Does the trimodal size distribution adequately describe aerosols from modern engines?

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Origin of using trimodal lognormal distribution to describe engine aerosols

Early submicron roadside measurements showed bimodal distribution

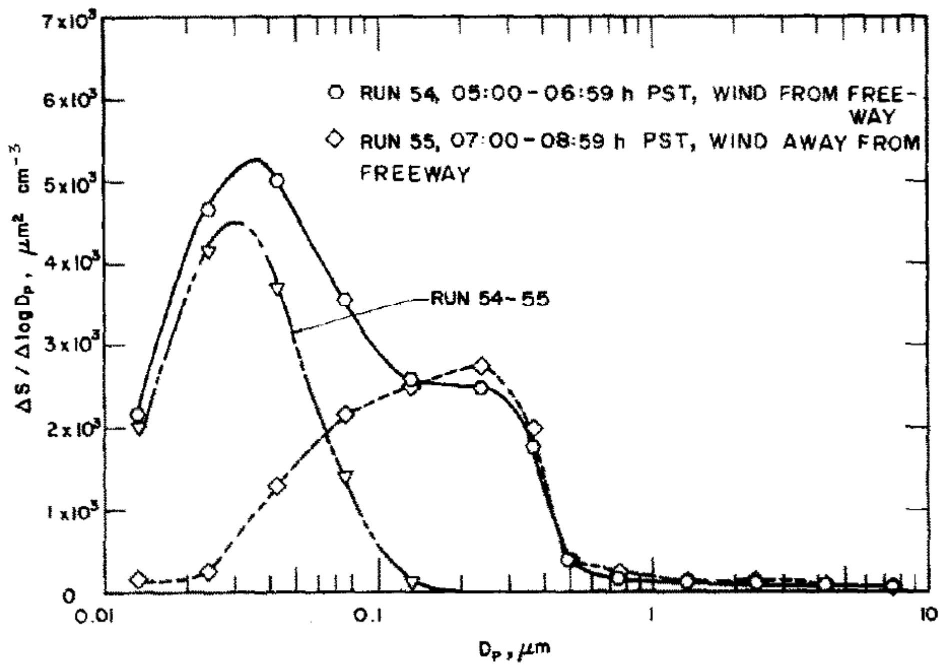

Fig. 7. Plot of the surface size distributions, $\Delta S/\Delta \log D_p$ for Run 54 when the wind was from the freeway, Run 55 when the wind was blowing toward the freeway, and the difference distribution, Run 54 minus Run 55 for D_p less than 0.15 μm.

DEPARTMENT OF MECHANICAL ENGINEERING

From: Characterization of California Aerosols-I. Size Distributions Of Freeway Aerosol, K. T. Whitby, W. E. Clark.,V. A. Marple, G. M. Sverdrup, G. J. Sem, K. Willeke, B. Y. H. LIU And D. Y. H. Pui, Atmospheric Environment Vol. 9. pp. 463-482. Pergamon Press 1975.

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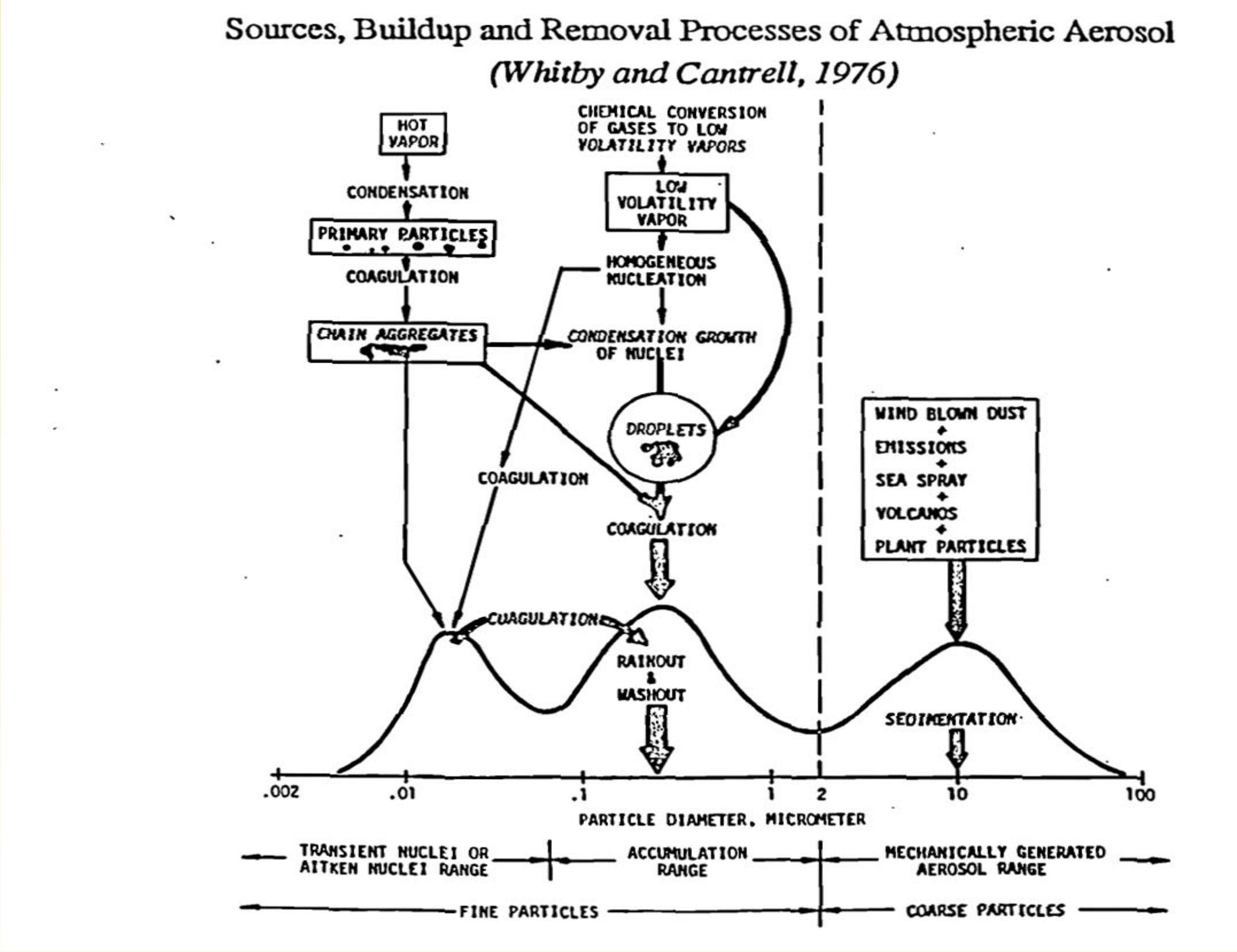
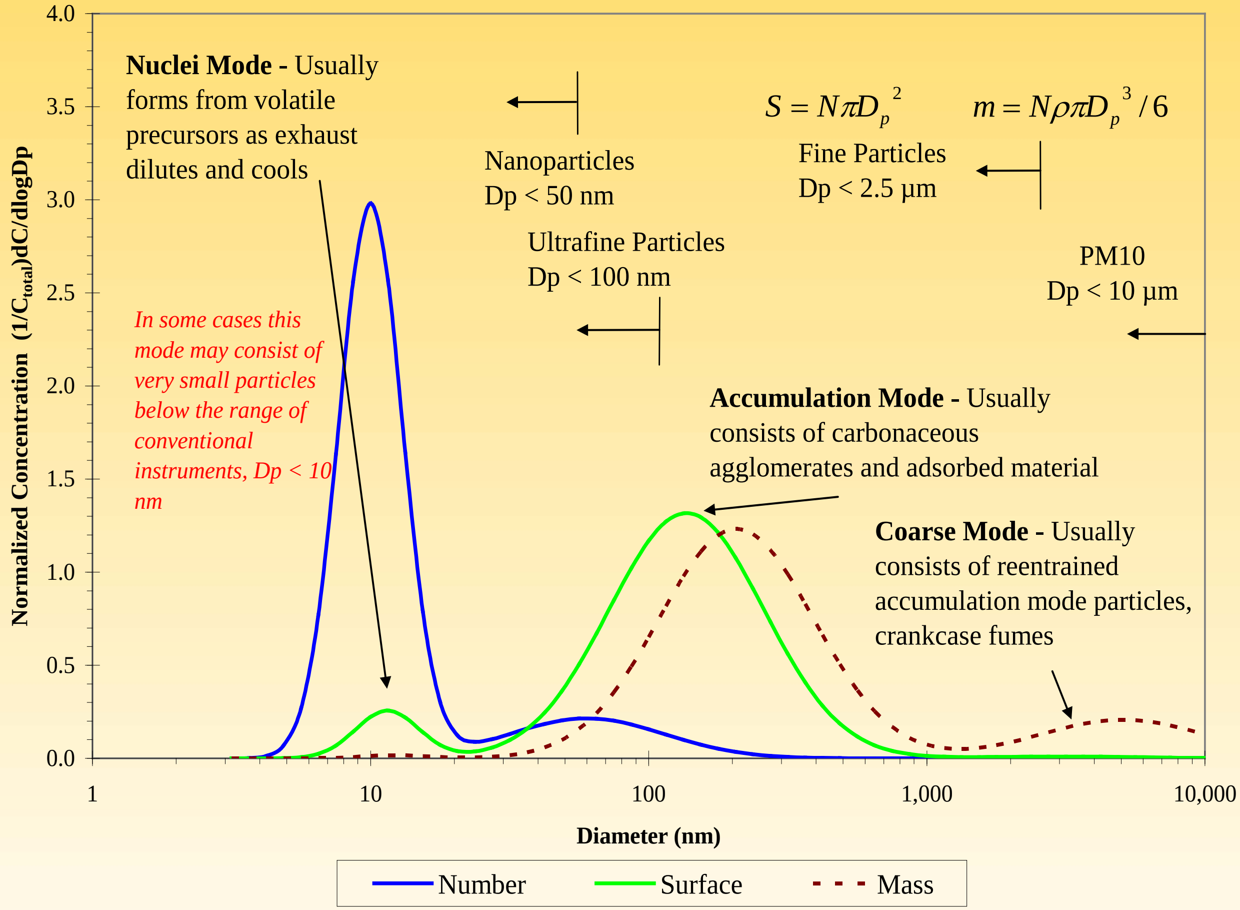
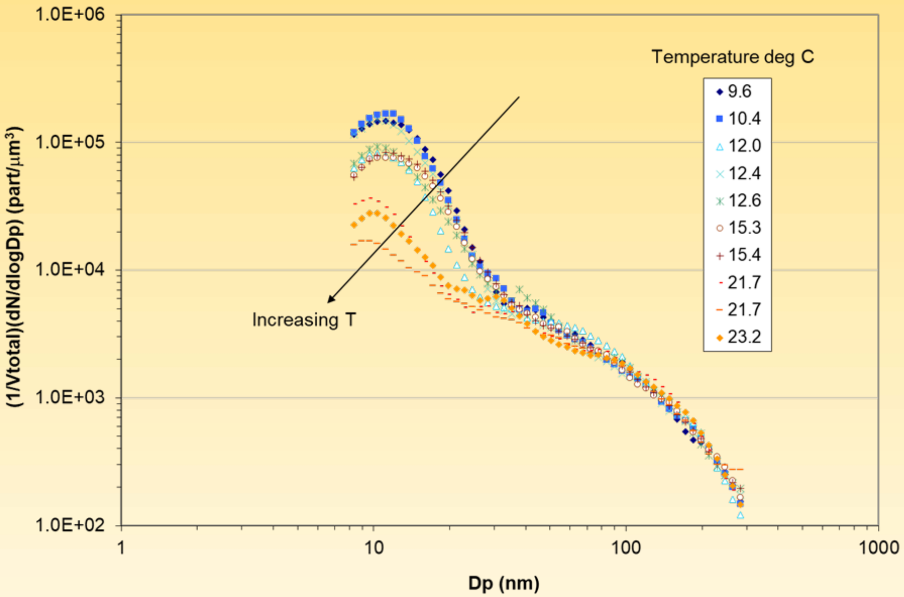
Atmospheric aerosols exhibit trimodal size distribution *linked to formation processes*

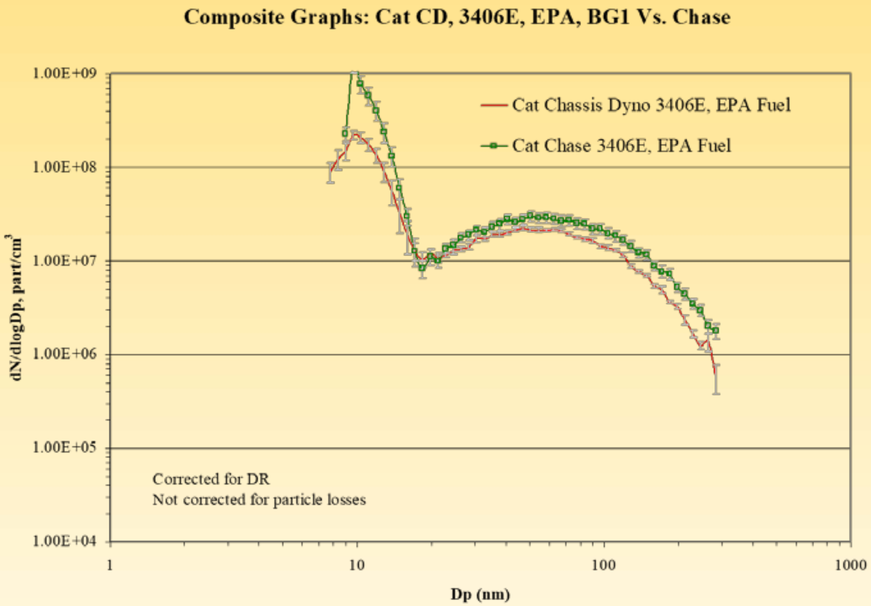


Diesel exhaust size distributions also typically trimodal, *modes linked to formation processes*



Many on-road measurements show clear modal structure





Kittelson, D. B., W. F. Watts, J. P. Johnson, M. L. Remerowki, E. E. Ische, G. Oberdörster, R. M. Gelein, A. C. Elder, and P. K. Hopke, 2004. "On-Road Exposure to Highway Aerosols: 1. Aerosol and Gas Measurements," Inhalation Toxicology, 16(suppl. 1):31–39

Kittelson, D. B., W. F. Watts, and J. P. Johnson 2006. "On-road and Laboratory Evaluation of Combustion Aerosols Part 1: Summary of Diesel Engine Results," Journal of Aerosol Science 37 (2006) 913–930.

