Fine Particle (Nanoparticle) Emissions on Minnesota Highways

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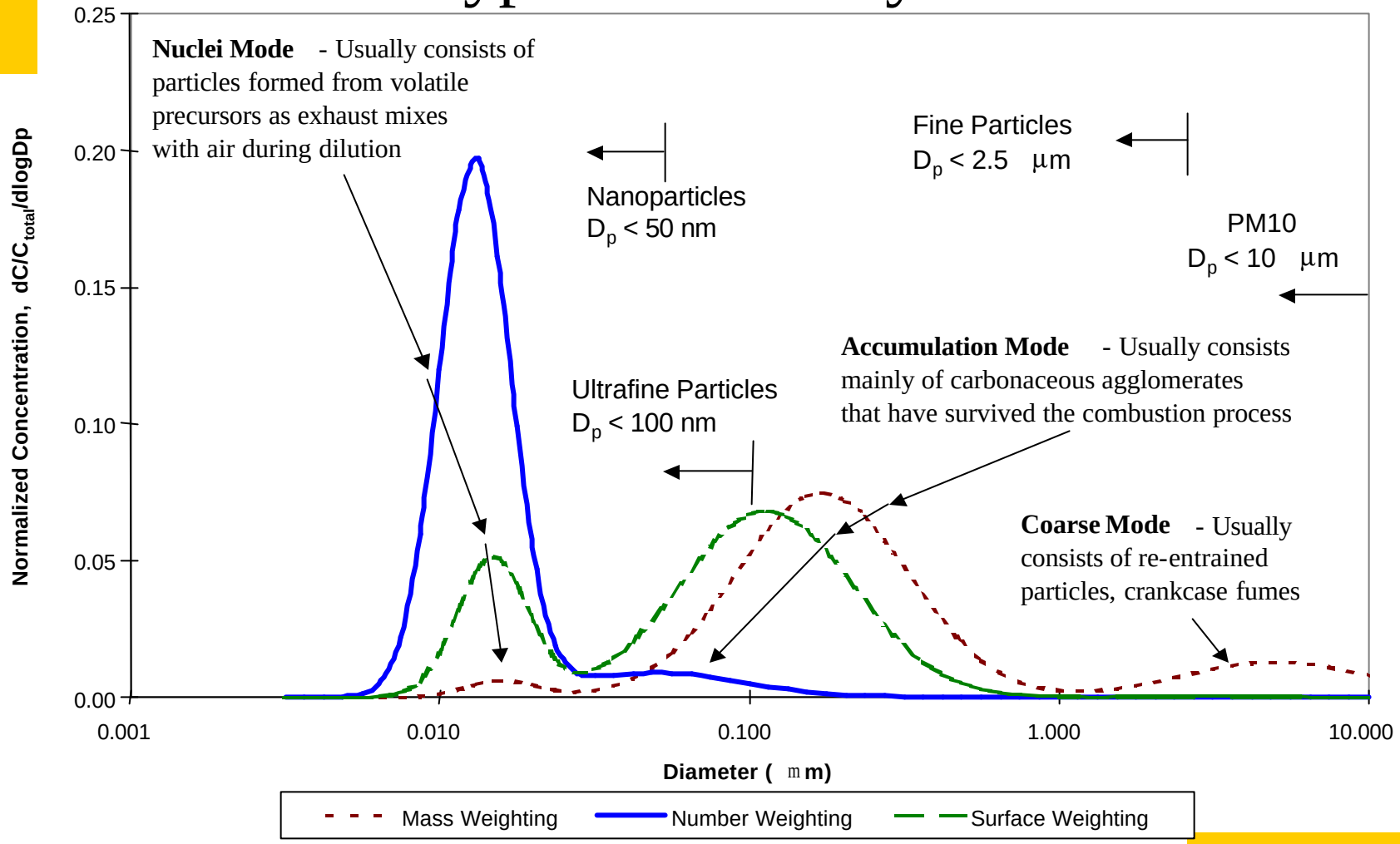
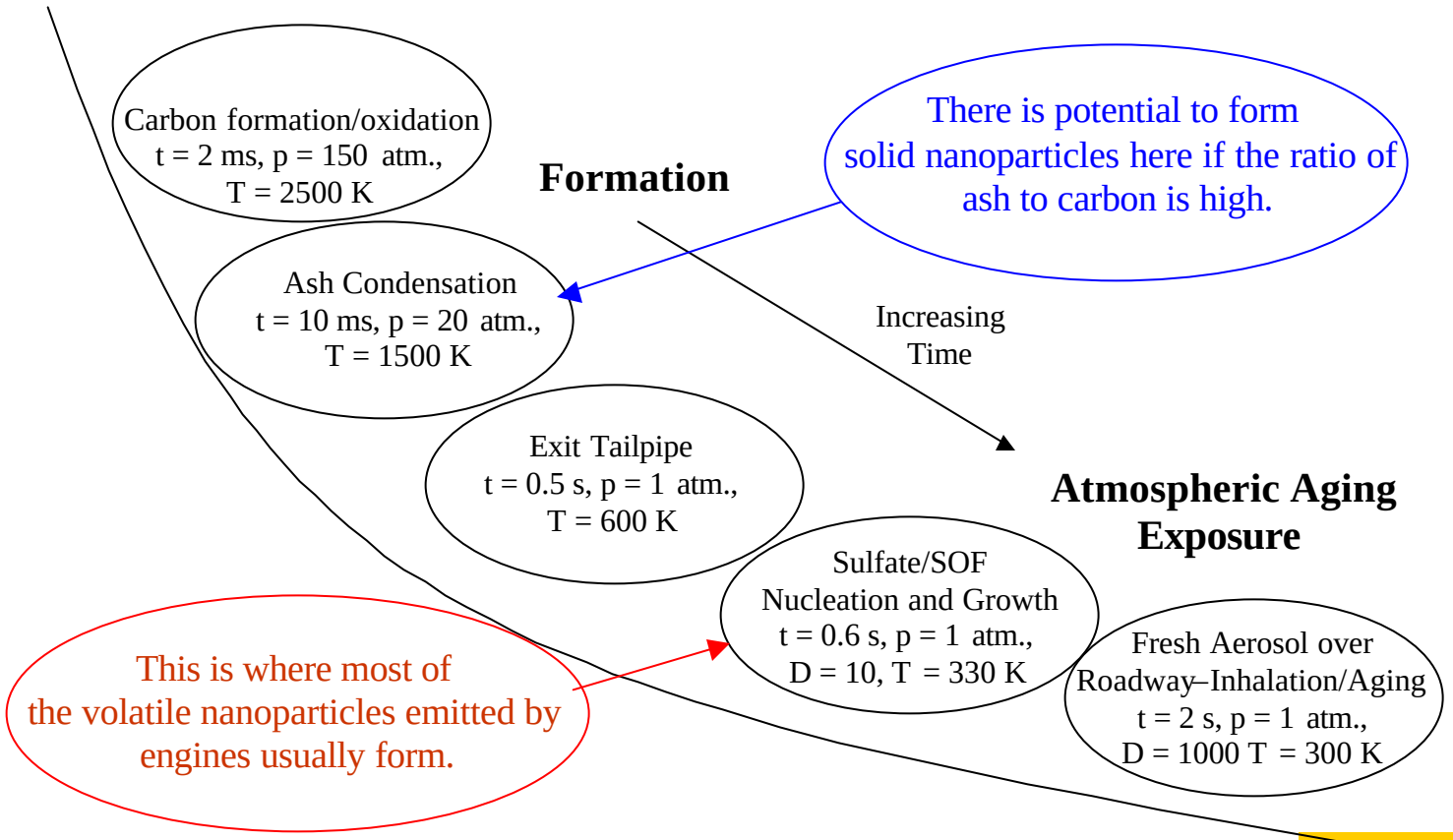
Background

