Particles from Soot Free Engines

David Kittelson, Anil Bika, Wei Fang, Luke Franklin, and Bin Huang
University of Minnesota, Center for Diesel Research

New engine technologies and fuels offer the promise of virtually soot free combustion. Some examples include: homogeneous charge compression ignition (HCCI) engines operate under lean, highly dilute low temperature conditions that results in little or no soot formation; Diesel engines operating on non-sooting fuels like dimethyl ether; and spark ignition engines operating on hydrogen, and if properly done, natural gas. However, all these non-sooting engines still produce particle emissions that are formed from the lubricating oil. The work described here is a study of particle emissions from a single cylinder conversion of a 5.2 liter Isuzu Diesel engine converted to operate under HCCI conditions using anhydrous ethanol, regular unleaded gasoline (ULG) AKI = 87, and hydrogen as fuels. Great care has been taken to ensure good mixing of the fuel and air so that combustion is as homogeneous as possible in order to avoid soot formation.

HCCI combustion was controlled using intake air heating and the test matrix was designed with the aid of CHEMKIN® modeling of the HCCI combustion process. It was necessary to run relatively light load conditions, IMEPs less than about 500 kPa in order to avoid excessive rates of pressure rise. Particle size and concentration were measured with a nano-SMPS configured to measure particles in the size range from 2 to 64 nm mobility diameter for most measurements but a limited number of measurements were made at diameters up to 160 nm. None of the fuels showed any evidence of soot formation or particles larger than about 60 nm under firing conditions. Engine exhaust was diluted using a two stage dilution system illustrated in Figure 1.

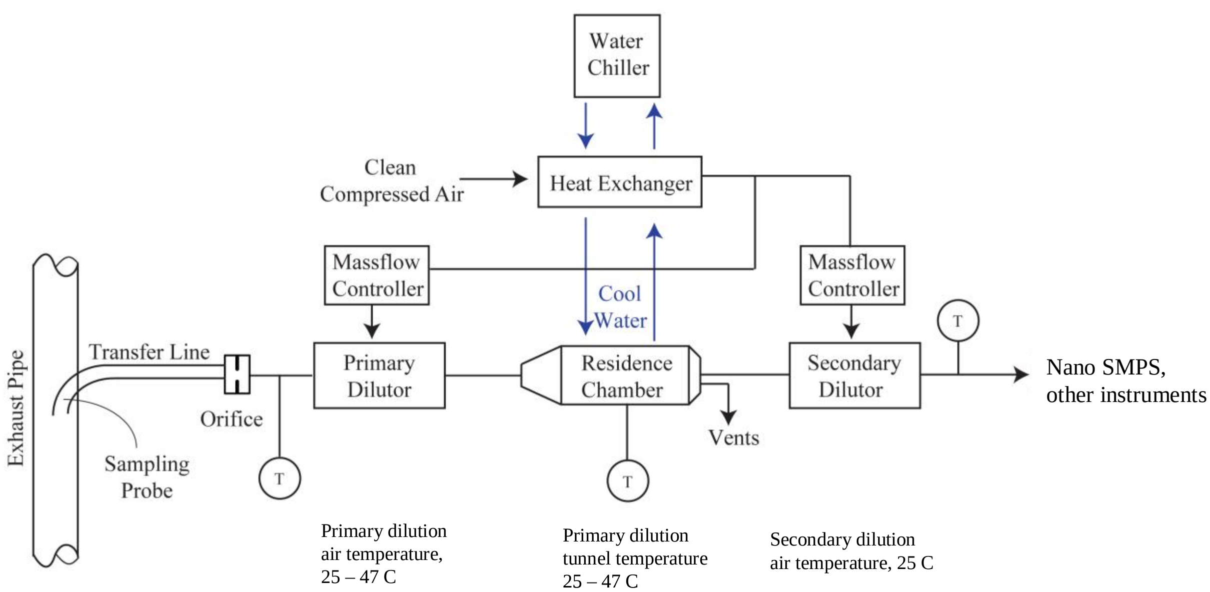


Figure 1 – Two-Stage Dilution System – Primary Dilution Temperature = 47 C. Primary Dilution Ratio = 15 Except in Dilution Sensitivity Tests, Secondary Dilution Ratio = 15, Primary Residence time ~ 1.5 sec

Past work has shown that conditions in the first stage (primary) of dilution strongly influence volatile nanoparticle formation (Abdul-Khalek, 1999). For this work the primary dilution temperature was fixed at 47 C and the primary dilution ratio was varied to determine dilution sensitivity. The emitted particles were found to be nearly 100% volatile material. This made them extremely sensitive to dilution conditions. Figure 2 shows total number and volume concentrations measured with an SMPS and corrected for dilution ratio plotted against primary dilution ratio. Both number and volume decrease markedly with dilution ratio and there is no obvious stable sampling condition. However the slopes with respect to dilution ratio are relatively flat at a ratio of 15 and this ratio was used in all subsequent tests.

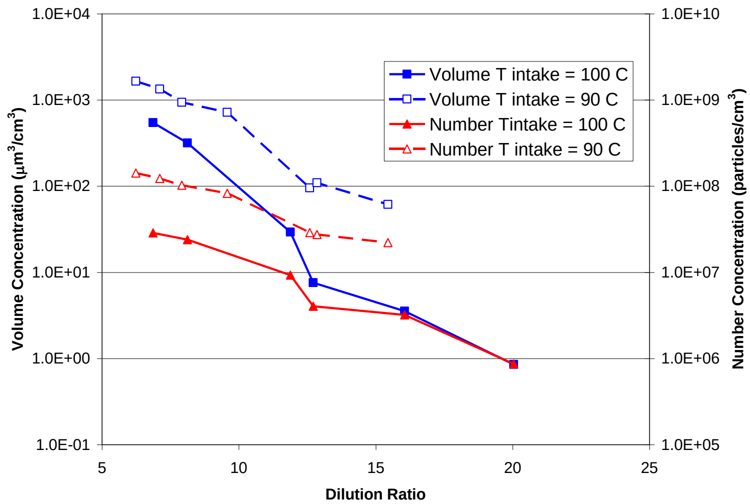


Figure 2 – Dilution Sensitivity - Total SMPS Number and Volume Concentrations Plotted against Primary Dilution Ratio, Primary Dilution Temperature = 47 C. Ethanol Fuel Energy Input 1.1 kJ/cycle

Figure 3 shows mass weighted size distributions for the engine running on ULG at a fuel energy input of 1.04 kJ per cycle which corresponds to an IMEP of approximately 250 kPa. Mass concentrations have been calculated from SMPS volumes assuming spherical oil droplets. Results are shown for three speeds. At each speed the intake temperature was adjusted to give maximum IMEP. Also shown are corresponding motoring tests with the engine running at the same speeds and intake temperatures. Figure 4 shows size distributions for the same engine conditions with a catalytic stripper placed upstream of the SMPS. Volume reduction in all cases is more than 99% indicating that the particles are nearly all volatile. The results of these size distribution measurements are summarized in Figure 5 which shows plots of total mass concentration measured with the SMPS for motored and fired conditions, with and without the catalytic stripper plotted against engine speed. In all cases the motored tests led to higher particle mass emissions than the fired tests. The solid fractions measured with the CS (indicated by the number next to the data points) were slightly higher for the fired tests but the volatility experiments described below showed that the particles produced under fired conditions were more volatile than those produced by motoring.

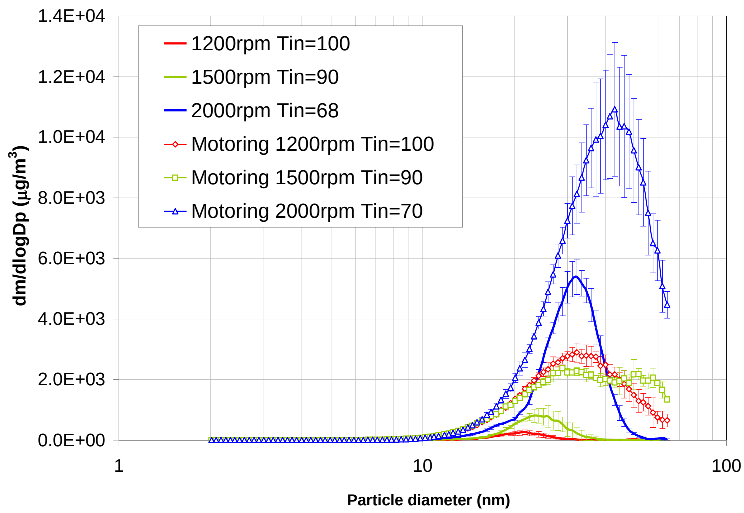


Figure 3 – Firing and Motoring Size Distributions at 1200, 1500, and 2000 RPM, Unleaded Gasoline, Fuel Energy Input 1.04 kJ/cycle. Intake Temperatures Adjusted to Give Maximum IMEP under Firing Conditions.

Volatility measurements of size selected particles were done using the tandem DMA method described in detail by Sakurai, et al., (2003). With this method two differential mobility analyzers (DMA) are used in series. The first is used to select particles in a narrow size range, in this case the mode of the number weighted size distribution. These particles are then passed through a heating tube and allowed to partially evaporate and subsequently sized by the second DMA. Figure 6 is a plot of diameter decrease against heater temperature for the engine running at 1500 RPM on ethanol fuel at low and medium loads ranging from 230 to 400 kPa IMEP. Also shown are the result for motoring at 1500 RPM and plots of predicted evaporation behavior of C28, C30, and C32 normal alkane particles. The latter plots were calculated using the approach suggested by Sakurai, et al. (2003), who observed that nanoparticles emitted from a heavy-duty Diesel engine exhibited evaporation behavior similar to these alkanes. They concluded that these Diesel nanoparticles were formed mainly from lubricating oil. Particles formed in the three fired cases all showed similar evaporation behavior but the particles formed by motoring, which would be expected to be mainly unburned lubricating oil were much less volatile. This suggests that lube oil particles produced under firing conditions have been partially broken down into more volatile components. The lower PM emissions under firing than motoring may be due to a combination of cracking and combustion of the lube oil and lower oil consumption associated with better sealing by the piston rings under fired conditions. Whatever the explanation, these results suggest that motoring experiments are unlikely to give a good estimate of the contribution of lubricating oil to PM emissions.

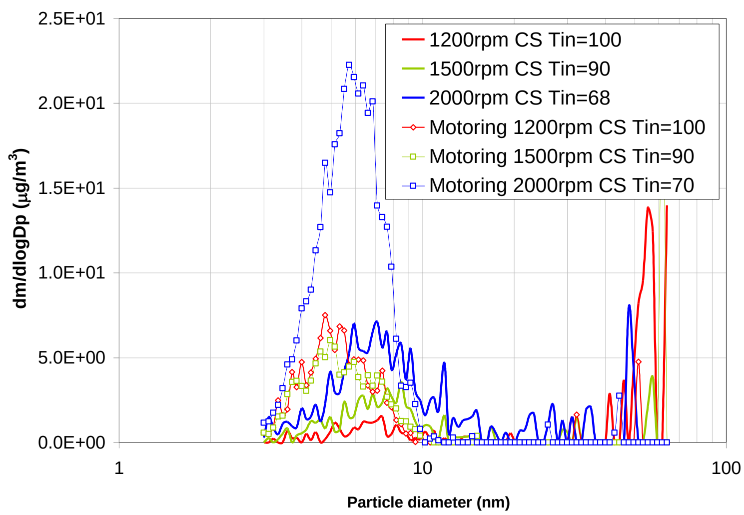


Figure 4 – Firing and Motoring Size Distributions with CS at 1200, 1500, and 2000 RPM, Unleaded Gasoline, Fuel Energy Input 1.04 kJ/cycle. Intake Temperatures Adjusted to Give Maximum Firing IMEP

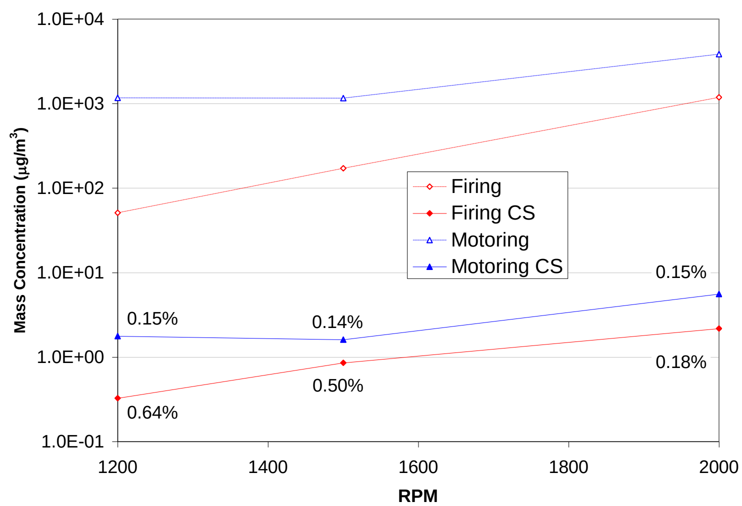


Figure 5 – Firing and Motoring Exhaust Mass Concentrations, ULG Fuel with Intake Temperature Adjusted to Maximum IMEP, Fuel Energy Input 1.04 kJ/cycle. Numbers on CS Plots Are Solid Fractions

The fuel used had only a minor influence on the particle emissions. Hydrogen fuel produced nearly the same emissions as ethanol or gasoline providing further evidence that the particles were formed from the lubricating oil.

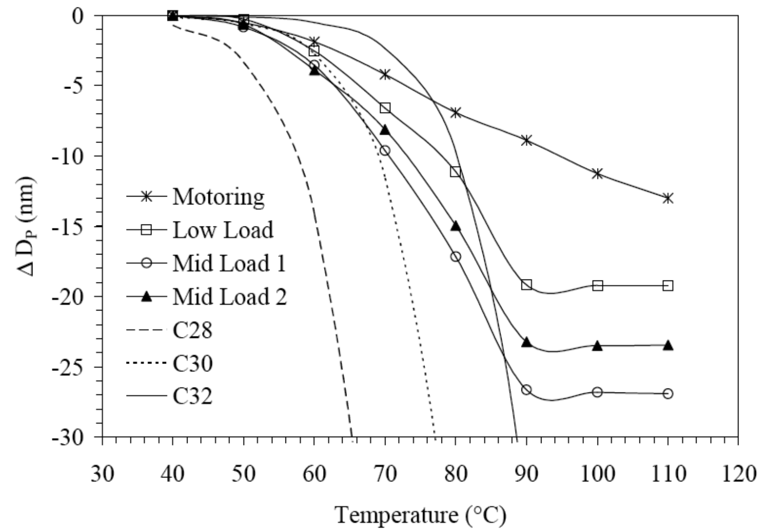


Figure 6 – Tandem DMA Volatility Tests, Particle Diameter Decrease vs. Heater Temperature. Engine Conditions: Ethanol Fuel, 1500 RPM, IMEPs of 230, 310, and 400 kPa. Theoretical Results Shown for 3 Normal Alkanes

The emissions were dependent upon the in-cylinder temperature history. Figure 7 shows plots of CO, NOx, and PM emissions against peak heat release rates (HRR). Emissions of all three pollutants are strongly correlated with peak HRR. Both NOx and PM increase with peak HRR. PM emissions are surprisingly high and exceed 0.1 g/kWh, more than 10 times the 2010 U.S. heavy-duty truck standard at the highest peak HRR.

