

ON-ROAD EXPOSURE AND EMISSION MEASUREMENTS

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We gratefully acknowledge support of on-road exposure by USEPA STAR
program grant #R827354, original construction of Mobile laboratory CRC, DOE,
E-43 Program, and donation of transport vehicle by Volvo Truck

ABSTRACT

The University of Minnesota's mobile emission laboratory (MEL) has been used to characterize on-road particle exposures and to determine engine emission factors under real world conditions. Particles emitted by modern Diesel and SI engines do not all form during combustion. Many particles, particularly those in the 3 to 30 nm diameter (nuclei mode) range, form from volatile materials as the exhaust dilutes and cools in the atmosphere. These particles may constitute 90% or more of the nanoparticle (< 50 nm) number emissions. Unfortunately, the formation of these particles is a nonlinear, gas-to-particle nucleation process that is extremely dependent upon dilution conditions. These conditions are difficult to simulate in the laboratory. Two types of experiments have been performed. In the first, the MEL was driven on an urban and rural route and, to the extent possible, on-road plumes from Diesel powered heavy-duty trucks were sampled. These measurements should give a good indication of the aerosol exposure of cross country truck drivers.

In the second type of experiment, a new sampling system was installed on the MEL that made it possible to sample the Mel's diluted exhaust plume. A 2000 model year engine that provides a particle signature, which is characteristic of modern engines, powers the MEL. This enabled us to study real world emissions from a modern engine under a variety of atmospheric conditions. We describe the characteristics of the aerosols observed these experiments along with the instrument array, calibration, particle losses, sampling artifacts, ambient conditions, traffic conditions, and sampling locations or routes. Using a carbon balance method, real world particle emission factors for heavy-duty Diesel engines have been determined. The instrument suite includes an SMPS to size particles in 9 to 300 nm size range, a UCPC with a lower limit 50% counting efficiency at 3 nm, instruments to measure total submicron particle surface area, a PAS to measure aerosol photoemission response, CO₂, CO, and NO_x analyzers, and a thermal denuder to distinguish between solid and volatile particles.

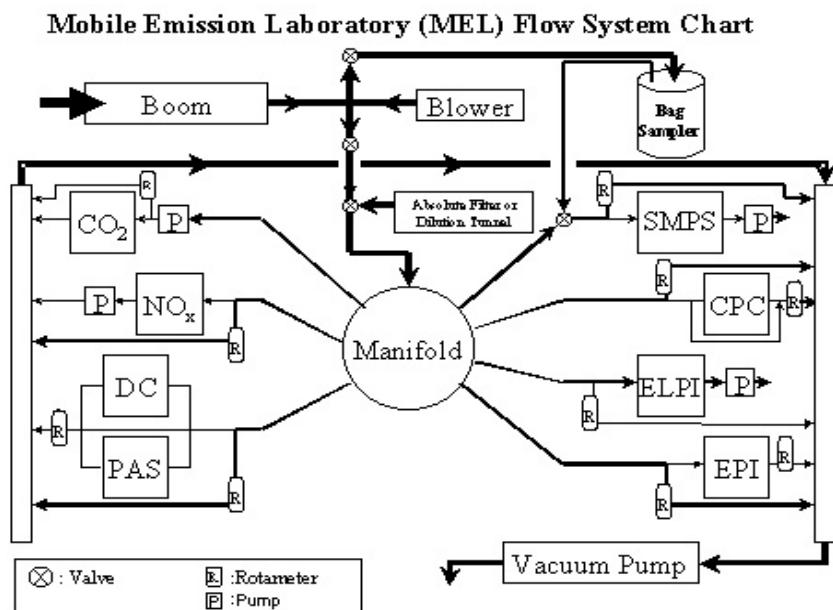
Exposure study goals and objectives

- Provide a stable platform for exposure experiments under real world conditions
 - Wanted high concentrations of nanoparticles in the nuclei mode
 - On road aerosol concentrations in the nuclei mode region greatly exceed roadside values and may equal or exceed those from particle concentrators without introduction of artifacts
- Two exposure studies will be discussed
 - The fall 2002 study used on-road aerosols from mixed Diesel traffic on a rural/urban freeway in New York State for exposure
 - The fall 2003 study used the naturally diluted plume of our mobile emissions laboratory for exposure
- Examine the relationships between different metrics of particle exposure under real world conditions
- Determine on-road emission factors
- Only physical measurements described here
 - Additional information on fall 2002 study available from Kittelson et al., 2004; Elder et al., 2004; both in special issue of Inhalation Toxicology)
 - Analysis of fall 2003 data in process

