Effects of the U.S. EPA Ultra Low Sulfur Diesel Fuel Standard on Heavy-Duty Fleet Average Nanoparticle Emissions in Minnesota

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Objectives
Background
Methods
Results
Future Research and Conclusions

Objectives

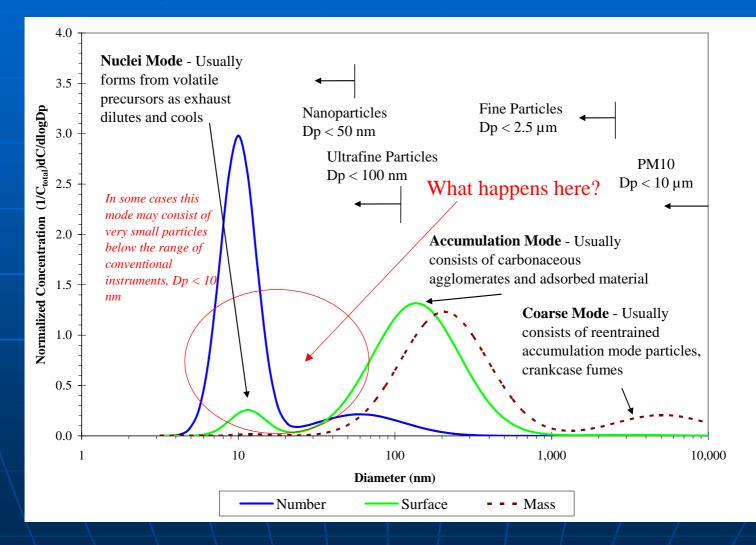
- The sulfur content of most Diesel fuel sold in the U.S for on-road use was required to be reduced from less than 500 ppm to less than 15 ppm (ultra low sulfur diesel, ULSD) by October 2006.
 - This was done to allow the use of catalyzed Diesel particle filters (DPF).
 - Question Has the introduction of ULSD led to a reduction on-road nanoparticle emissions from HD vehicles in Minnesota? Very few have DPFs.
 - Answer Yes, for the summertime urban freeway conditions tested

Method

- Make on-road particle and gas measurements in 2006 and 2007 and measure fuel sulfur levels for test periods
 - 2006 33 ppm
 - 2007 8 ppm
- Determine volume of HD and LD traffic on test routes
- Apportion results to calculate vehicle specific and fuel specific emissions of HD and LD vehicles

Compare results to past studies and other apportionment methods

Nanoparticles in the Environment



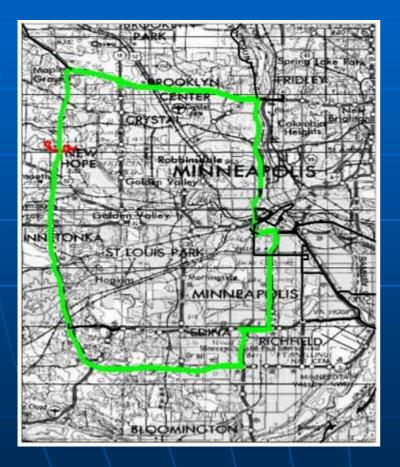
MEL Capabilities and Features



The MEL was originally built for the CRC/DOE E43 Project

- Sample air from front of truck, either at above cab level or at street level
- Used above cab level for Diesel study
- GPS for location and speed, time synchronization
- Particle Instruments, with bypass flows to minimize losses
 - CPCs, Leaky Filters
 - SMPS
 - EEPS
 - DC and PAS
 - EAD/NSAM
 - DustTrak
- Gas Instruments
 - CO₂
 - CO
 - NO_x (NO, NO₂)
- Calibration with HEPA filters, nebulized DOS, span and zero gases

Approach and Test Route



Typical Test Route

- Morning calibration
- 2-3 loops around freeway route between rush hours
- Sample particle and gases on a second-by-second basis
- Daily average of all data average over roadway
- Slightly different route from one year to the next because of construction
- Comparable speeds and vehicles

