Particle number measurements: How small should we go?

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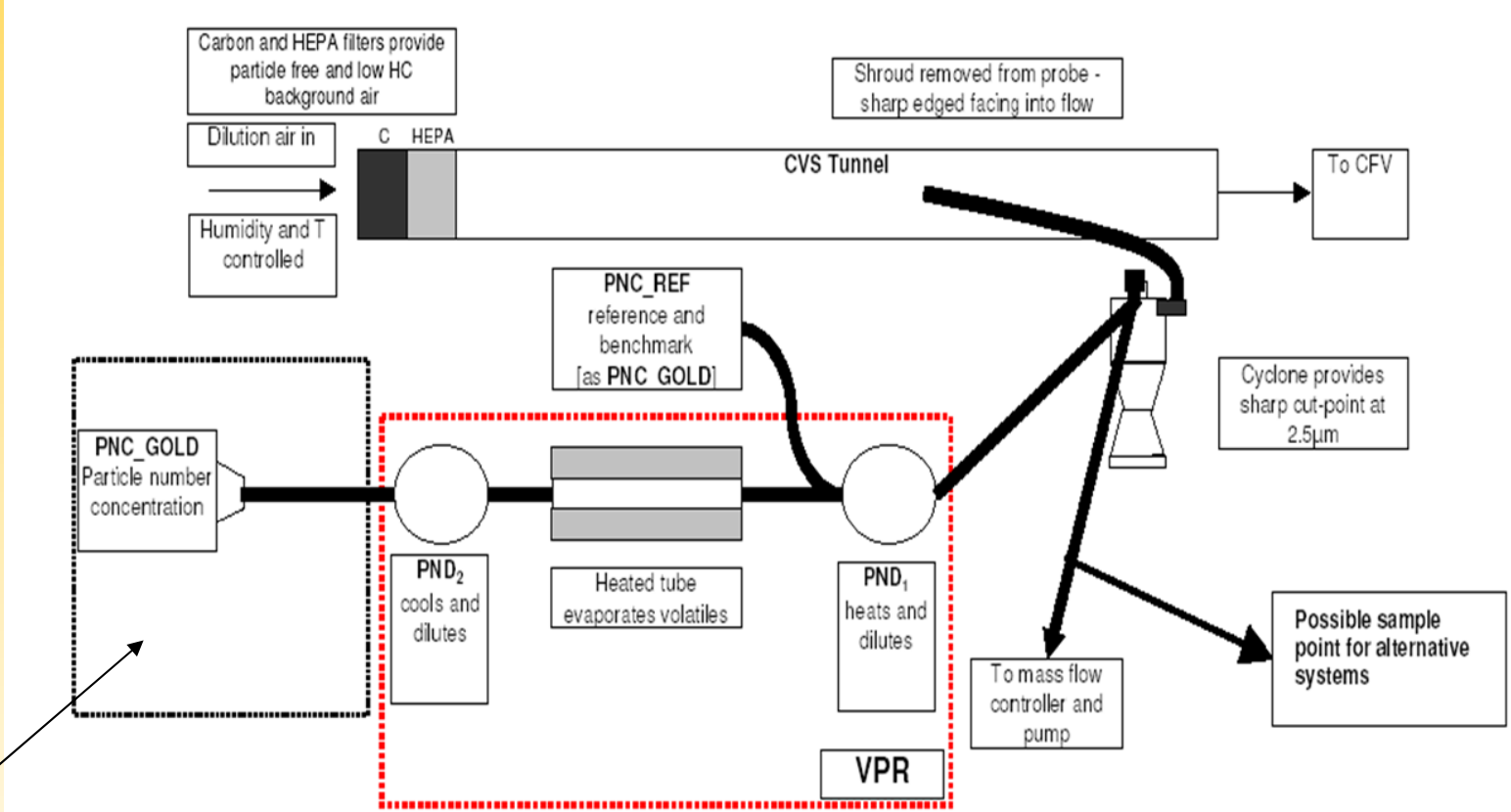
Solid particle number emissions

- In the EU “solid” particles larger than 23 nm from Diesel and SI engines (ground vehicles) are regulated
- Many engine technologies emit particles smaller than 23 nm
- “Solid” (BC) particle mass and “solid” particle number larger than 10 nm from aircraft turbine engines will be regulated worldwide starting in 2020
- Should the lower size cutoff for “solid” particle number emissions from ground vehicles be lowered to 15 nm, 10 nm, or lower?