- Typical Roadway Aerosol - Particle Size Distribution
- Nanoparticle Formation - History
- Typical Engine Exhaust - Particulate Matter Constituents
- Nanoparticle Formation - Model
- Nanoparticle Formation - Practical Implications

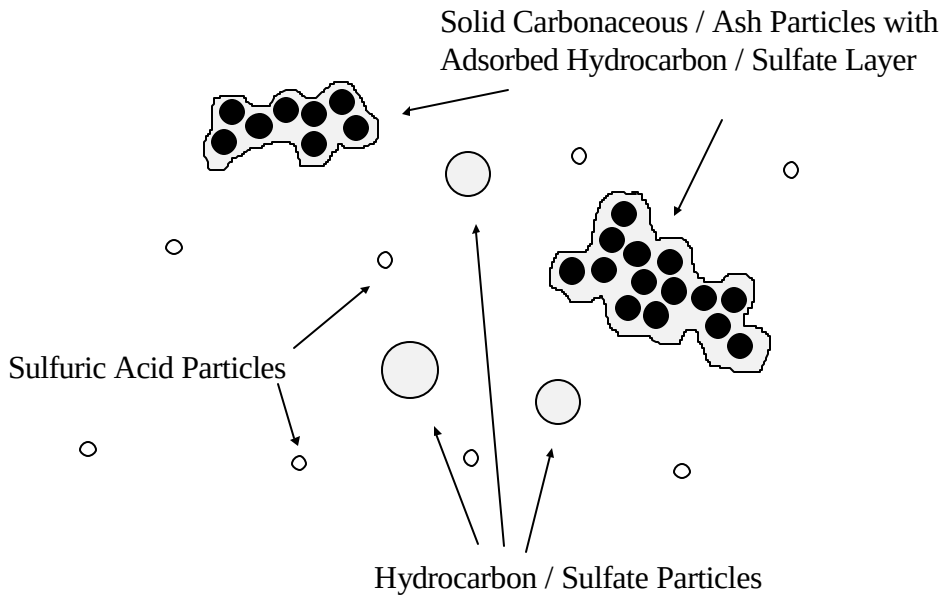
Particle Size Distribution of Typical Roadway Aerosol



Nanoparticle Formation History
From the Start of Combustion to the Nose



Particulate Matter Constituents of Typical Engine Exhaust



- Solid particles are typically carbonaceous chain agglomerates and ash and usually comprise most of the particle mass.
- Volatile and semi-volatile materials are formed through gas-to-particle conversion as exhaust cools and dilutes.

Nanoparticle Formation Model based mainly on Diesel Studies

- Most of the particles (90 % of the number and 30% of the mass) are formed from volatile precursors (H_2SO_4, H_2O, Fuel and Oil residues) during exhaust dilution in the atmosphere.
- A simple model of nanoparticle formation gives qualitative results that are consistent with experimental observations:
  - Initial growth due to binary nucleation of H_2SO_4 and H_2O.
  - Subsequent growth due to adsorption of the Soluble Organic Fraction (SOF) by sulfuric acid nuclei.
  - Additional growth may be due to nucleation of high molecular weight Hydrocarbons (HC).

Nanoparticle Formation Model Practical Implications

- Nonlinear Particle Dynamics can produce large changes in particle number due to small changes dilution and sampling conditions.
- Sampling Systems should mimic atmospheric dilution in order to obtain representative aerosol measurements - this has not been accomplished to date.
- Factors that may promote volatile nanoparticle formation include:
  - Low Exhaust Carbon Content
  - High Fuel Sulfur Content
  - Fast (Highway) Dilution
  - High Engine Exhaust Gas Temperature (Engine Load)
  - Low Ambient Temperature
  - Low Ambient Relative Humidity

Mn/DOT - CTS Project Goals and Objectives

- Funding was provided by the Minnesota Department of Transportation (Mn/DOT) through the University of Minnesota Center for Transportation Studies (CTS)

- Goals and Objectives
  - Determine the relationship between traffic congestion and nanoparticle concentrations on Minnesota roadways.
  - Estimate fuel specific emissions factors.
  - Determine the concentrations of nanoparticles in neighborhoods near major highways.

Mn/DOT - CTS Outline

- Aerosol Sampling Methodology
- Mobile Emissions Laboratory (MEL)
  - Physical Description
  - Aerosol Instrumentation
  - Transient Response
- Spark Ignition (SI) Aerosol Distribution Statistics
  - Number
  - Diameter of Average Surface
  - Volume

Mn/DOT - CTS Outline Continued

- SI and Diesel Size Distribution Comparisons
- Fuel Specific Emission Indexes
  - Number
  - Mass
- Neighborhood Aerosol Measurements
  - Downwind/Upwind Comparison
  - Far-Field/Near-Field Comparison

Aerosol Sampling Methodology

- Data collected in November 2000
  - Highways: County and Interstate with high traffic volume
  - Residential Areas: Downwind/Upwind & Far-Field/Near-Field
- Ambient Environment
  - Temperature Range: 1-13°C (34-55°F)
  - Relative Humidity Range: 40-60%.
- Aerosol Sampling Parameters
  - Sample Line Length: 10 m
  - Total Transport Flowrate: 150 lpm
  - Residence Time: 25 seconds