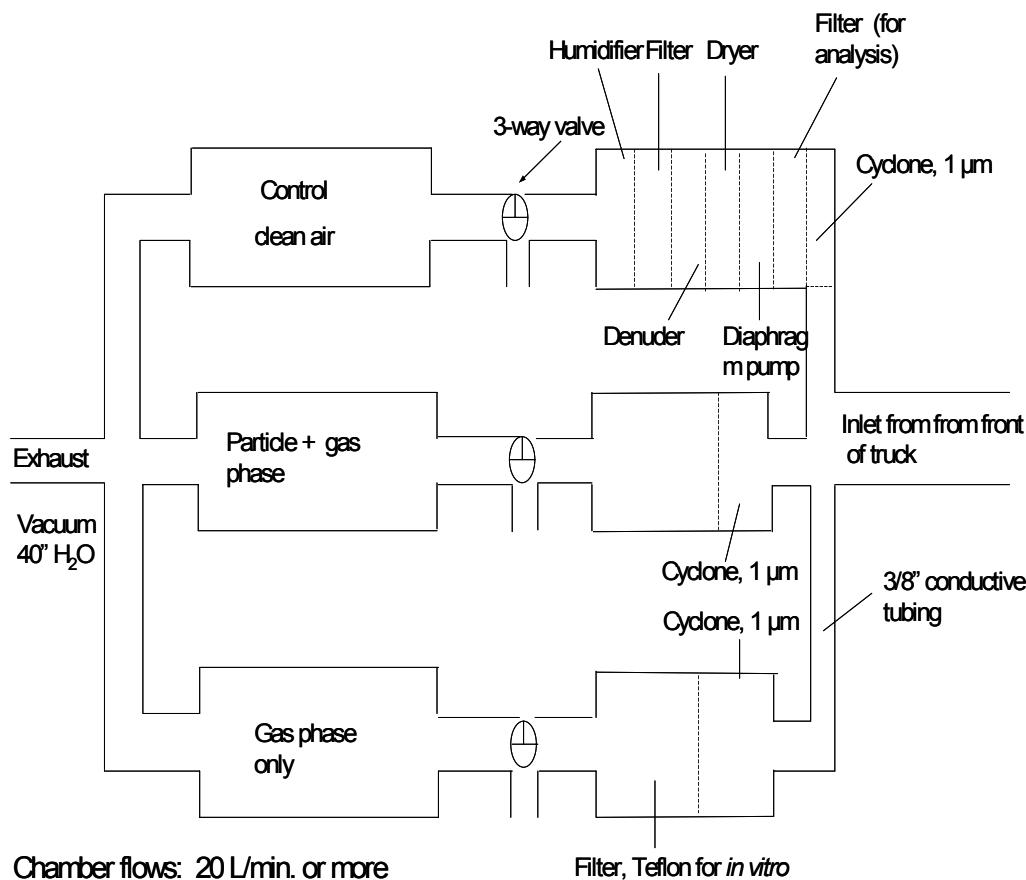
U of M Mobile Laboratory built to study real-world formation of nanoparticles for CRC E-43 project



- Instruments (primary instruments highlighted in blue)
 - SMPS to size particles in 9 to 300 nm size range
 - ELPI to size particles in 30 to 2500 nm size range
 - CPC to count all particles larger than 3 nm
 - Diffusion Charger to measure total submicron particle surface area
 - PAS to measure total submicron surface bound PAH equivalent
 - CO₂, CO, and NO analyzers for gas and dilution ratio determinations



Modifications to laboratory for exposure experiments



- Install exposure chambers in place of sampling bag
- Three exposure chambers provided with
 - Roadway aerosol
 - Filtered roadway aerosol
 - Clean temperature and humidity controlled air for control group
- Additional instruments – not in all studies
 - Aethalometer for black carbon measurements
 - TSI electrical aerosol detector (EAD) for particle length concentration measurements
 - Second SMPS with thermal denuder to measure solid fraction of on road particles
 - PM2.5 filter sampling for EC/OC measurements