Traffic Counting

- Used MN/DOT Traffic Camera Monitoring System, recorded on video tape
- Random Sample of 5-Minute Camera Windows, 10-20 per day
- Counted manually from tapes by students
 - Heavy Duty: Multi-Axle Trucks, Buses, RVs Delivery Trucks, Flatbed Pickup Trucks
 - Light Duty: Passenger Cars, Vans, SUV's

Apportionment Method

Weekend/Weekday

- Developed in Summer 2002 study* for USDOE
- Assume form of equation for weekends and weekdays, linear combination of traffic volume times contribution plus nonvarying daily "background"
- Solve system of equations
- Generally requires background correction
- Multi-Variable Linear Regression Used in this Study
 - Assume matrix form of same scenario using daily average and daily traffic volumes
 - Multi-variable regression (least-squares)
 - Solve by matrix methods
 - Currently, estimate error based on percentage error in traffic and particle/gas measurements
 - Does not require background correction

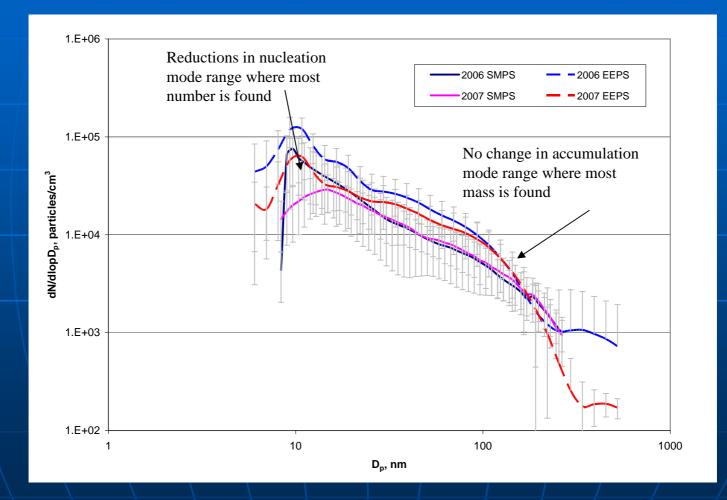
Results

Traffic Volume and Operating Parameters Focus on Heavy Duty Results Roadway Size Distributions Apportioned Size Distribution (per unit traffic volume) Fuel Specific Results for Heavy Duty Comparison to Previous Studies

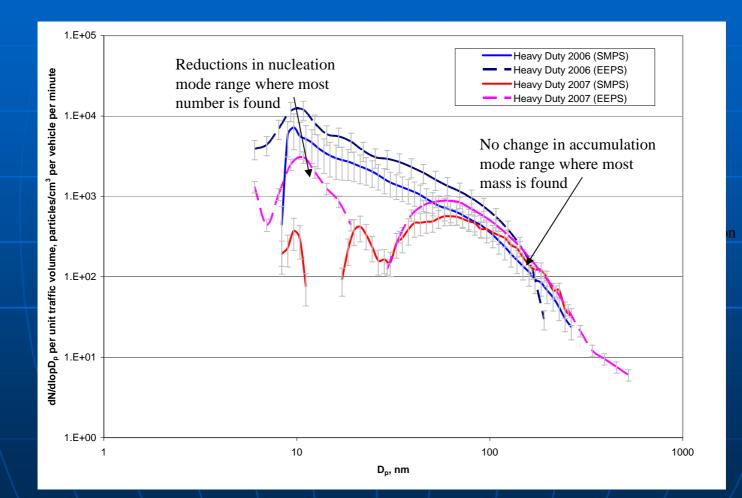
Average Values of Traffic Volume and Operating Parameters