Interpreting modal structure

- Each mode suggests a distinct formation mechanism
  - Nucleation mode
    - Nucleation of semi-volatile during exhaust dilution and cooling
    - Ash nucleation during expansion stroke
    - Nascent soot
    - All of these are suppressed by existing particles in accumulation mode
  - Accumulation or soot mode
    - Formed on burning fuel jet early in engine cycle
    - Fractal like structure
    - May be masked by semi-volatile material (nucleation mode) in low soot engines/fuel
  - Coarse mode – not considered here

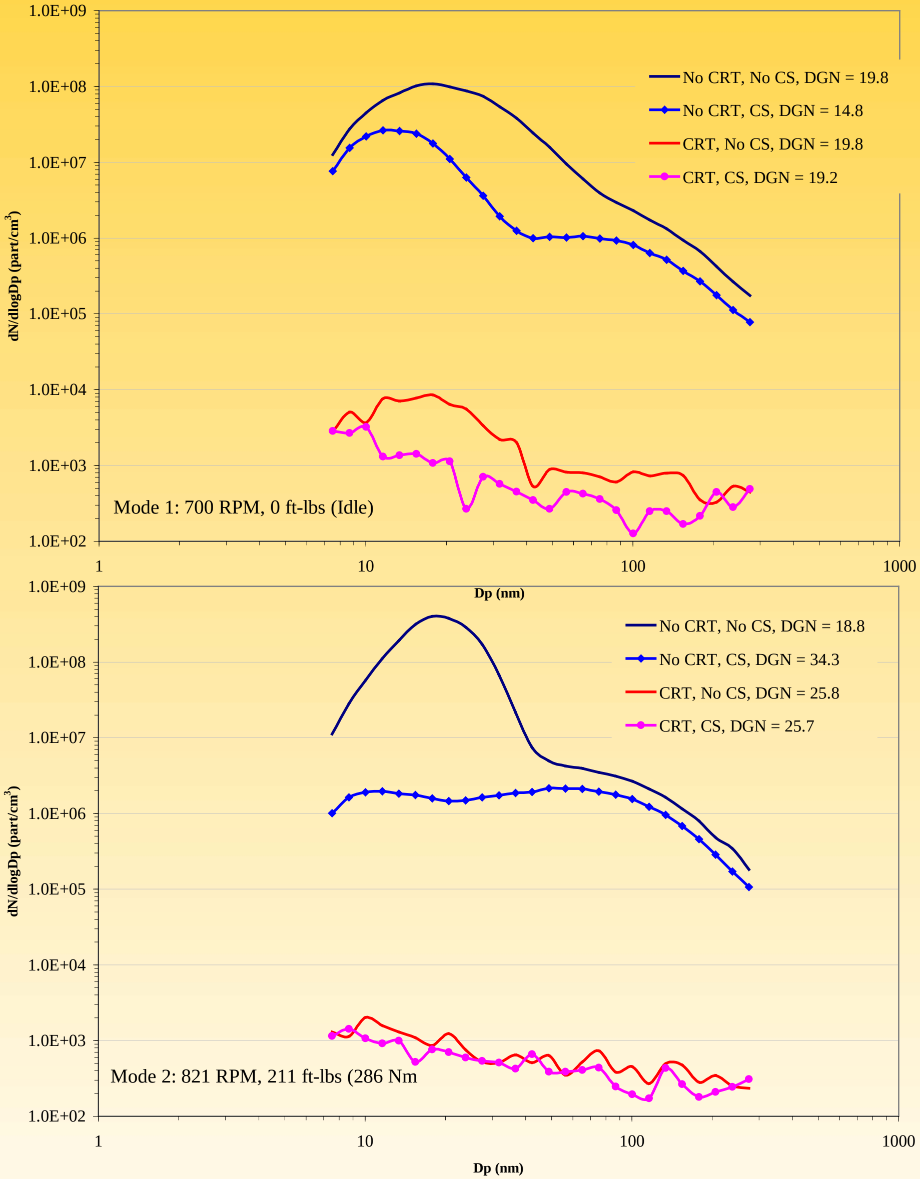
Masking modal structure

- Multi-source ambient aerosols may lead mixtures with many modes
- Average size distributions in transient cycle with modes moving around
- Removal or partial removal of semi-volatile material during sampling
- Low soot engines may produce mainly nucleation mode
- High soot engines may produce mainly accumulation mode

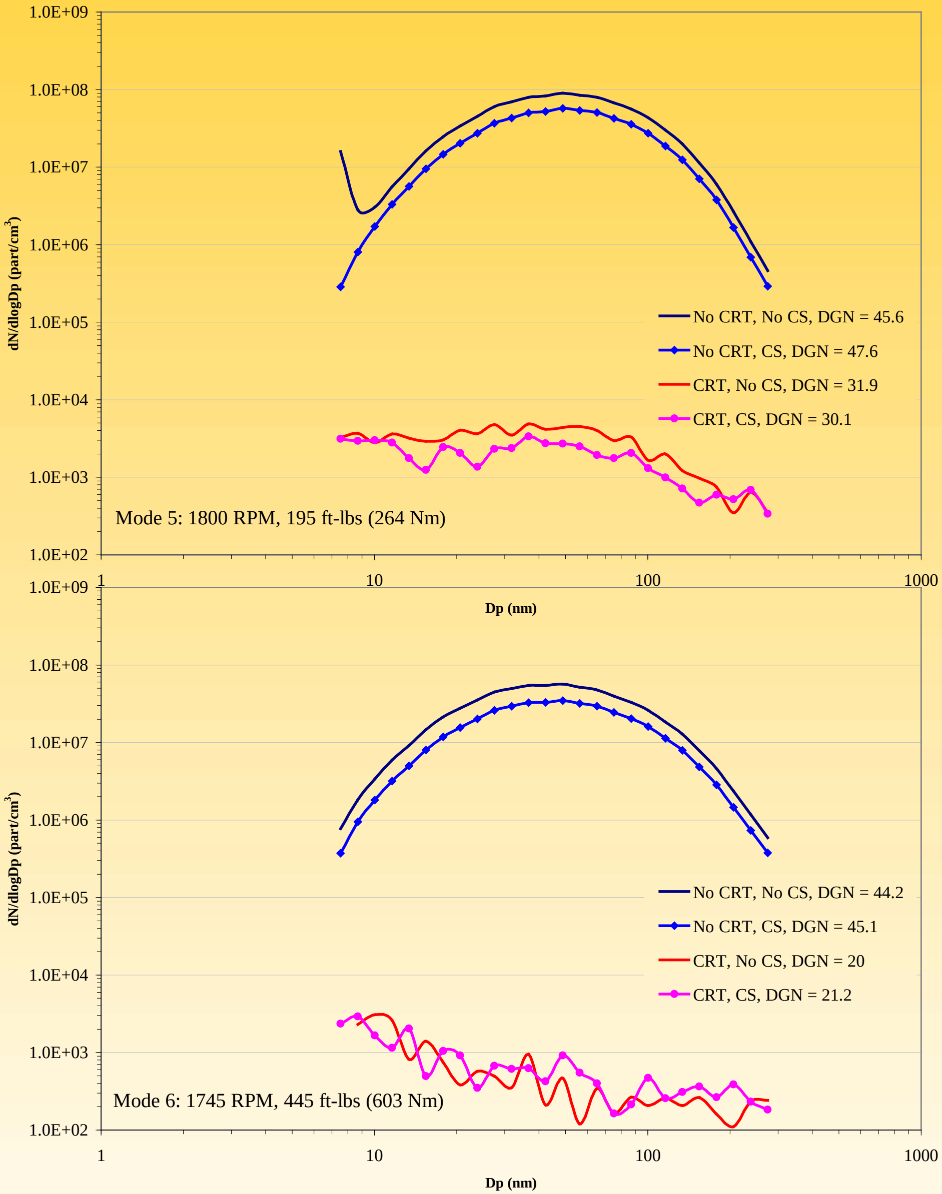
Example: Diesel exhaust size distributions with and without catalyzed exhaust filter

- Test were run using AVL steady state test modes designed to simulate the U.S. heavy-duty transient test – 6 of the 8 AVL modes were run – ***operating modes, not size modes***
  - Mode 1: 700 RPM, 0 ft-lbf (Idle)
  - Mode 2: 821 RPM, 211 ft-lbf (286 Nm)
  - Mode 5: 1800 RPM, 195 ft-lbf (264 Nm)
  - Mode 6: 1745 RPM, 445 ft-lbf (603 Nm)
  - Mode 7: 1745 RPM, 767 ft-lbf (1040 Nm)
  - Mode 8: 1800 RPM, 1024 ft-lbf (1388 Nm)
- SMPS measurements were made with a catalytic stripper that allows solid volatiles to be differentiated

Lab studies with and without filtration

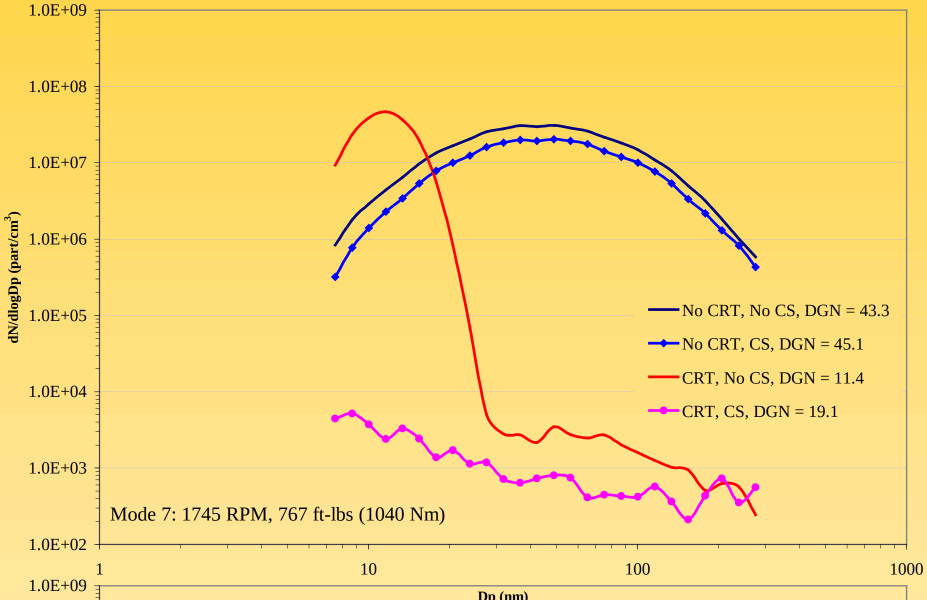


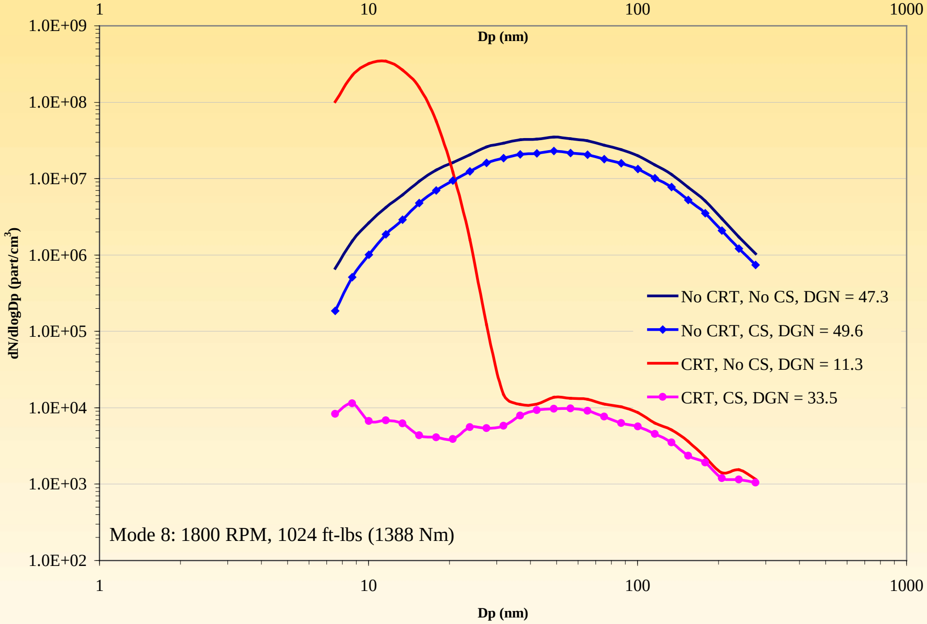
- Mode 1 and 2 are cases that show a nucleation mode without filtration but none with
- Mode 1 shows a clear nucleation mode even after volatile materials are removed by the CS. This suggests that it consists of nonvolatile materials – lube oil ash, very heavy HCs??
- Mode 2 also shows a small nonvolatile nucleation mode



Lab studies with and without filtration

- Modes 5 and 6 give essentially unimodal distributions without filtration – large accumulation mode suppressed nucleation
- With filtration the concentrations are 3 to 5 orders of magnitude lower and near the noise floor of the measurement





Lab studies with and without filtration

- Modes 7 and 8 give unimodal distributions without filtration
- Both of these modes form a nucleation mode downstream of the catalyzed exhaust filtration system with concentrations 1 to 2 orders of magnitude higher than without
- In the accumulation mode range the exhaust filtration system reduces concentrations by 3 to 5 orders of magnitude
- It appears that most of the material in the nucleation mode is sulfate

Are spark ignition engines different from diesel?