Glossary

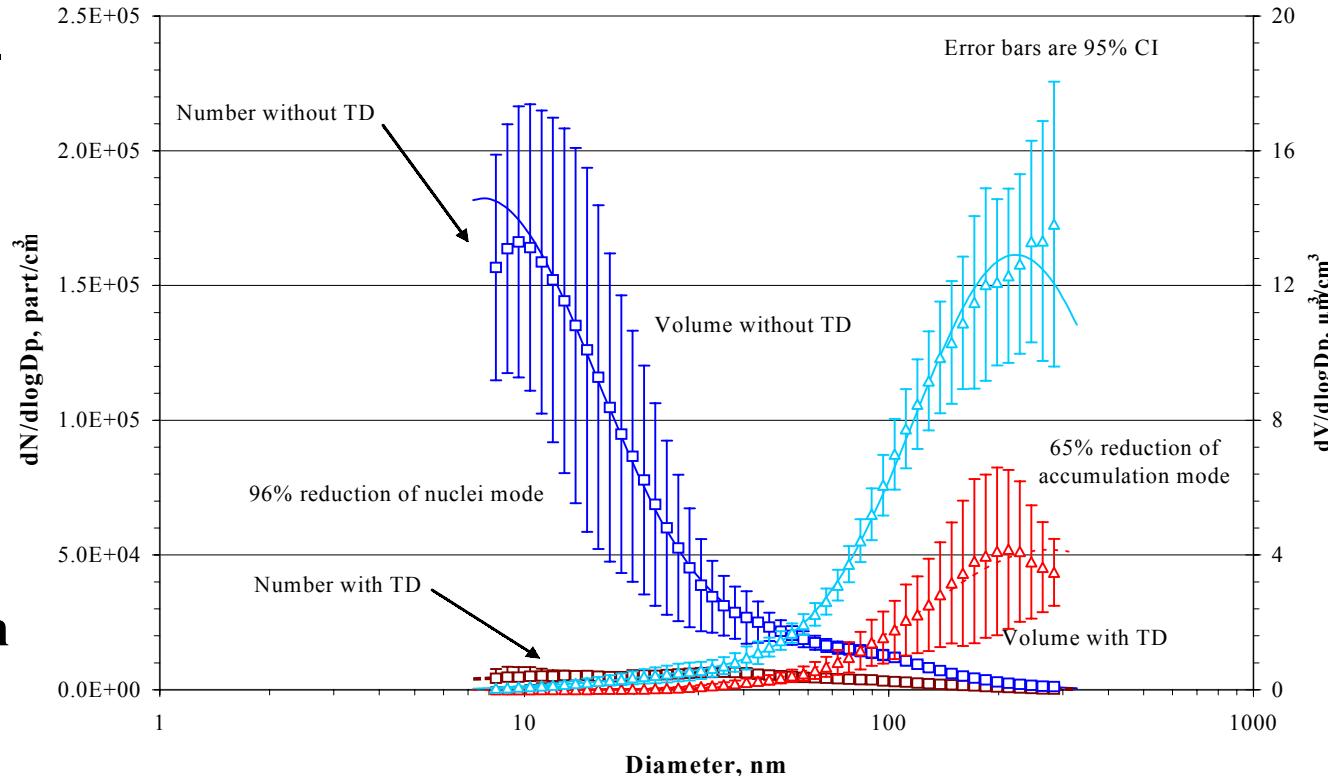
- V_{nuc} – fitted volume in nuclei mode, best estimate of nuclei mode (nanoparticle) volume or mass, $\mu\text{m}^3/\text{cm}^3$, ($1 \mu\text{m}^3/\text{cm}^3 = 1 \mu\text{g}/\text{m}^3$ for $\rho = 1 \text{ g}/\text{cm}^3$)
- V_{30} – volume of particles smaller than 30 nm measured by SMPS, estimate of V_{nuc}
- V – total submicron volume
- V_{acc} – fitted volume in accumulation mode
- SMPS N – total number of particles measured with SMPS with effective lower limit between 10 and 15 nm for SMPS as used here, particles/ cm^3
- CPC N – total number measured with CPC – all particles above about 7 nm for CPCs used here, particles/ cm^3
- Source – measurements made at entry to MEL
- Cage – measurements made in breathing zone of exposure chamber

Much of the on road aerosol is volatile, especially in the nuclei mode region

Overall average fall 2002 on-road data

- Sampled 6 hours per day for 10 days on New York State Throughway between Rochester and Buffalo
- Tried to drive in Diesel truck traffic and sample aerosol to which truckers are exposed