	2006		2007	
	Avg	SD	Avg	SD
Fuel Sulfur, ppm	33		8	
Temperature, C	27	0.6	26.1	0.5
MEL Speed, mph	56.9	3.6	57.7	3.8
Weekday Heavy Duty by Vehicle, %	9.8	1	12	0.8
Weekday Light Duty by Vehicle, %	90.2	1	88	0.8
Weekend Heavy Duty by Vehicle, %	1.9	0.9	2.3	0.1
Weekend Light Duty by Vehicle, %	98.1	0.9	97.7	0.1

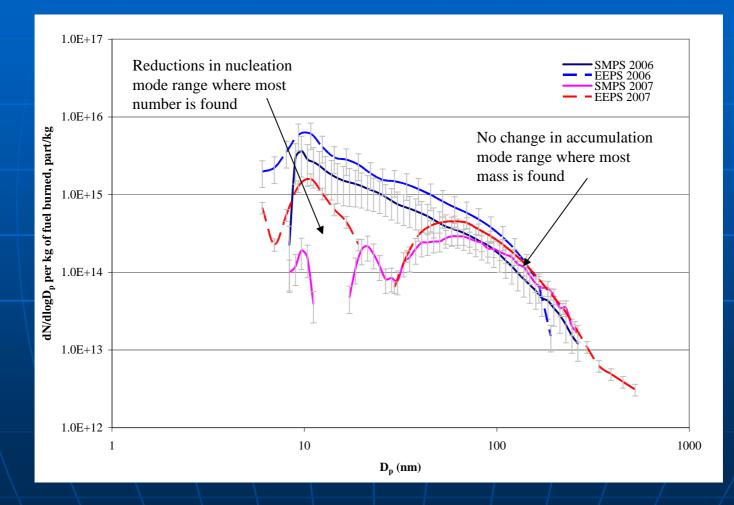
Roadway Size Distributions



Apportioned Heavy-Duty Size Distributions (per unit traffic volume)