Port fuel injected SI engine - influence of additives – size modes?

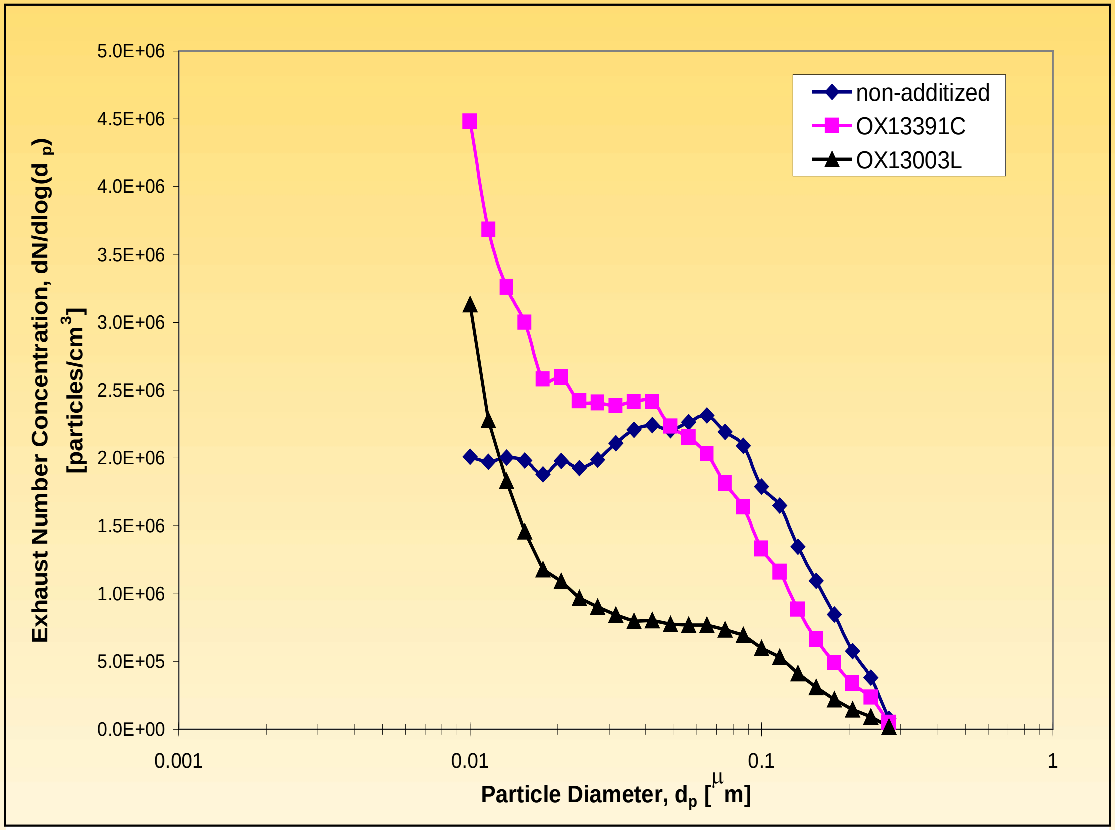
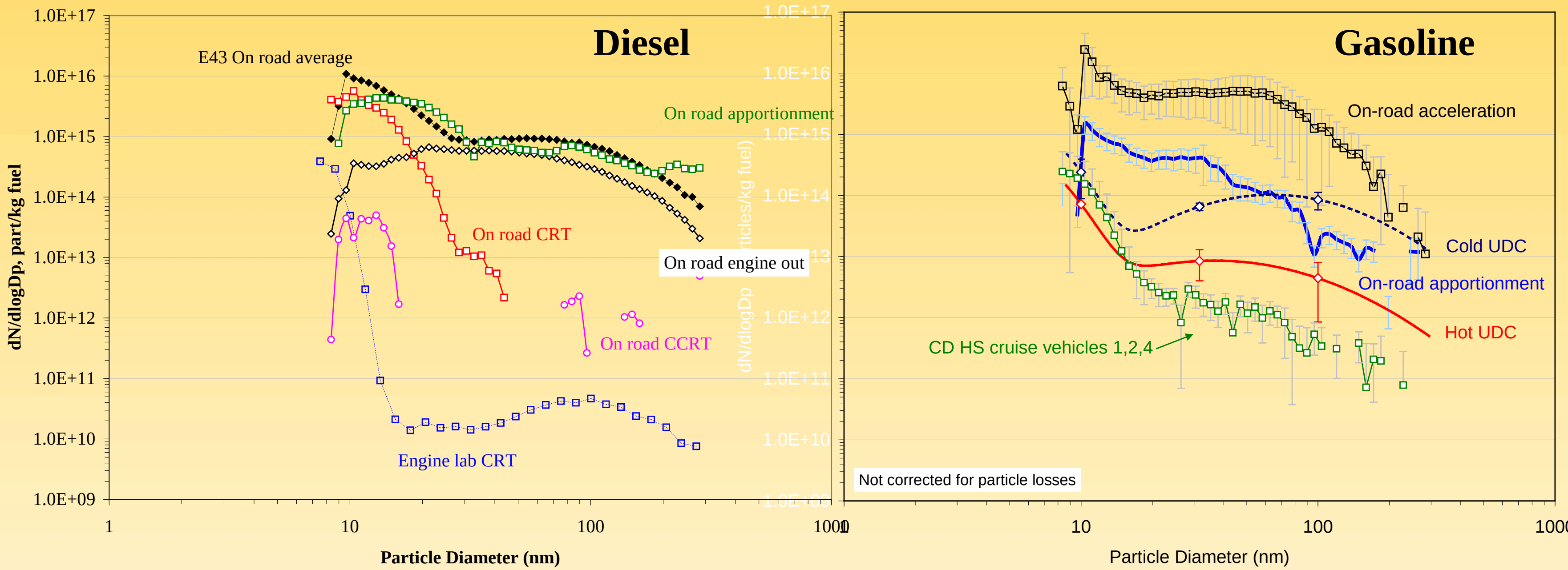


Figure 3. Representative Baseline Size Distributions for the OX13391 and OX13003 Additives [2500 RPM, 90 kPa]

Graskow, B.R., D.B. Kittelson, M.R.Ahmadi, and J.E. Morris. 1999. “Exhaust Particulate Emissions from Two Port Fuel Injected Spark Ignition Engines,” SAE Paper No. 1999-01-1144, 1999.

On-road and lab experiments – Diesel and PFI gasoline size distributions



- During highway cruise PFI emissions are significantly lower than Diesel
- PFI emissions are much more load dependent than Diesel
- Modal structure less obvious with PFI

Johnson, Jason P., David B. Kittelson, Winthrop F. Watts, 2005. "Source Apportionment of Diesel and Spark Ignition Exhaust Aerosol Using On-Road Data from the Minneapolis Metropolitan Area," Atmospheric Environment 39, 2111–2121. - Kittelson, D. B., W. F. Watts, J. P. Johnson, J. Schauer, and D. R. Lawson 2006. "On-road and Laboratory Evaluation of Combustion Aerosols Part 2: Summary of Spark Ignition Engine Results," Journal of Aerosol Science 37, 931 – 949. - Kittelson, D. B., W. F. Watts, J. P. Johnson, C. J. Rowntree, S. P. Goodier, M. J. Payne, W. H. Preston, C. P. Warrens, M. Ortiz, U. Zink, C. Goersmann, M. V. Twigg and A. P. Walker, 2006. "Driving Down On-Highway Particulate Emissions," SAE paper number 2006-01-0916

Particles from PFI engines highest for cold start, hard acceleration, fuel rich

Hard acceleration

Cold start

Very little in between

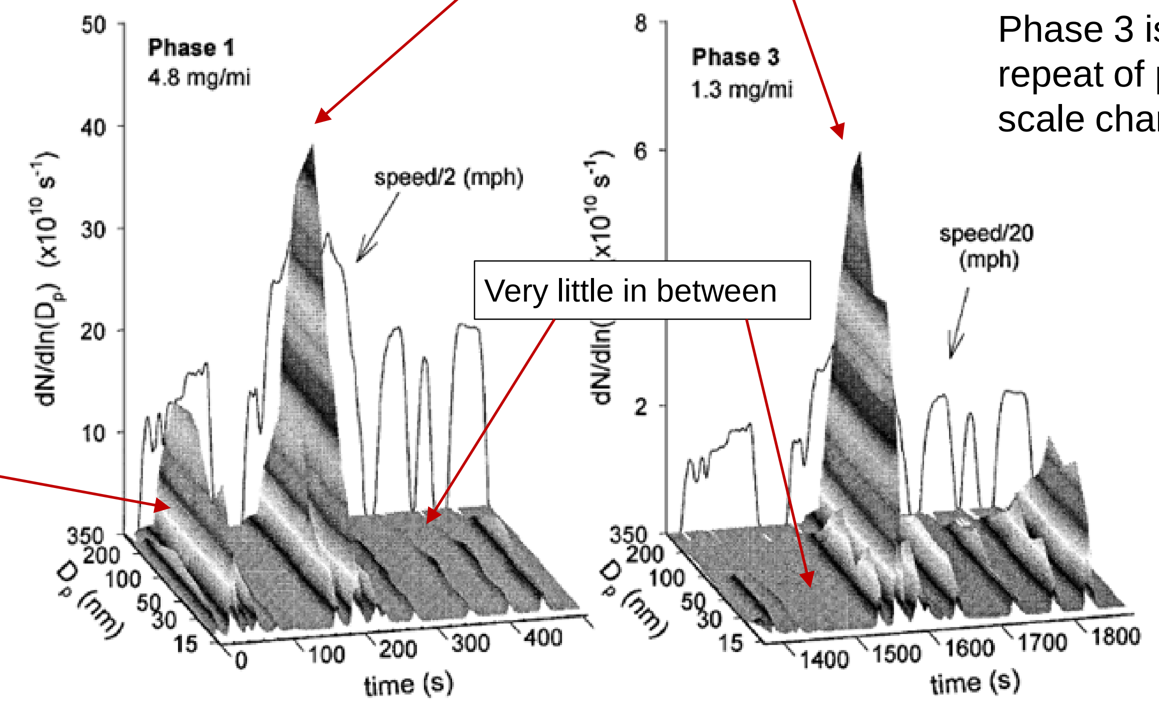


FIGURE 3. Comparison of size-resolved particle emissions from a gasoline light-duty truck (T5) during the cold start (phase 1) versus the hot start (phase 3) of the FTP. Note the change in vertical scale from phase 1 to phase 3. Vehicle speed is shown by the traces along the rear walls of the plots.

Phase 3 is warm start repeat of phase 1, note scale change, 5x

Bimodal, trimodal?