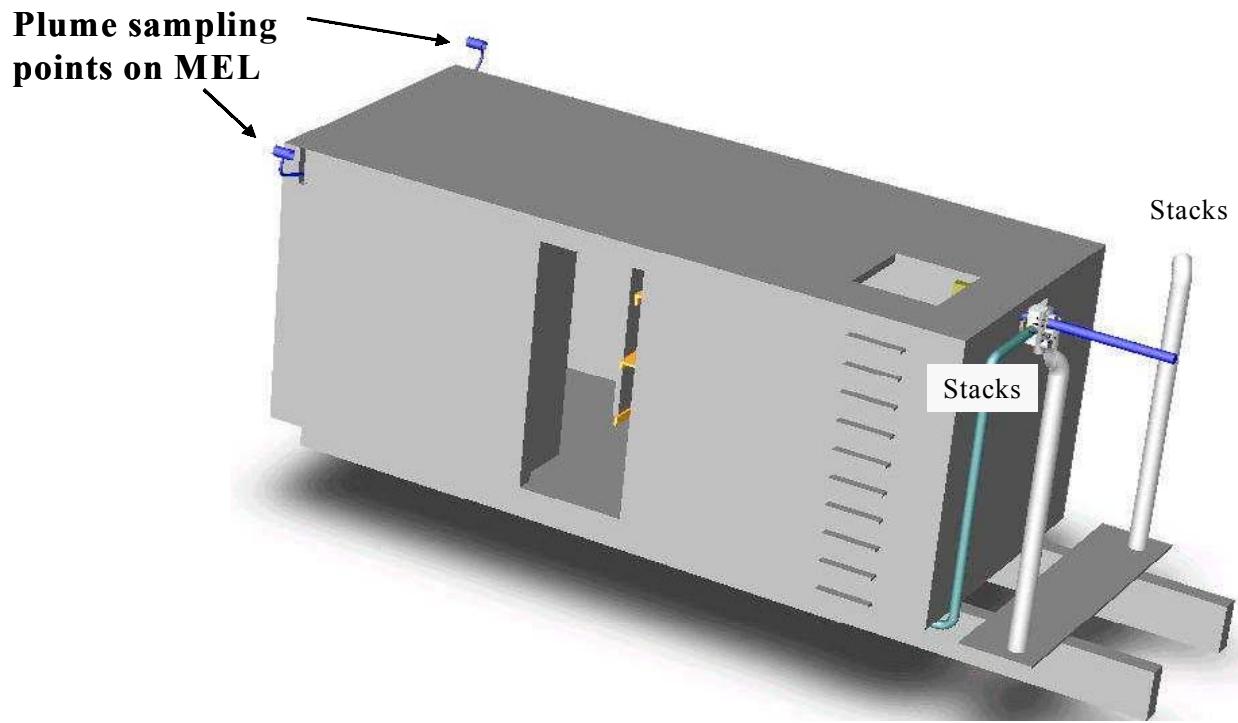
Clear bimodal structure with consistent formation of nuclei mode



Thermal denuder (TD) used to remove volatile particles

- 96% reduction in nuclei mode region
- 65% reduction in accumulation mode region
- Nuclei mode or nanoparticles are nearly all volatile – as expected from laboratory tests