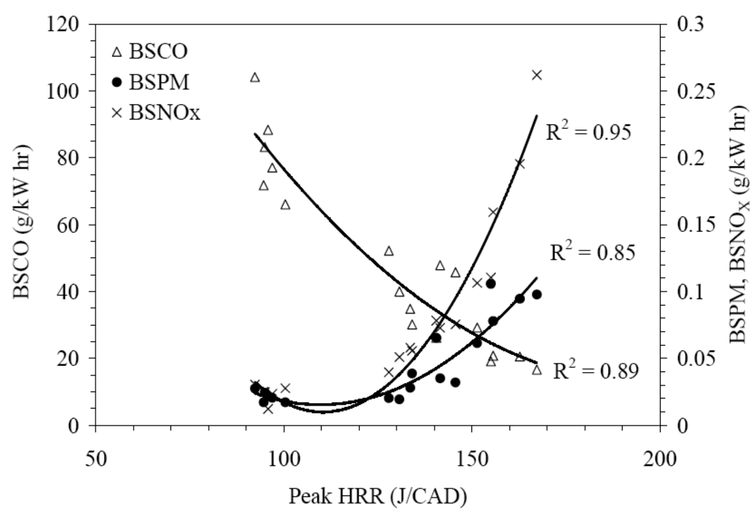


Figure 7 – CO, PM, and NOx Emissions Plotted against Peak Heat Release Rate for Different Fueling Rates, 1500 rpm, Ethanol And Hydrogen Fuels

In conclusion, these results show that even with ultra clean low temperature combustion, significant PM emissions were formed from lubricating oil. It appears that these particles were formed by a combination of atomization and evaporation of the lubricating oil and subsequent thermal processing in the combustion chamber.

References

Abdul-Khalek, I., D.B. Kittelson, and F. Brear. 1999. "The Influence of Dilution Conditions on Diesel Exhaust Particle Size Distribution Measurements," SAE Paper No. 1999-01-1142, 1999.

Sakurai, Hiromu, Kihong Park, Peter H. Mcmurry, Darrick D. Zarling, David B. Kittelson, and Paul J. Ziemann, 2003, "Size-Dependent Mixing Characteristics Of Volatile And Non-Volatile Components In Diesel Exhaust Aerosols," Environ. Sci. Technol., 37, 5487-5495

Particle Emissions from a Soot Free Engine

Anil Bika, Wei Fang, Luke Franklin, Bin Huang, and David Kittelson
Center for Diesel Research
Department of Mechanical Engineering
University of Minnesota

15th ETH-Conference on Combustion Generated Nanoparticles

June 26th – 29th 2011
Zurich, Switzerland

Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

Soot free and low soot engines / fuels

- Low temperature combustion – there are a variety of so called low temperature combustion concepts including:
  - HCCI, Homogeneous Charge Compression Ignition)
  - PCCI or PCI (Premixed Charge Compression Ignition)
  - RCCI (Reaction Controlled Compression Ignition)
  - *All rely on control of the temperature – mixing history to avoid passing through regions of soot and NOx formation. However only very well mixed, lean HCCI has the potential to completely eliminate soot emissions.*
- There are also fuels that lead little or no soot formation including:
  - Natural gas – if you do it right
  - DME (dimethyl ether) – it is nearly impossible to do it wrong
- But all of these processes and fuels still emit PM, especially in the nanoparticle range
- ***The work presented here is for a converted Diesel engine running HCCI with ethanol, gasoline, and hydrogen fuels***
- ***Number and mass emissions of particles were of the same order as those from contemporary Diesel engines without aftertreatment but the particles were nearly all volatile***

Why are we interested?

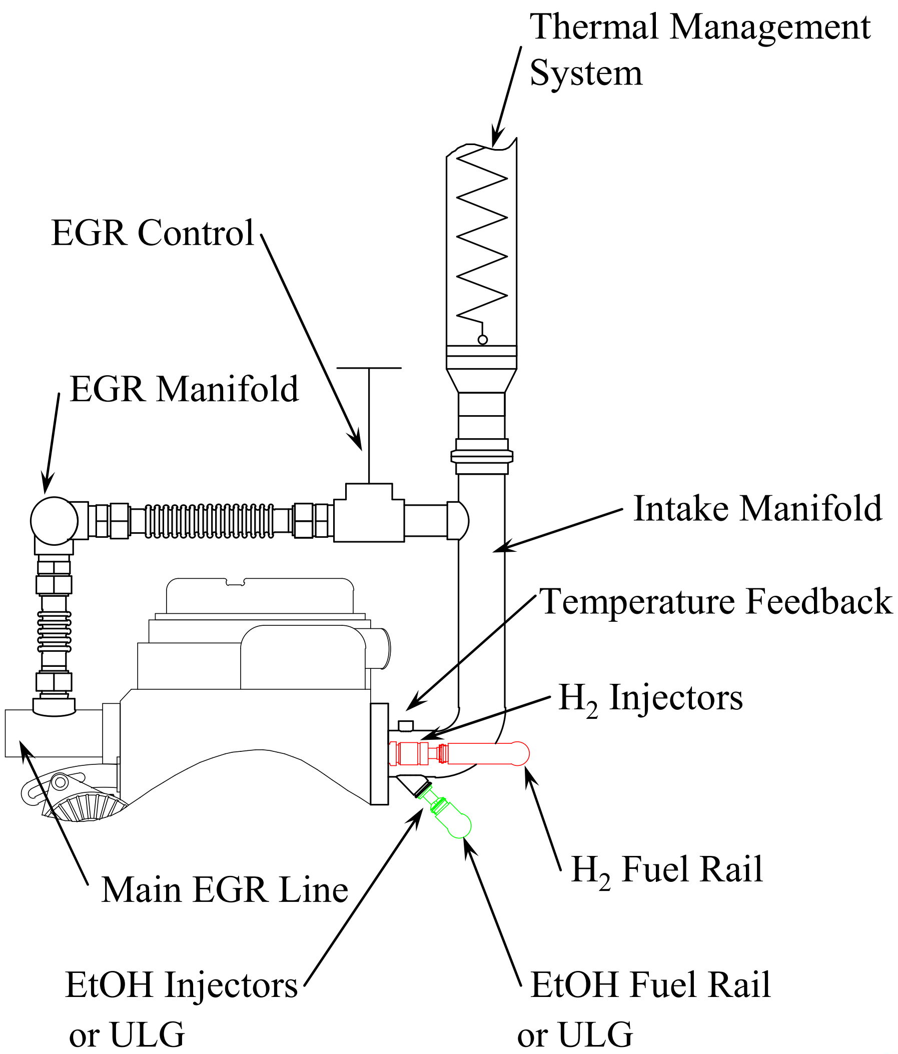
- To determine the source PM emissions from low temperature combustion
  - Engine modifications to minimize emissions
  - Identification of appropriate aftertreatment, exhaust temperatures are low
- To examine the use of soot free engine combustion to improve our understanding of the contribution of lubricating oil PM emissions
  - Oil and related ash emissions impact performance of exhaust filters
  - Typically emissions under fired conditions are much lower than under motored conditions
  - Emissions related to heat release rate and internal cylinder temperatures and pressures and their influence on lubricating oil evaporation and atomization
  - Very sensitive to engine conditions and history

Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

Engine modifications for HCCI

- The test engine is a modified 2005 Isuzu 4 cylinder, 5.3 L, medium-duty Diesel engine.
- Turocharger and aftercooler removed
- Common rail Diesel fuel injection not used
- Primary fuel ethanol or unleaded gasoline preheated to improve atomization
- Independent control of EGR, air temperature, hydrogen, ethanol or unleaded gasoline
- Closed loop controlled thermal system capable of maintaining temperatures of 150 °C
- Intelligent Controls IC 5620 engine management system used for fuel injector control



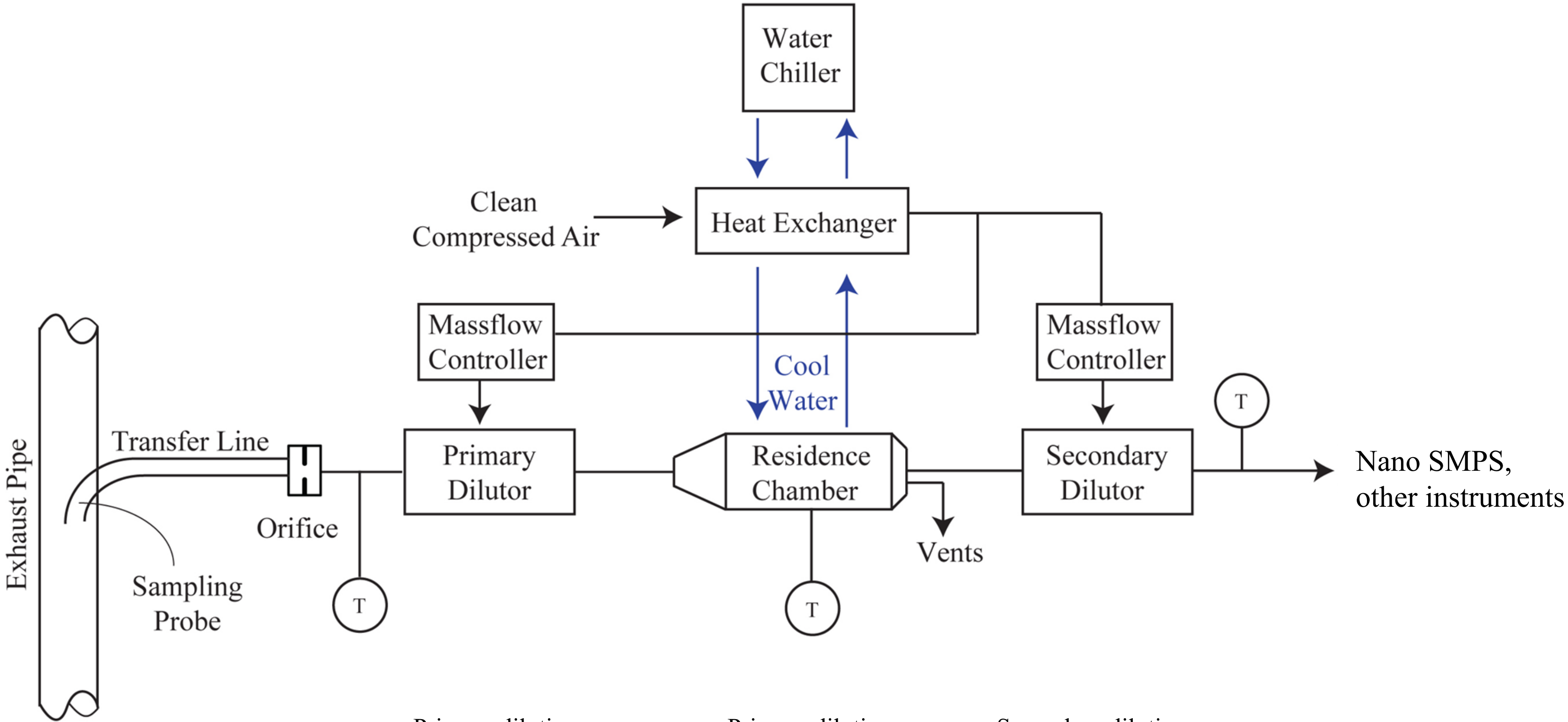
Test conditions

- Engine combustion was controlled in three ways
  - Variable intake temperature
  - Variable EGR rate
  - Changing fuel blend by substituting H_2 for ethanol
- Engine speed was varied from 1200 to 2000 rpm but most tests were done at constant engine speed of 1500 rpm
- The table below shows variable temperature test conditions for ethanol tests
- Variable EGR and fuel blending test were done at essentially same load and equivalence ratio ranges
- Tests with gasoline and hydrogen have been done mainly in the low and mid-1 range

	Engine Load		
	Low	**Mid-1**	**Mid-2**
λ range	5.0 - 4.2	4.0 - 3.5	3.2 – 3.0
Fueling Rate (g_{EtOH}/sec)	1.43	1.84	2.24
Fuel Input Energy Rate (kW)	42.5	54.6	66.5
Intake Temperature Range (°C)	110-160	90-130	90-110
Load Range (N•m)	48-55	59-93	118-128
IMEP Range (kPa)	221- 236	233 - 318	383 - 403

Sampling and dilution system

Dilution ratios, primary = 18 early work, 15 later work, secondary = 15
Residence time ~ 1.5 sec



Primary dilution
air temperature,
25 – 45 C

Primary dilution
tunnel temperature
25 – 45 C

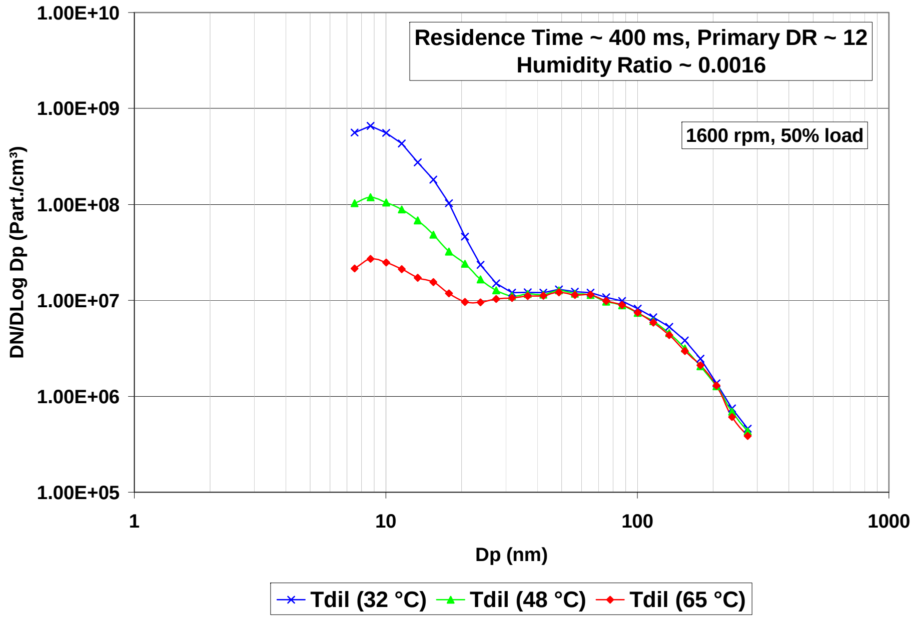
Secondary dilution
air temperature, 25 C

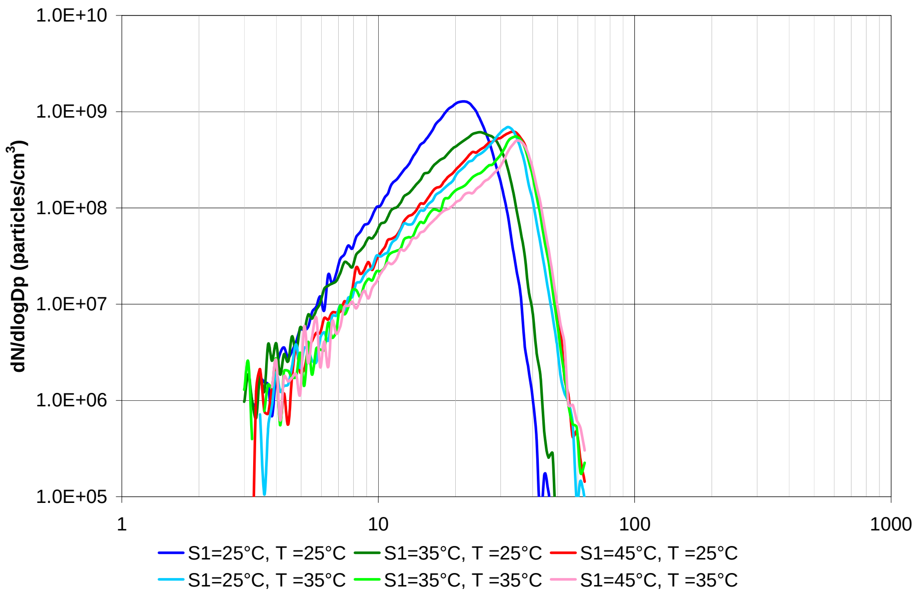
Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

Nanoparticle formation is very sensitive to dilution conditions - comparison with Diesel dilution sensitivity

The sensitivity to dilution temperatures is similar the that observed with Diesel nanoparticles. Our first set of experiments were done with primary tunnel and dilution air temperatures, 35 and 35 C, respectively, recently we have moved to 47 and 35 C to make the tunnel temperature compliant with EPA filter sampling temperature.