Examination of the Size-Resolved and Transient Nature of Motor Vehicle Particle Emissions, M. Matti Maricq, Diane H. Podsiadlik, and Richard E. Chase, Environ. Sci. Technol., 1999, 33 (10), 1618-1626• DOI: 10.1021/es9808806

GDI car, PN sensitive to fuel composition, diesel like size distributions, clear modes

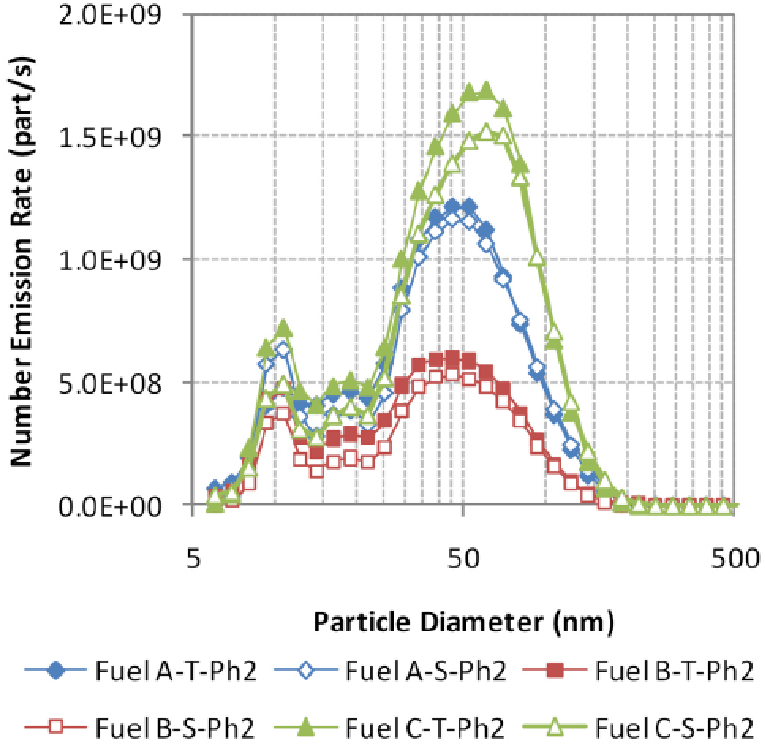


Figure 13. Phase 2 Total and Solid Particle Number-Weighted Size Distribution for Fuels A, B, and C

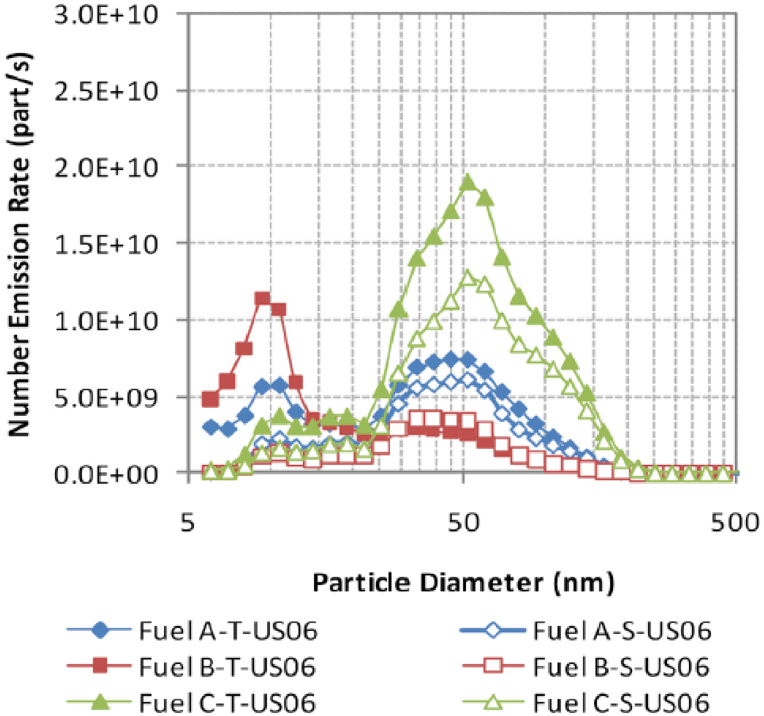


Figure 15. US06 Total and Solid Particle Number-Weighted Size Distribution for Fuels A, B, and C

From: Khalek, Imad A., Thomas Bougher, and Jeff J. Jetter, 2011. Particle Emissions from a 2009 Gasoline Direct Injection Engine Using Different Commercially Available Fuels, SAE paper number 2010-01-2117.

Highest numbers from GDI also cold start, high load but doesn’t drop off as much, *modes unclear*

Cold start and run

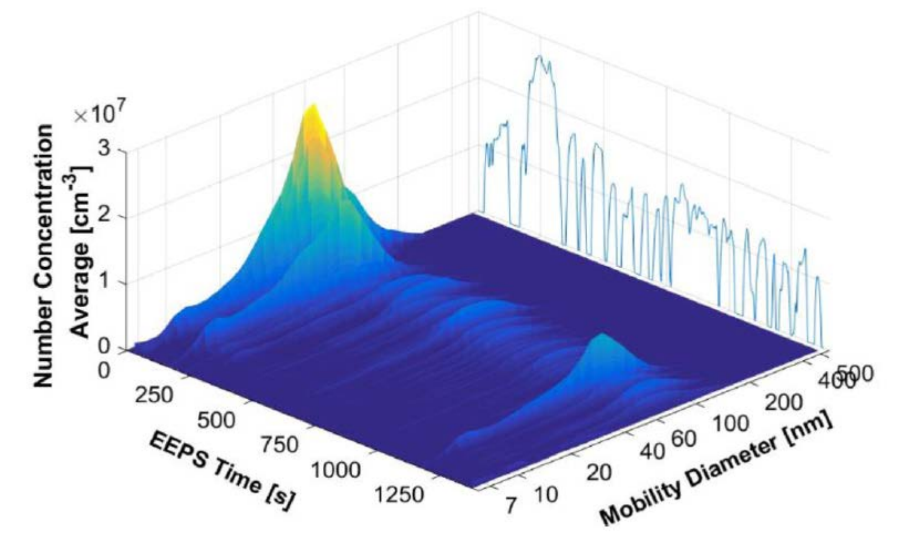


Figure 2. The PSD for phases 1 and 2 of the FTP cycle. The drive trace is included for visual reference.

Warm start

Scale change 5x

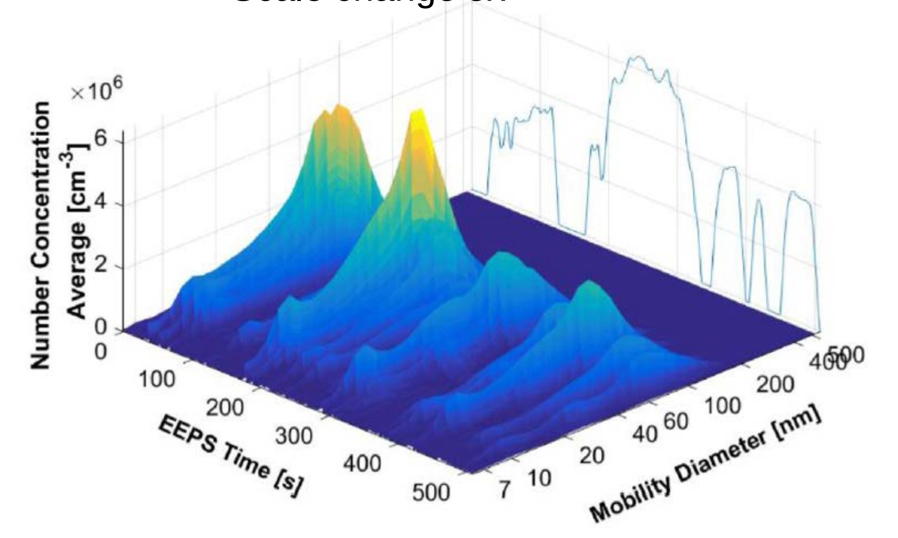


Figure 3. The PSD for phase 3 of the FTP cycle. The drive trace is included for visual reference.

Koczak, J., Boehman, A., and Brusstar, M., "Particulate Emissions in GDI Vehicle Transients: An Examination of FTP, HWFET, and US06 Measurements," SAE Technical Paper 2016-01-0992, 2016, doi:10.4271/2016-01-0992.

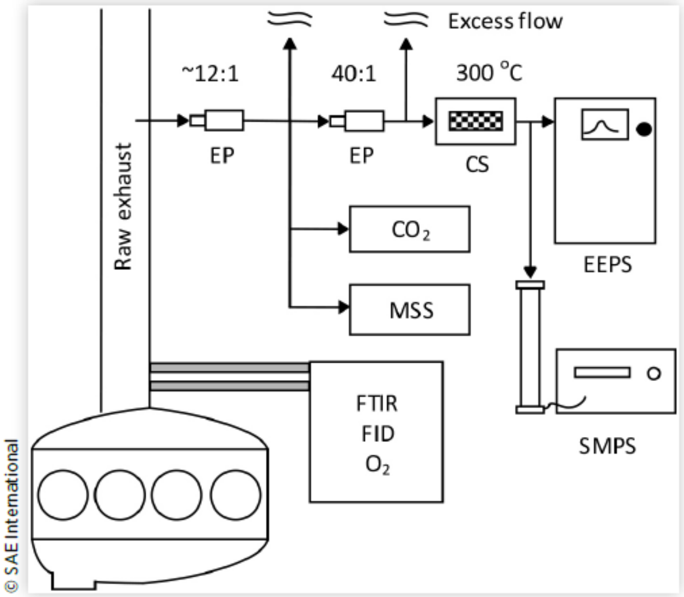
Recent U of M work: Particle emissions from *lean* and stoichiometric burn GDI

BMW N43B20, four-cylinder, 2.0 L, naturally aspirated engine

TABLE 1 Engine specifications

Model Number	N43B20
Displacement (cc)	1995
Bore x Stroke (mm)	84 x 90
Compression Ratio	12:1
Rated Power (kW)	125 @ 6700 rpm
Rated Torque (Nm)	210 @ 4250
Induction	Naturally Aspirated
Injection	Central Spray Guided Piezo Injectors
Max Rail Pressure (bar)	200

FIGURE 1 Engine and instrumentation schematic showing dilution system and instruments used in the experimental study.



All measurements made with catalytic stripper to eliminate semi-volatile modes

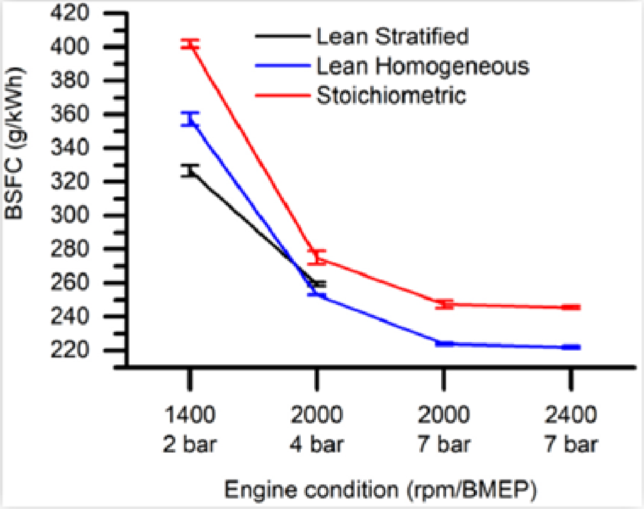
Adapted from: Bock, N., Jeon, J., Kittelson, D., and Northrop, W.F., “Solid Particle Number and Mass Emissions from Lean and Stoichiometric Gasoline Direct Injection Engine Operation,” SAE Technical Paper 2018-01-0359, 2018, doi:10.4271/2018-01-0359.

Significant fuel savings associated with lean operation

TABLE 2 Engine conditions including operating mode and equivalence ratio. S = stoichiometric, LH = lean homogeneous, and LS = lean stratified.

Condition	Speed (rpm)	BMEP (bar)	Mode	φ
SS 1	1400	2	S	1.0
	1400	2	LH	0.67
	1400	2	LS	0.5
SS 2	2000	4	S	1.0
	2000	4	LH	0.65
	2000	4	LS	0.65
SS 3	2000	7	S	1.0
	2000	7	LH	0.69
SS 4	2400	7	S	1.0
	2400	7	LH	0.73
Load steps	2000	2-7	S	1.0
	2000	2-7	LH	0.73-0.67
Engine start	1000	0	S	1.0

FIGURE 2 Average BSFC for the 10 steady state engine conditions. Error bars represent one standard deviation.

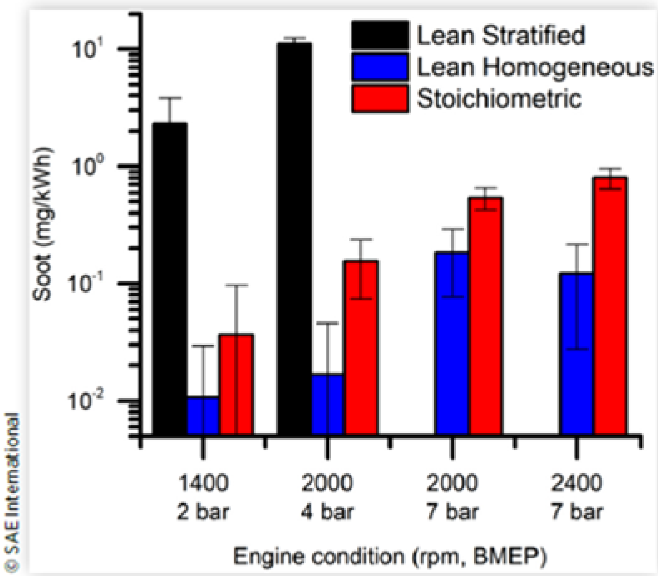


Adapted from: Bock, N., Jeon, J., Kittelson, D., and Northrop, W.F., “Solid Particle Number and Mass Emissions from Lean and Stoichiometric Gasoline Direct Injection Engine Operation,” SAE Technical Paper 2018-01-0359, 2018, doi:10.4271/2018-01-0359.