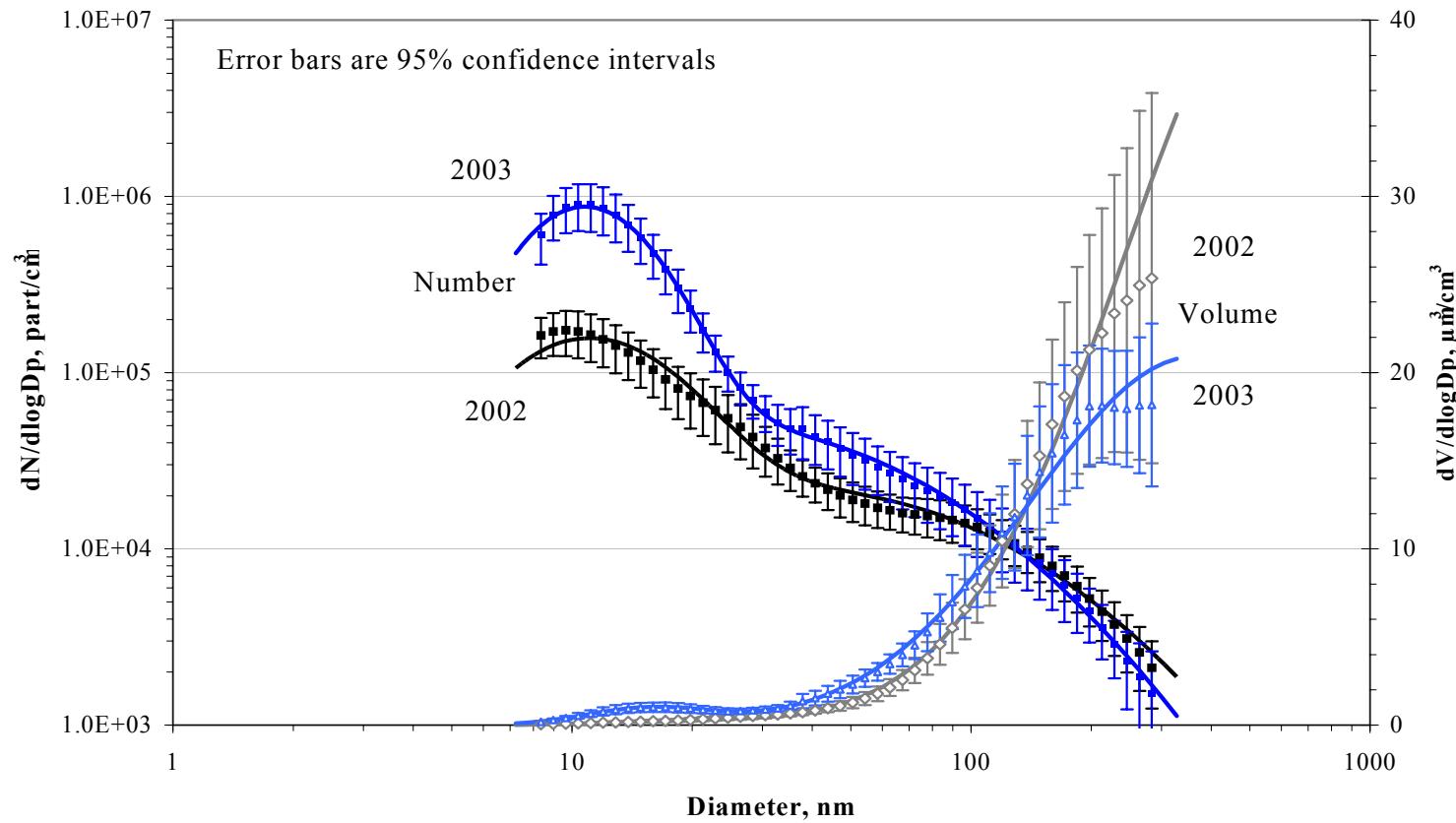
For the fall 2003 study we wanted higher and more consistent exposure levels



- MEL modified so that could sniff our own plume
- MEL is powered by modern Diesel engine meeting 2000 model year emission standards
- Sampling from rear of the MEL allowed natural atmospheric dilution to a dilution ratio of about 500
- This is high enough to freeze the size distribution for exposure experiments