Sampling Methodology Continued

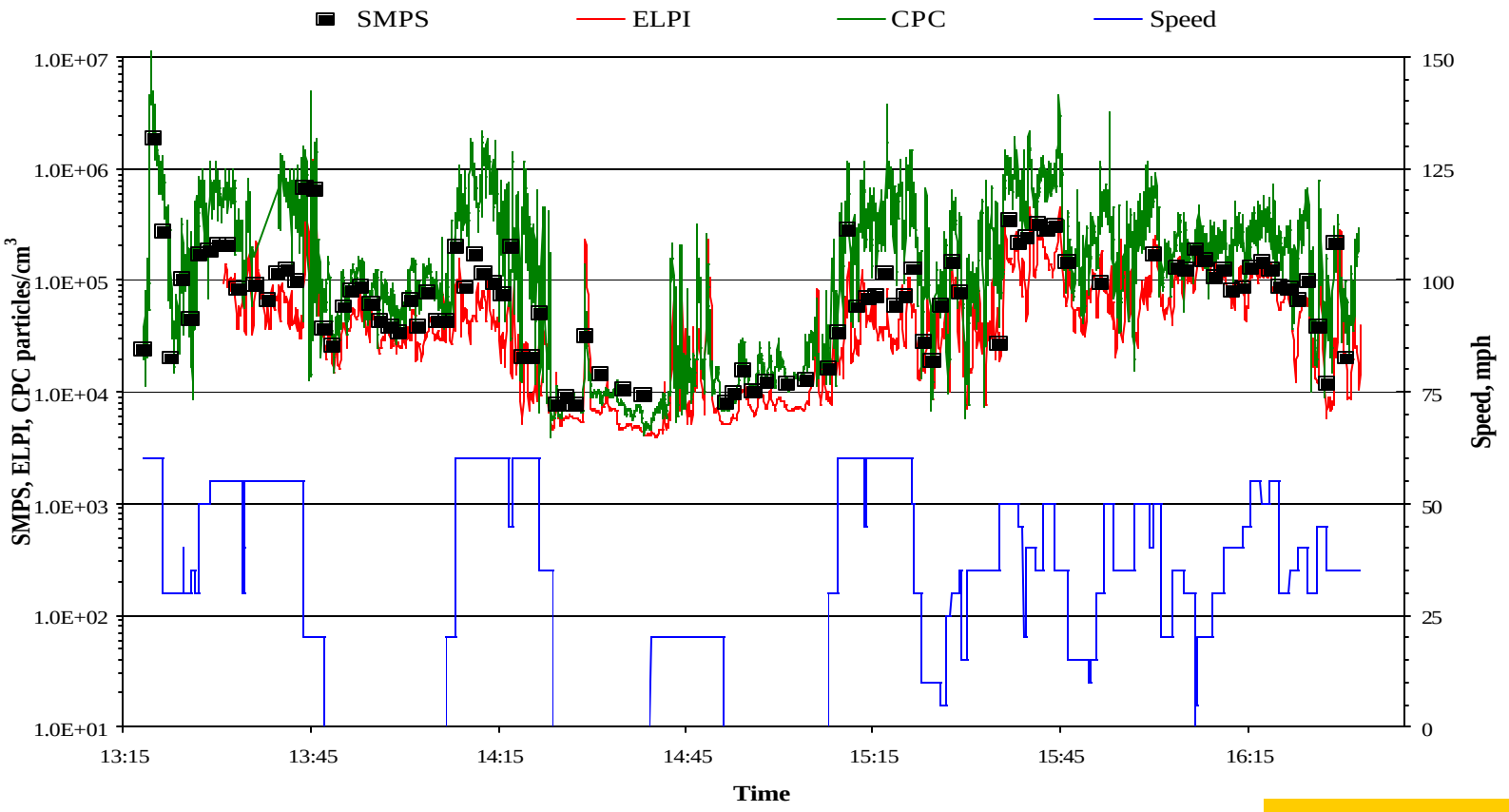
- Aerosol Distribution Measurements
  - Time Integrated (Continuous SMPS Scan)
  - Near Real Time (Bag Sample SMPS Scan, ELPI)
- MEL Vehicle Operation
  - Video/Event Log maintained by monitor in front of MEL.
    - Traffic Counts (SI/Diesel)
    - Time/Location Data
  - Traffic Speed Estimate made relative to speed of MEL
  - Minimum Following Distance: 10 m

Equipment
Storage

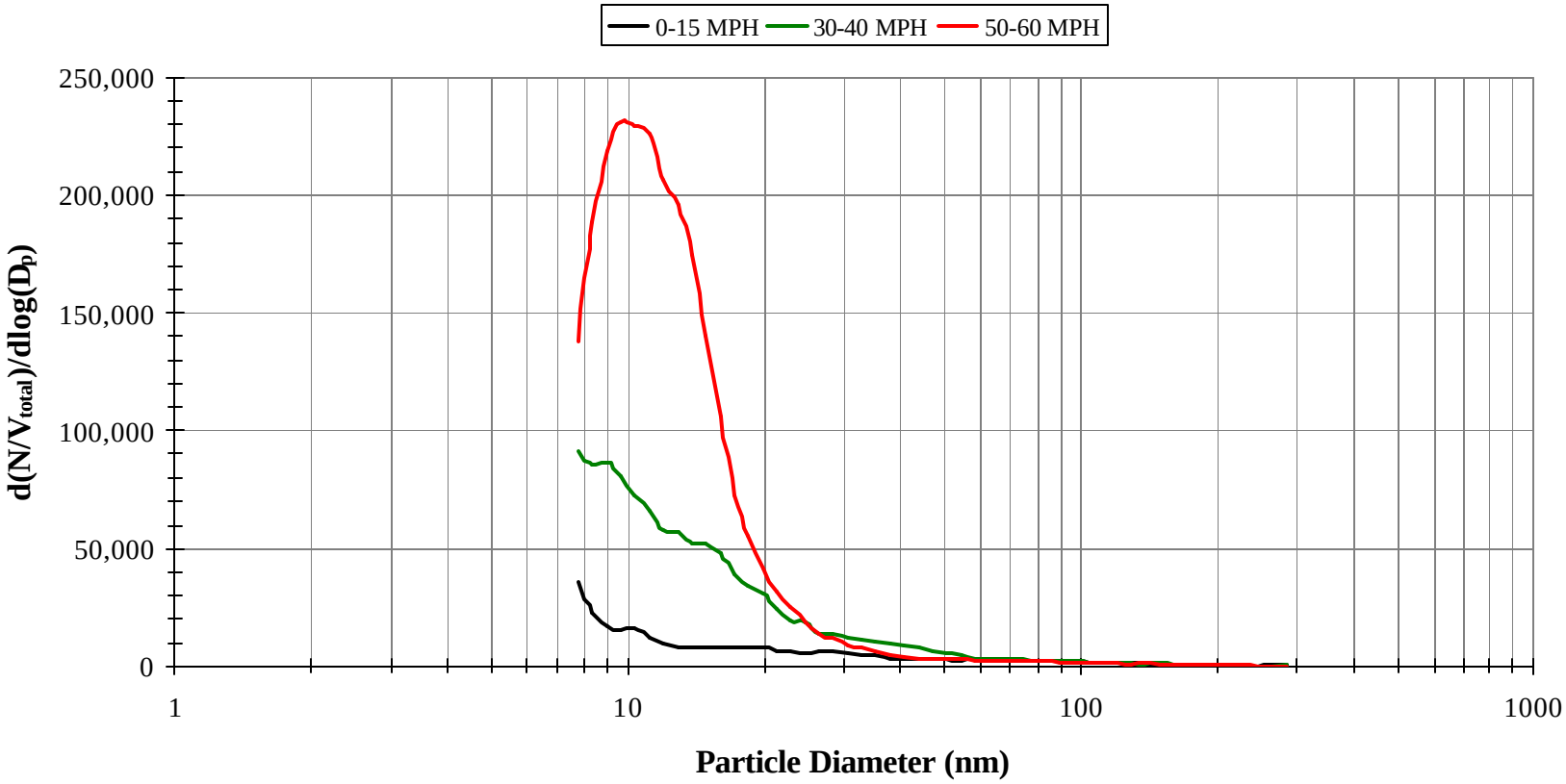
MEL Principal Instruments

- Total Particle Number
  - UCPC - Ultrafine Condensation Particle Counter
    (D_p > 3 nm, N < 10^5 particles/cm^3)
- Particle Size Distribution
  - SMPS - Scanning Mobility Particle Sizer (7 to 300 nm)
  - ELPI - Electrostatic Low Pressure Impactor (30 to 2,500 nm)
- Particle Surface Area
  - DC - Diffusion Charger (Total submicron particle surface area)
  - PAS - Photoelectric Aerosol Sensor
    (Total submicron bound PAH equivalent surface area?)
- Gas Concentration
  - CO (0-1100 ppm)
  - CO_2 (0-2500 ppm)
  - NO (0-10 ppm)