Lean stratified shows much higher emissions than other modes

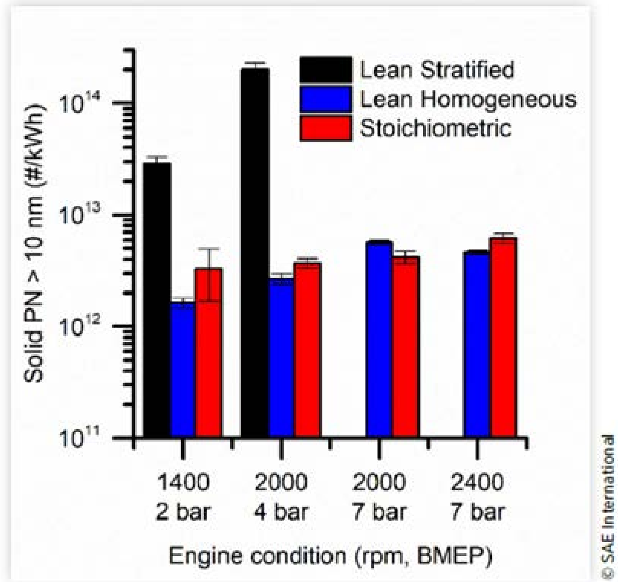
Lean stratified soot mass emissions approach US HD diesel standard

FIGURE 4 Average brake specific soot emissions as measured by MSS for the 10 steady state engine conditions. Error bars represent one standard deviation.



Shown here are PN>10nm, PN>23nm about half, but PN>23 still at or above 6×10^{11} EU HD diesel limit

FIGURE 3 Average brake specific solid PN > 10 nm emissions as measured by EEPS for the 10 steady state engine conditions. Error bars represent one standard deviation.

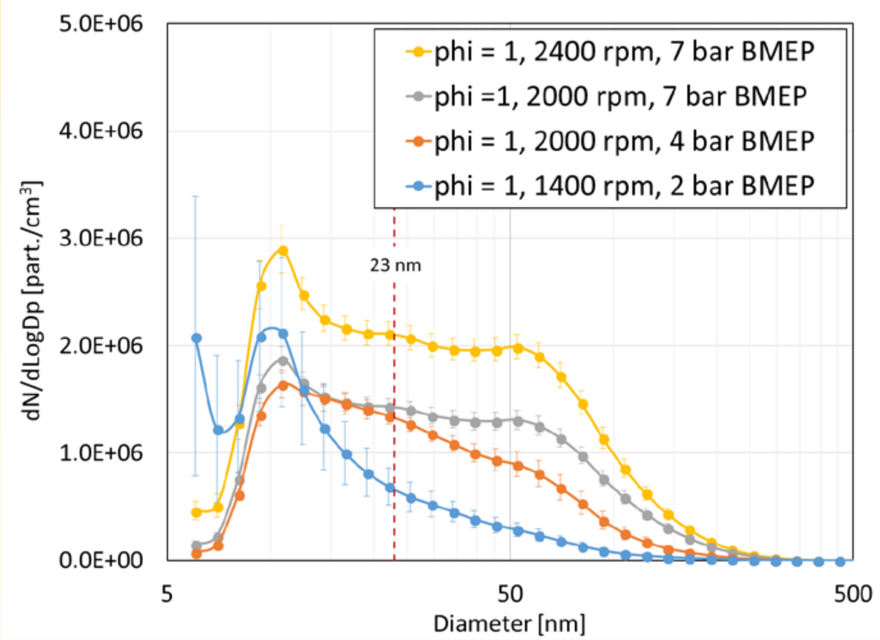


Adapted from: Bock, N., Jeon, J., Kittelson, D., and Northrop, W.F., “Solid Particle Number and Mass Emissions from Lean and Stoichiometric Gasoline Direct Injection Engine Operation,” SAE Technical Paper 2018-01-0359, 2018, doi:10.4271/2018-01-0359.

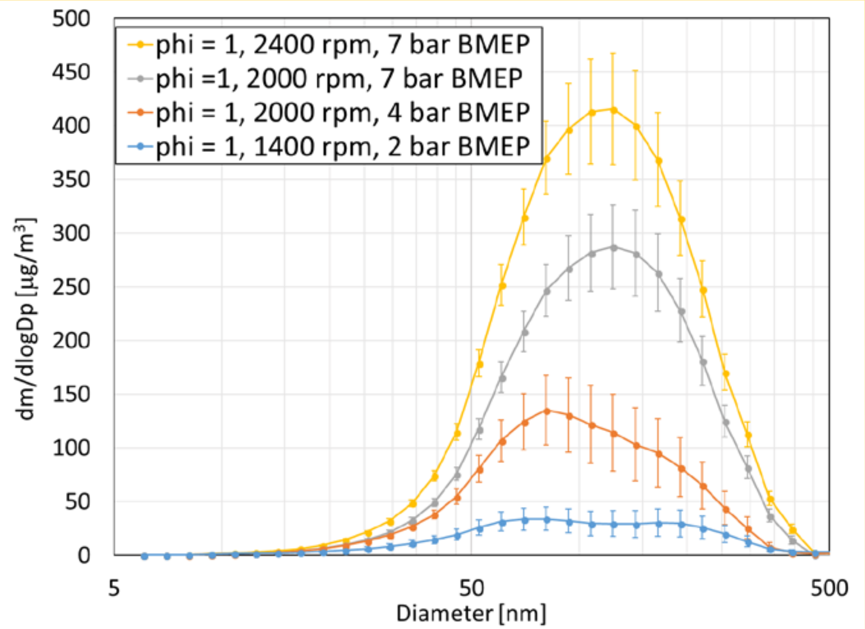
Stoichiometric operation:

Odd size distributions – unresolved modes, emissions, especially mass, very load dependent

Number



Mass



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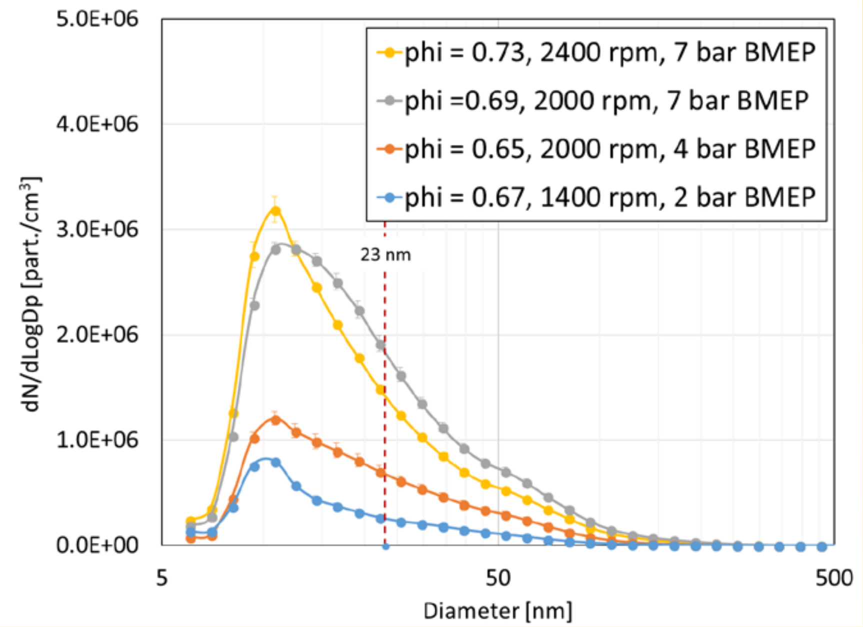
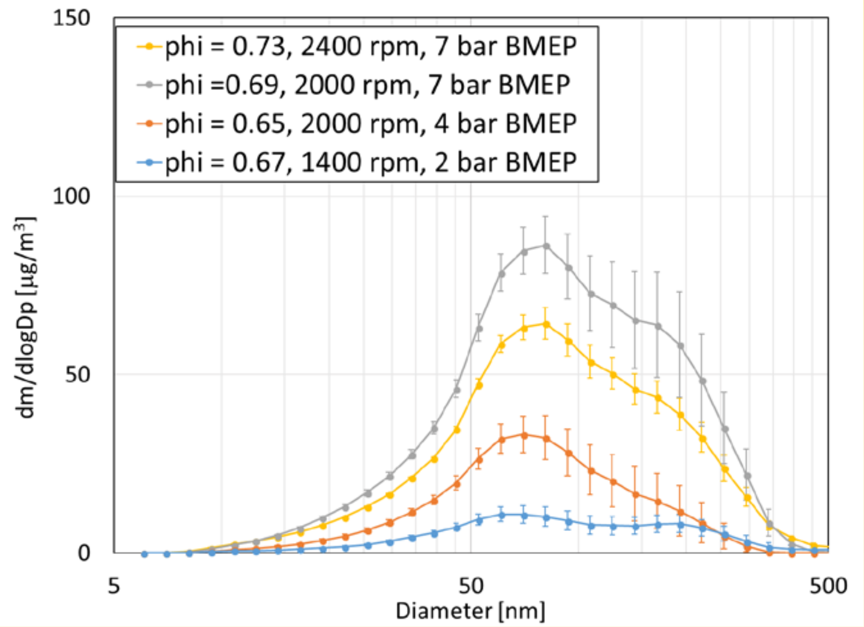
Lean homogeneous operation:

Smaller particles, unresolved modes, much lower mass emissions than stoichiometric

Number



Mass



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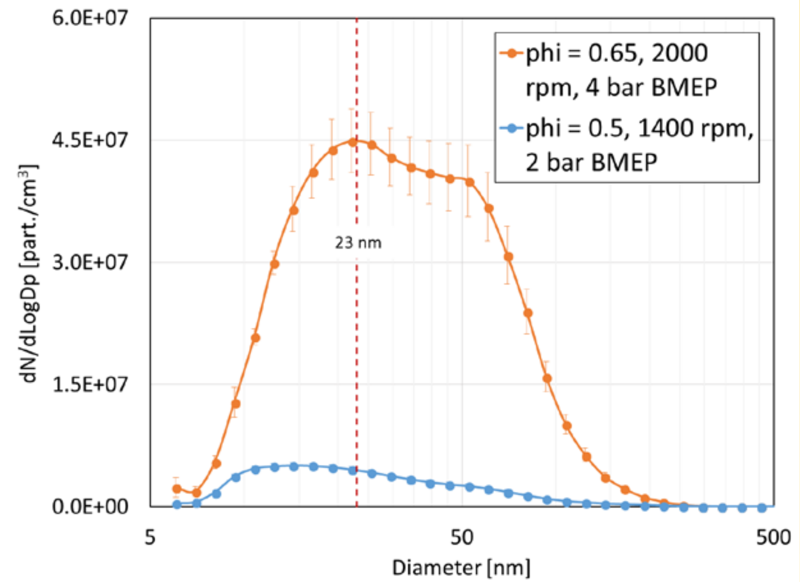
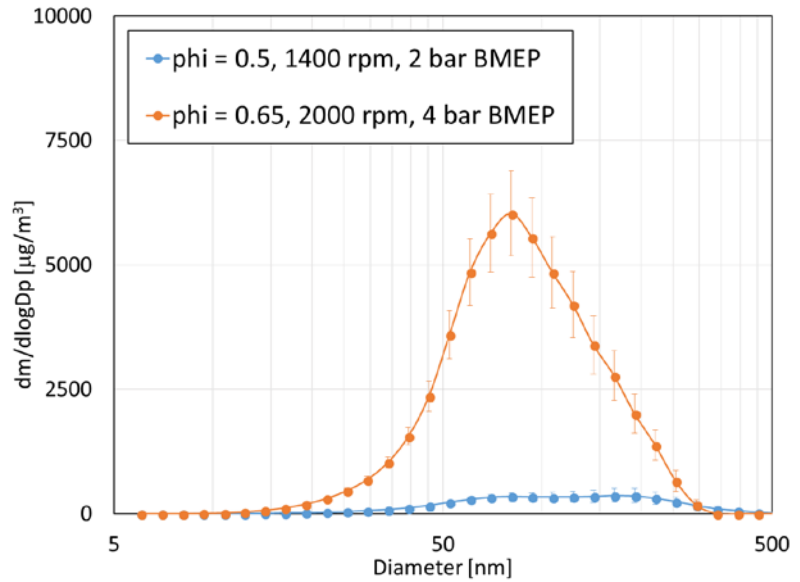
Lean stratified operation:
Odd flat size distributions, unresolved modes, much higher number and mass emissions