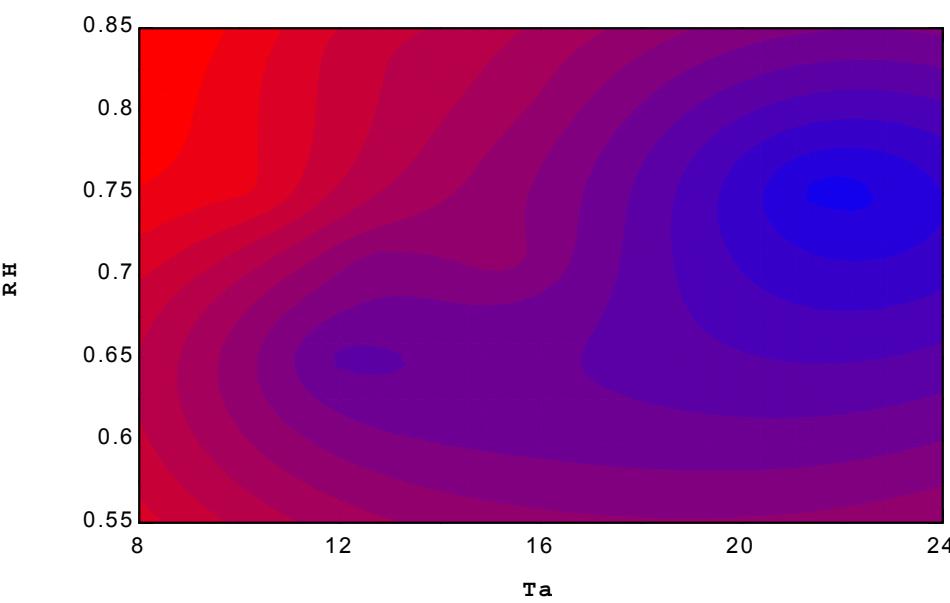
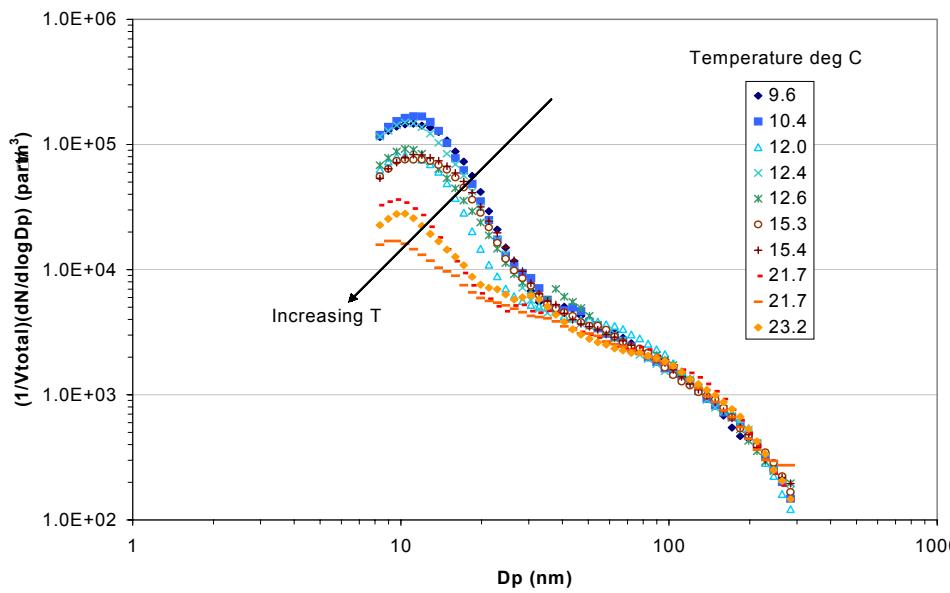
Overall average size distributions for 2002 and 2003 studies

Using the plume sniffing method led to reduced effective dilution ratios, higher exposure to ultrafine particles, and less daily variation in the shape of the size distribution and concentration of particles



- The number in the nuclei mode increased by a factor of 4.3
- The volume (mass) in the nuclei mode increased by a factor of 2.3
- Total volume (mass) above 100 nm decreased despite lower dilution because the engine had lower PM mass emissions than the on-road fleet of the 2002 study

Ambient temperature (mainly) and humidity influence size of nuclei mode



Results shown here are for the fall 2003 study and represent 60 hours of highway cruise operation on New York State Throughway