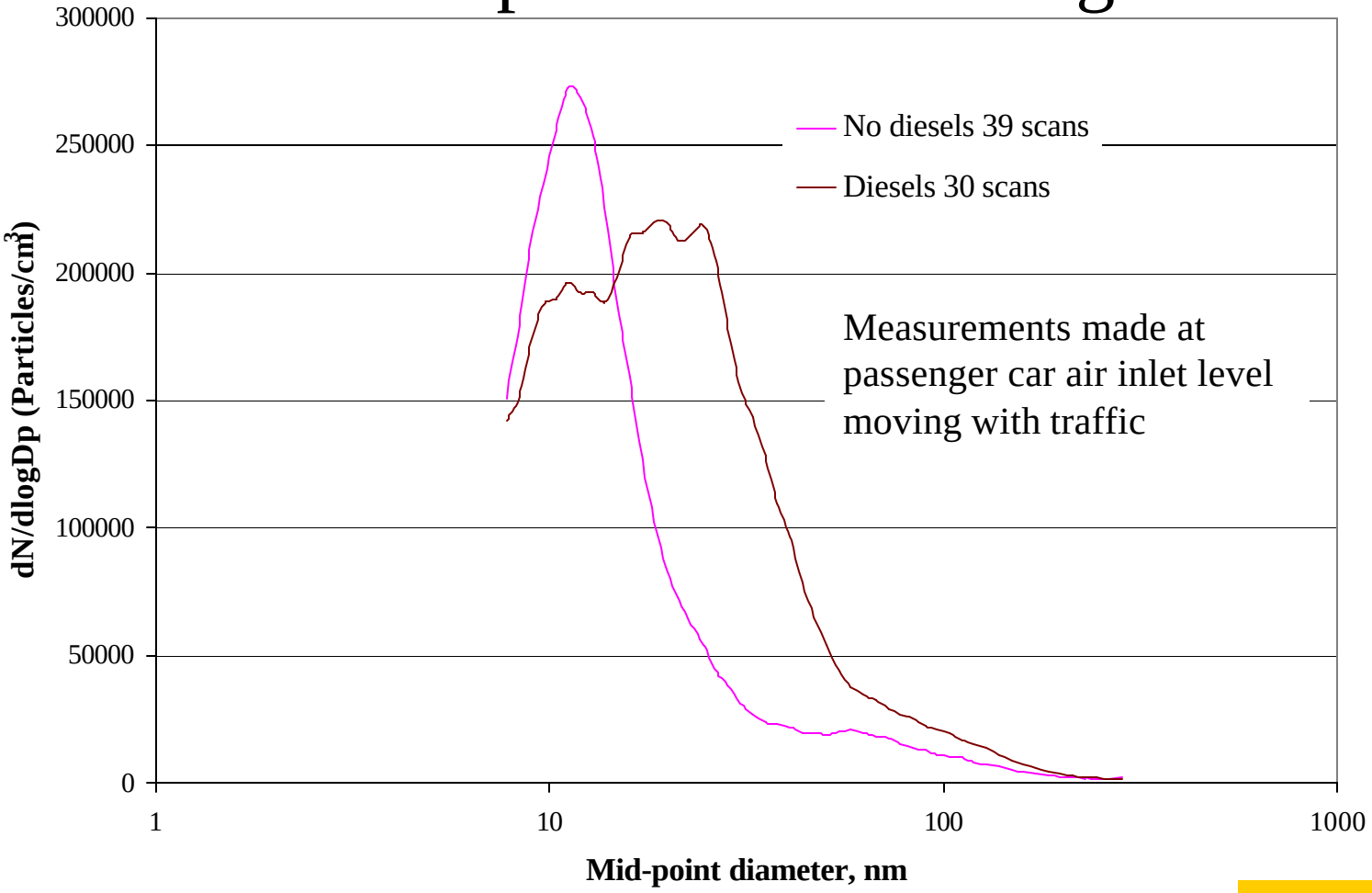
MEL Instruments respond to Transient Aerosol Distribution



SI Particle Number Concentration Increases with Vehicle Speed



SI Engines produce Smaller Particles compared to Diesel Engines



Measurements (Färnlund et al., 2001) show SI and Diesel in same Number Emission Range