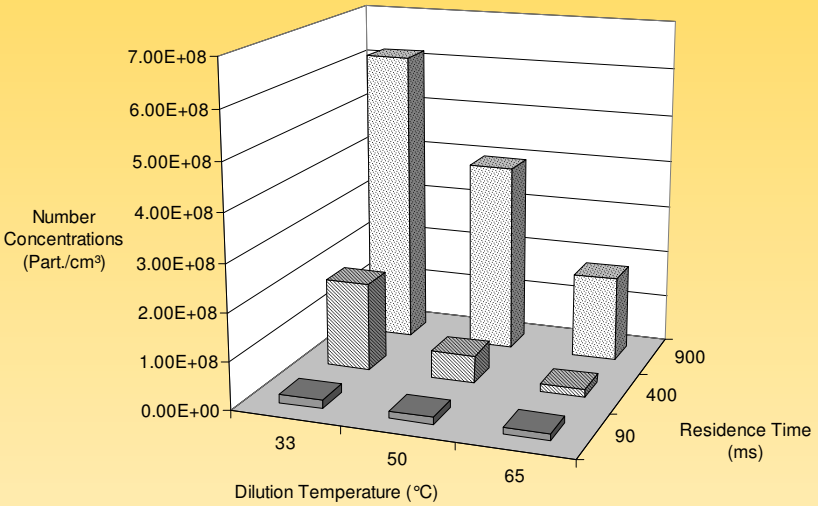
PMP “solid” number measurement

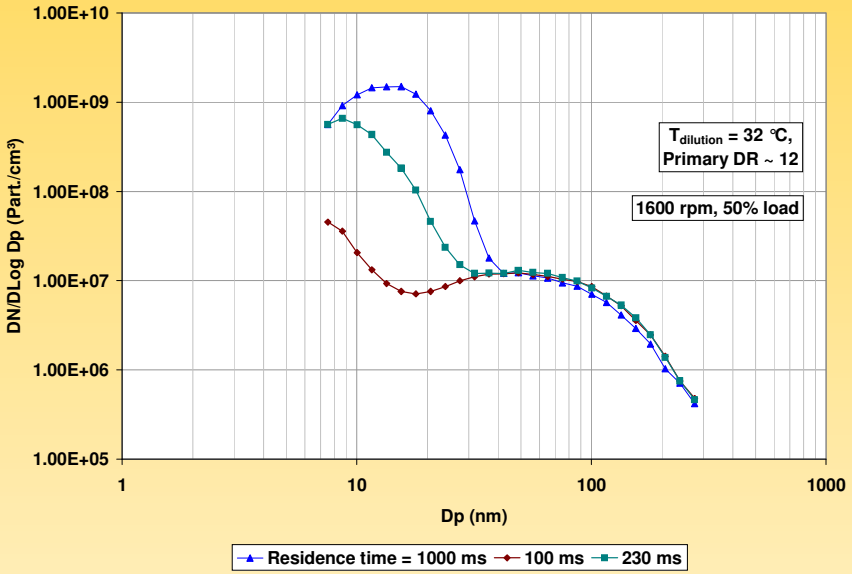


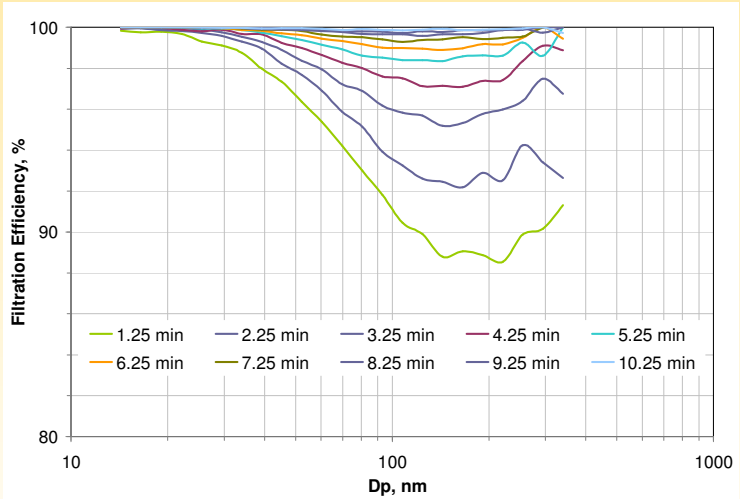
A specially designed CPC with a lower size cutoff (50%) of 23 nm is used

Solid particles are defined as those measured with a 23 nm cut size CPC in a diluted exhaust stream that has passed through a heated diluter and a volatile particle remover (VPR). It is an operational definition.

Why solid, why only larger than 23 nm?



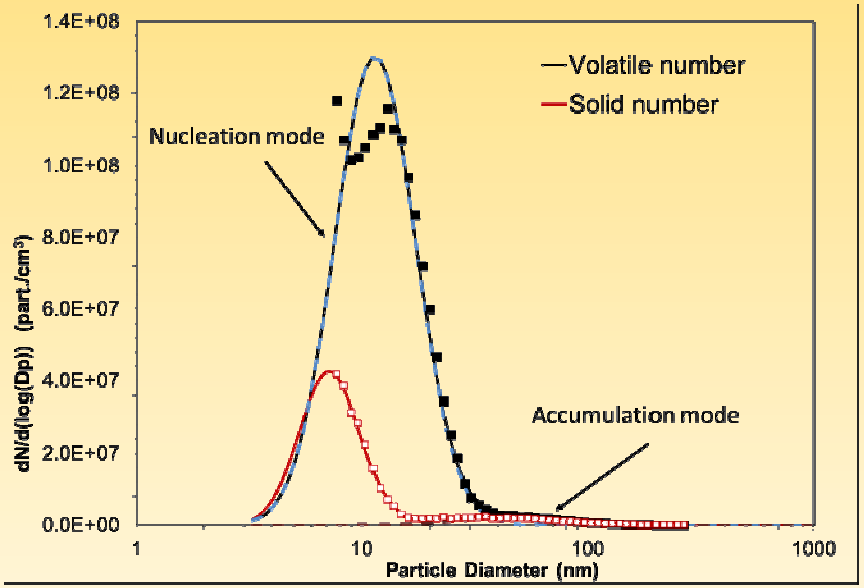




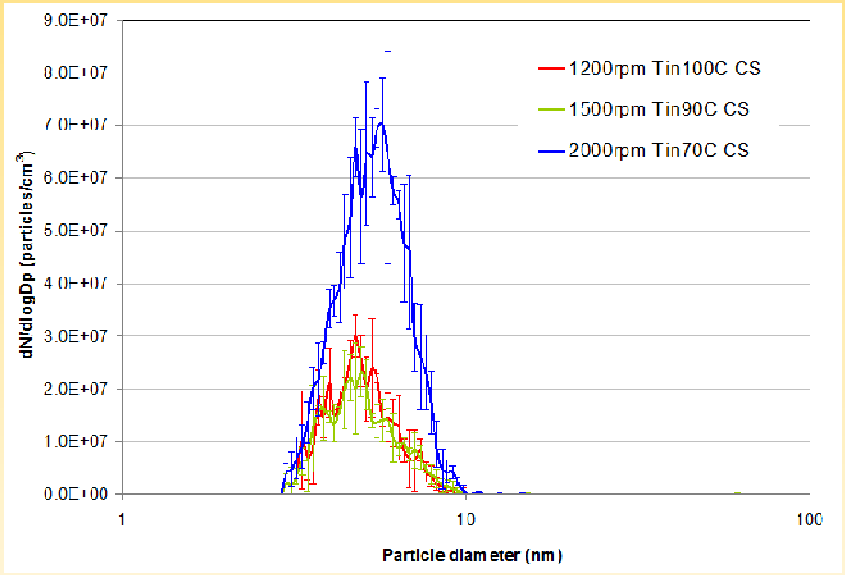
- The concentration of volatile nucleation mode particles is very dependent on sampling conditions
- Most of these particles are smaller than 23 nm
- If the engine is fitted with a particle filter, particles below 50 nm or so are very effectively removed
- Thus regulating solid particles above 23 nm is effectively regulating all particles for a trap equipped engines
- ***Without a trap the story is different***

Many current and advanced technology engines emit many, sometimes nearly all, solid particles below 23 nm

Interim tier IIIB / tier IV engine designed for certification with SCR only, no DPF, light load