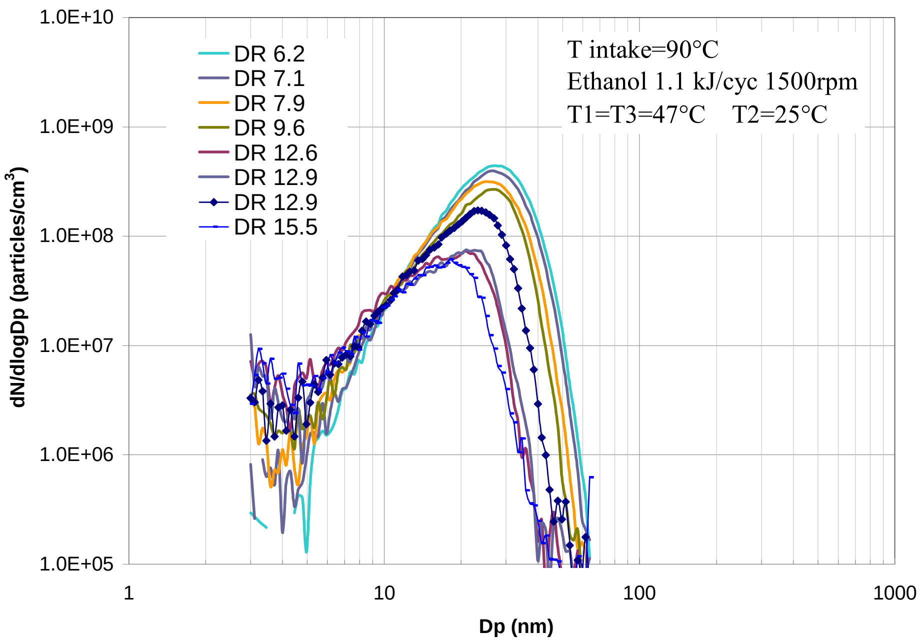


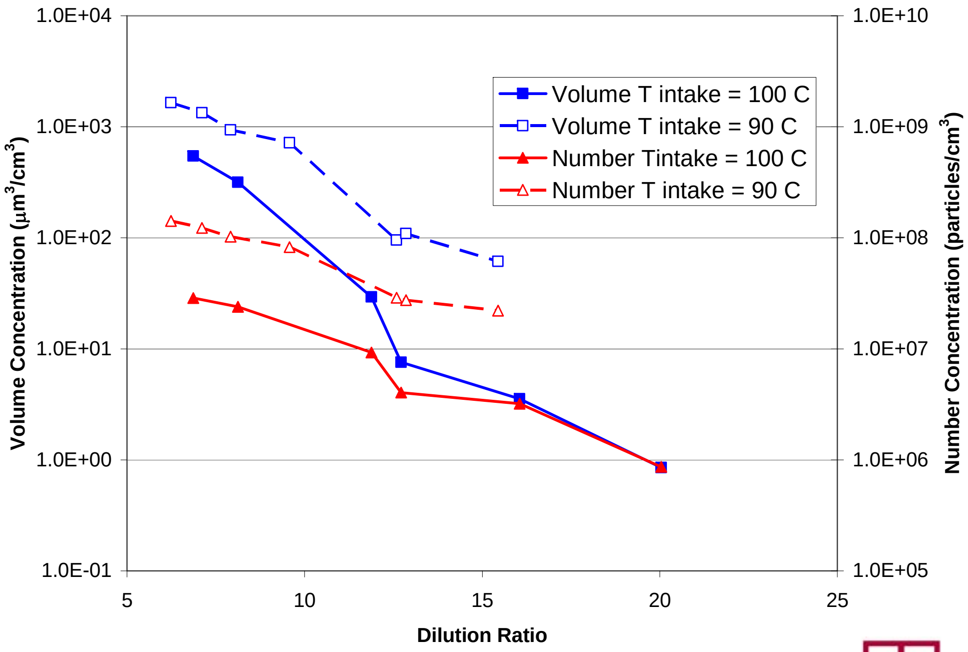


Abdul-Khalek, I., D.B. Kittelson, and F. Brear. 1999. "The Influence of Dilution Conditions on Diesel Exhaust Particle Size Distribution Measurements," SAE Paper No. 1999-01-1142, 1999.

Further examination of dilution sensitivity showed strong dependence on dilution ratio

- We decided to standardize first stage dilution temperature – which is the critical one, to 47 C
- Both total number and total volume are very sensitive to dilution conditions but volume is most sensitive due to effect of both changing number and size
- There is no stable region for DR but the curve is “relatively” flat at DR = 15 so that is where we have mainly tested

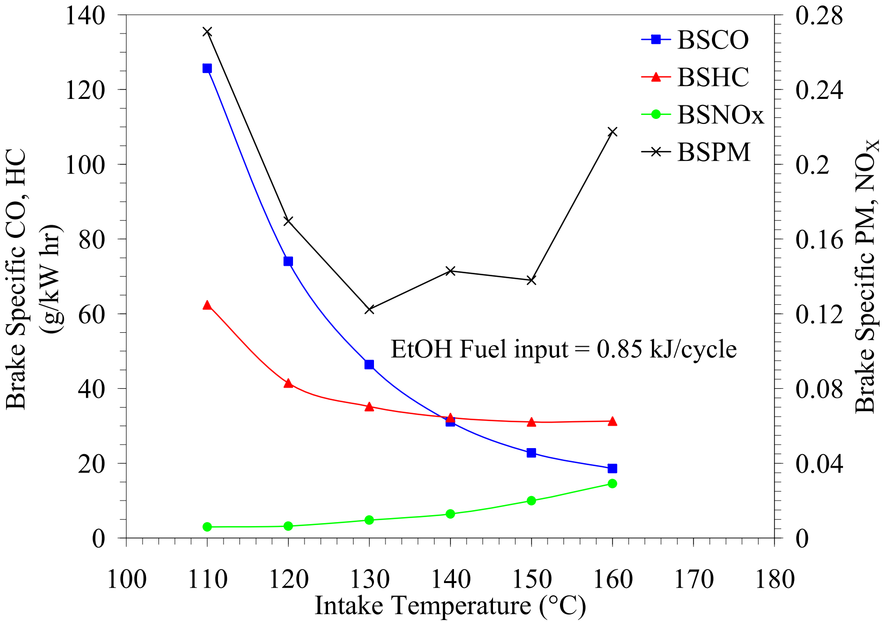


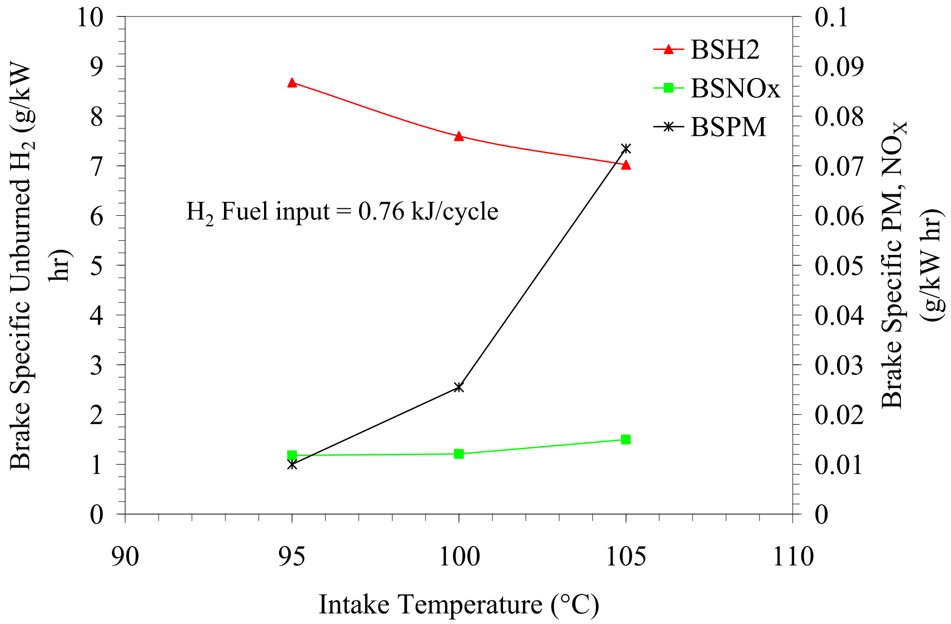


Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

Influence of intake temperature (ignition timing) on emissions with pure ethanol and hydrogen fuels



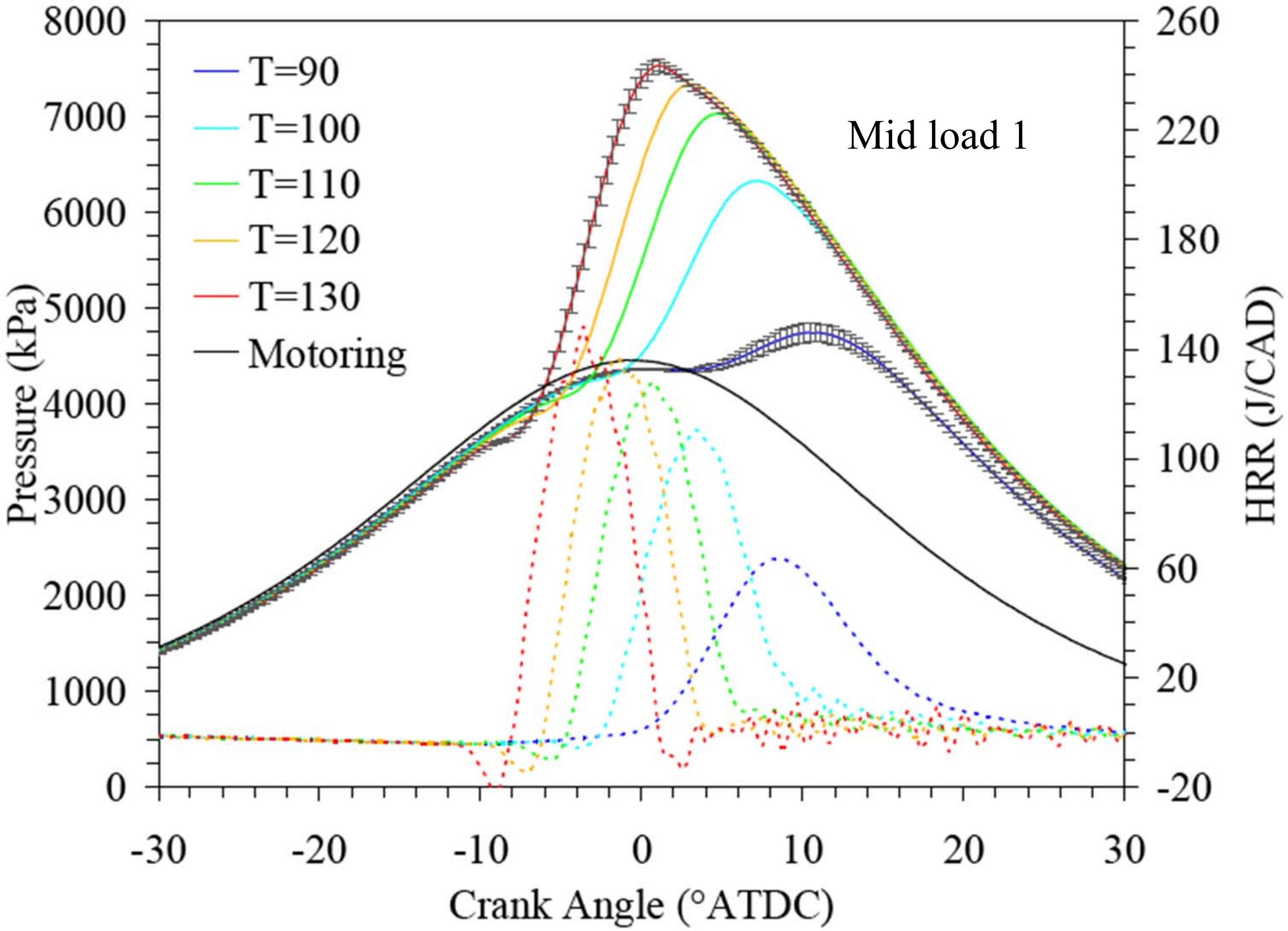


- In these tests maximum IMEP was achieved at inlet temperatures of 130 and 100 °C for ethanol and hydrogen, respectively
- Very low NOx emissions, < 0.02 g/kWh
- Surprisingly high PM emissions, but nearly 100% volatile
- EtOH has higher PM than H_2 but we believe this is mainly due different burning rates and average in-cylinder temperatures influencing oil evaporation and atomization