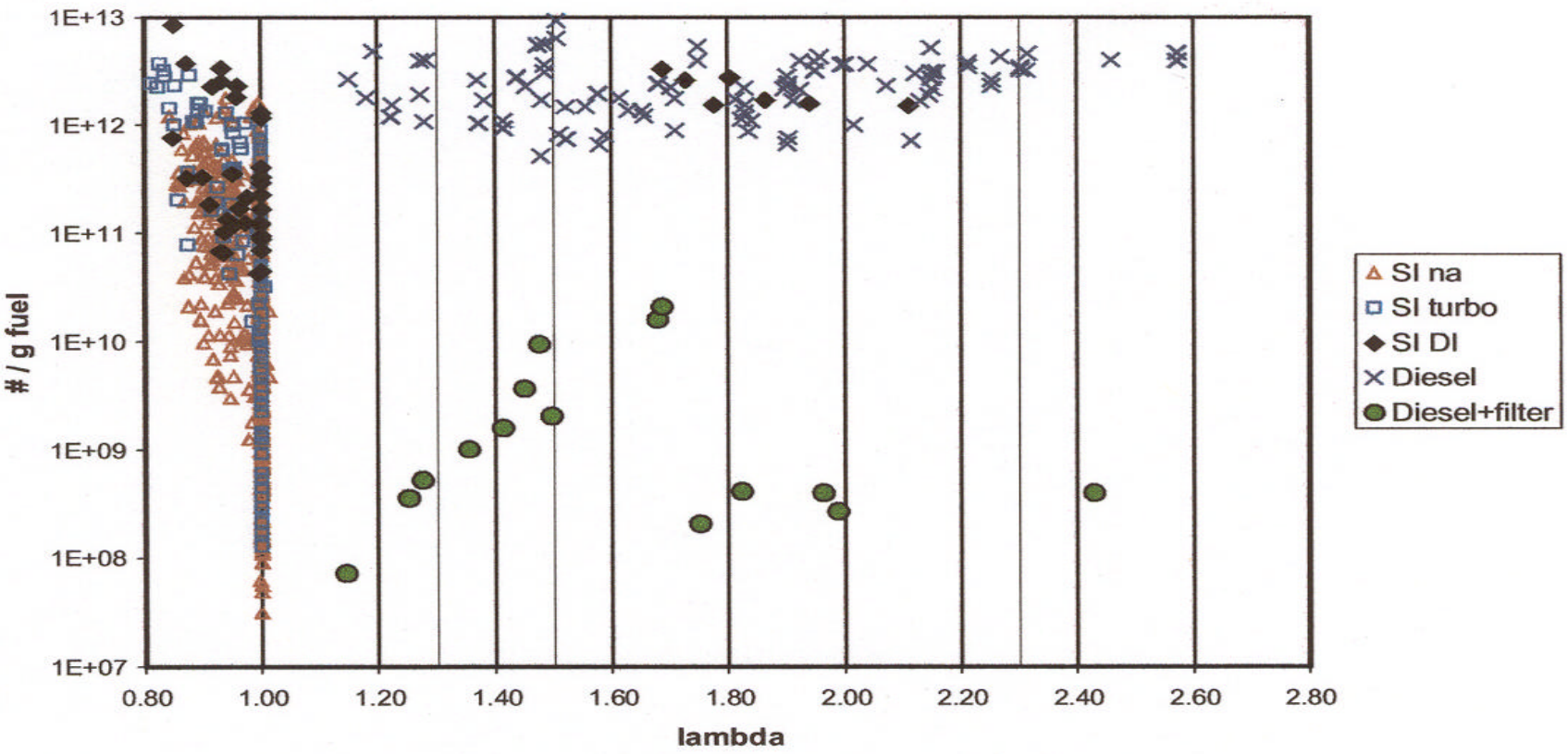


Figure 3 - Total particle number per fuel mass related to lambda

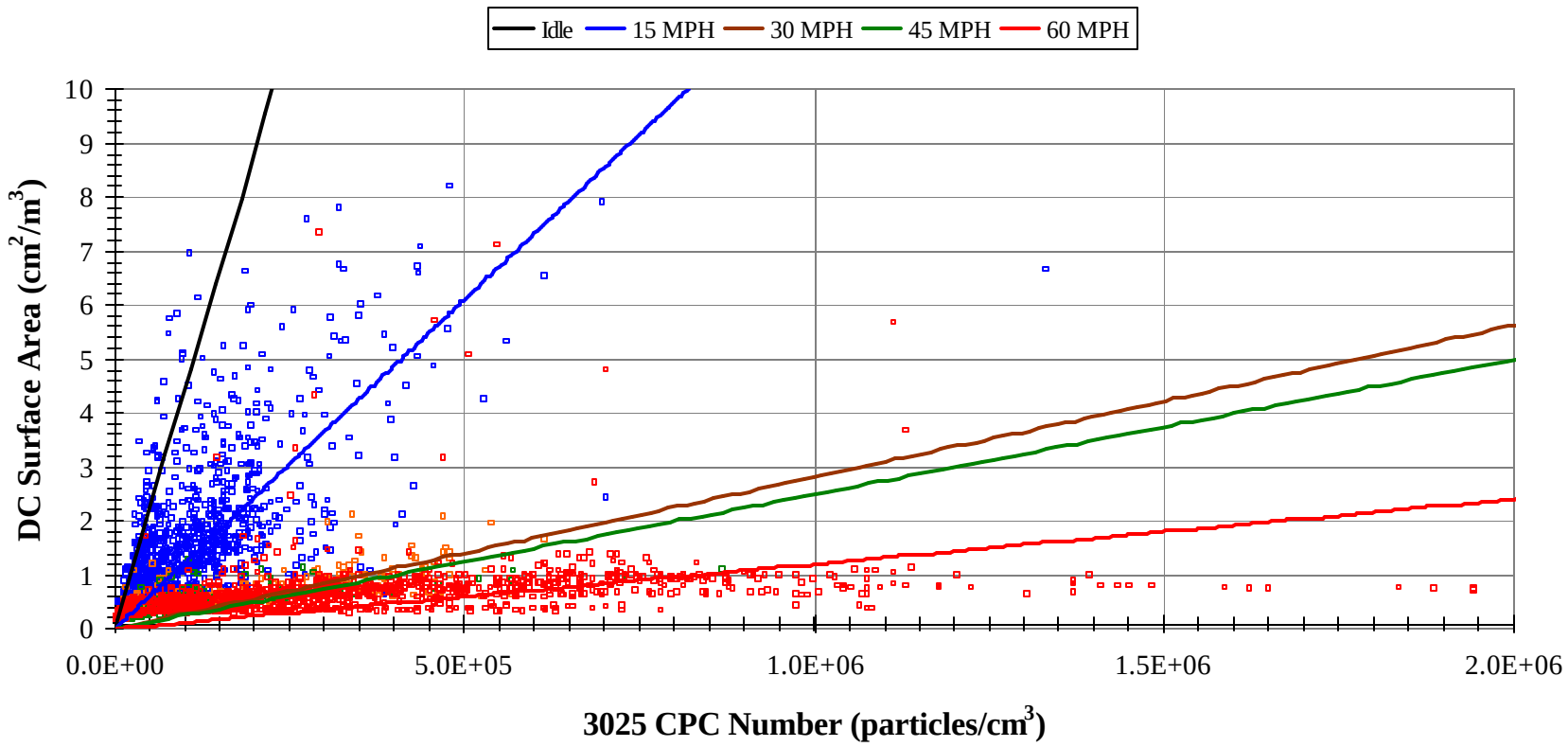
Diameter of Average Surface Relation to Surface/Number Slope