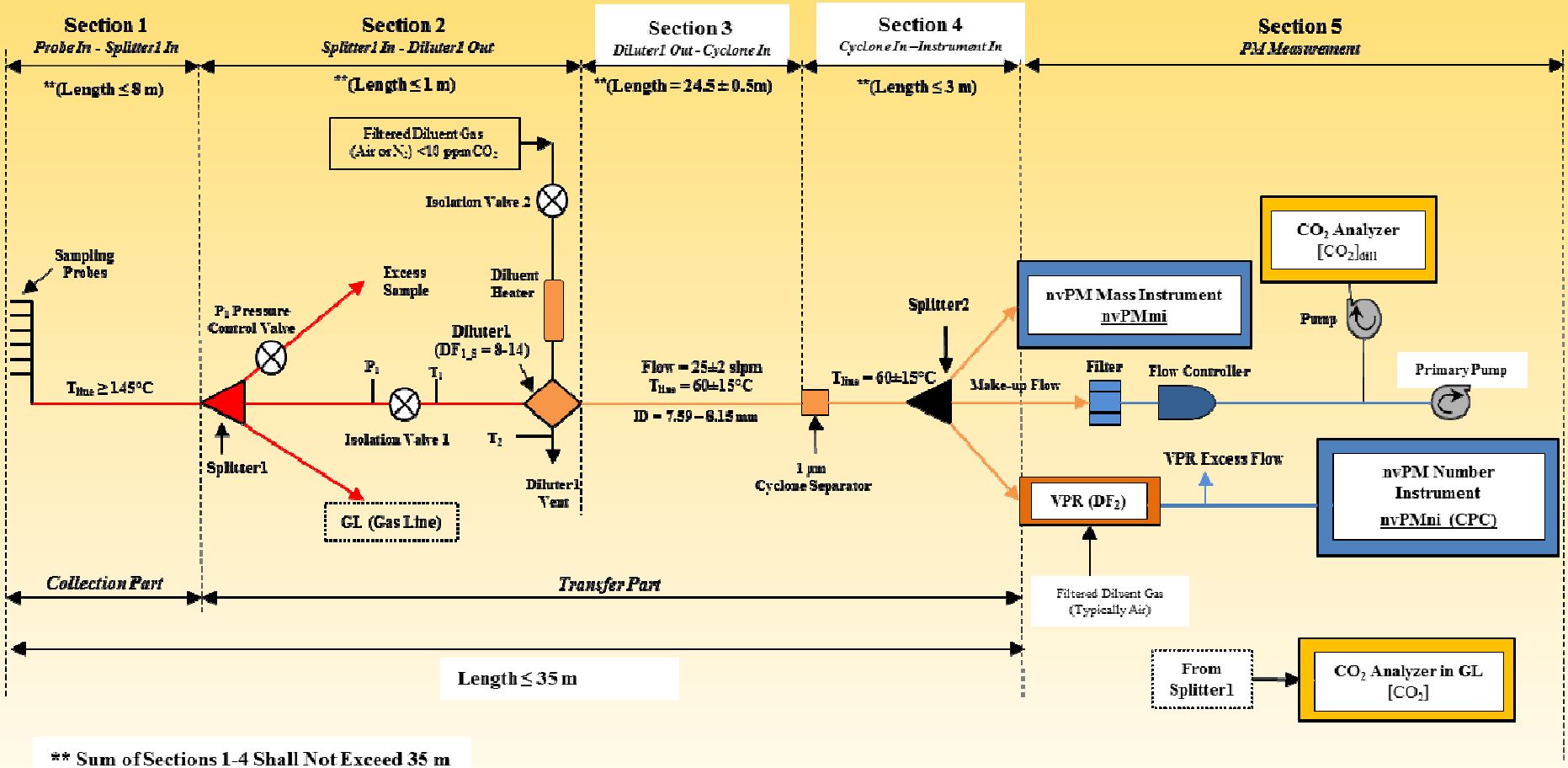


Medium-duty Diesel engine converted to operate in HCCI mode, no solid particles **above** 10 nm



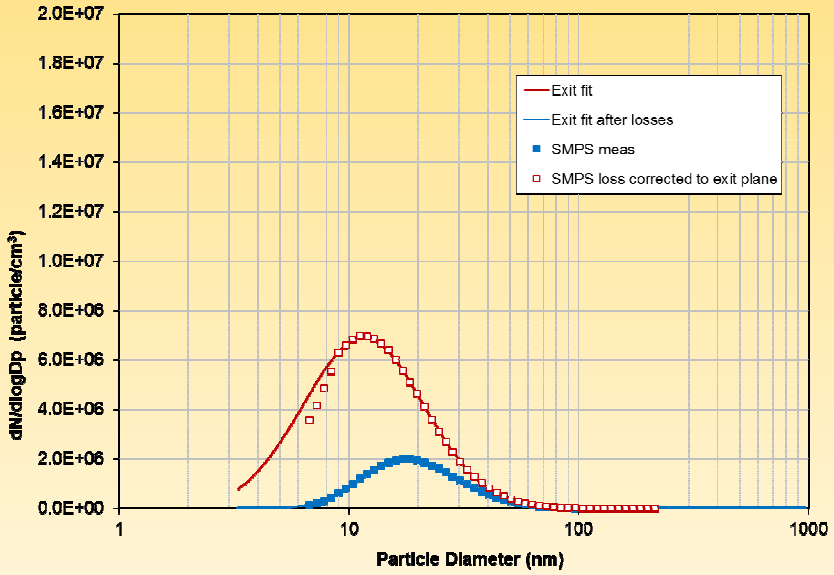
There are many other examples of modern engines without exhaust filters that emit many solid particles smaller than 23 nm, e.g., both PFI and GDI, and with fuels like CNG, LNG, DME..

Recommended aircraft sampling line configuration

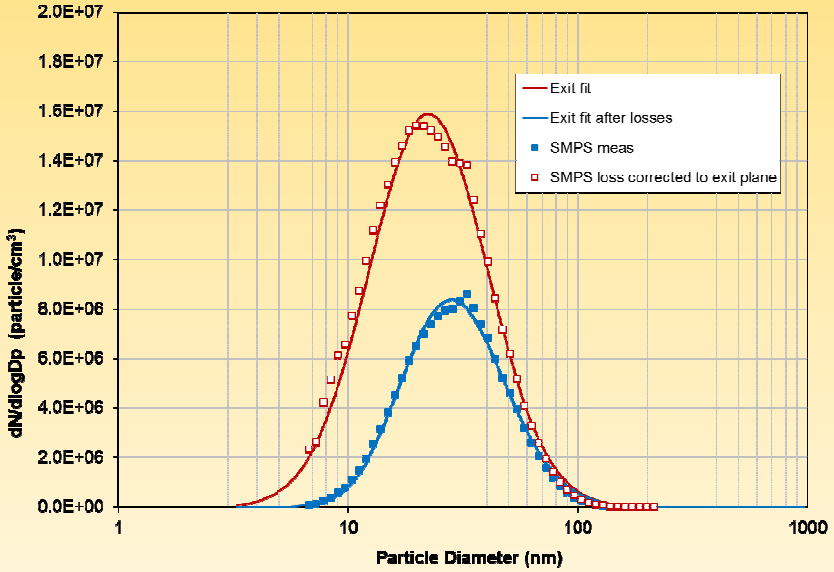


Jet engine aircraft exhaust particle size distributions, very small particles, mainly EC at measurement point