Outline

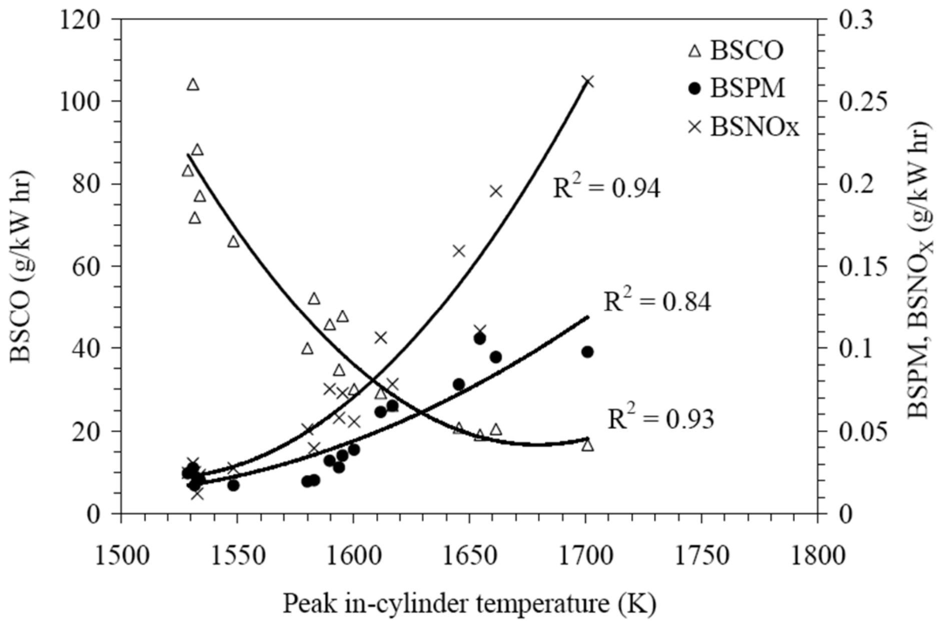
- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

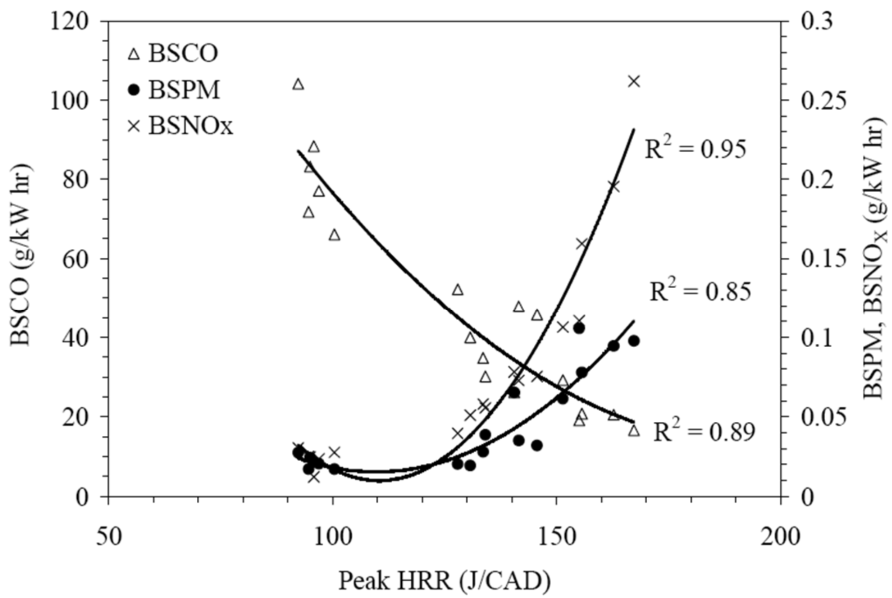
In cylinder pressure measurements were used to calculate bulk temperatures, cycle work, and heat release rate



Dependence of PM and other pollutants depend upon combustion conditions, thermal processing

In Diesel engines there is usually a strong positive correlation between CO and PM formation, here the opposite trend is apparent.

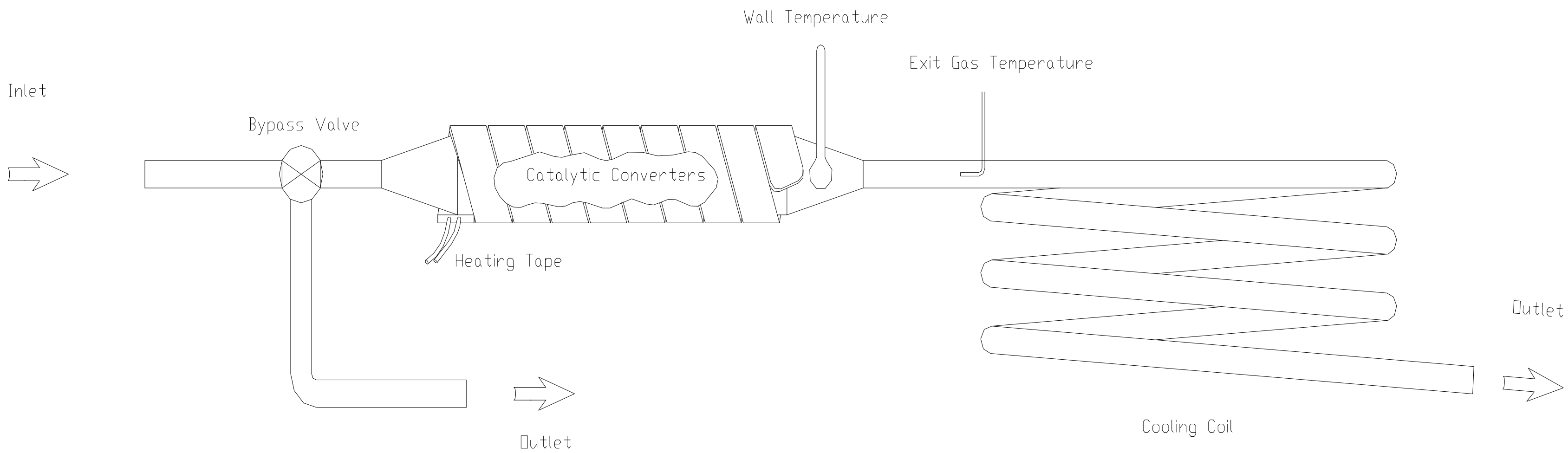




Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  – Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

A catalytic stripper was used to differentiate volatile and solid particles

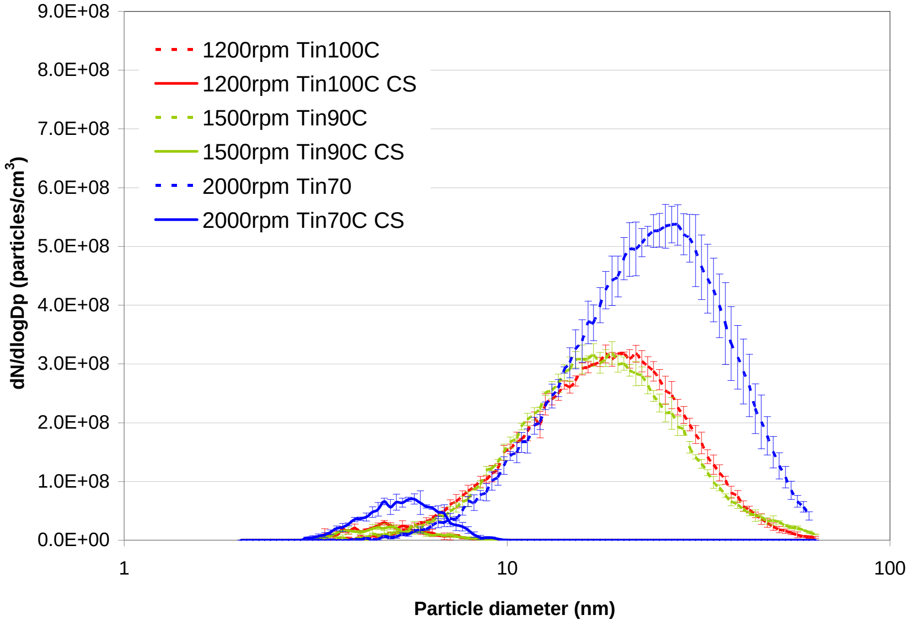


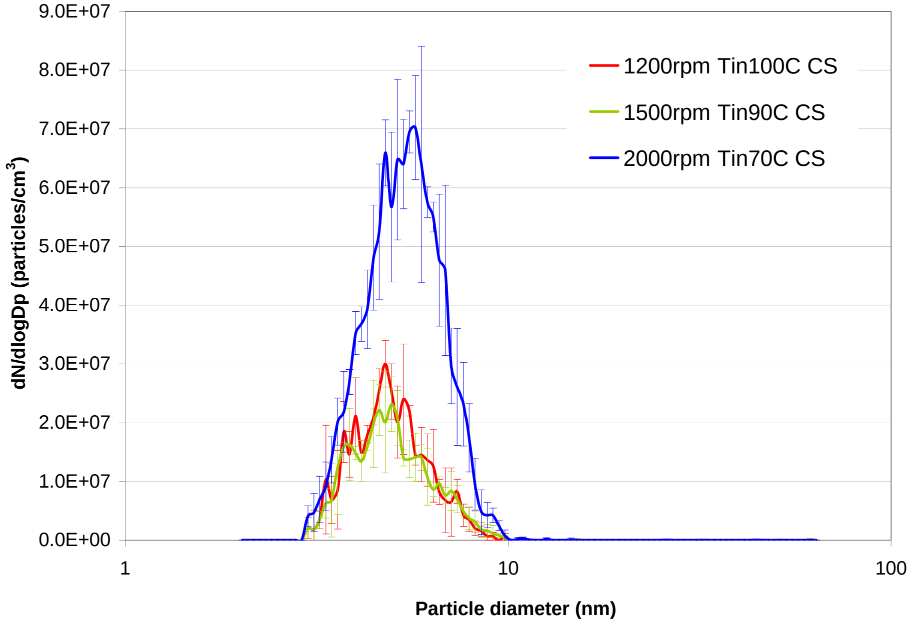
- Recent stripper design
  - Stripper consists of a 2 substrate catalyst* followed by a cooling coil
  - The first substrate removes sulfur compounds
  - The second substrate is an oxidizing catalyst
  - Diffusion and thermophoretic losses present but well defined

*Catalysts were provided by Johnson-Matthey

Kittelson, D. B.; Watts, W. F.; Savstrom, J. C.; Johnson, J. P. Influence of catalytic stripper on response of PAS and DC. *J. Aerosol Sci.* ***2005***, *36*, 1089–1107.

Measurements of solid particles with catalytic stripper (CS)

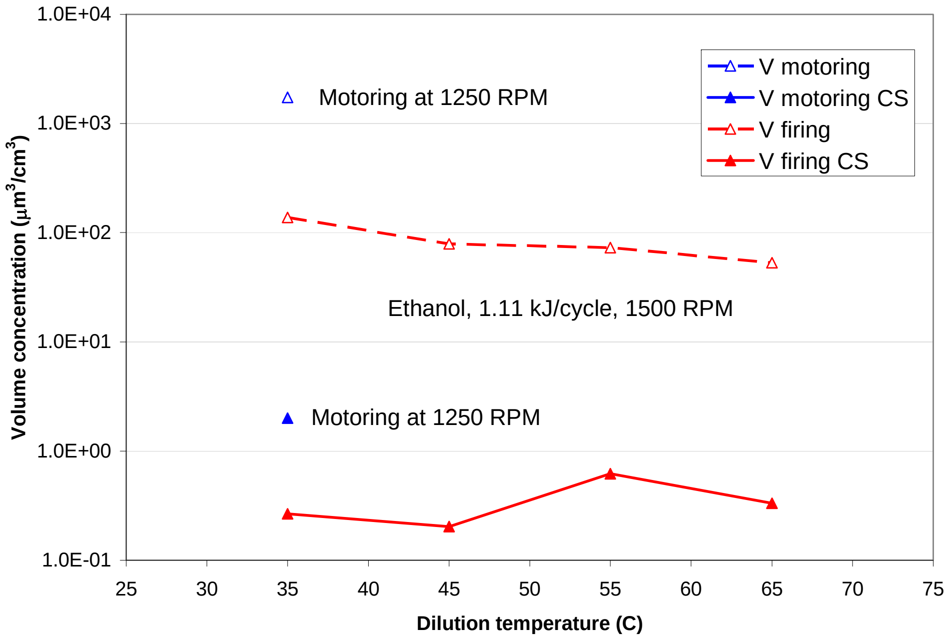


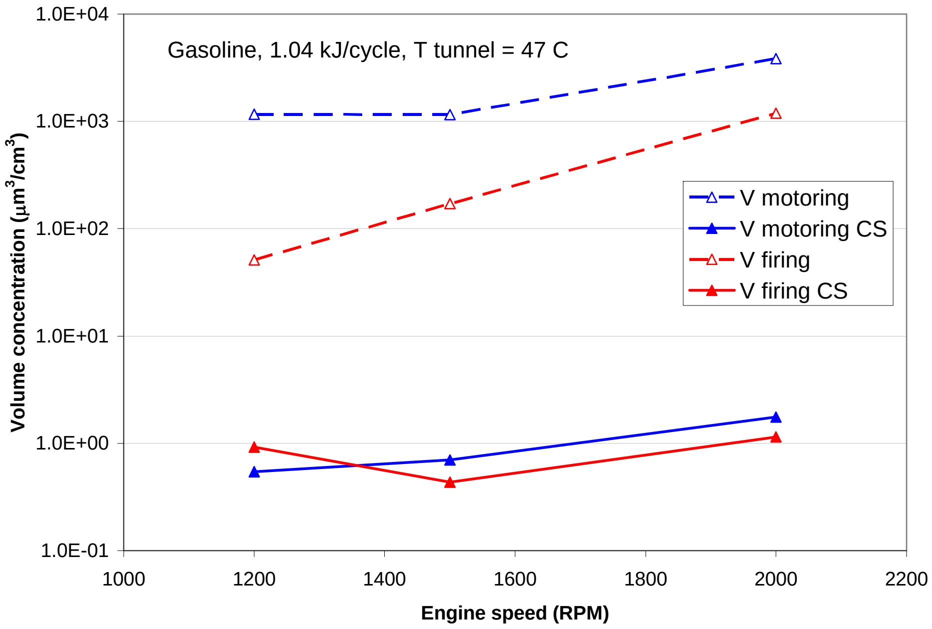


- Right plot shows solid fraction on 10x expanded scale
- Very small solid fraction present
- Depends upon speed, load, temperature – thermal processing

Total and solid particle volume emissions, motoring and firing

- Gasoline produces slightly higher total and solid emissions than ethanol
- Particle emissions are usually much lower under fired conditions than under motored conditions – hot motoring does not give reliable estimates of lube oil related particles
- Solid particle emissions may be slightly lower under fired conditions than motored conditions