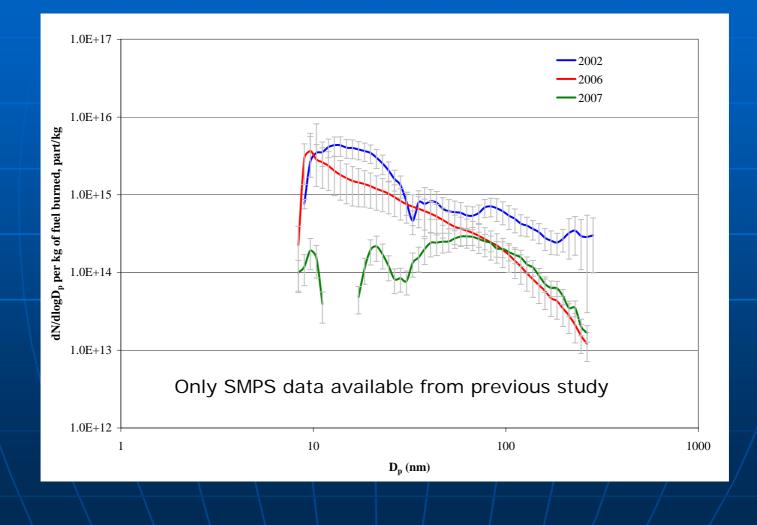


Apportioned Heavy Duty, Fuel Specific Size Distributions*

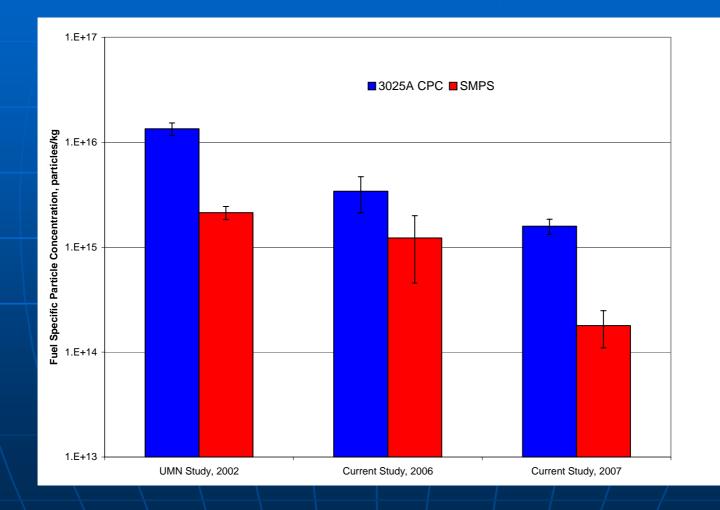


*Problem with CO₂ analyzer discovered late in study, CO₂ for 2007 is estimated value

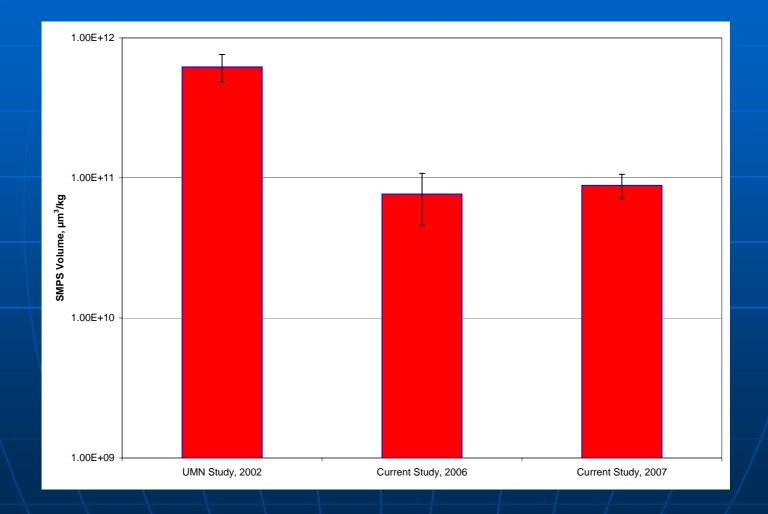
Comparison to Previous UMN Study, SMPS Size Distributions



Fuel Specific Number Concentrations 2002 and Current Study



Fuel Specific Volume Concentrations 2002 and Current Study



Comparison to Other Apportionments

Study	Particle Count	Size Range	Fuel Specific Particle Number (# km ⁻¹)			
Suuy	Instrument	Size Kange	Unapportioned	HD/Diesel		
Current Study, Year 2007*	CPC	>3 nm		$5.0\pm 0.8 x 10^{14}$		
Current Study, Year 2007*	SMPS	8-300 nm		$5.6 \pm 2.2 x 10^{13}$		
Current Study, Year 2006*	CPC	>3 nm		$1.1\pm 0.4 x 10^{15}$		
Current Study, Year 2006*	SMPS	8-300 nm		$3.8\pm 2.4 x 10^{14}$		
UMN 2002 Study, Johnson, et. al. (2005)*	CPC	>3 nm		$4.2 \pm 0.6 x 10^{15}$		
UMN 2002 Study, Johnson, et. al. (2005)*	SMPS	8-300 nm		$6.6 \pm 1.0 \mathrm{x10}^{14}$		
Imhoff et al. (2005) Birrhard Location (motorway, 120 km hr ⁻¹)	CPC	>7 nm		7.3×10^{15}		
Imhoff et al. (2005) Humlikon Location (highway, 100 km hr^{-1})	CPC	>7 nm		6.9×10^{15}		
Imhoff et al. (2005) Weststrasse Location (urban main road, 50 km hr ⁻¹)	CPC	>7 nm		5.5×10^{15}		
Abu Allaban et al. (2002)	SMPS	approx 10 to 400 nm	5.16 to 21.0×10^{13}			
Gidhagen et al. (2003)	DMPS	>10nm		5.88×10^{15}		
Gidhagen et al. (2003)	DMPS	Nuc. mode +>10nm		7.33×10^{15}		
Jamriska and Morawska (2001)	SMPS	17 to 890 nm	$1.75 \pm 1.18 x 10^{14}$			
Ketzel et al (2003)	DMPS	10-700 nm	$2.8 \pm 0.5 x 10^{14}$			
Kirchstetter et al. (1999)	CNC	>10nm		2.49×10^{15}		
Kittelson et al. (2004)	CPC	>3nm	1.9 to 9.9×10^{14}			
Kittelson et al. (2004)	SMPS	>8nm	8.7 to 22.4x10 ¹³			
Kristensson et al. (2004)	DMPS	3-900 nm	$4.6 \pm 1.9 {x10}^{14}$			
*Based on 3.2 km/kg fuel economy						