Turbojet, low thrust, Jet A fuel, exit plane DGN, 12 nm, measured DGN,19 nm, FacN = 6.6



Turbojet, high thrust, Jet A fuel, exit plane DGN, 22 nm, measured DGN, 29 nm, FacN = 3.7



It is very hard to put particle measuring instruments near the exhaust, imagine, the GE90, 120,000 lbf thrust, exhaust at ~900 K, Mach 1. Thus very long sample lines must be used and large corrections for sampling loss must be made. FacN = $N_{exit\ plane}/N_{measured}$

Number measurement sampling systems for aircraft jet engines and for ground vehicles (Diesel, SI)

Aircraft

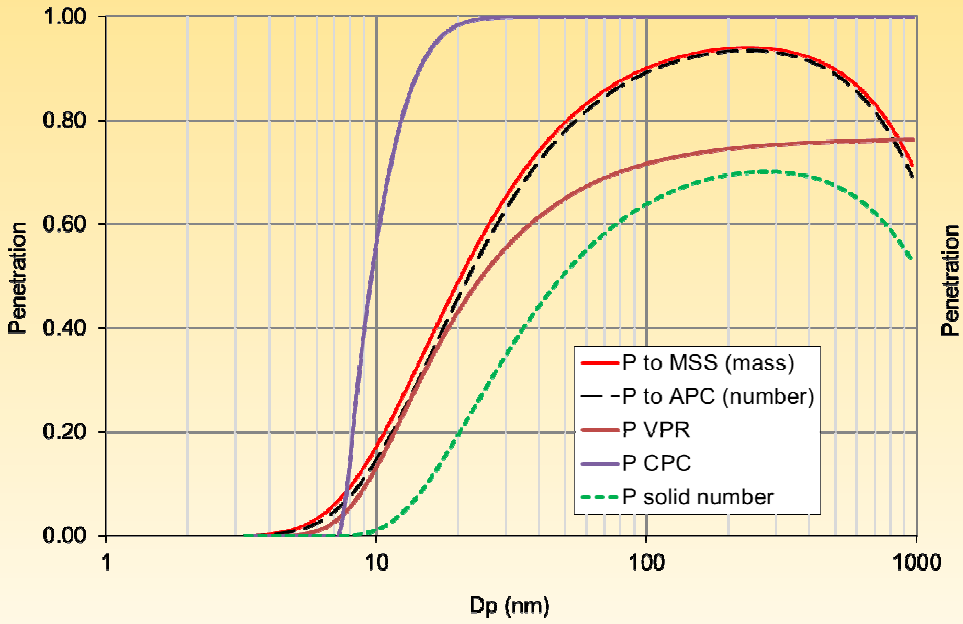
- Sampling from exhaust streams, T may be > 900 K, often near Mach 1
- Undiluted heated sampling lines ~ 8 m at 160 C
- Dilute ~ 10:1
- Heated sample line to instruments: 25 m at 60 C
- Mass instruments measure black carbon ~= elemental carbon
- For number measurements must remove all semi-volatiles, only measure above 10 nm
  - VPR, volatile particle remover
  - CPC, condensation particle counter with 50% cut at 10 nm
- Requires size dependent loss correction for both mass and number

Ground vehicle

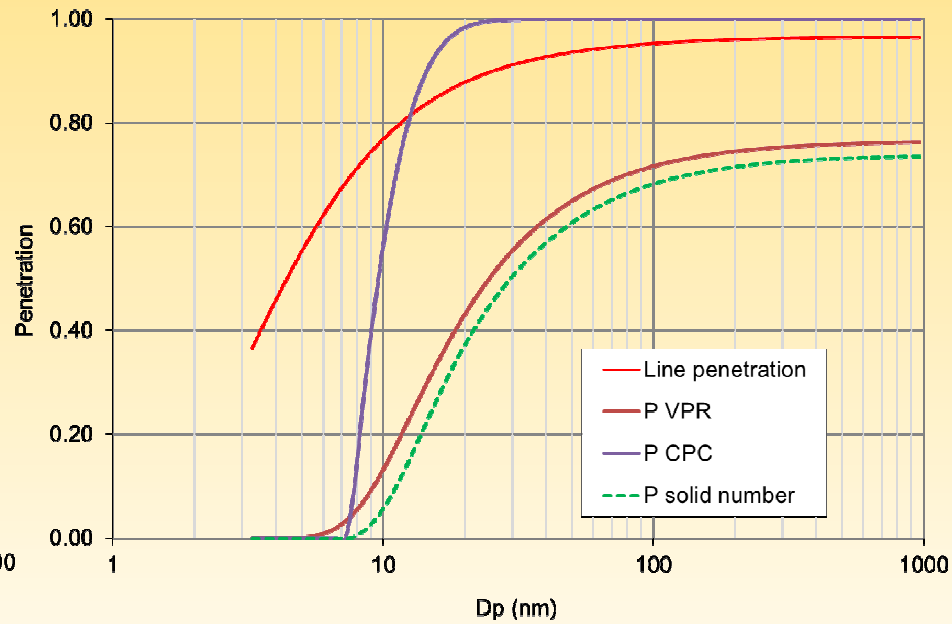
- Sampling from diluted steam in CVS tunnel modest temperatures and velocities
- Relatively short line to filters, instruments ~ 3 m
- Gravimetric filter mass
- For number measurements must remove all semi-volatiles, only measure above xx nm
  - VPR, volatile particle remover
  - CPC, condensation particle counter with 50% cut at ?? nm
- Surely with these short lines we won't require loss correction … or will we?