Tandem DMA used for detailed volatility measurements

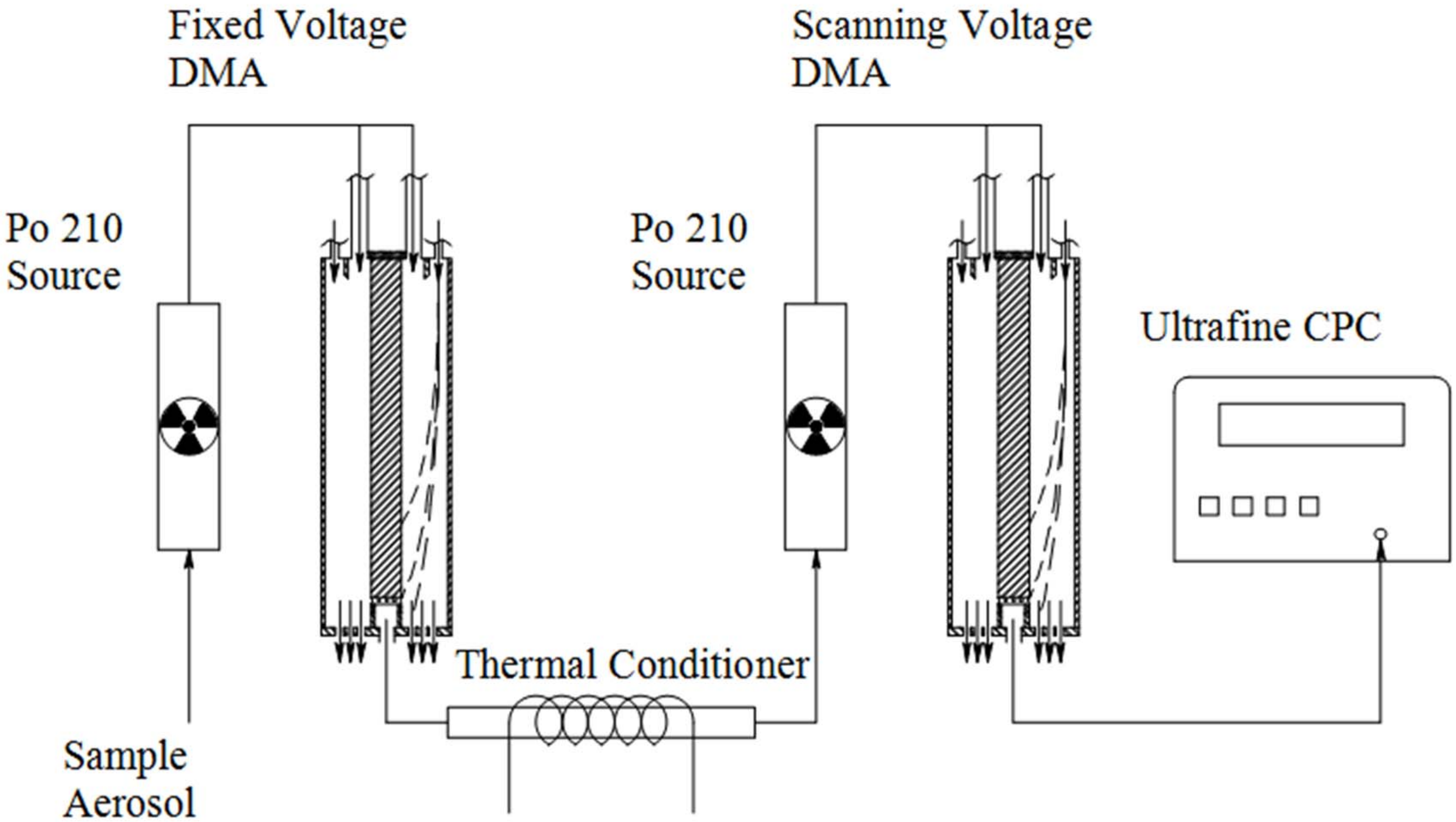
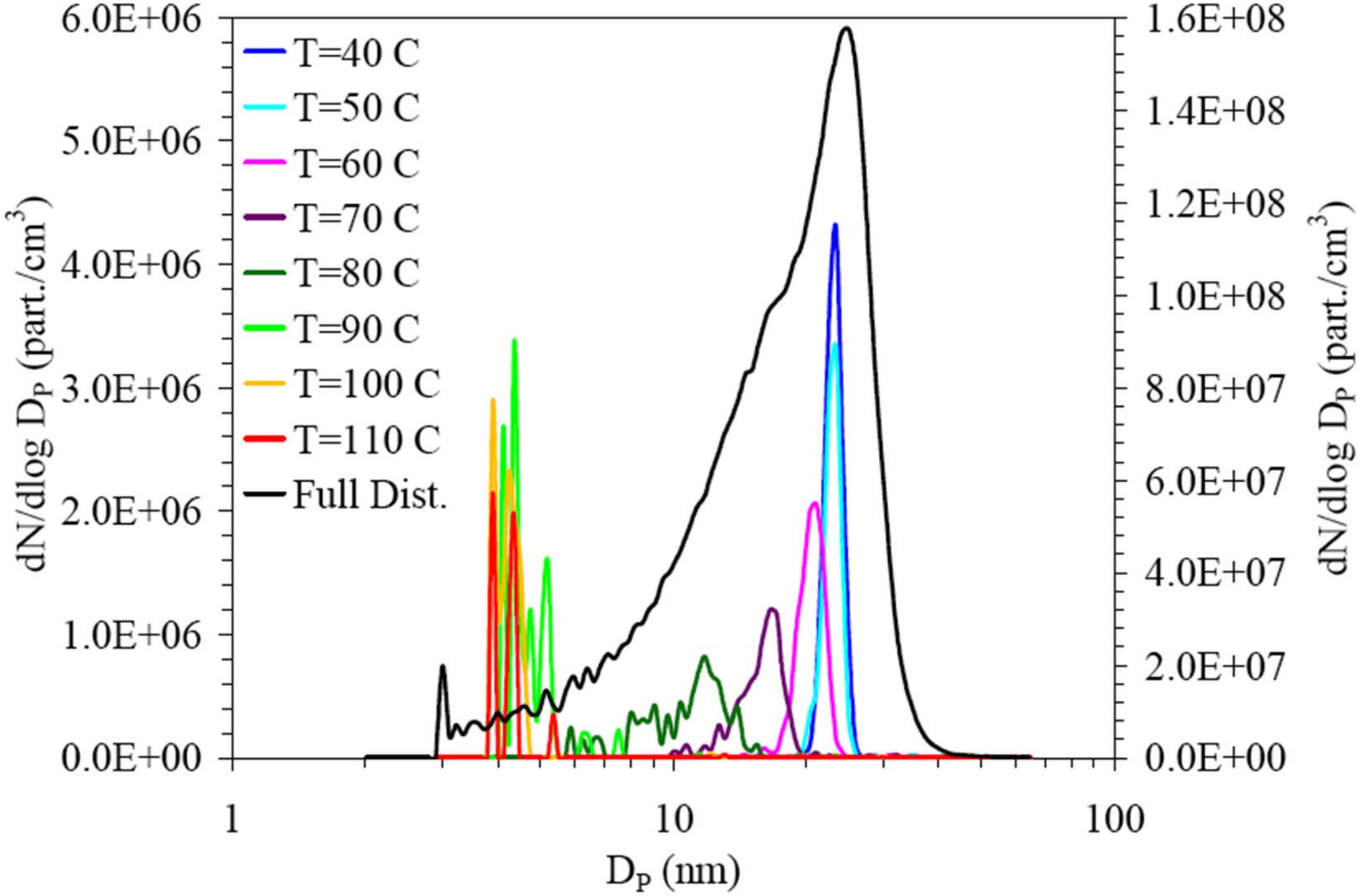


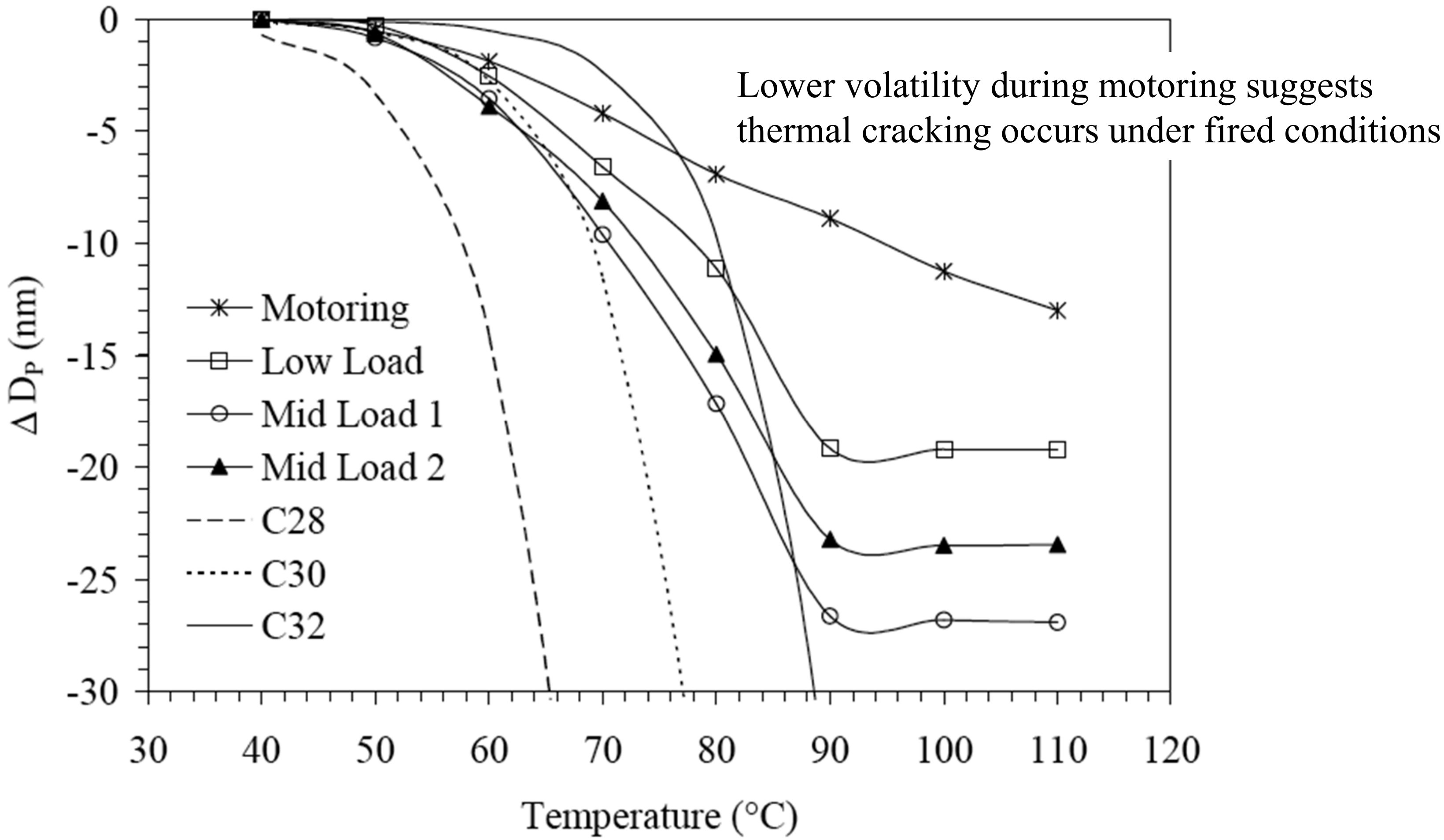
Figure 11: TDMA Apparatus

Tandem DMA measurements of particle volatility

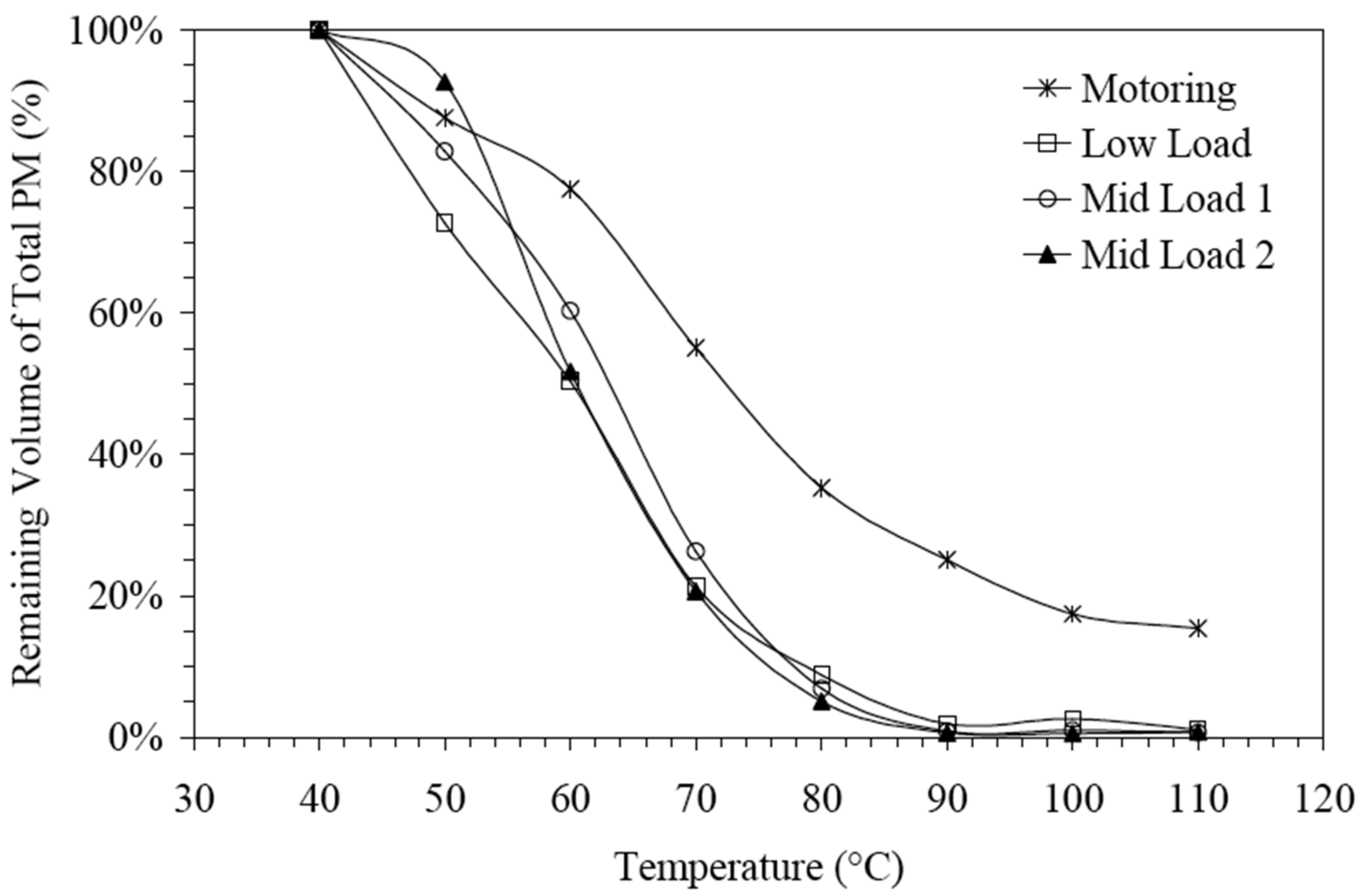


Light load, ethanol fuel, no EGR

Evaporation profiles for fired conditions are similar to C30 – C32 normal alkanes.