Conclusions

- Substantial Reduction in on-road nanoparticles (nuclei mode) with reduction in fuel sulfur
- Insignificant change in accumulation mode volume (mass) 2006 to 2007
- Substantial Reduction in accumulation mode volume from 2002 to 2006-2007

Acknowledgements

 Engine Manufacturers Association (EMA)
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Supplementary Material

Previous apportionment methods
Fuel sulfur levels
Future directions
Apportioned results on linear plot
References

Existing Apportionment Methods

- Road tunnel and roadside measurements, in which pollutant flux into and out of a confined space is controlled and traffic levels are directly measured (e.g. Pierson and Brachaczek, 1983; Pierson, et al., 1996; Weingartner, et al., 1997, Kirschstetter, et al, 1999; Abu-Allaban, et al., 2002; Sturm, et al., 2003; Kristensson, et al., 2004, Imhoff, et al., 2005)
- Inverse modeling of street canyon measurements, which uses a numerical model combined with street level and background measurements of particles (Wahlin, et al., 2001; Ketzel, et al., 2003).
- Mathematical models used in conjunction with stationary roadside measurements, such as the mass-balance box models (Jamriska and Morawska, 2001) and the emissions factor models of Gramotnev, et al. (2004)

From Johnson, Kittelson, Watts, 2005

Fuel Sulfur

EPA required that on-road Diesel fuel sulfur content be reduced from <500 ppm to <15ppm by October 15, 2006 Mixture of 0.4 gallons of Diesel taken from each of 10 locations on major routes into and out of Minneapolis/St. Paul metropolitan area Summer 2006, prior to regulation change: 33 ppm Sulfur by Mass Summer 2007, after regulation change: 8 ppm Sulfur by Mass

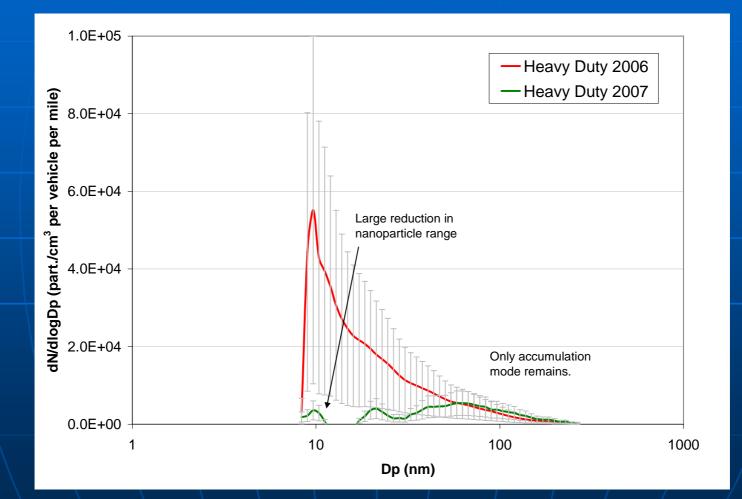
Future Directions and Future Research

- Advanced regressions that account for error in both traffic and particle/gas measurements
- Improve estimates based on regressions
- Study accumulation mode reduction as fleet changes and aftertreatment is adopted
- Study volatility and composition of nuclei mode
- Improve signal to noise ratio for light-duty vehicles
- Study effect of traffic and weather conditions