Number



Mass



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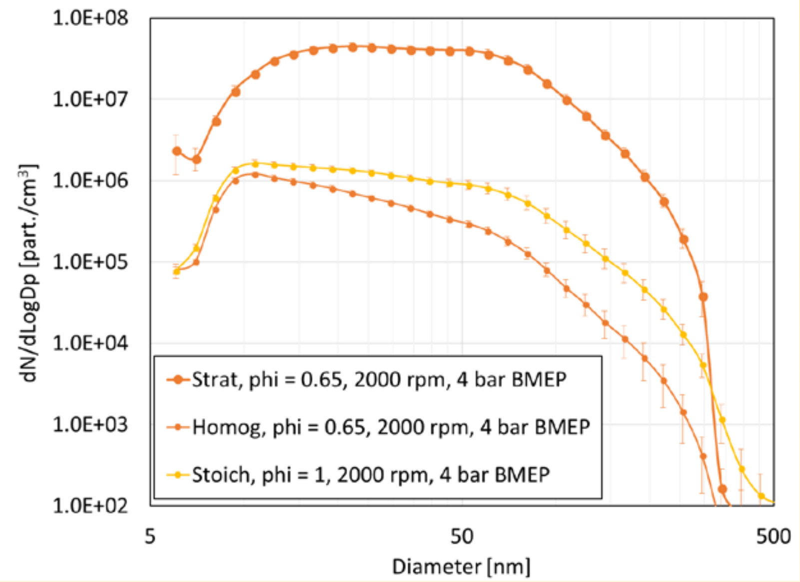
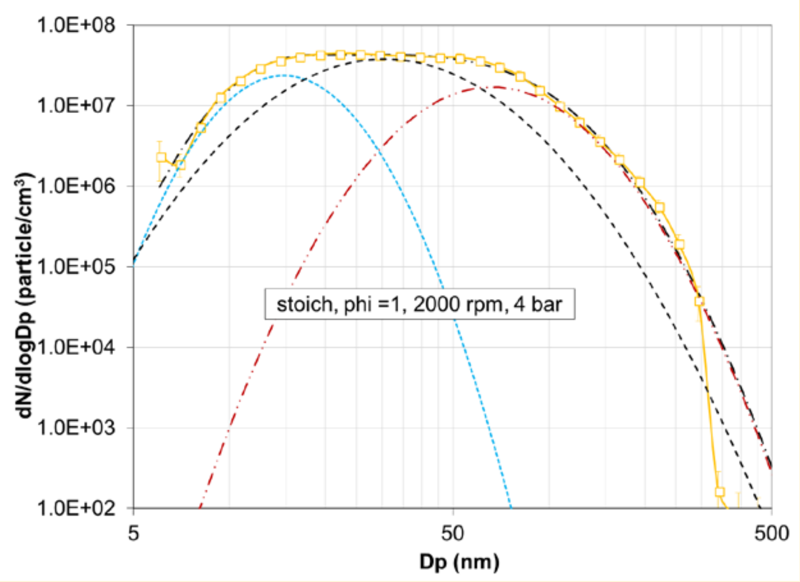
Examine modal structure, 2000 rpm, 4 bar bmep

3 distinct modes, apparently solid, CS used





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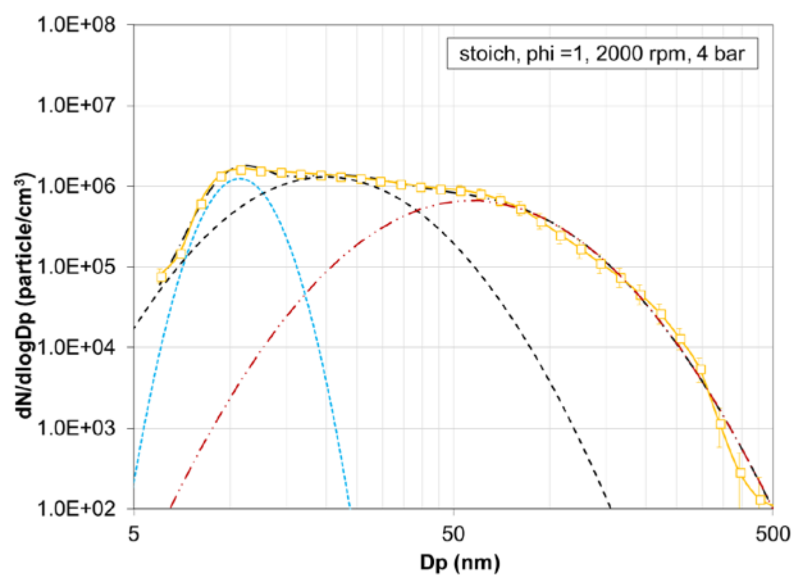
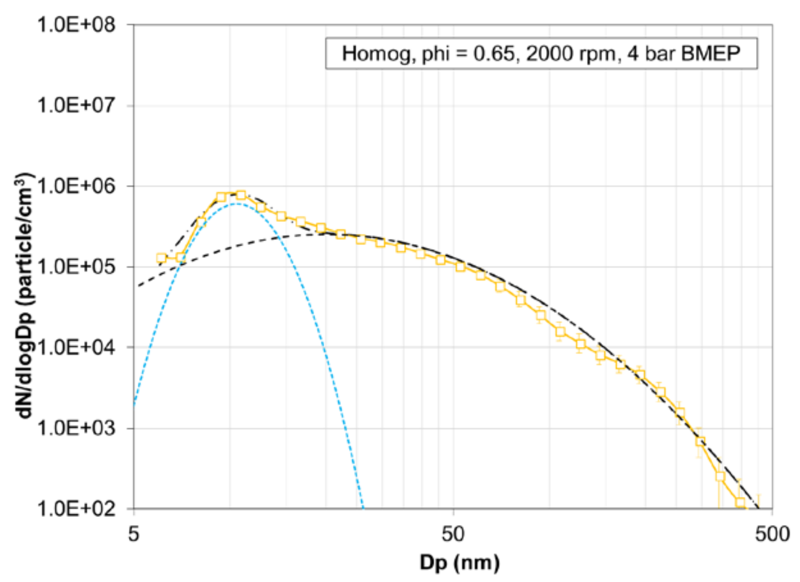


2 distinct modes, apparently solid, CS used



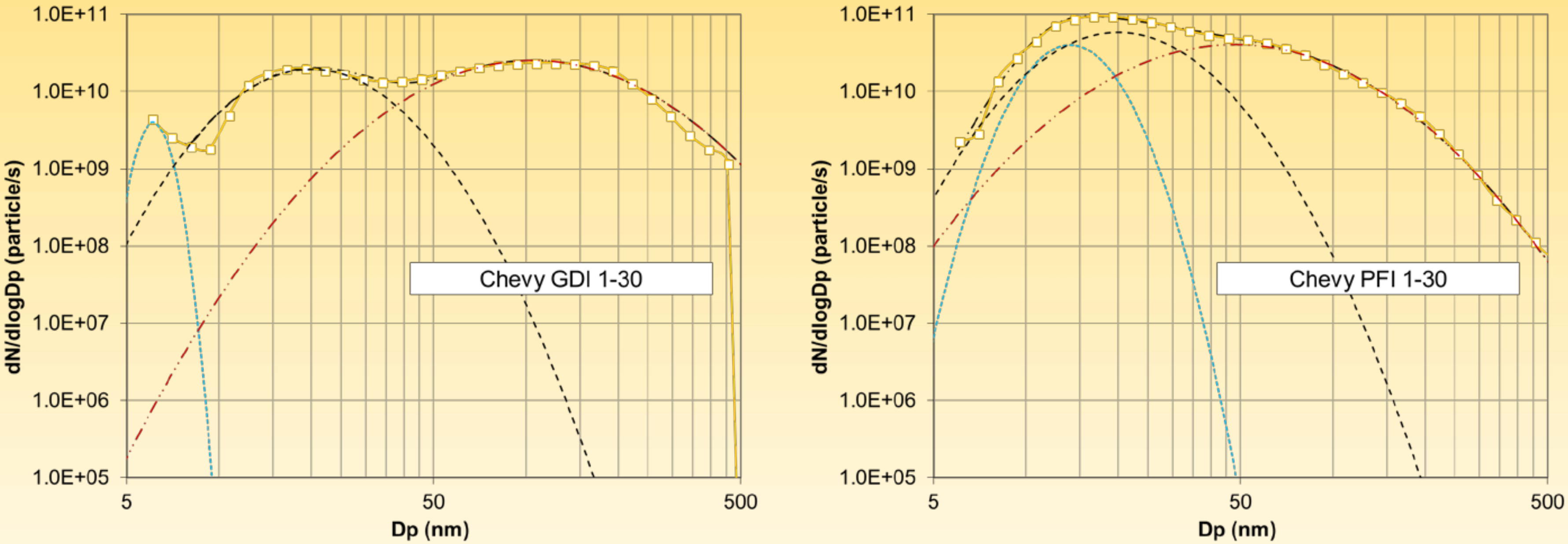
Modal structure of cold start emission rates of PFI and GDI engines - based on data from:

Particle Emissions from Light Duty Vehicles during Cold-Cold Start, Huzeifa I. Badshah, William F. Northrop, and David B. Kittelson, *SAE Int. J. Engines 9(3):2016*

Particle Emissions from Light Duty Vehicles during Cold-Cold Start and Identified from Ambient Measurements, Huzeifa Ismail Badshah, M.S. Thesis, University of Minnesota, 2015

Typical modal structures of cold start emission rates, average first 30 s (1-30)

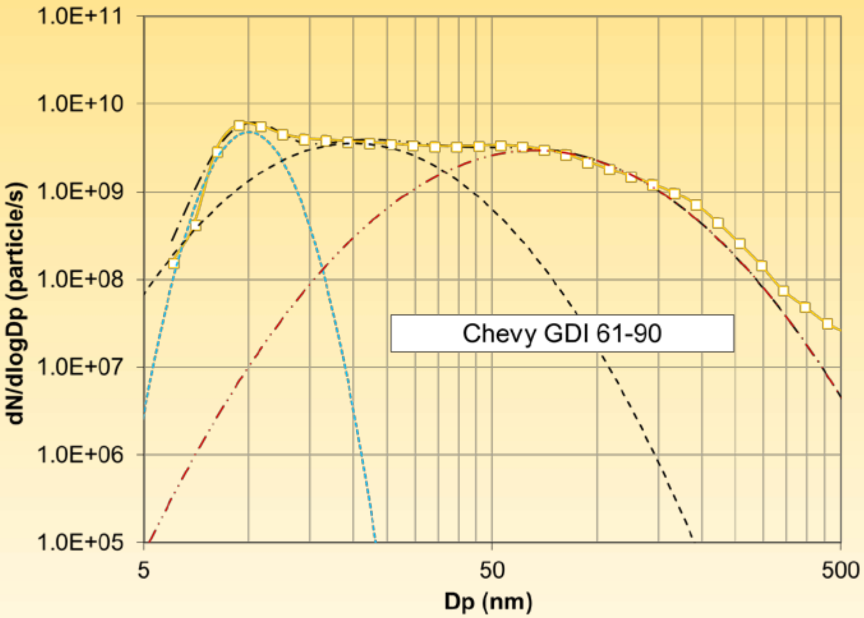
Often not much difference in emission rates between PFI and GDI during first 30 s

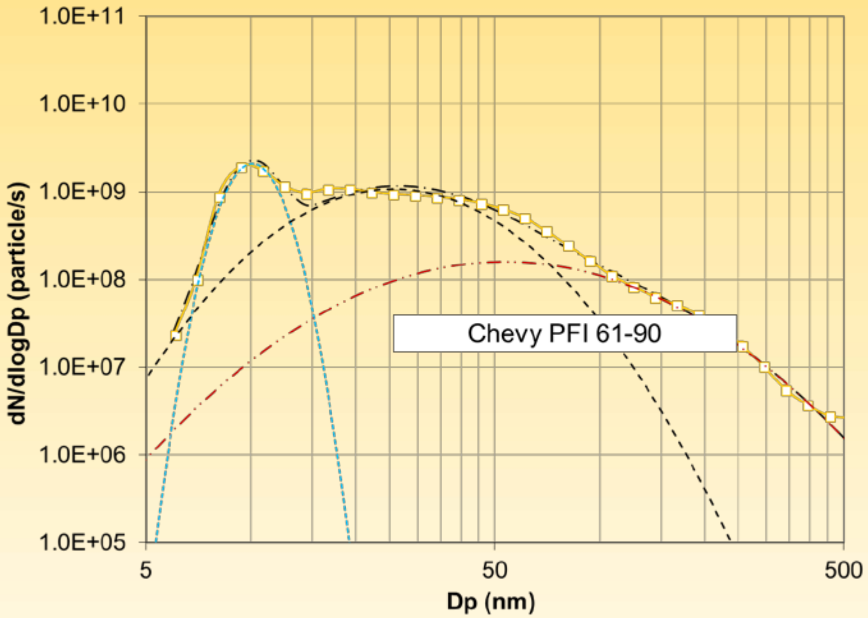


These are solid particle emission rates measured downstream of catalytic stripper

Typical modal structures of cold start emission rates, average 60-90 s

PFI emission rates fall much faster than GDI, especially in the 3rd (largest) mode where most of the mass is found





These are solid particle emission rates measured downstream of catalytic stripper

Observed submicron modal structures, solid modes measured with catalytic stripper

- Diesel – 1 or 2 solid modes
  - Nucleation mode
    - Ash
    - Nascent soot
  - Accumulation or soot mode
  - Formed at different times, different processes
- Gasoline spark ignition – multiple solid modes
  - Nucleation
    - Ash
    - Nascent soot
  - Multiple soot modes?
  - Formed at different times, different processes

Modes linked to formation processes

- Diesel
  - Accumulation mode formed by well defined burning fuel jet
  - Nucleation modes form later
    - Unscavenged ash or nascent soot
    - Unscavenged Semi-volatiles
- Gasoline
  - Accumulation modes formed by
    - Pool burning
    - Injector dribble (GDI)
    - Valve dribble (PFI)
    - Local rich pockets
  - Nucleation modes form later
    - Unscavenged ash or nascent soot
    - Unscavenged Semi-volatiles