Sampling line, VPR and CPC particle penetration: UTRC loss model, actual CPC (10 nm cut) and VPR data

Typical aircraft jet engine sampling system

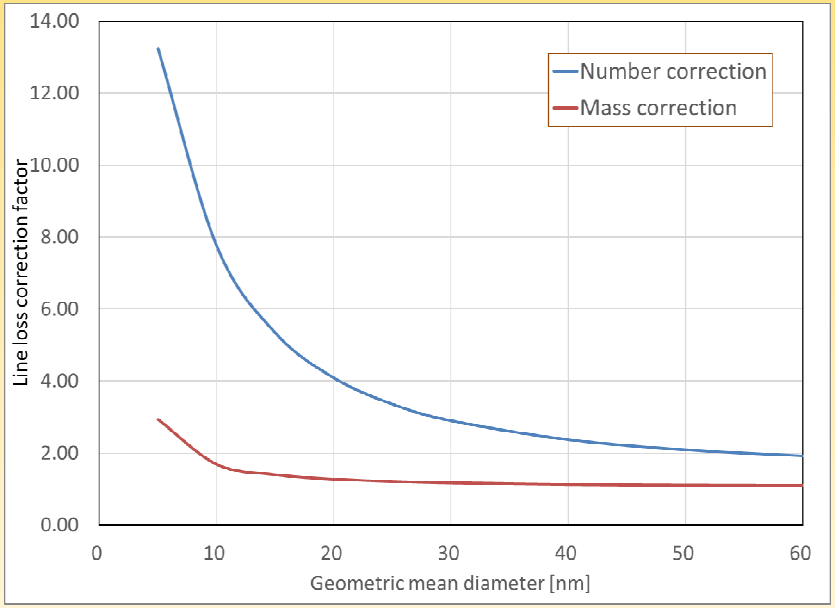


Assumed ground vehicle sampling system

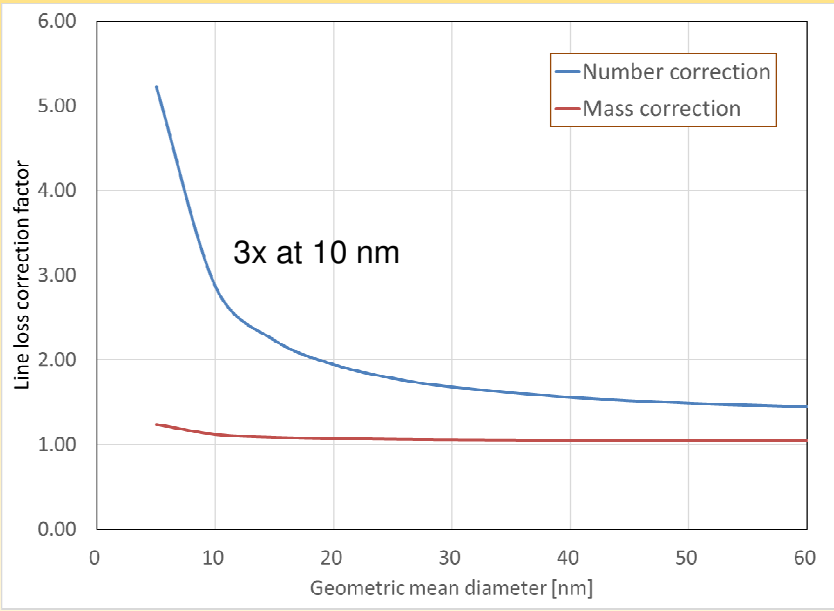


Sampling loss corrections associated measuring very small particles

Typical aircraft jet engine sampling system



Assumed ground vehicle sampling system



- Significant size dependent loss corrections necessary for solid particle number measurements, even with relatively short sampling lines that would likely be used for ground vehicle engine testing
- Why? Mainly due to losses in volatile particle remover
- Mass correction small, no VPR, CPC

Aircraft line loss method: assumptions and approach

- The exit plane solid size distribution is lognormal
- The exit plane particle density, ρ_p, and geometric standard deviation, σ_g, are known
- The size dependent penetration through the sample line, volatile particle remover, and CPC are known
- Solid mass and number are measured at the end of the sampling system and N/m is determined
  - The N/m ratio is a well defined function of the line losses, ρ_p, σ_g, and the geometric mean diameter, DGN.
  - All the above values except DGN and known or assumed so we solve for DGN
  - DGN, σ_g, ρ_p, and the system loss functions allow the number and mass loss correction factors to be determined
- Critical assumptions
  - Lognormal and the exit plane
  - Known σ_g and ρ_p
  - N and m are measured simultaneously with similar time response
  - Any semi-volatile material present does not change the line solid particle line losses

Ground vehicle loss method: assumptions and approach?

- The exit plane solid size distribution is lognormal – likely to be bimodal?
- The exit plane particle density, ρ_p, and geometric standard deviation, σ_g, are known – maybe, problem if bimodal? Would take lots experience
- The size dependent penetration through the sample line, volatile particle remover, and CPC are known, OK
- Solid mass and number are measured at the end of the sampling system and N/m is determined – solid m not measured filter only – alternatives:
  - Determine nonvolatile filter mass but not real time? Likely no
  - Measure BC mass as with aircraft but ash interferes? Likely no
  - Measure solid active surface, S, with CS plus diffusion charger? Possible but sensitivity might be issue.

Ground vehicle loss method: possible approaches:

- Measure solid active surface, S, with CS plus diffusion charger
  - The N/S ratio is a well defined function of the line losses, ρ_p, σ_g, and DGN.
  - All the above values except DGN and known or assumed so we solve for DGN
  - DGN, σ_g, ρ_p, and the system loss functions allow the number and mass loss correction factors to be determined
  - Critical assumptions
    - Lognormal and the exit plane – aircraft work suggest small error due to bimodal
    - Known σ_g and ρ_p
    - N and S are measured simultaneously with similar time response
    - Any semi-volatile material present does not change the line solid particle line losses.
- Use PCRF as in current PMP?
  - Large losses at 10 nm ~ x3 make this extreme compromise
  - Undercount small particles, overcount large
- Measure downstream size distribution with real time particle sizer
  - Works with any size distribution, density
  - Expensive instruments
  - Sensitivity could be issue
  - Semi-volatile particles could bias results

Ground vehicle loss method: Instruments

- CPCs with 50% cut at 23 and 10 nm available
- Most CPCs with 50% cut below 10 nm use internal flow splits complicating calibration
- Many varieties of low cost diffusion chargers available but sensitivity might be issue
- Fast response sizing, EEPS, DMS, available but expensive and sensitivity could be issue
- PMP Evaporation tube may not adequately suppress nucleation for particles 10 nm or smaller. Likely need to use catalytic stripper like that used for aircraft