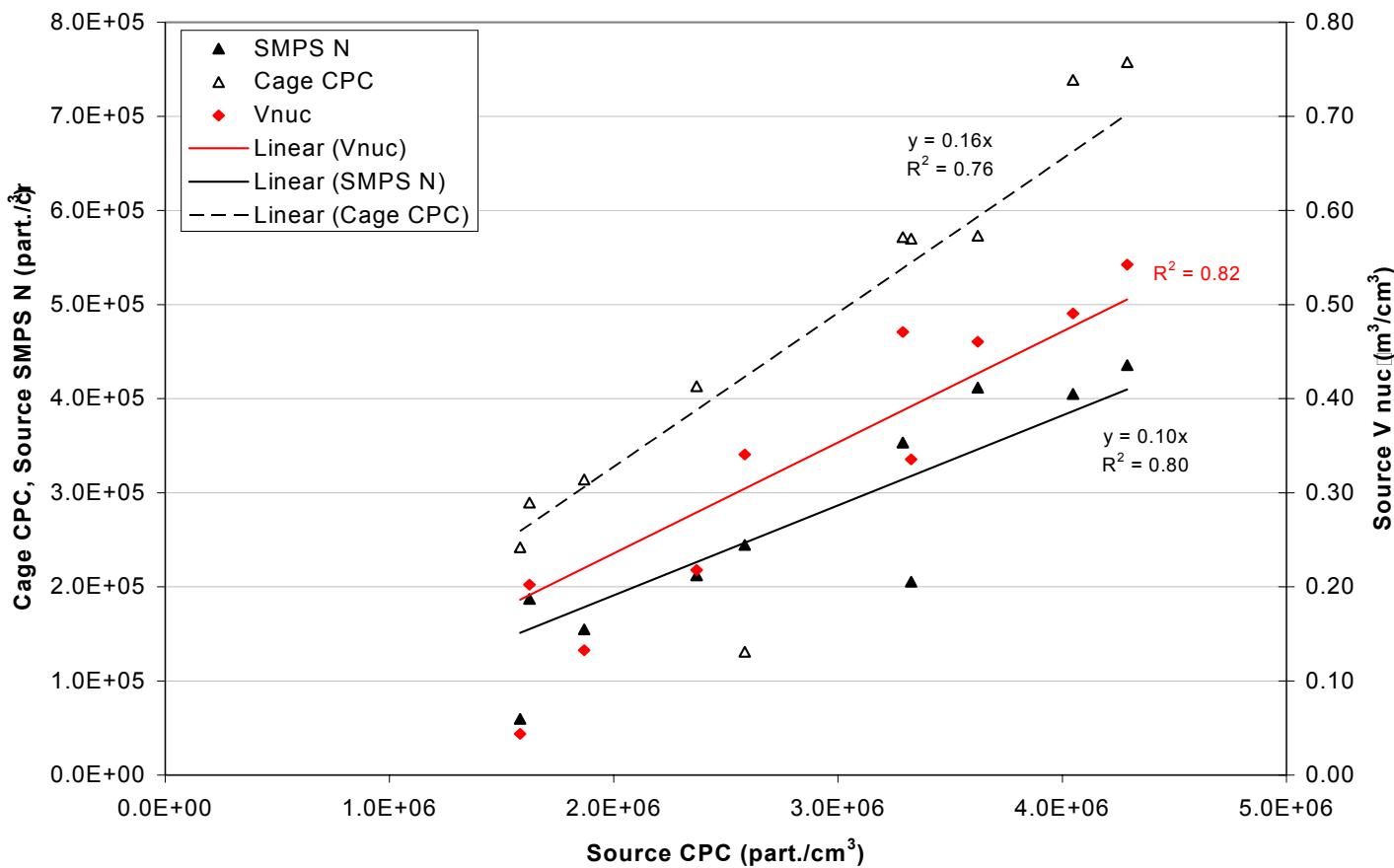
Temperature, humidity, and size distributions are daily averages
 V_{30}/V is the fraction of aerosol volume smaller than 30 nm and is an estimate of the volume (and mass) fraction in the nuclei mode

V_{30}/V increases by an order of magnitude as ambient temperature falls from about 23 to about 10 C