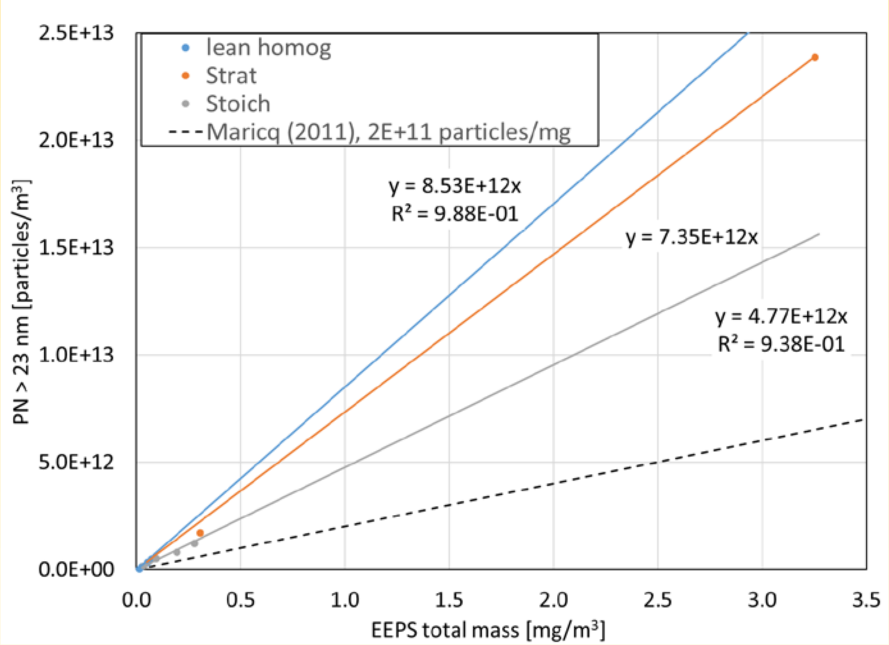
Conclusion

- Modal structure of size distribution linked to formation mechanisms
- Understand the modes, understand PM and PN formation

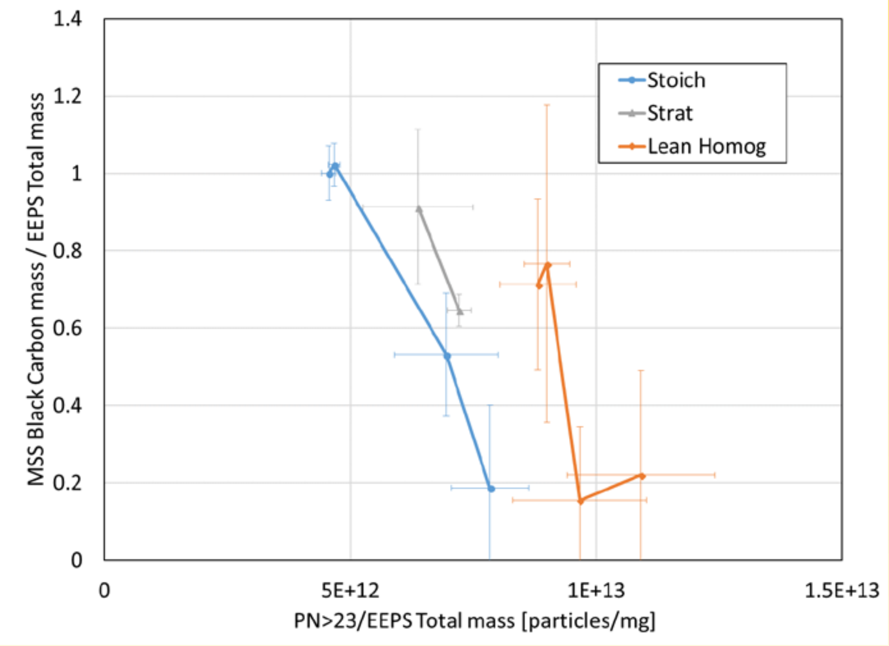
Thank you
Questions?

Size and composition trends

Higher particle number to mass ratio, smaller particles than reported elsewhere



Lower black carbon content in smaller particles - ash, tightly bound OC?



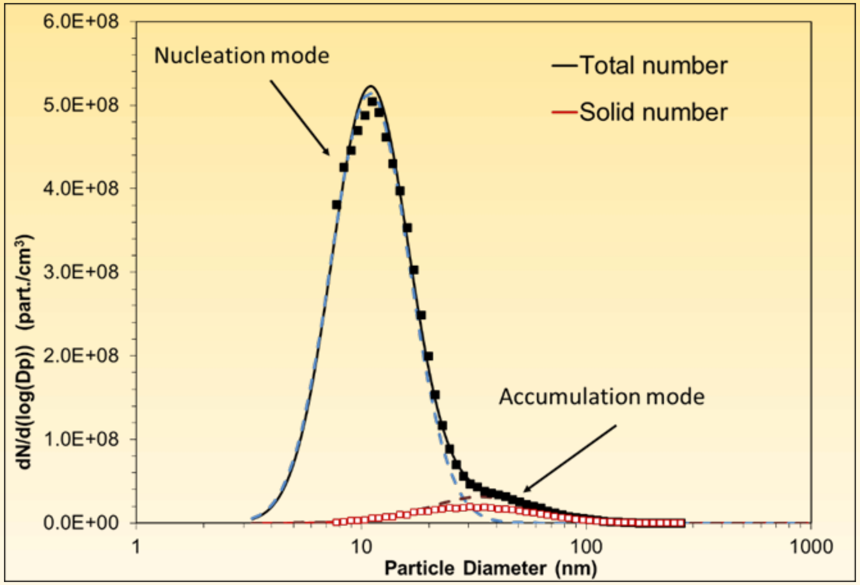
Maricq, M.M., Szente, J., Loos, M., and Vogt, R., “Motor Vehicle PM Emissions Measurement at LEV III Levels,” SAE Int. J. Engines 4(1):597-609, 2011

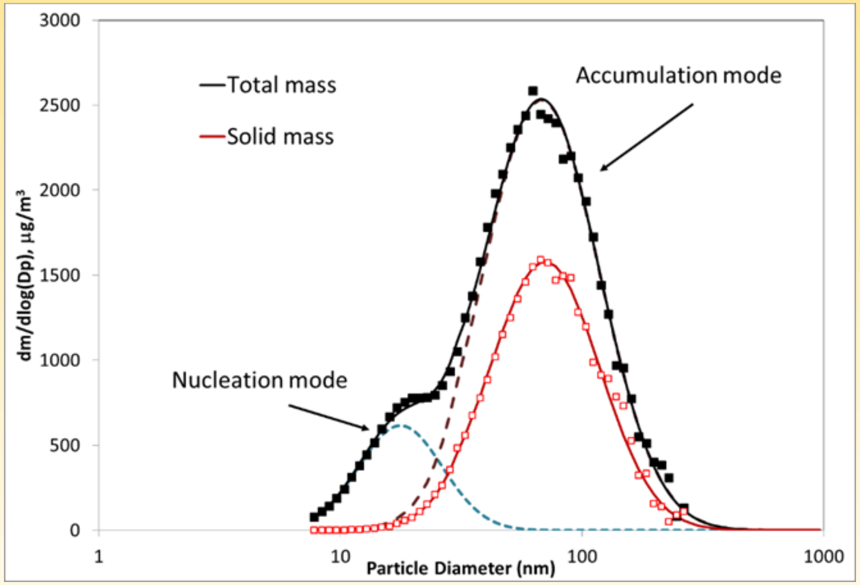
Adapted from: Bock, N., Jeon, J., Kittelson, D., and Northrop, W.F., “Solid Particle Number and Mass Emissions from Lean and Stoichiometric Gasoline Direct Injection Engine Operation,” SAE Technical Paper 2018-01-0359, 2018, doi:10.4271/2018-01-0359.

Additional slides

Total and solid (CS) particle number and mass distributions, Tier 4/Interim IIIB engine, 2400 rpm, 175 N-m

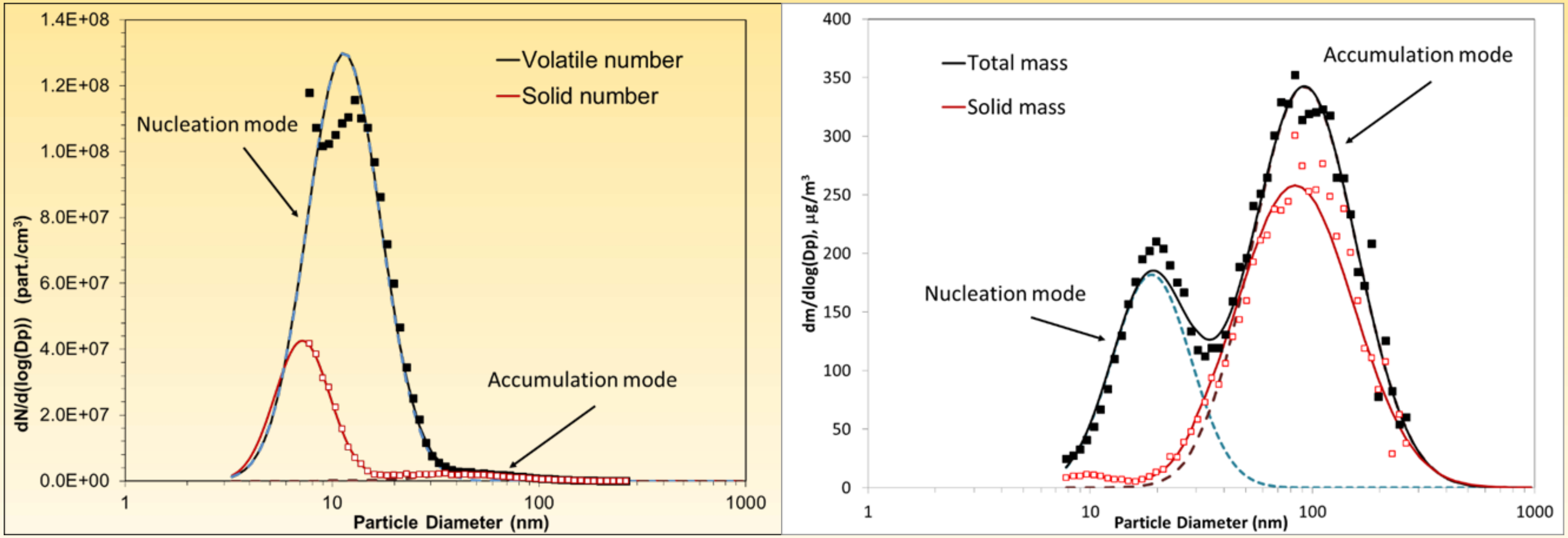
- Number distribution on left, mass on right
- This condition forms large volatile nucleation mode, mainly < 23 nm, containing nearly all the number and significant mass
- On the other hand, nearly all the solid mass and much of the solid number is in the accumulation mode, mainly > 23 nm, consistent with the PMP approach





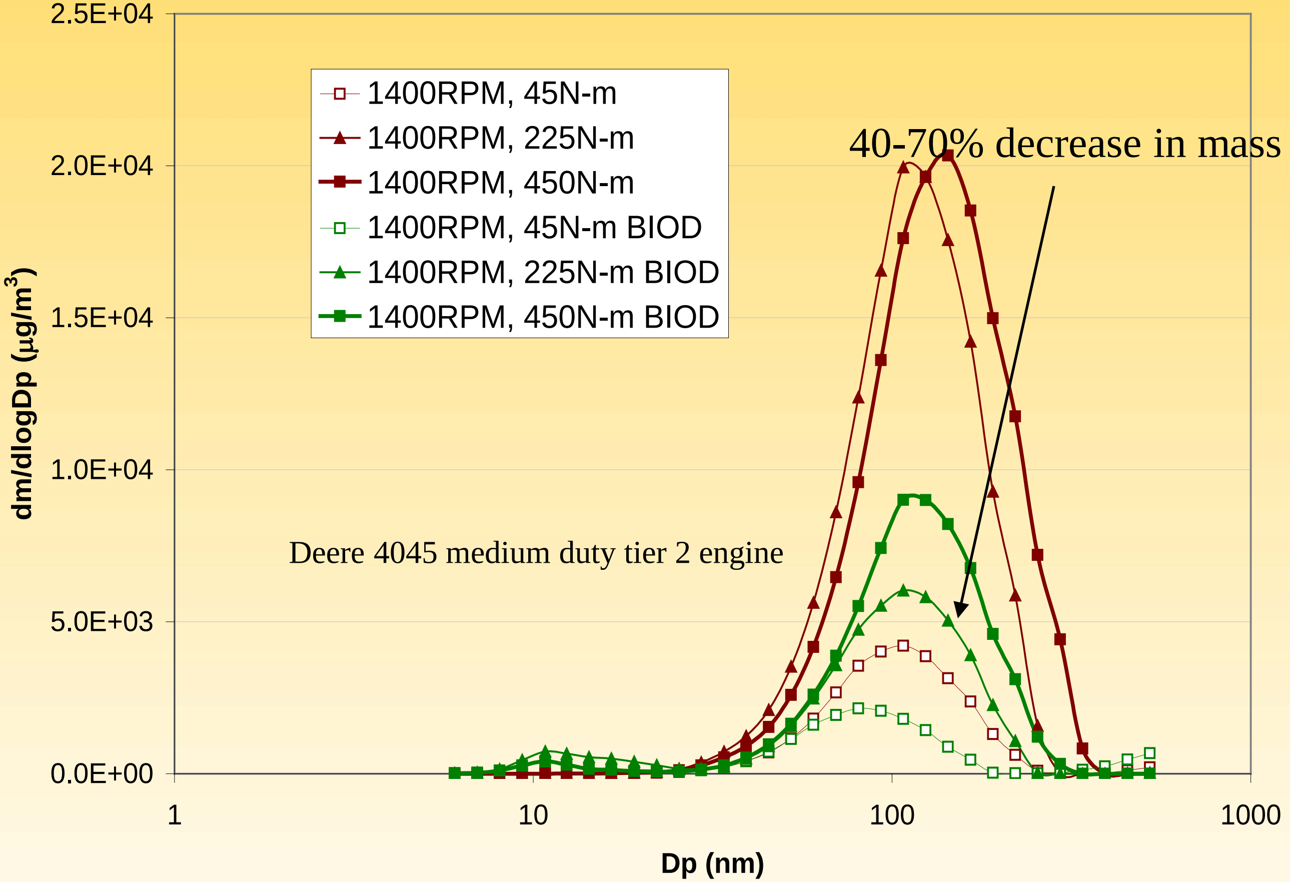
Total and solid (with CS) particle number and mass distributions, Tier 4/Interim IIIB engine, 900 rpm, 25 N-m