Ground vehicle loss method: Conclusions

- Moving lower counting limit to 10 nm or below will be challenging and possibly more difficult than for aircraft
- Several approaches possible but adding downstream size measurement would be by far the most accurate approach, but expensive

Acknowledgements - VARIAnT 1 and 2 Test Campaign Participants

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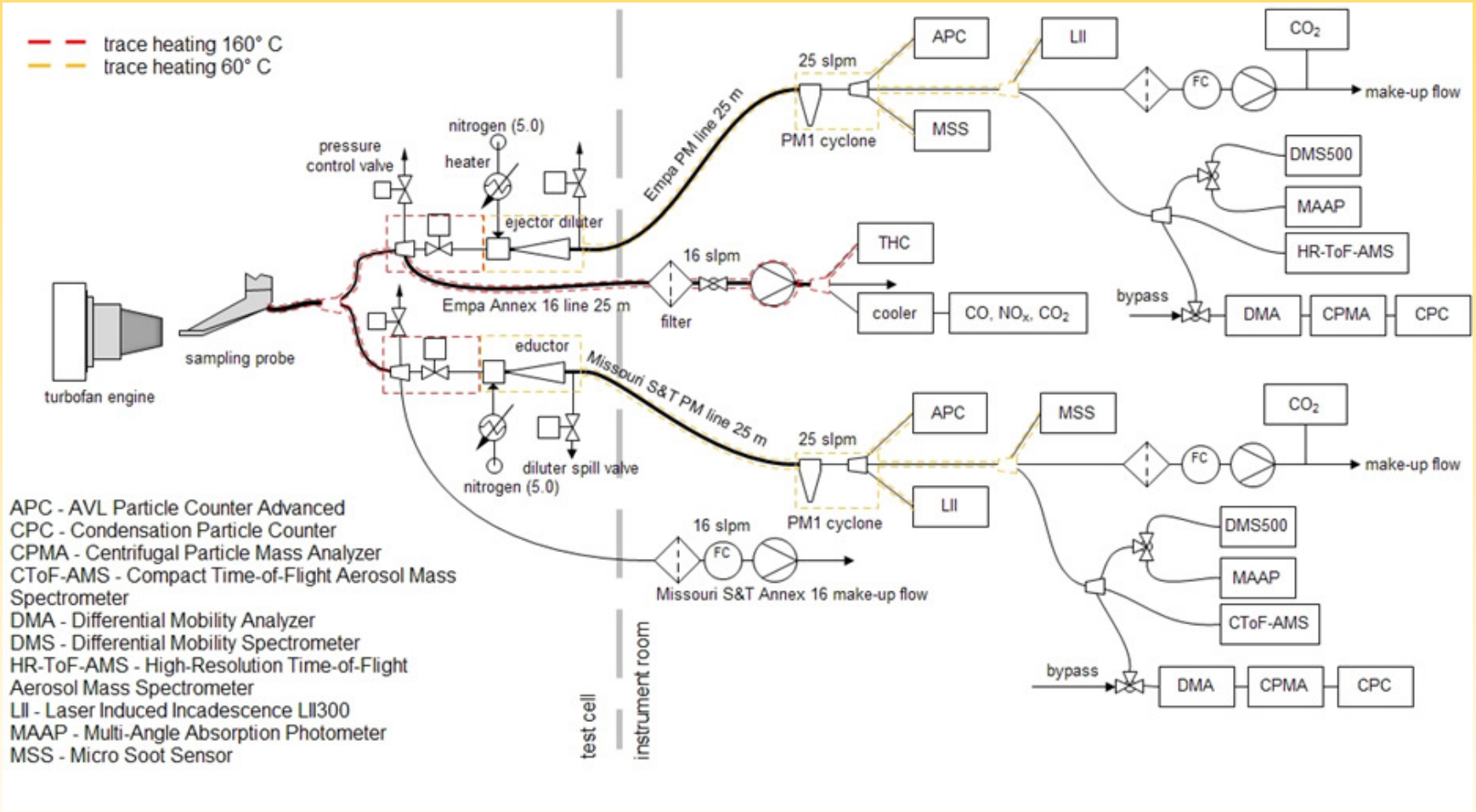
Variable Response in Aircraft nvPM Testing (VARIAnT): Sponsored by U. S. Environmental Protection Agency and Conducted Jointly by EPA and AEDC

Thank you - Questions

Extra slides

Detailed aircraft sampling line configuration



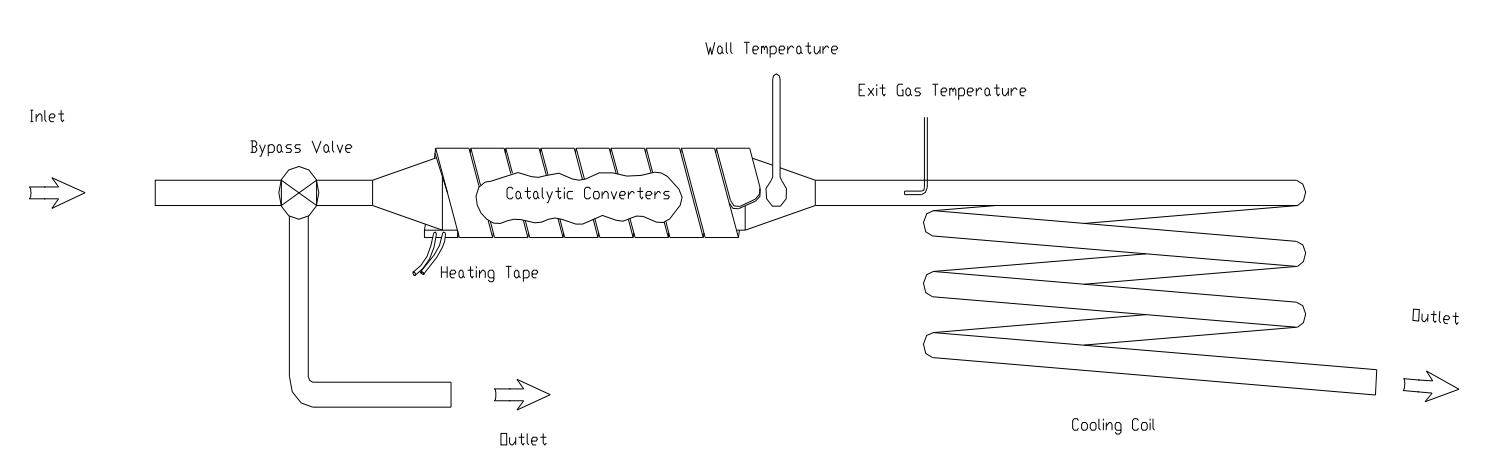
From: Lobo, et al., Aerosol Science and Technology, 49:472–484, 2015