$$S_C = N_C\left(\frac{p}{4}D_{S,Avg}^2\right)$$

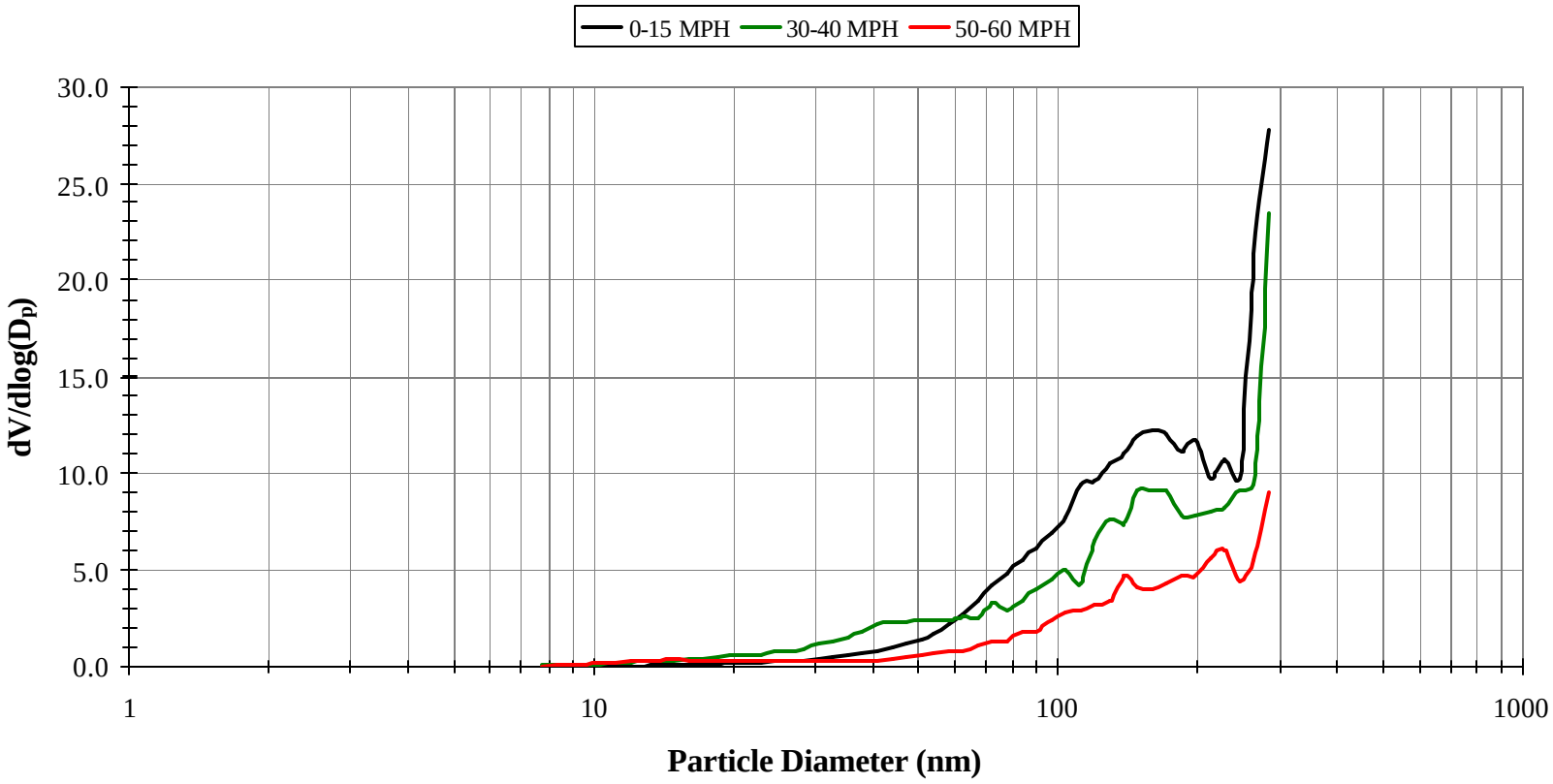
$$D_{S,Avg} = \sqrt{\frac{4}{p}\left(\frac{S_C}{N_C}\right)}$$

- S_C is the Surface Area Concentration obtained by using the DC
- N_C is the Number Area Concentration obtained using the CPC
- $D_{S,Avg}$ is directly proportional to the slope of the DC/CPC response.

Diameter of Average Surface Decreases with Vehicle Speed



SI Particle Volume Concentration Decreases with Vehicle Speed



Aerosol Distribution Measurements Summary of Results

- High Speed Traffic was generally associated with:
  - High Particle Number Concentration
  - Smaller Diameter of Average Surface
  - Lower Particle Volume
- SI Engines produce aerosol distributions with mean particle diameter less than that produced by Diesel Engines.
- Recent European Measurements show that SI and Diesel Engines produce nanoparticles with similar number concentrations.

Fuel Specific Emissions Calculation Procedure

- Fuel Specific Number Emission (particles/kg_{fuel}):

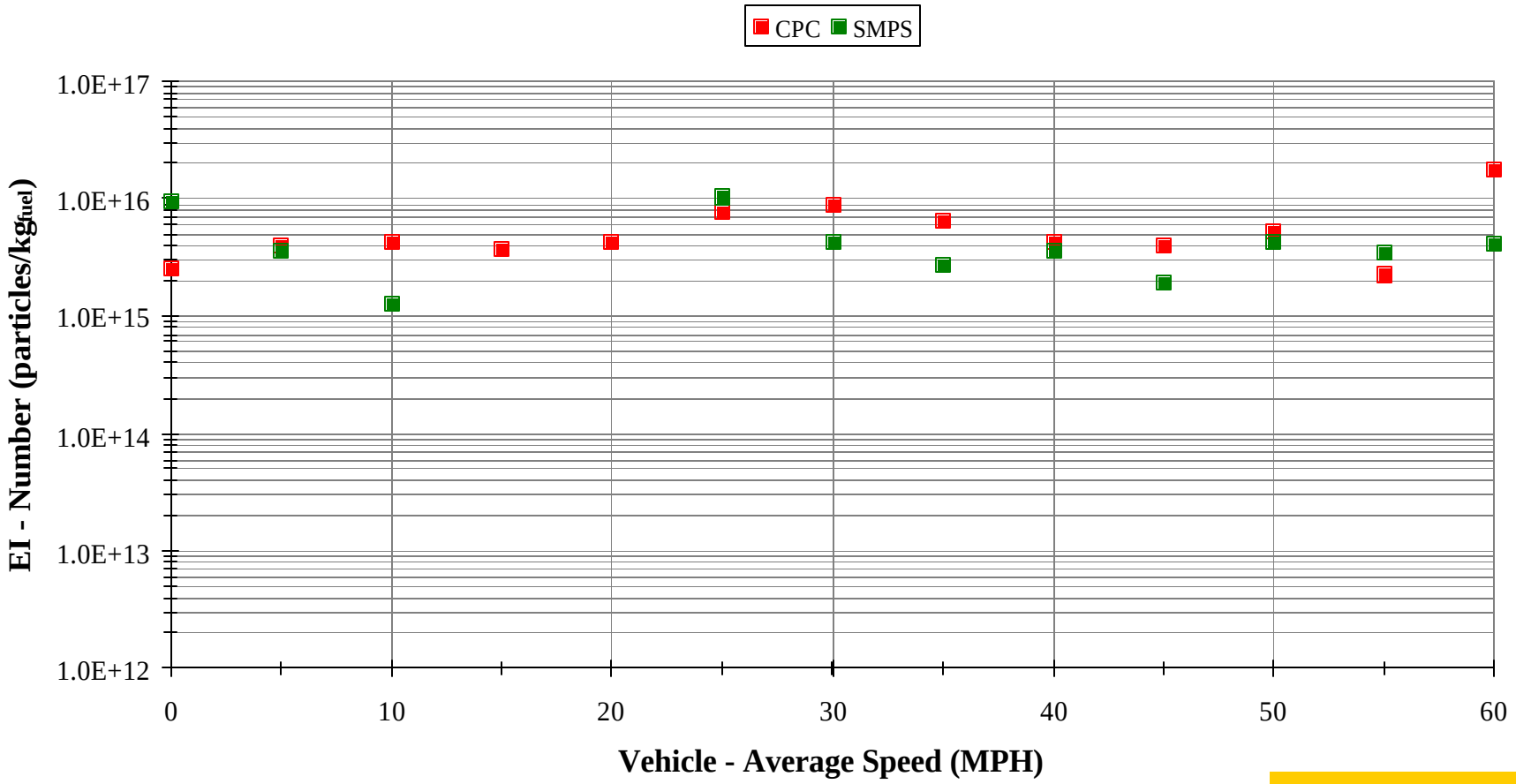
$$EI_N = \frac{N}{\left(x_{CO} + x_{CO_2}\right)\left(M_C / M_{air}\right) y_{C_{fuel}} \, r_{air}}$$