- Number distribution on left, mass on right
- This condition forms large volatile nucleation mode, mainly < 23 nm, with nearly all the number and nearly half the mass
- A large solid nucleation mode is present with nearly all the solid number < 23 nm, likely a solid ash mode, these would not be counted by the PMP method



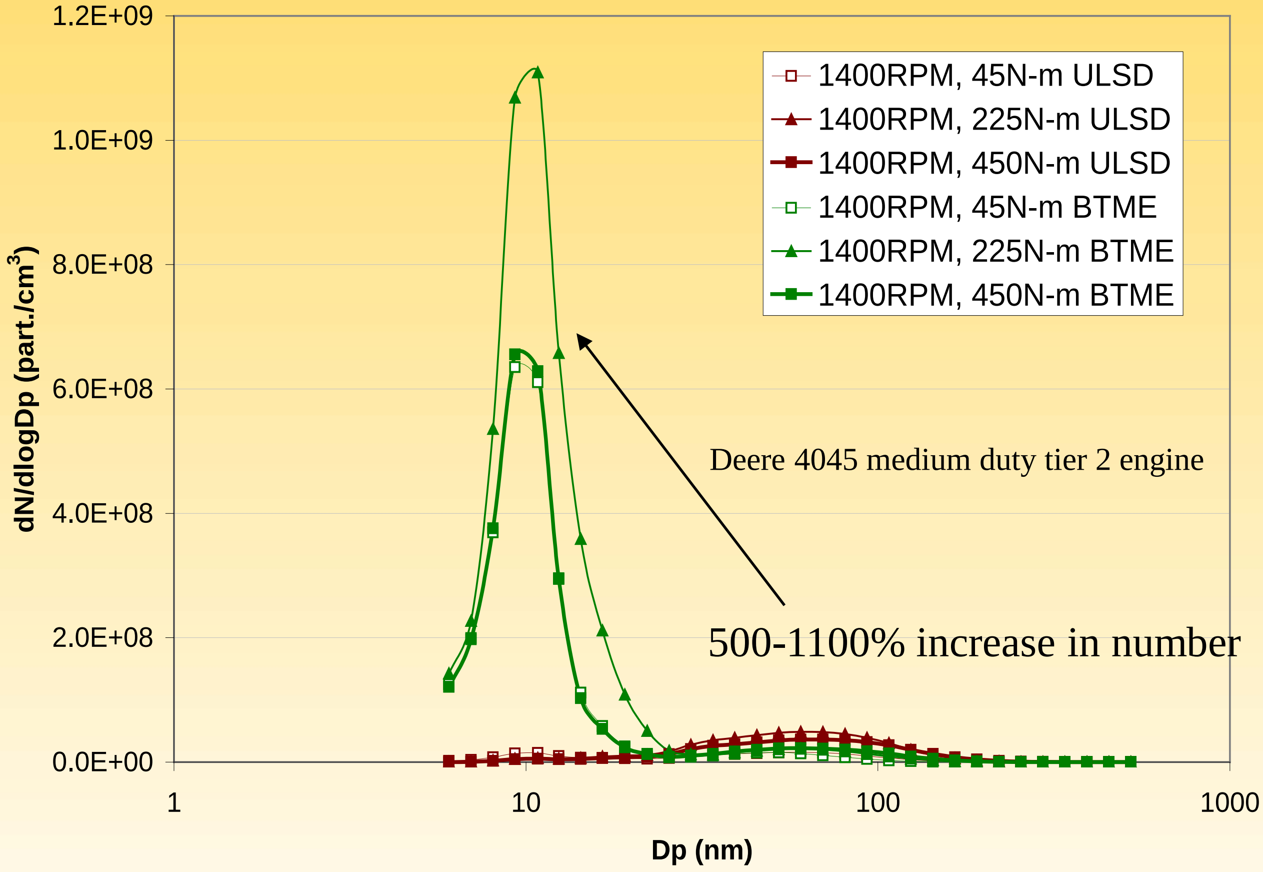
Lucachick, Glenn, Aaron Avenido, Winthrop Watts, David Kittelson, and William Northrop, 2014. Efficacy of In-Cylinder Control of Particulate Emissions to Meet Current and Future Regulatory Standards, SAE paper number 2014-01-1597.

Mass distributions, medium-duty engine, ULSD and Beef Tallow Methyl Ester (BTME)



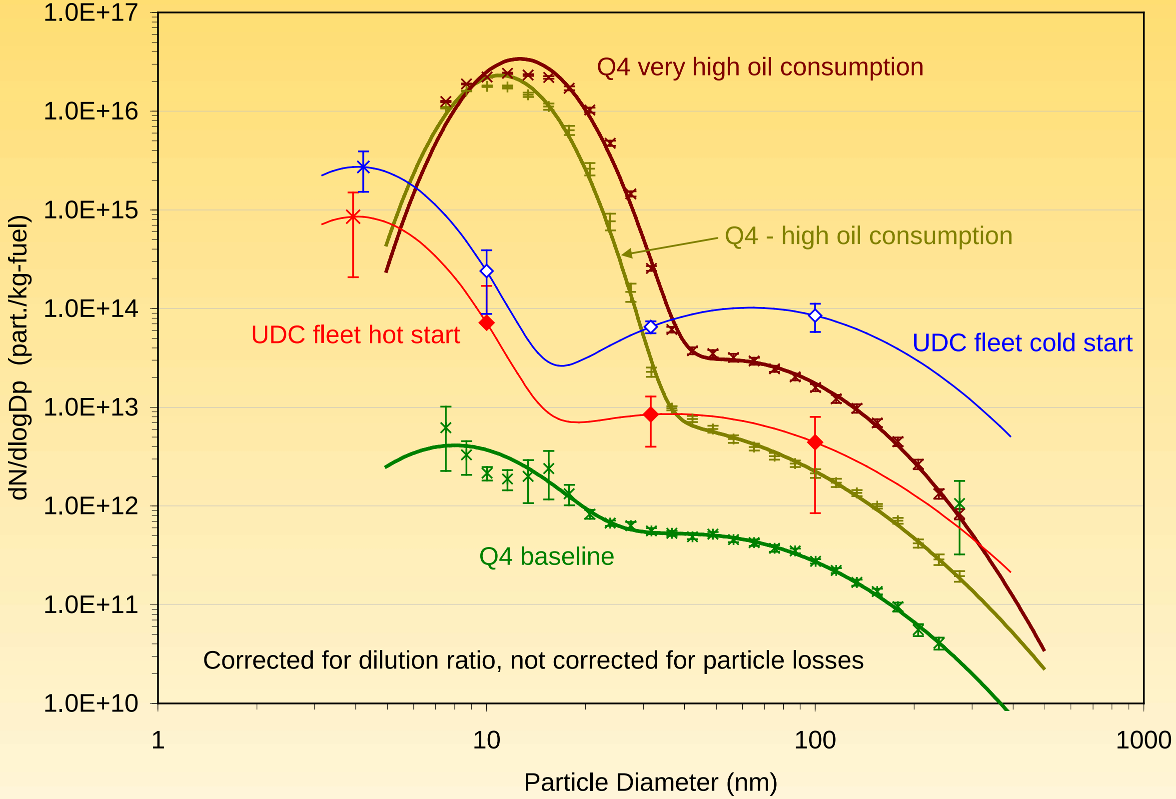
*BTME and USLD results obtained by ME 4431 lab, Spring Semester 2008, Aaron Collings, TA

Number distributions, medium-duty engine, ULSD and Beef Tallow Methyl Ester (BTME)



*BTME and USLD results obtained by ME 4431 lab, Spring Semester 2008, Aaron Collings, TA

PFI engine emissions strongly influenced by cold starts and oil consumption



Q4 - Quad 4 engine dyno tests with varied oil consumption, steady state cruise

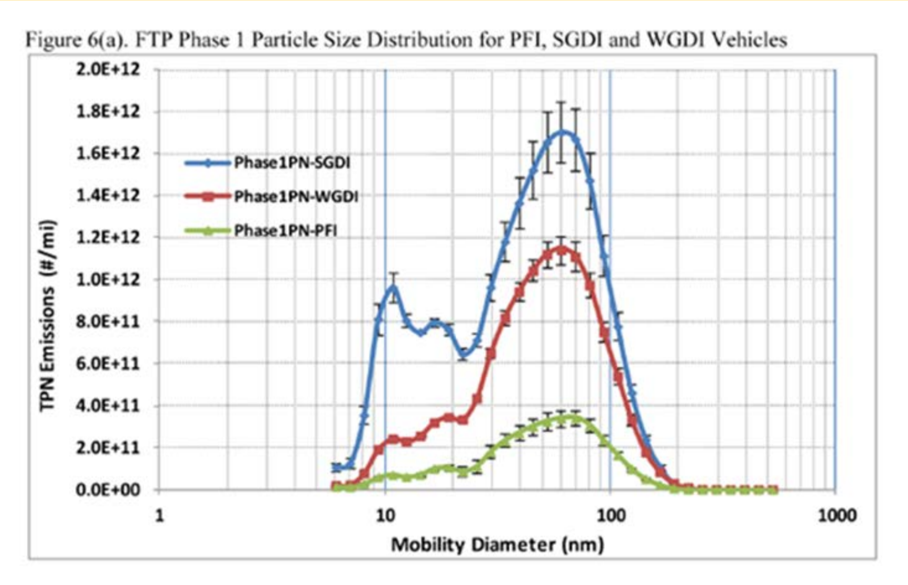
UDC – Chassis dyno tests of small fleet on UDC cycle

These results are shown on fuel specific basis, particles added per unit fuel burned.

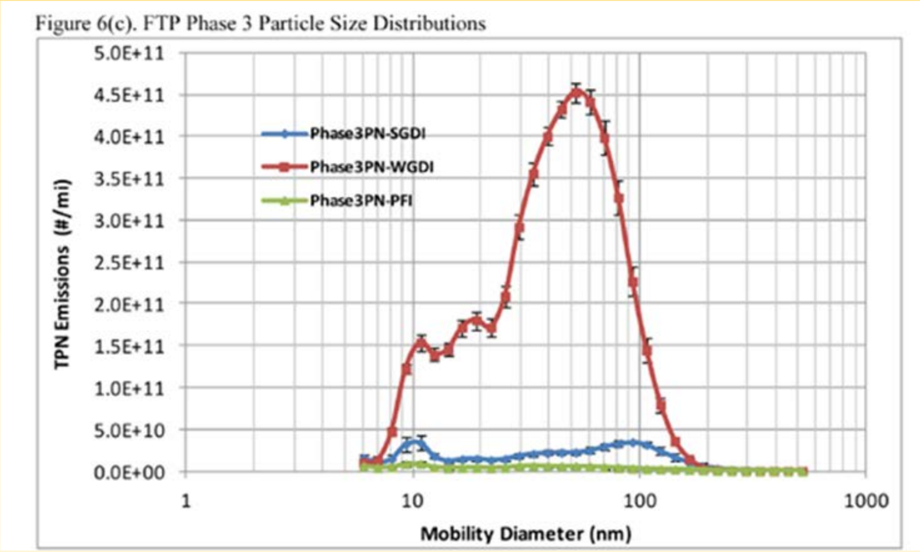
Kittelson, D. B., W. F. Watts, J. P. Johnson, D. Zarling, A. Kasper, U. Baltensperger, H. Burtscher, J. J. Schauer, C. Christenson, and S. Schiller. 2003. Gasoline vehicle exhaust particle sampling study. Contract Final Report U. S. Department of Energy Cooperative Agreement DE-FC04-01AI66910.

PFI engines emit mainly during cold phase, GDI continue even warmed up

Cold start, note scale 4x



Warm start, the two GDIs switch order



Department of Mechanical Engineering

Zhang, S. and McMahon, W., "Particulate Emissions for LEV II Light-Duty Gasoline Direct Injection Vehicles," SAE Int. J. Fuels Lubr. 5(2):637-646, 2012, doi:10.4271/2012-01-0442.