Nearly all the volume (and mass) of these particles is volatile



Outline

- Introduction
- Experimental apparatus and procedure
- Dilution sensitivity
- Results
  - Emissions
    - Influence of fuel
    - Influence of thermal processing
    - Volatility measurements
- Conclusions

Conclusions – PM emissions from pure HCCI

- Significant mass and number emissions observed
  - Most of material between 10 and 50 nm
  - Nearly all volatile
  - Particle emissions strongly associated with in-cylinder thermal history
  - Significant particle formation even with pure H_2 fuel
    - Particles apparently formed from thermal processing of lube oil
    - Should explore other lube oil formulations and oil vaporization / atomization mechanisms
  - It is likely that most of these particles could be removed by an oxidizing catalyst at sufficiently high exhaust temperatures
- It is likely that particle formation mechanisms will be similar in other non-sooting engine / fuel concepts like other low temperature combustion modes, and engines running on DME, CNG, H_2
- Lube oil related particles are one of the last remaining problems to be understood as we move to ever cleaner engines and combustion systems

Questions?

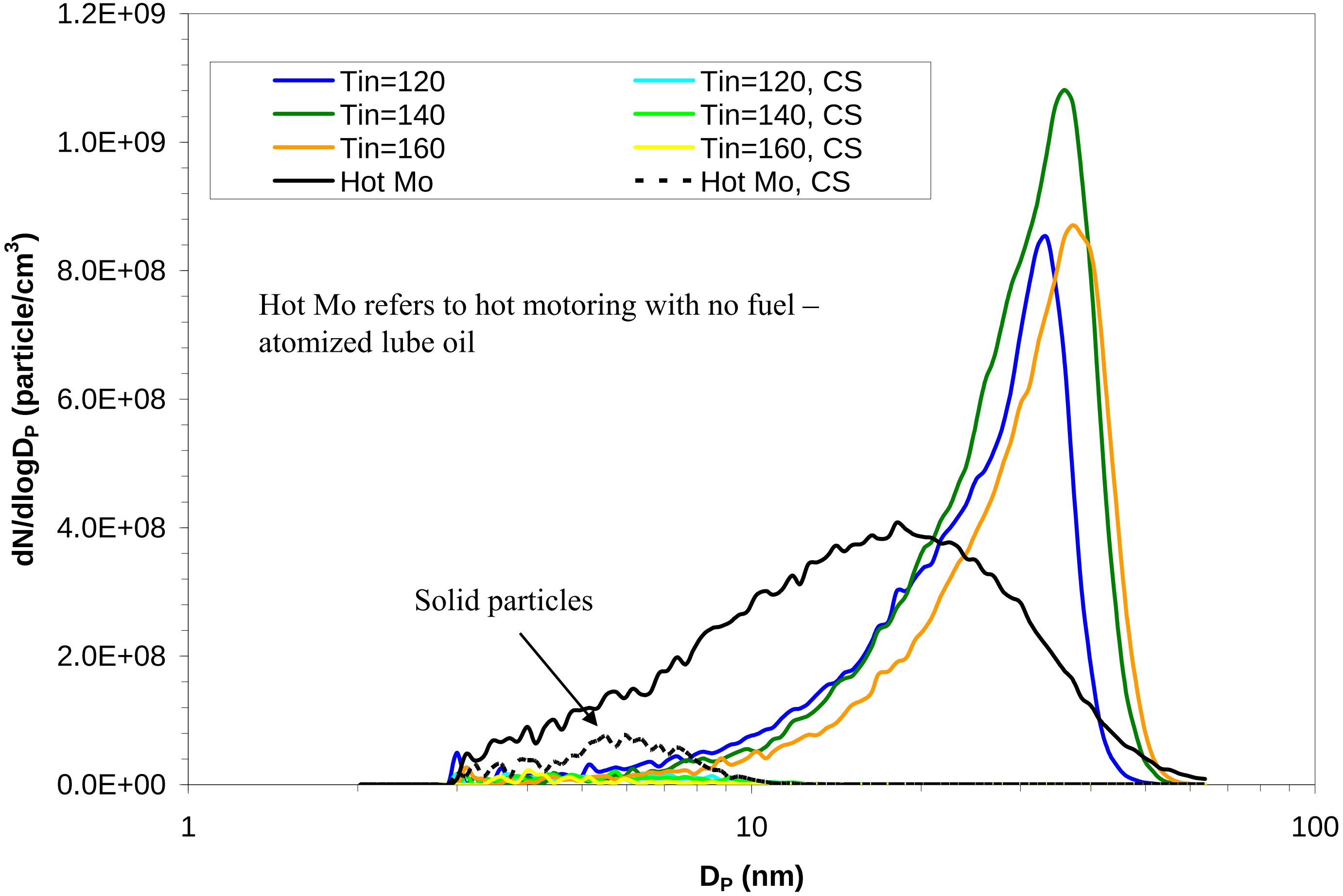
Additional slides

Related work on HCCI particle emissions

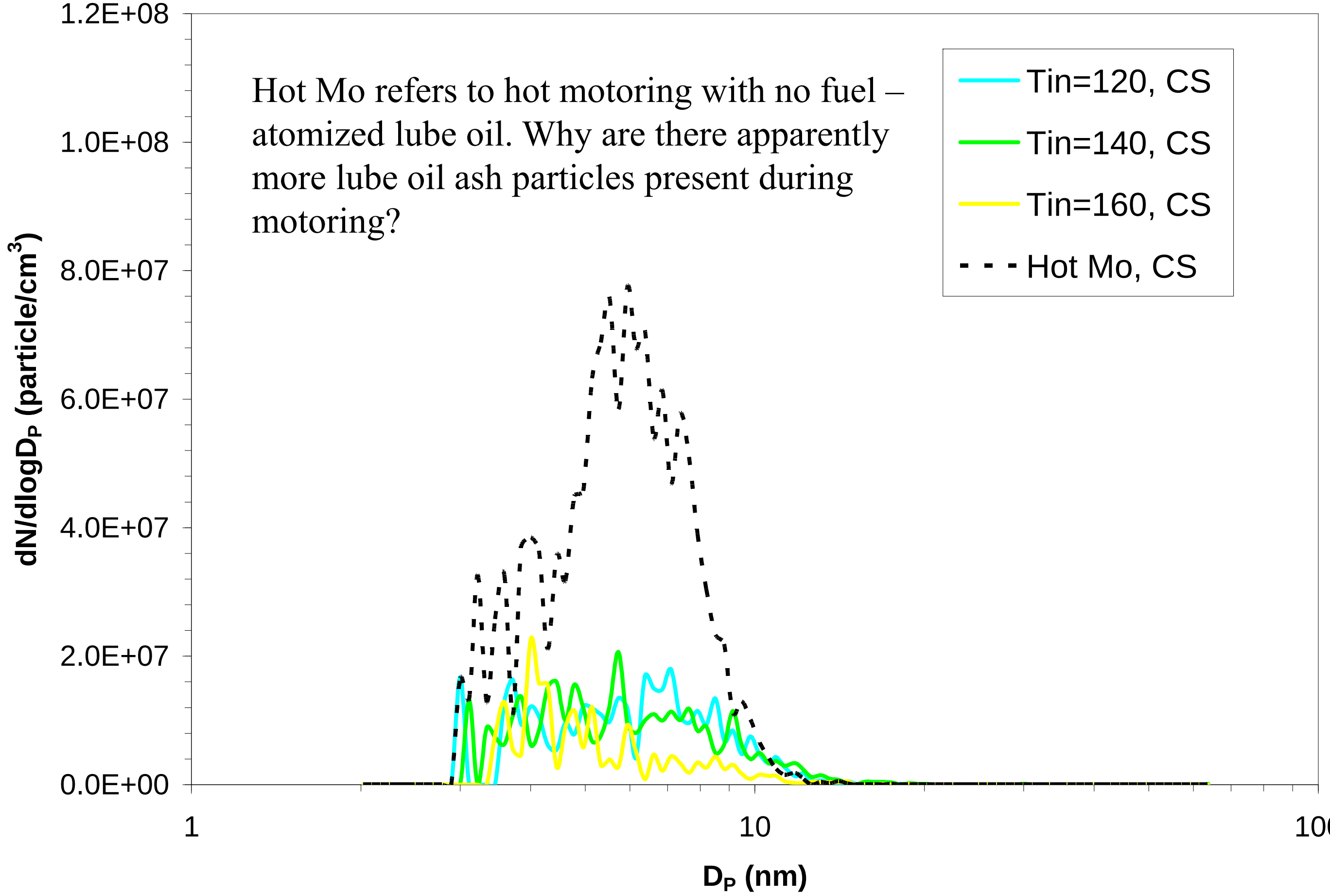
	Kaiser *et al* (2002)	Price *et al* (2007)	Misztal *et al* (2009)	Zinola & Lavy (2009)
Engine	DI, Intake heating, CR=15.2:1,	DI, 19 valve timings, λ=1 only,	Mixed hot/cold intake streams, variable valve timing	2.2 liter, DI, CR=14:1, boost, cool/hot EGR mixing
Fuel	Gasoline	Gasoline	Gasoline	low sulfur(<10ppm) diesel, CN =56.1
Instrumentation	SMPS, 2 stage dilution	DMS500	DMS500	SMPS 3071A ,with 3022 CPC
Findings	-Mid load HCCI yielded more and larger accum. mode PM than DISI operation	-HCCI showed more accum. mode PM and less nucl. mode PM than DISI	-Increased EGR-decreased total PM -Lack of dilution monitoring/control reported	-NO_2:NO_X ≈12-17% -VOF 75-90% for low load HCCI -no nucleation mode PM present -no dilution conditions reported

Solid particle measurements

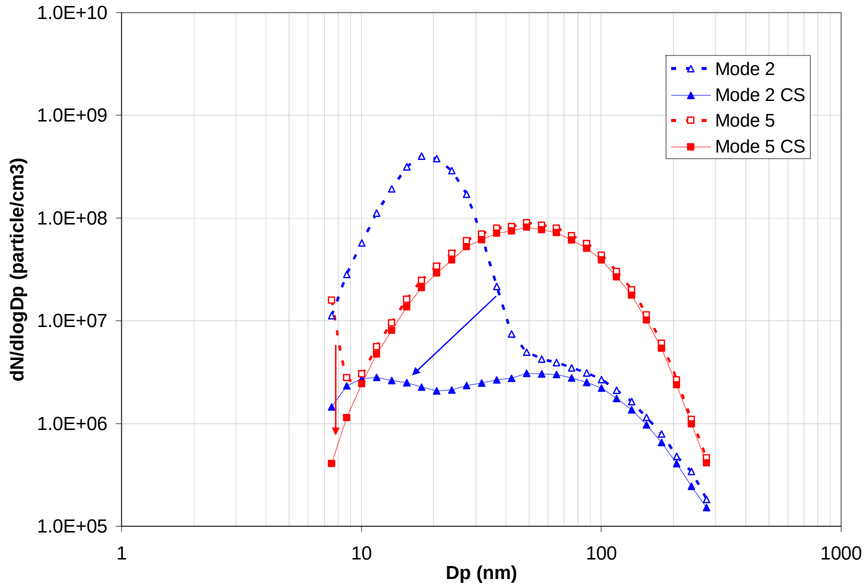
What about solid particles – here are some results with and without the catalytic stripper at light load