Mass and number emission standards

The EU has set a number based emission standards for light and heavy duty Diesel vehicles

- The standards are based on “solid” particles larger than 23 nm
- Light-duty, Euro 5b/6, September 2011/2014
  - The standard is 6×10^{11} particles/km
  - The mass emission standard is 4.5 mg/km, but the number standard corresponds to about 0.15 to 0.7 mg/km, depending on DGN – a much tighter standard!
  - An interim standard of 6×10^{12} has been set for gasoline vehicles, through 2017, after that they must meet diesel standard
  - US/CARB standards are still mass based – 2017: 1.8 mg/km, 2025: 0.6 mg/km
- Heavy-duty, Euro VI, January 2013
  - The standards are 6×10^{11} and 8×10^{11} particles/kWh on the WHTC and the WHSC, respectively
  - The mass emission standard is 10 mg/kWh, but the number standard corresponds to about 0.2 to 0.9 mg/kWh, depending on DGN – again a much tighter standard!
- Meaningful filter mass measurements are very difficult at levels corresponding to these number standards
- CARB 2025 light-duty standard of 0.6 mg/km may be difficult to measure by traditional filter sampling but corresponds to 5×10^{11} to 3×10^{12} particles/km, easily measured

The University of Minnesota alternative to the evaporation tube VPR: the catalytic stripper (CS)



- Our strippers consists of a 2 substrate catalyst (provided by Johnson-Matthey) 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

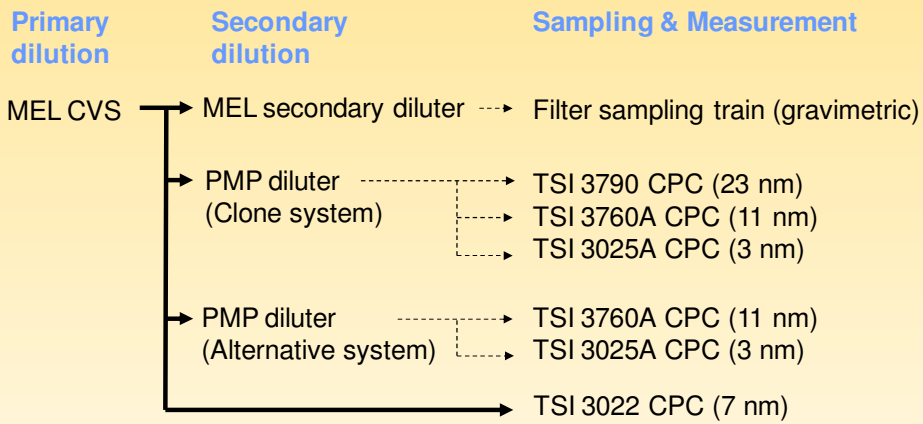
Kittelson, D. B., W. F. Watts, J. C. Savstrom, J. P. Johnson, 2005. "Influence of Catalytic Stripper on Response of PAS and DC," Journal of Aerosol Science 36 1089–1107.

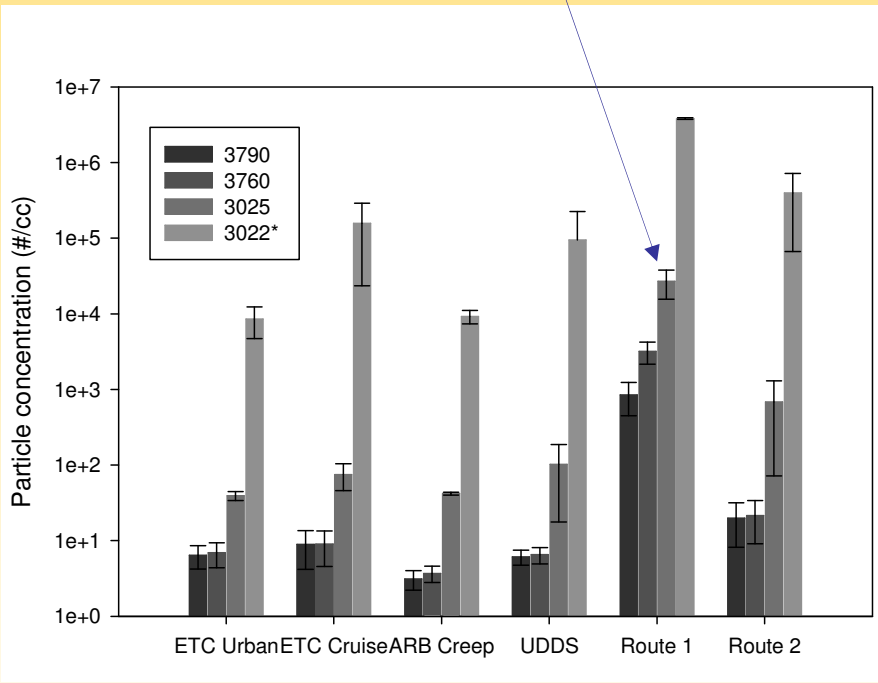
Swanson, Jacob and David Kittelson, 2010. Evaluation of thermal denuder and catalytic stripper methods for solid particle measurements, Journal of Aerosol Science, Volume 41, Issue 12, Pages 1113-1122.

On road tests using PMP protocol show unexpected “solid” particles many below 23 nm

A heavy-duty truck equipped with a CRT was tested on road and on a chassis dynamometer

- It showed large concentrations of “solid” particles below 23 nm at high load conditions
- These conditions favor sulfate particle formation.
- Filtration efficiency for particles below 23 nm is very high.