Comparison between instruments – fall 2003 study

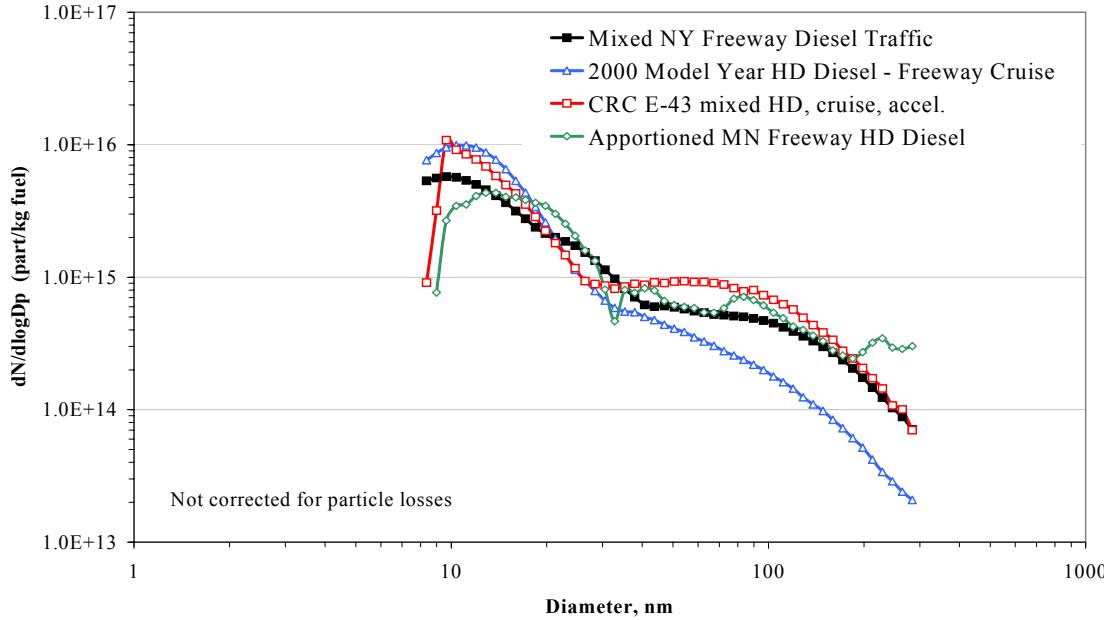
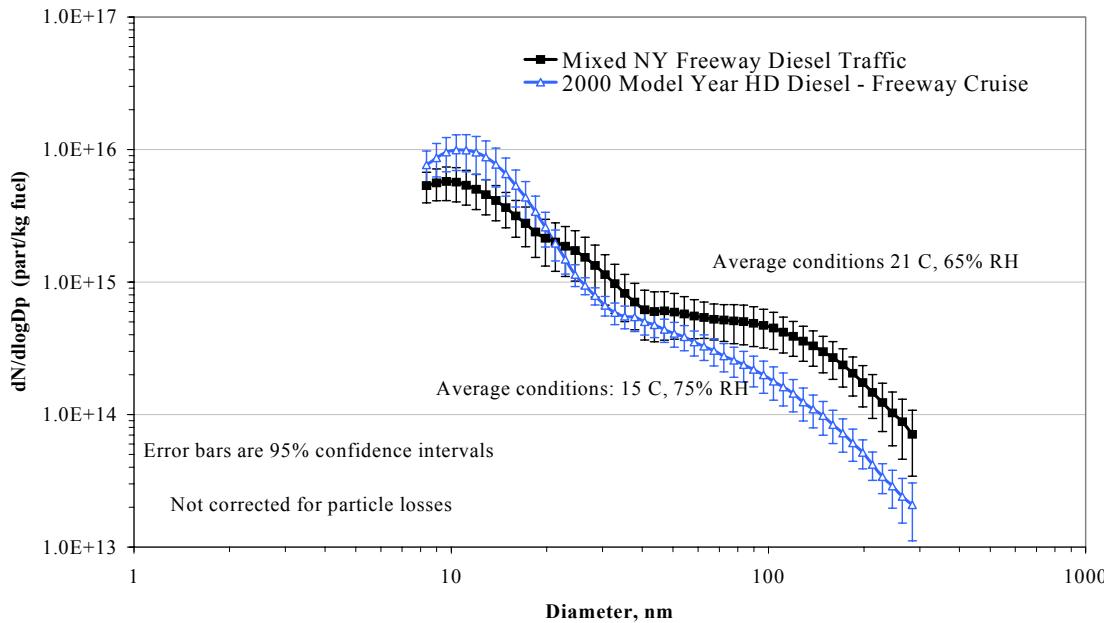
This plot compares the measurements of the CPC and SMPS used to monitor the exposure source and the CPC used to monitor the exposure chamber

- All three number measurements agree to within 20% on 50 nm calibration aerosol.
- The SMPS N is only 10% the CPC indicating many particles below the SMPS sizing range.
- The volume in the nuclei mode tracks very closely with the source CPC as expected.
- The exposure chamber CPC reads only 16% of the source CPC indicating significant losses in the chambers.



Clearly there are large losses in exposure chambers. Care must be taken to measure actual concentrations in breathing zones.

Fuel Specific Emission Factors



Particles/kg fuel =
((particles added)/volume exhaust)/
(fuel burned)/volume exhaust)

(fuel burned)/volume exhaust is
calculated from carbon balance

- The upper plot shows the on-road exposure data converted to fuel specific form
- The lower plot shows a comparison earlier studies
- Surprisingly consistent results between different fleets

Conclusions

- Large concentrations of nanoparticle, mainly in the nuclei mode are present over rural/urban freeways
 - Many of these particles are extremely small and not detected by conventional long column SMPS
 - On road aerosols are mainly volatile, especially in nuclei mode region
 - Large nuclei mode concentrations were associated with reduced responses of the PAS and aethalometer
 - The fraction of particles in the nuclei mode depends upon ambient conditions
 - Increases with decreasing temperature
 - Increases with increasing humidity
 - More than an order of magnitude change with modest changes in fall weather
- The MEL provides an effective means of measuring fuel specific emission factors of real world fleets of thousands of vehicles

Conclusions

- The MEL provided a stable platform for doing exposure experiments with real world aerosols
- Losses in the exposure chamber used here were significant
 - Number concentrations in the exposure chamber were about six times lower than in the source aerosol
 - Great care should be taken to measure actual concentrations in the breathing zones of exposure chambers
- If laboratory inhalation studies are to mimic real-world exposure from the perspective of providing an aerosol with similar physical and chemical characteristics then more data are required to characterize on-road aerosols so that laboratory dilution systems can be developed that produce real-world exposure.