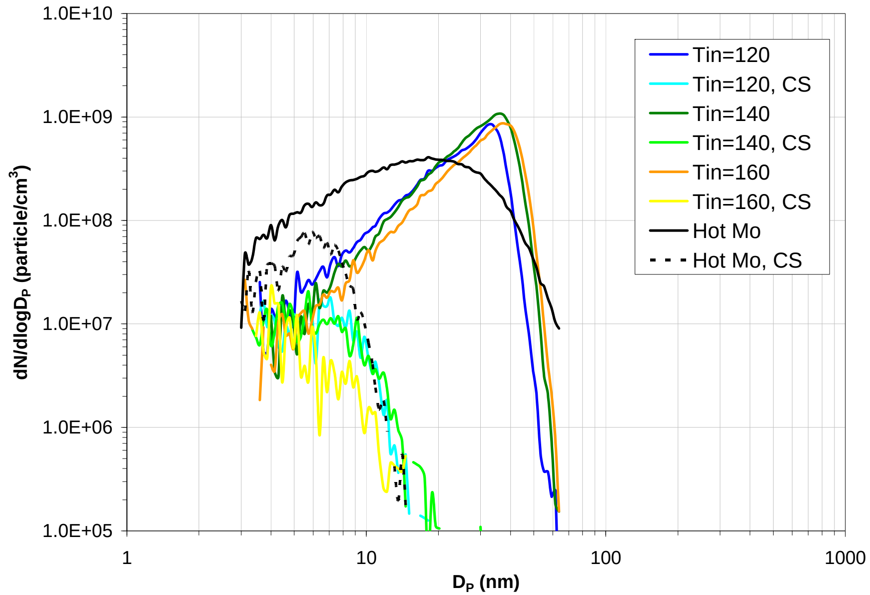


Solid particles from previous slide 10x scale



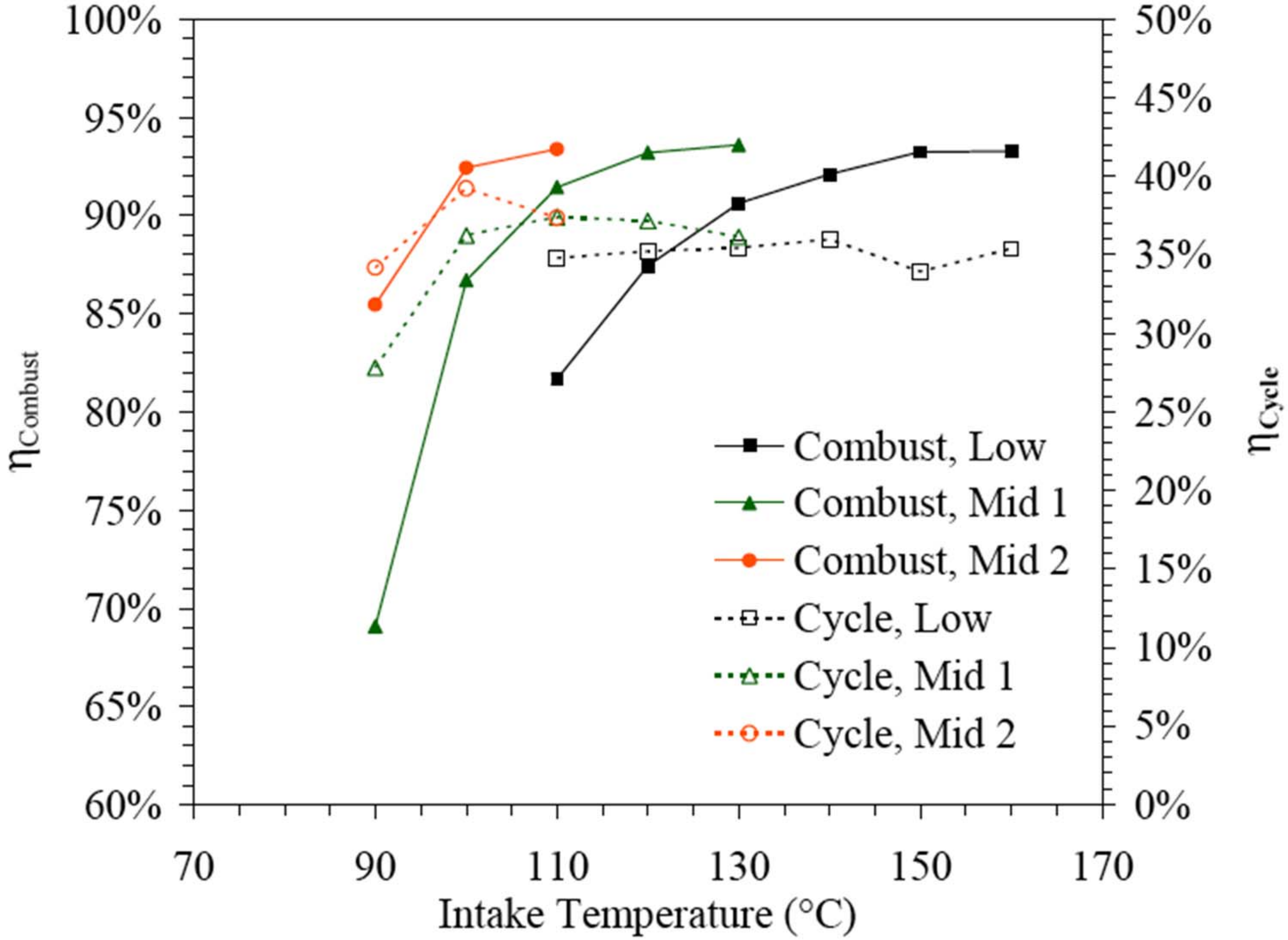
Comparison with solid particles from a modern Diesel. HCCI nucleation mode particles much smaller but in higher concentration.



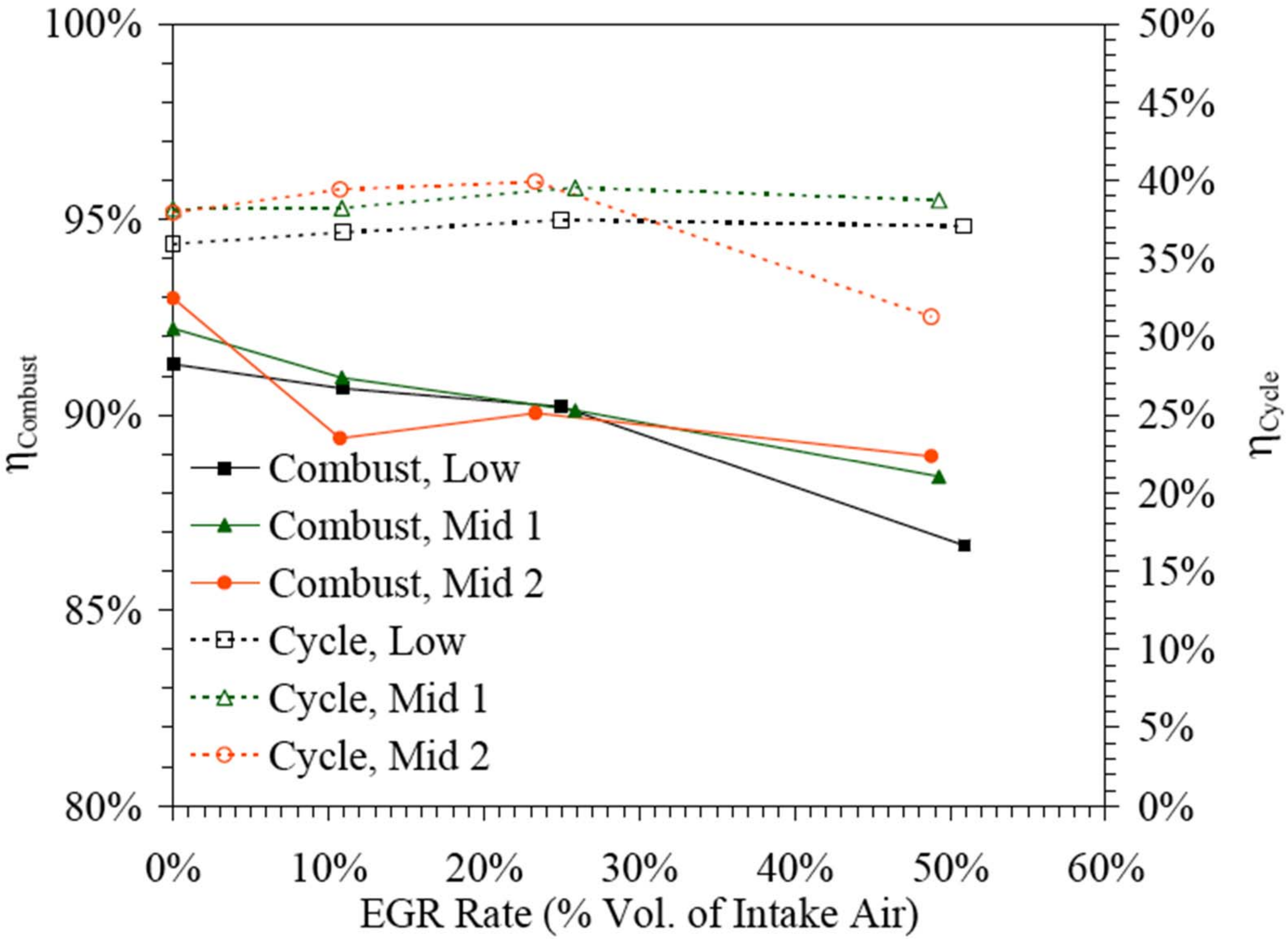


Additional combustion data

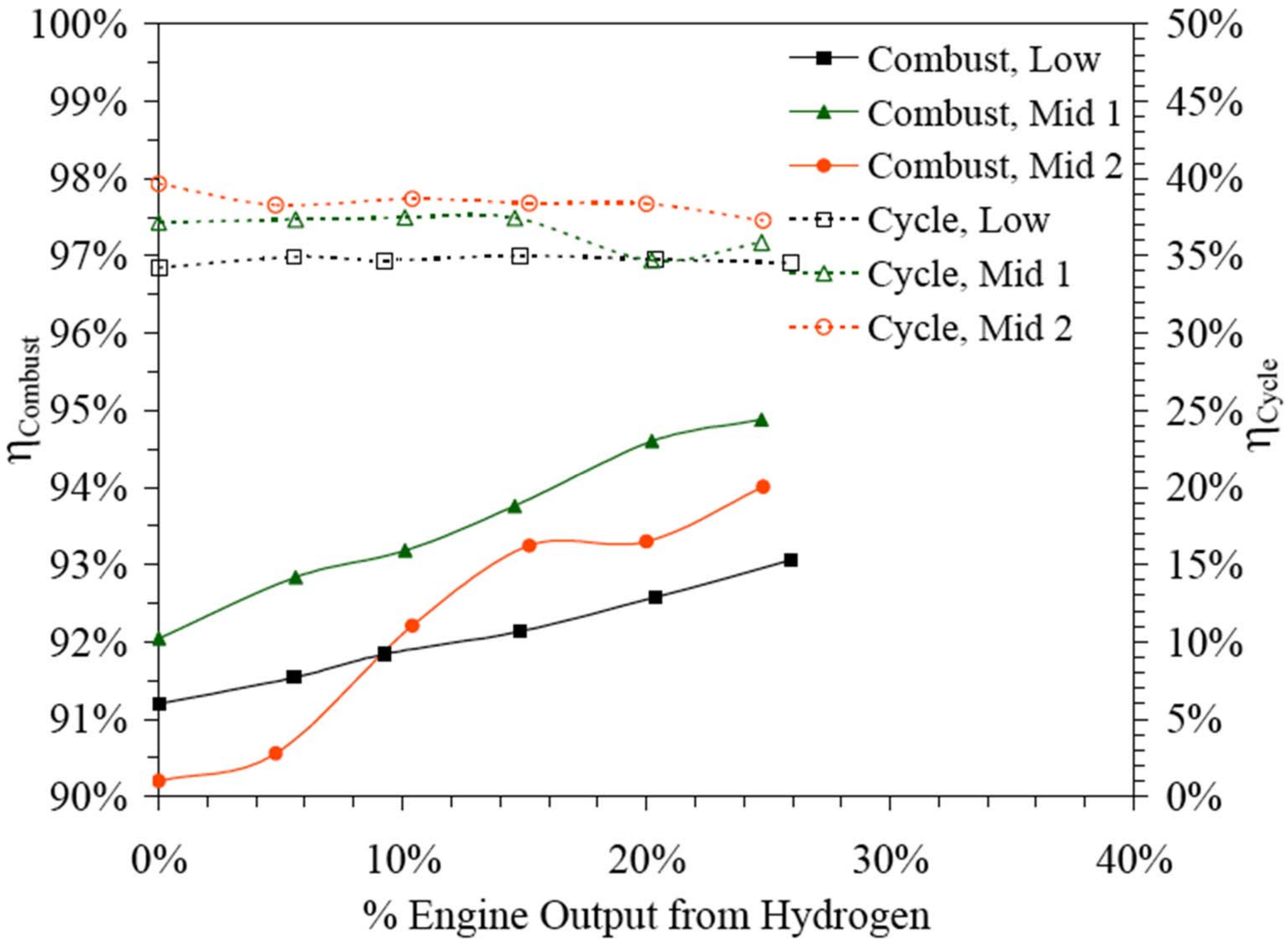
Typical engine performance, cycle efficiency and combustion efficiency, ethanol fuel



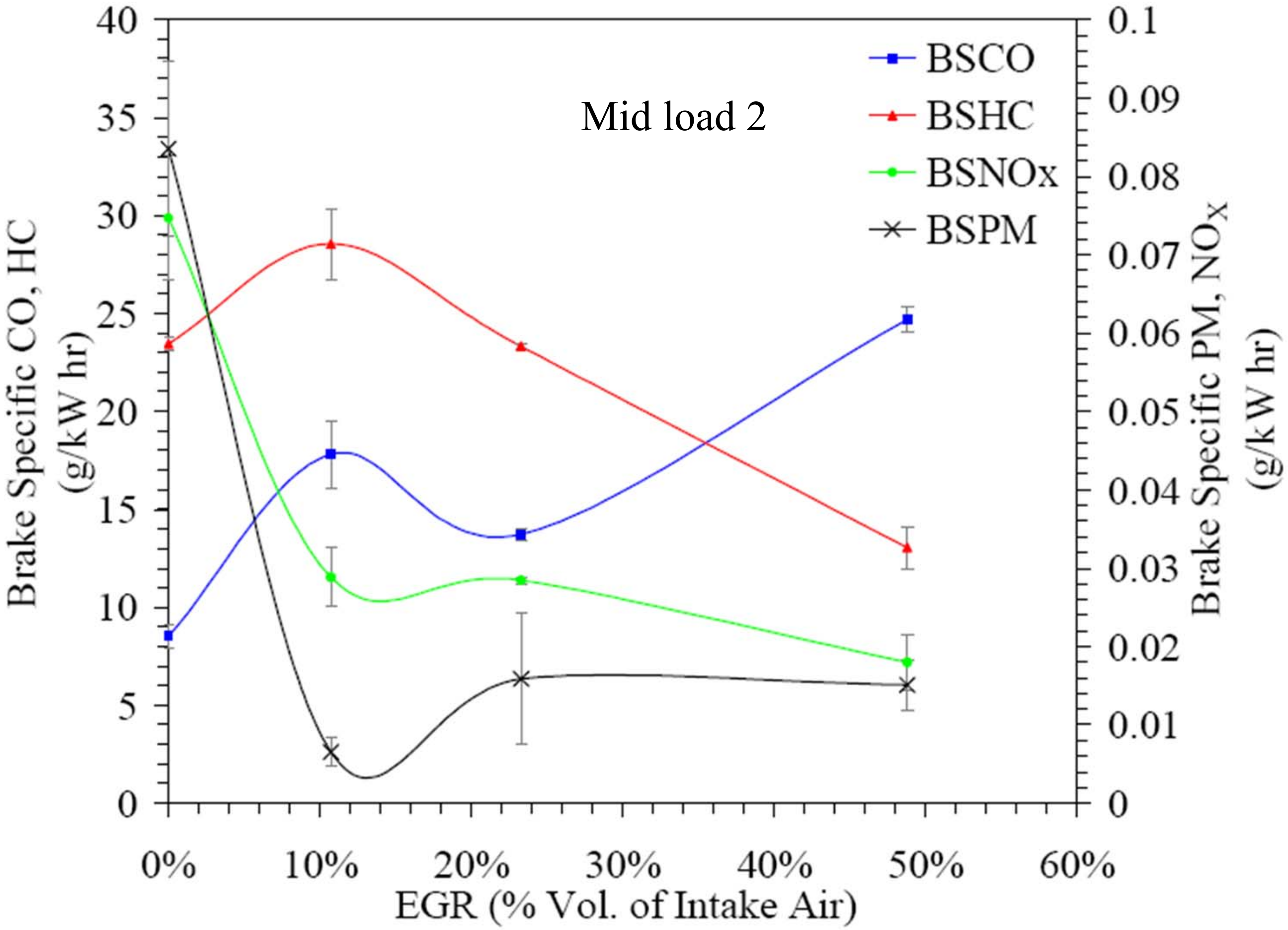
Changing EGR rate at constant load changes combustion timing and efficiency