References

- Abu-Allaban, M., W. Coulomb, A. W. Gertler, J. Gillies, W. R. Pierson, C. F. Rogers, J. C. Sagebiel, and L. Tarnay. 2002. Exhaust particle size distribution measurement at the Tuscarora mountain tunnel. Aerosol Science Technology. 36, 771-789.
- Gidhagen, L., C. Johansson, J. Strom, A. Kristensson, E. Swietlicki, L. Pirjola, H.-C. Hansson. 2003. Model simulation of ultrafine particles inside a road tunnel. Atmospheric Environment 37, 2023-2036.
- Gramotnev, G., Ristovski, Z.D., Brown, R.J., Madl, P. 2004. New methods of determination of average particle emission factors for two groups of vehicles on a busy road. Atmospheric Environment 38, 2607-2610.
- Imhof, D., Weingartner, E., Ordóñez, C., Gehrig, R., Hill, H., Buchmann, B., Baltensperger, U. 2005. Real-World Emission Factors of Fine and Ultrafine Aerosol Particles for Different Traffic Situations in Switzerland. Environ. Sci. Technol., 39 (21), 8341 -8350.
- Jamriska, M. and L. Morawska. 2001. A model for determination of motor vehicle emission factors from on-road measurements with a focus on submicrometer particles. The Science of the Total Environment. 264(3), 241-255.
- Johnson, J. P., D. B. Kittelson, W. F. Watts, 2005. Source Apportionment of Diesel and Spark Ignition Exhaust Aerosol Using On-Road Data from the Minneapolis Metropolitan Area. Atmos. Environ. 39(11):2111-2121.
- Ketzel, M., P. Wahlin, R. Berkowicz, and F. Palmgren. 2003. Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations. Atmospheric Environment 37, 2735-2749.
- Kittelson, D. B., W. F. Watts and J. P. Johnson. 2004. Nanoparticle emissions on Minnesota highways. Atmospheric Environment 38, 9-19.

Apportioned Size Distributions (per unit traffic volume)



References

- Kirchstetter, E. W., R. A. Harley, N. M. Kreisberg, M. R. Stolzenburg, S. V. Herring. 1999. On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles. Atmospheric Environment 33, 2955-2968.
- Kristensson, A., C. Johansson, R. Westerholm, E. Swietlicki, L. Gidhagen, U. Wideqvist, and V. Vesely. 2004. Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden. Atmospheric Environment 38:657-673.
- Pierson, W. R. and W. W. Brachazek. 1983. Particulate matter associated with vehicles on the road. 11. 1983. Aerosol Science and Technology 2, 1-40.
- Pierson, W. R., A. W. Gertler, N. F. Robinson, J. C. Sagebiel, B. Zielenska, G. A. Bishop, D. H. Stedman, R. B. Zweidinger, and W. D. Ray. 1996. Real-world automotive emissions – summary of studies in the Fort McHenry and Tuscarora mountain tunnels. Atmospheric Environment 30, 2233-2256.
- Sturm, P.J., Baltensperger, U., Bacher, M., Lechner, B., Hausberger, S., Heiden, B., Imhof, D., Weingartner, E. Prevot, A.S.H., Kurtenback, R., Wiesen, P. 2003. Roadside measurements of particulate matter size distribution. Atmospheric Environment 37, 5273-5281.
- Wahlin, P., F. Palmgren, and R. Van Dingenen. 2001. Experimental studies of ultrafine particles in streets and the relationship to traffic. Atmospheric Environment 35(S1), S63-S69.
- Weingartner, E., C. Keller, W. Stahel, H. Burtscher, and U. Baltensperger. 1997. Aerosol emission in a road tunnel. Atmospheric Environment 31, 451-462.