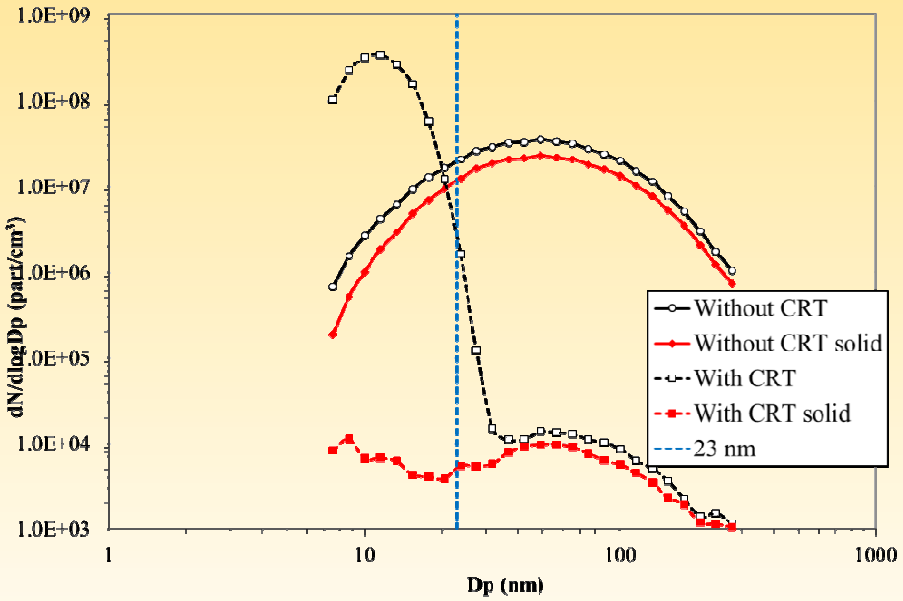


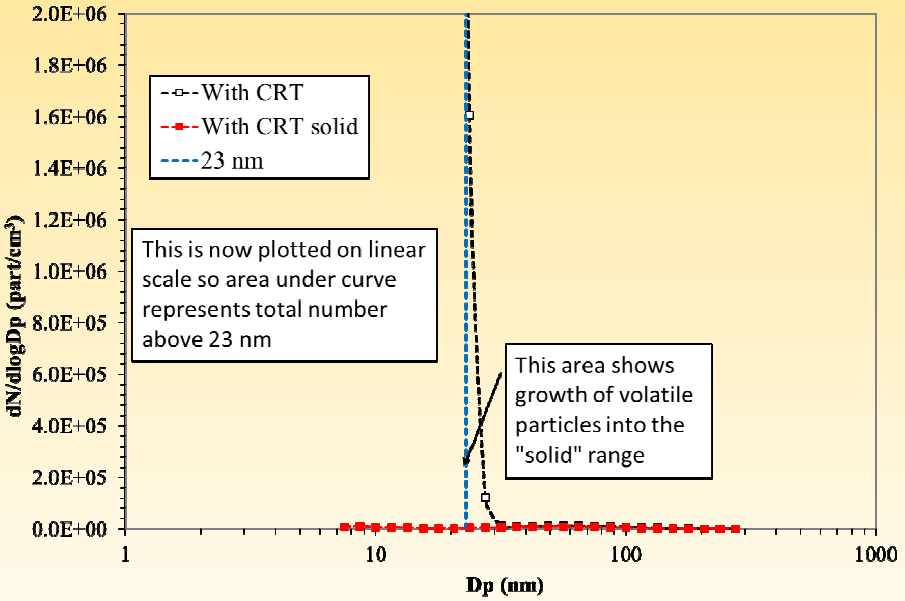
Johnson, Kent C., Thomas D. Durbin, Heejung Jung, Ajay Chaudhary, David R. Cocker III, Jorn D. Herner, William H. Robertson, Tao Huai, Alberto Ayala, and David Kittelson, 2009. Evaluation of the European PMP Methodologies during On-Road and Chassis Dynamometer Testing for DPF Equipped Heavy Duty Diesel Vehicles, Aerosol Science and Technology, 43:962–969, 2009.

Under high load conditions a catalyzed soot filter may produce a large sulfuric acid mode

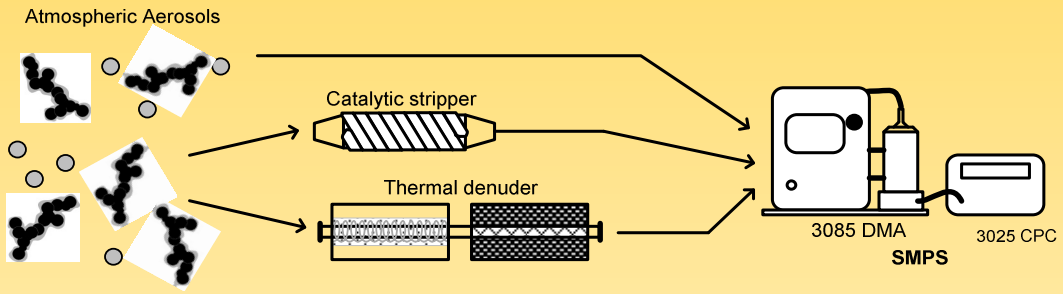
Cummins 2004 ISM engine, BP 50 fuel, AVL mode 8, Total and solid particles with and without CRT

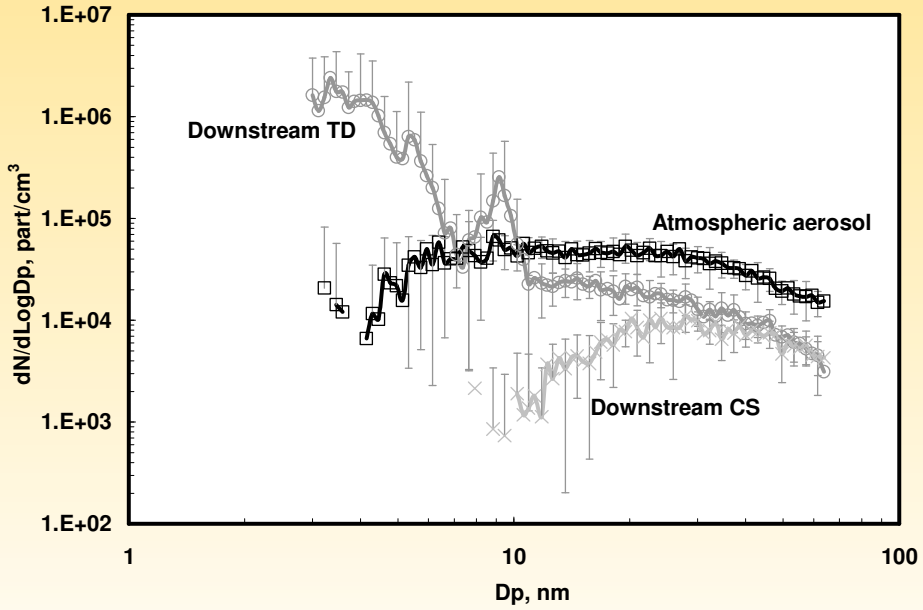
Here I have switched to a linear scale to show breakthrough of semi-volatiles might bias "solid" N





Evaporation of semi-volatiles without total removal may re-nucleate particles





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