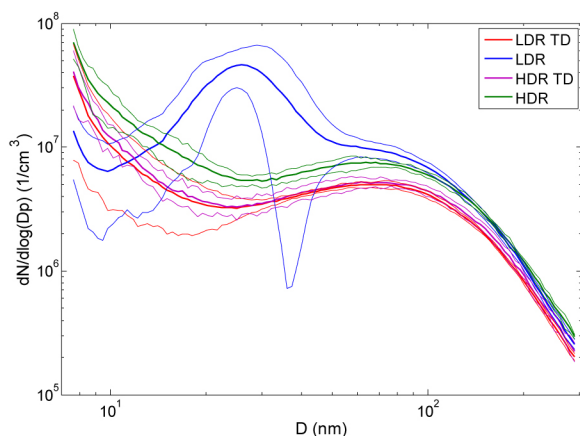


Figure 4. CDC Mode 2 (1490 rpm/4.2 bar IMEP) PSD curves: DR/TD investigation; Solid lines for PSD, dashed lines for 95% confidence interval for each color-matched cases.

Figure 4 shows the PSD curves acquired for the different particulate sample conditioning tested for CDC Mode 2. The parameters for the dilution process are given in Table 3. The solid lines stand for the PSD under different sampling condition, and the dashed lines represent the 95% confidence interval. The LDR data without the TD show a strong bimodal character with a large nucleation mode. Increasing the dilution

ratio greatly reduces the size of the nucleation mode and shifts it to lower particle diameters. For the LDR the nucleation mode peak of the PSD occurs around 25 nm, when using the HDR the peak is pushed to the smallest diameter resolved (7 nm). Despite the obvious change in the small diameters, the PSDs of the accumulation mode particles match each other very well, indicating the DR does not have a substantial effect on larger particles, except to possibly reduce the amount of organics absorbed on the surface of these particles.

As mentioned earlier, PSDs are very sensitive to the temperature profile (T_1 and T_3) within the micro diluter. Since the change in the DR is achieved by changing the flowrates of the primary (heated) dilution air and the secondary (unheated) dilution air, the ratio of heated/unheated dilution air has an effect on temperatures during the dilution process. As is shown in Table 3, the change in the dilution ratio increases T_1 by 5% and T_3 by 16%. This is due to the larger flowrate of heated primary dilution air for the HDR cases. The combination of increased dilution and increased temperature for the HDR condition appears to be almost as effective at eliminating volatile particles as applying the TD. The effect of TD is less obvious, but it appears that with the TD, some additional amount of small particles (<20 nm) are removed. Utilizing the TD also seems to give more consistent results that are less sensitive to dilution conditions, as indicated by the two TD curves in Figure 4 that are nearly identical even though the dilution conditions are measurably different.

Table 3. Parameters for the dilution process for CDC Mode2, temperature units [°C]. TD set temp is the set point temperature for the thermodenuder heater, T1 is the primary dilution air temperature measured in the micro diluter, T3 is the temperature after the primary diluter, T_{sample} is the temperature in the exhaust at the sampling point, Valve settings refer to the setting for the valves in the FPS control unit.

TD set temp	T1	T3	T_{sample}	Valve setting	DR
-	272	142	141	2,5	17.5
250	272	143	143	2,5	17.3
-	287	166	140	2,6	105.3
250	287	167	140	2,6	105

There is a reasonably large discrepancy between the HDR data and the data with the TD. This is assumed to be due to losses in the TD itself or within the flowpath which the TD is installed. The loss correction for the TD applied to the data is insufficient to correct the TD to the number seen with the HDR which has a distribution similar to the TD data. The extra losses with TD seem to have a slightly larger effect on smaller diameter particles (potentially due to diffusion losses). The lower particle number seen with the TD was similar for all of the data presented. Since it is believed that the TD provides the most robust removal of volatile particles the data presented in the rest of the paper is from the HDR cases with the TD.

Therefore, comparisons are made based on the non-volatile particles generated for each combustion mode. The basis for this comparison is based on the fact that oxidation catalysts