- Fuel Specific Mass Emission (mg/kg_{fuel}):

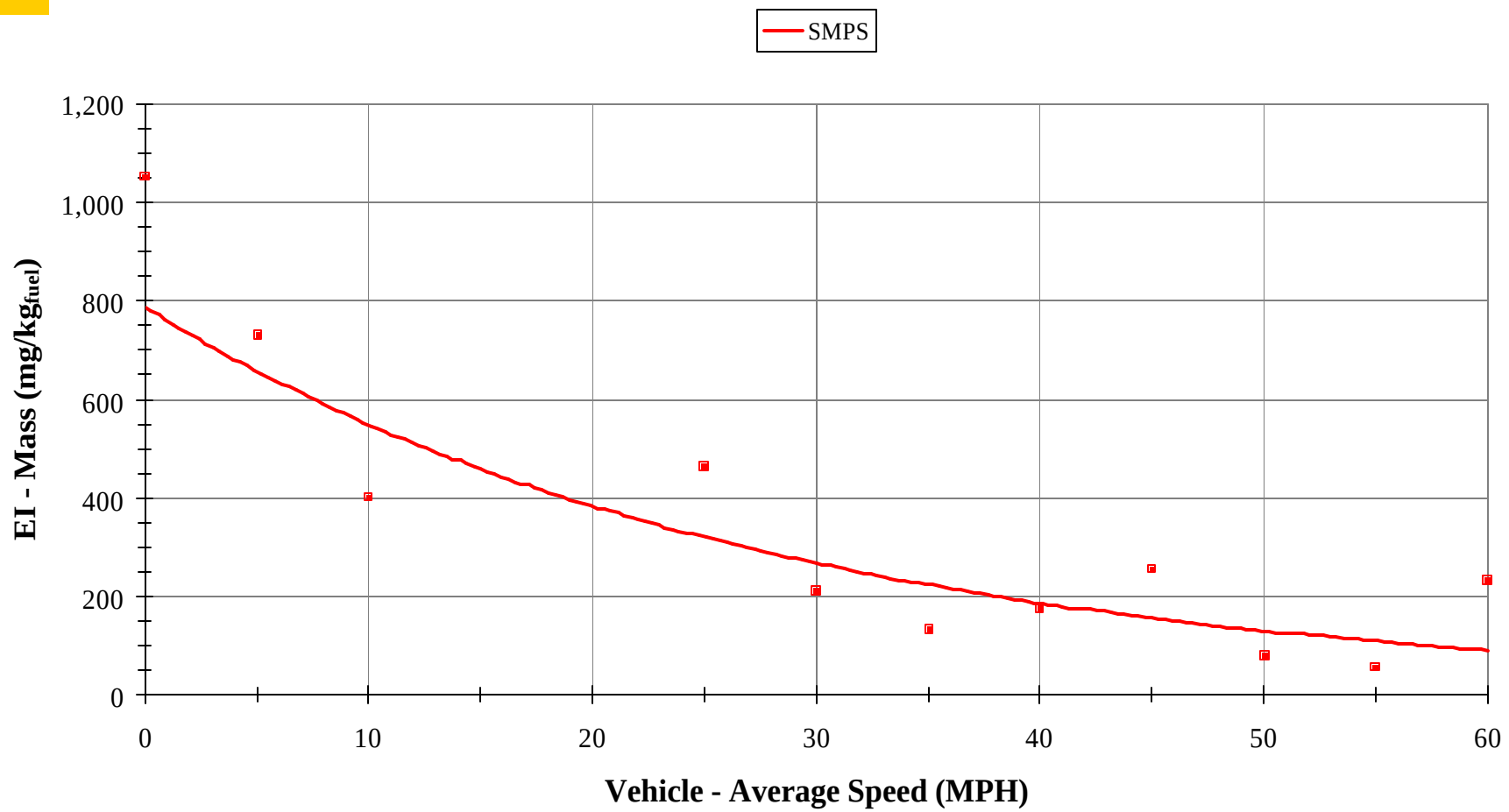
$$EI_m = \frac{m}{\left(x_{CO} + x_{CO_2}\right)\left(M_C / M_{air}\right) y_{C_{fuel}} \, r_{air}}$$

- Measured carbon compound content above background levels is assumed to come from combustion of fuel.