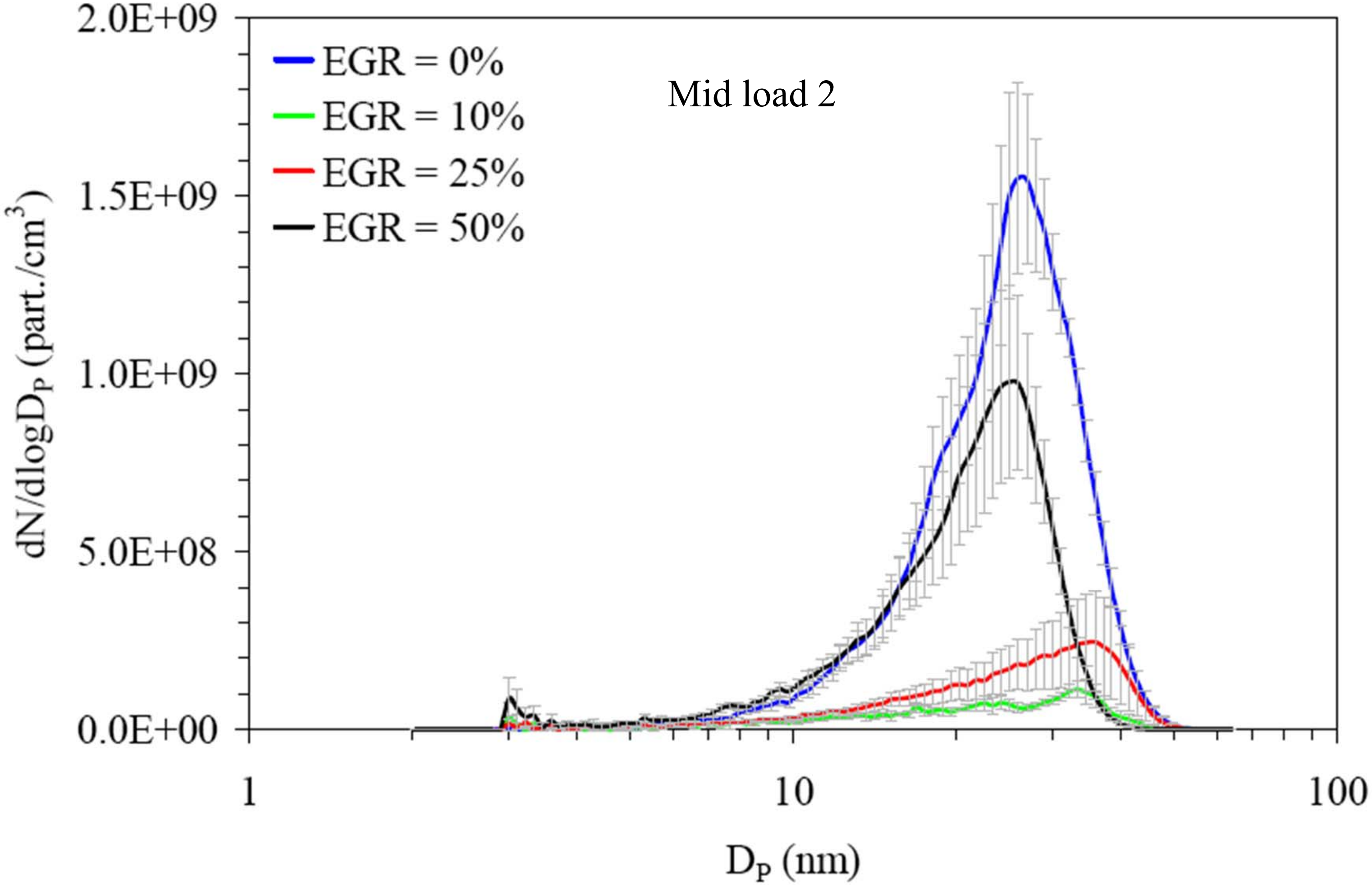
Changing H_2 rate at constant load changes combustion timing and efficiency



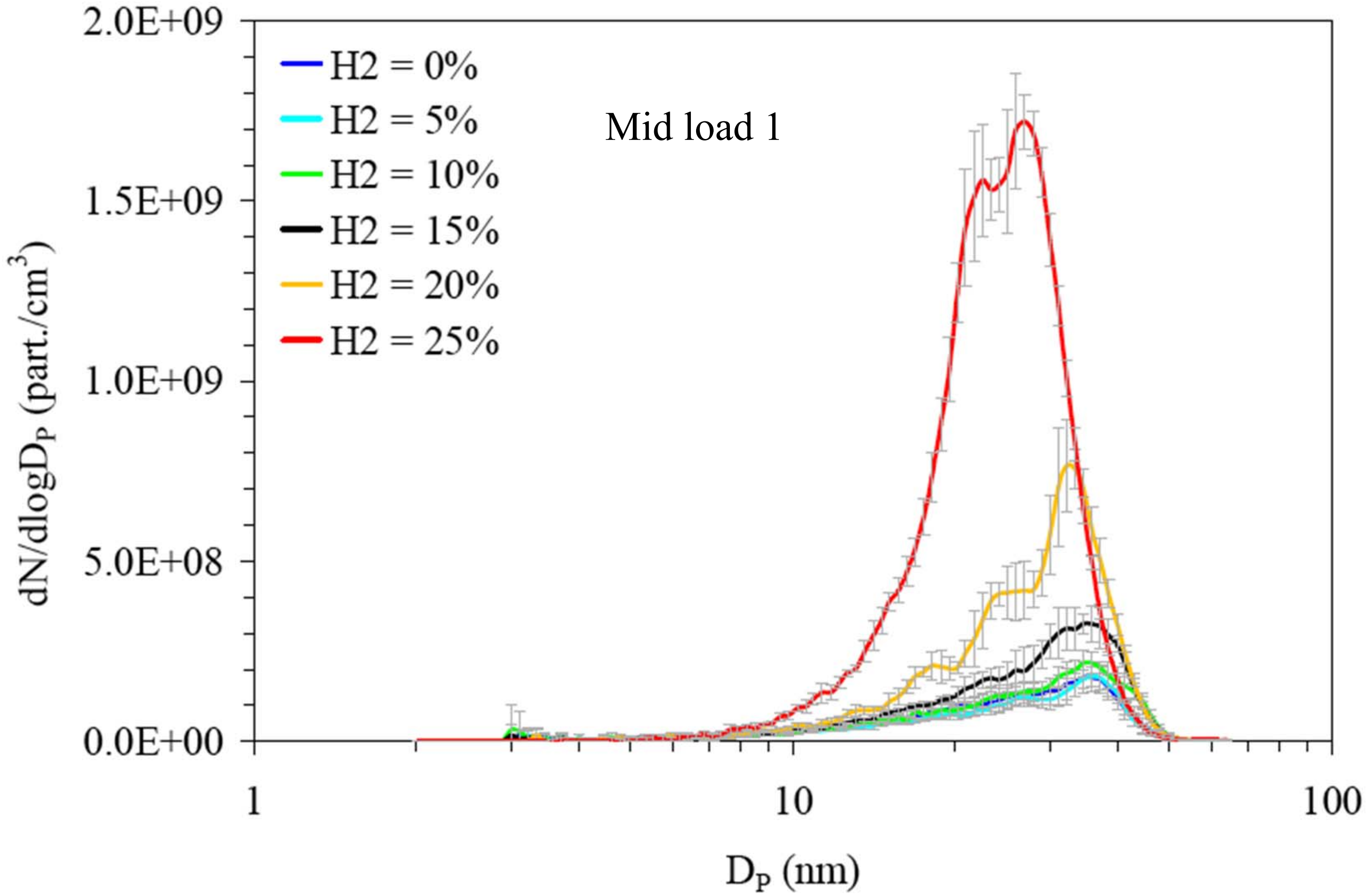
Influence of EGR on emissions



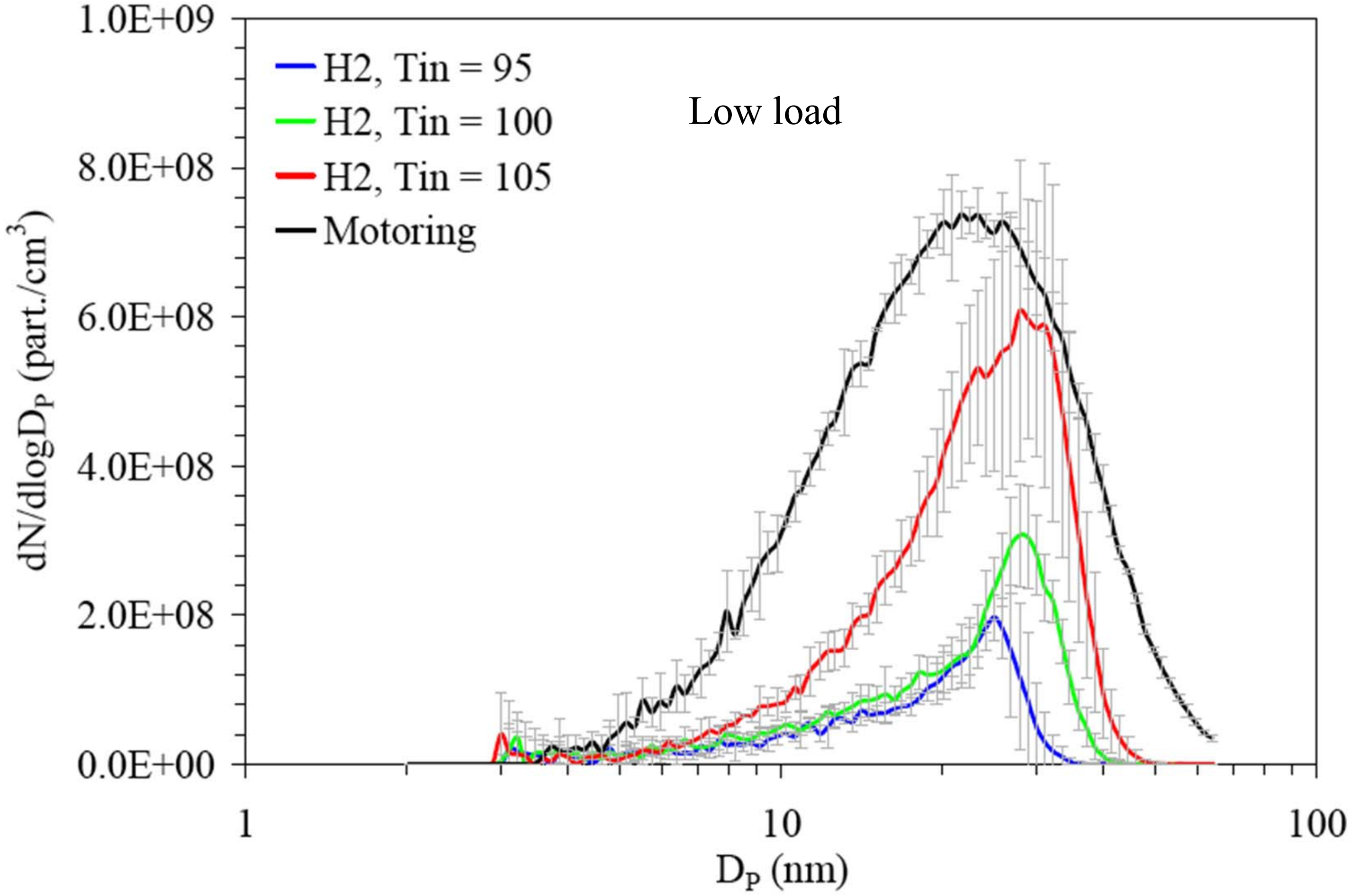
The particle size distribution is unimodal and in the nucleation mode range – concentrations decrease with EGR



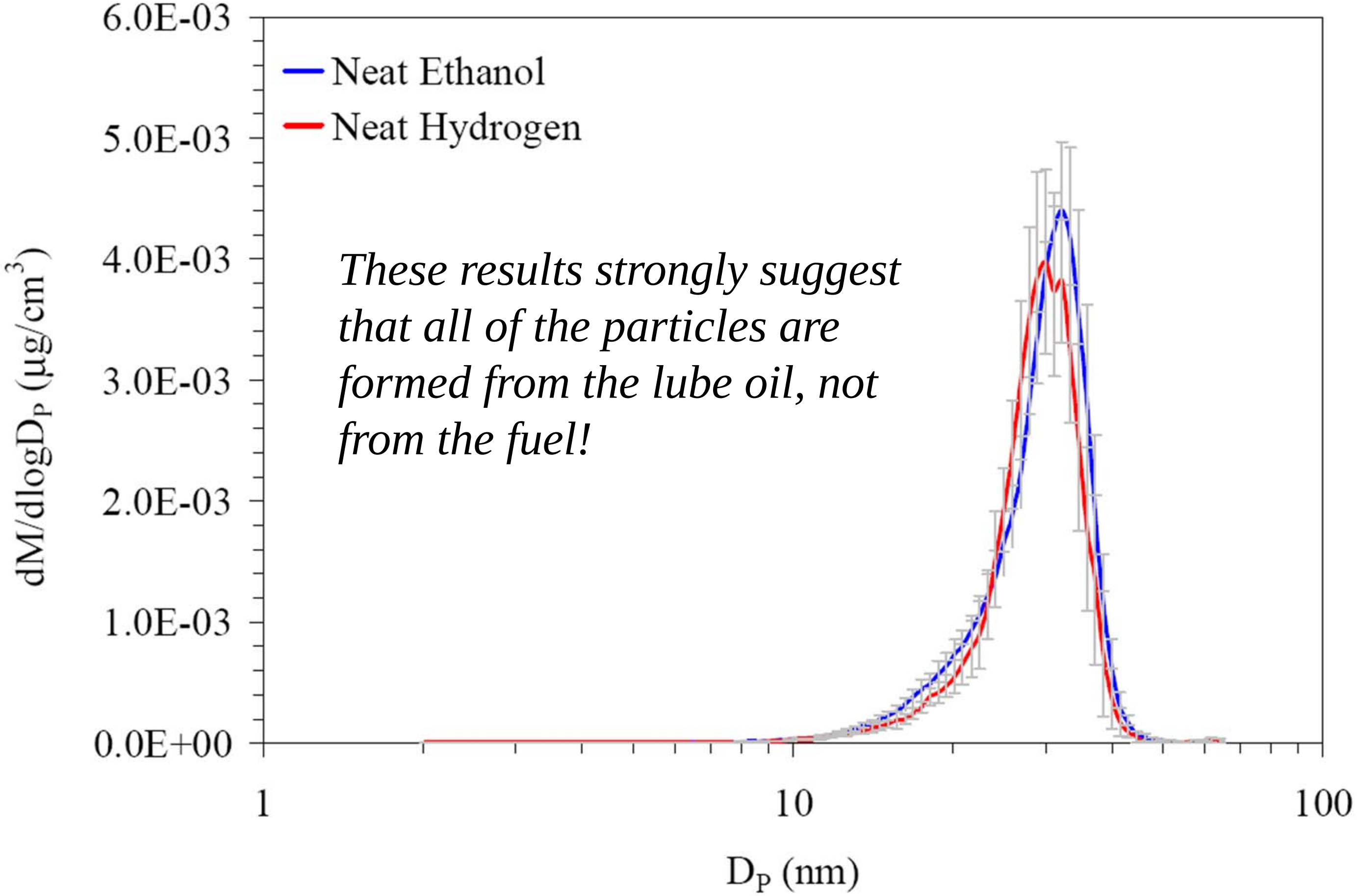
The particle emissions increase with increasing H_2 addition at constant load



For pure H_2 particle concentrations increase with inlet temperature, hot motoring result also shown



PM formation with ethanol and pure H_2 fuels, matched combustion conditions



These results strongly suggest that all of the particles are formed from the lube oil, not from the fuel!