Fuel Specific Number Emissions Index



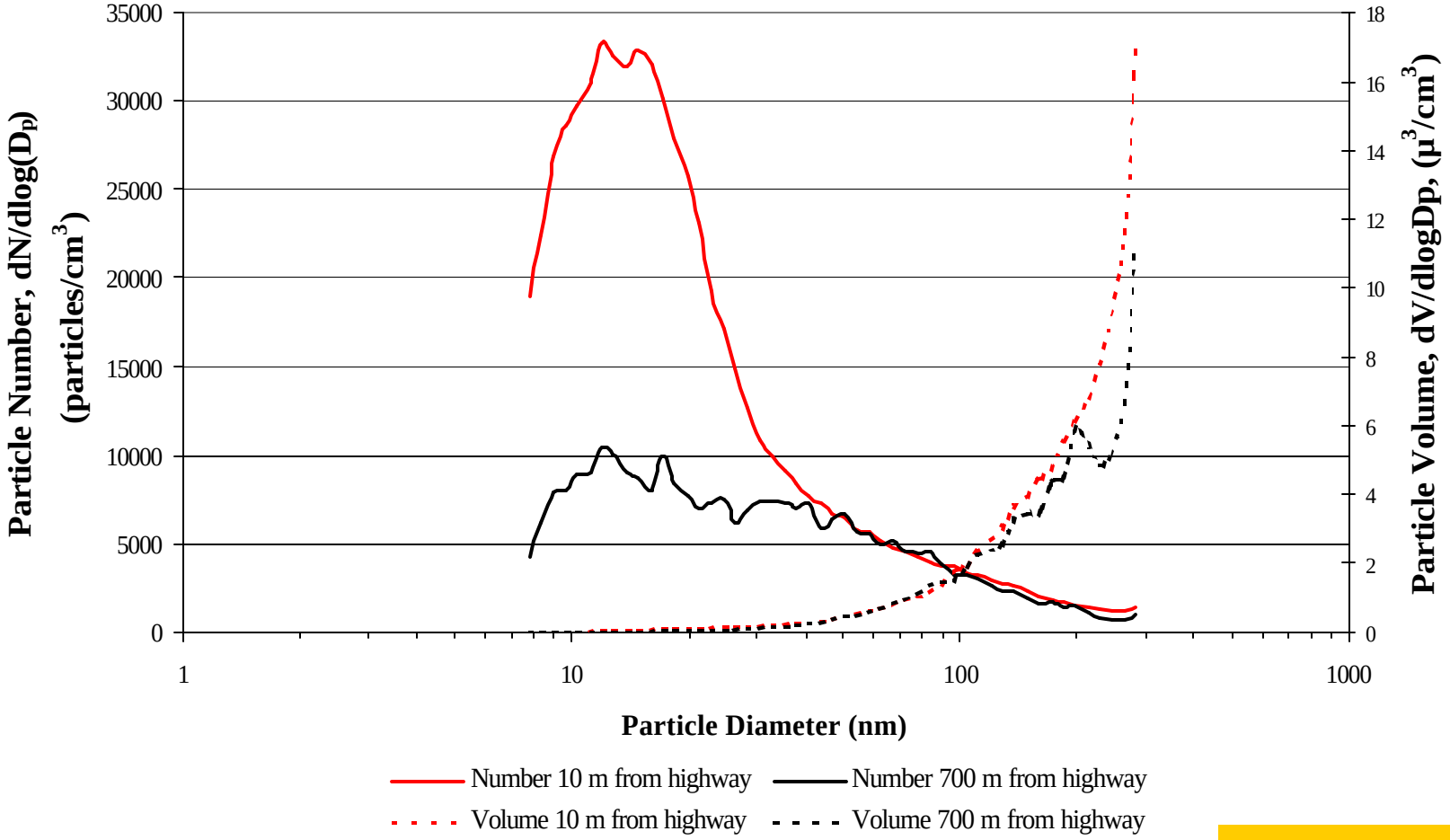
Fuel Specific Mass Emissions Index



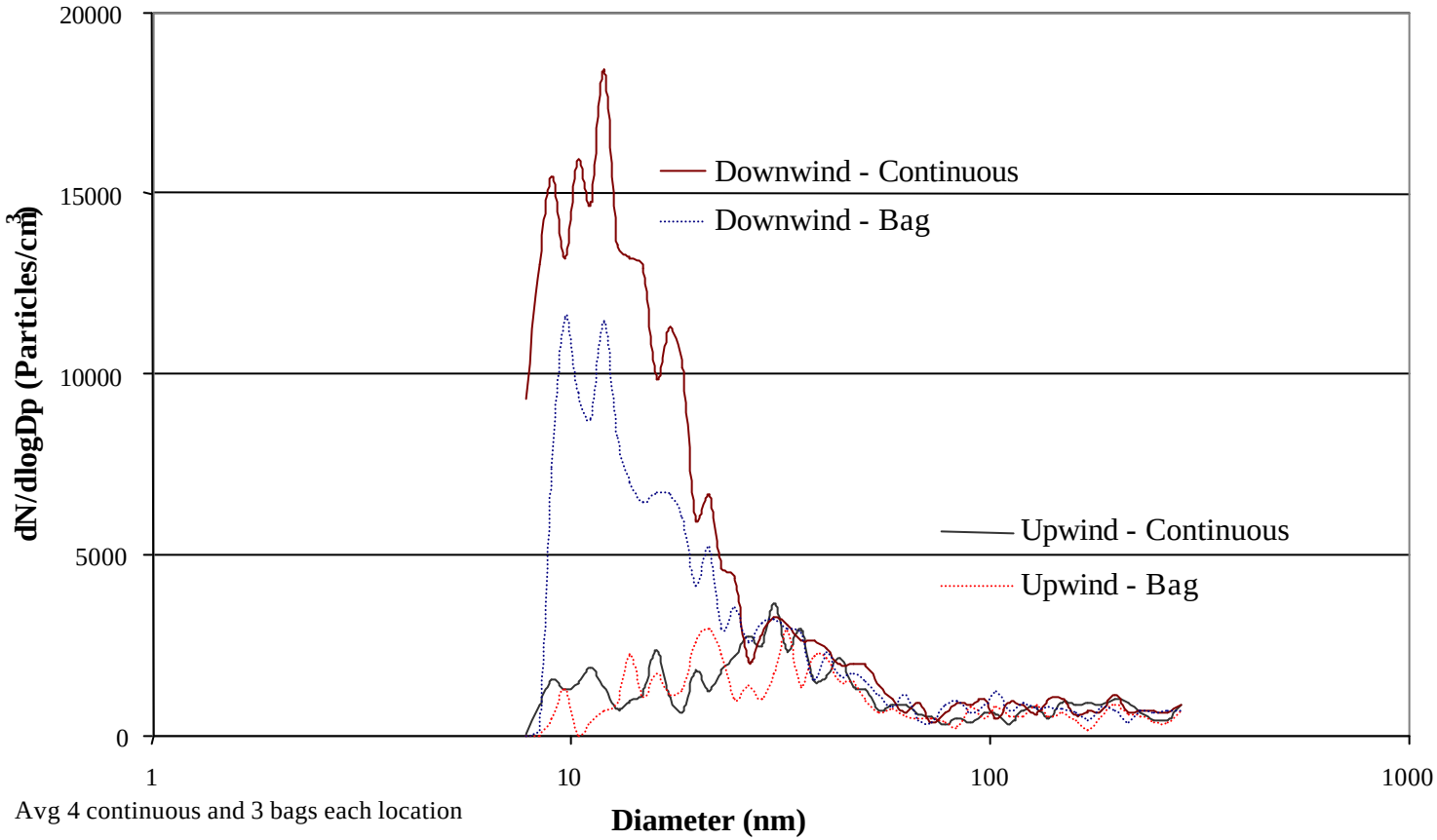
Fuel Specific Emissions Indexes Summary of Results

- Number emission factors obtained are an order of magnitude higher than the maximum values found in the literature (>10^{14} particles/mile).
  - Measurements made during cold weather favor nanoparticle formation.
  - Typical Laboratory Dilution Tunnels used to determine regulated emissions may not replicate Real World Dilution.
  - UCPC and SMPS were optimized for nanoparticle dectection.
- The inverse relationship between Mass Emission Index and vehicle speed (increased fuel consumption) is consistent with the increased production of nanoparticles (lower mass).

Neighborhood Aerosol Measurements Near-Field/Far Field Comparison



Neighborhood Aerosol Measurements Downwind/Upwind Comparison



Avg 4 continuous and 3 bags each location

Neighborhood Aerosol Measurements Summary of Results

- Measurements made in residential areas demonstrate that aerosol concentrations can approach on-road concentrations for homes located 10 to 30 m from highways.

- Much lower concentrations and larger particle sizes were observed in residential areas located 500 to 700 m from the highway.

- Nanoparticles can exist for significant distances downstream of highways.

Conclusions

- Spark ignition engines typically emit smaller particles than diesel engines and are an important source of fine particles and nanoparticles.
- SI Engines can can emit particle number concentrations comparable to Diesel Engines when:
  - Operating in “fuel enriched” conditions (cold start, high power).
  - Designed for Direct Injection (DI)
- Particle number concentration increases with vehicle speed due to storage and release of volatile hydrocarbons deposited in the exhaust system.
- The collection of additional data during warmer periods in needed to expand the comprehensiveness of the results.

Acknowledgements

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