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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Overview

- 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

What happens here?

Nuclei Mode - Usually forms from volatile precursors as exhaust dilutes and cools.

Accumulation Mode - Usually consists of carbonaceous agglomerates and adsorbed material.

Coarse Mode - Usually consists of reentrained accumulation mode particles, crankcase fumes.

In some cases this mode may consist of very small particles below the range of conventional instruments, Dp < 10 nm.
MEL Capabilities and Features

- 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
  - $CO_2$
  - $CO$
  - $NO_x$ (NO, NO$_2$)
- Calibration with HEPA filters, nebulized DOS, span and zero gases

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

Typical Test Route
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 non-varying 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

<table>
<thead>
<tr>
<th></th>
<th>2006 Avg</th>
<th>2006 SD</th>
<th>2007 Avg</th>
<th>2007 SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel Sulfur, ppm</td>
<td>33</td>
<td>--</td>
<td>8</td>
<td>--</td>
</tr>
<tr>
<td>Temperature, C</td>
<td>27</td>
<td>0.6</td>
<td>26.1</td>
<td>0.5</td>
</tr>
<tr>
<td>MEL Speed, mph</td>
<td>56.9</td>
<td>3.6</td>
<td>57.7</td>
<td>3.8</td>
</tr>
<tr>
<td>Weekday Heavy Duty by Vehicle, %</td>
<td>9.8</td>
<td>1</td>
<td>12</td>
<td>0.8</td>
</tr>
<tr>
<td>Weekday Light Duty by Vehicle, %</td>
<td>90.2</td>
<td>1</td>
<td>88</td>
<td>0.8</td>
</tr>
<tr>
<td>Weekend Heavy Duty by Vehicle, %</td>
<td>1.9</td>
<td>0.9</td>
<td>2.3</td>
<td>0.1</td>
</tr>
<tr>
<td>Weekend Light Duty by Vehicle, %</td>
<td>98.1</td>
<td>0.9</td>
<td>97.7</td>
<td>0.1</td>
</tr>
</tbody>
</table>
Roadway Size Distributions

Reductions in nucleation mode range where most number is found

No change in accumulation mode range where most mass is found
Apportioned Heavy-Duty Size Distributions (per unit traffic volume)

Reductions in nucleation mode range where most number is found

No change in accumulation mode range where most mass is found
Apportioned Heavy Duty, Fuel Specific Size Distributions*

Reductions in nucleation mode range where most number is found

No change in accumulation mode range where most mass is found

*Problem with CO₂ analyzer discovered late in study, CO₂ for 2007 is estimated value
Comparison to Previous UMN Study, SMPS Size Distributions

Only SMPS data available from previous study
Fuel Specific Number Concentrations 2002 and Current Study

UMN Study, 2002
Current Study, 2006
Current Study, 2007

Fuel Specific Particle Concentration, particles/kg

3025A CPC SMPS
Fuel Specific Volume Concentrations
2002 and Current Study

SMPS Volume, µm³/kg

URN Study, 2002
Current Study, 2006
Current Study, 2007
## Comparison to Other Apportionments

<table>
<thead>
<tr>
<th>Study</th>
<th>Particle Count Instrument</th>
<th>Size Range</th>
<th>Fuel Specific Particle Number (# km⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Current Study, Year 2007*</td>
<td>CPC</td>
<td>&gt;3 nm</td>
<td>5.0 ± 0.8x10¹⁴</td>
</tr>
<tr>
<td>Current Study, Year 2007*</td>
<td>SMPS</td>
<td>8-300 nm</td>
<td>5.6 ± 2.2x10¹³</td>
</tr>
<tr>
<td>Current Study, Year 2006*</td>
<td>CPC</td>
<td>&gt;3 nm</td>
<td>1.1 ± 0.4x10¹⁵</td>
</tr>
<tr>
<td>Current Study, Year 2006*</td>
<td>SMPS</td>
<td>8-300 nm</td>
<td>3.8 ± 2.4x10¹⁴</td>
</tr>
<tr>
<td>UMN 2002 Study, Johnson, et. al. (2005)*</td>
<td>CPC</td>
<td>&gt;3 nm</td>
<td>4.2 ± 0.6x10¹⁵</td>
</tr>
<tr>
<td>UMN 2002 Study, Johnson, et. al. (2005)*</td>
<td>SMPS</td>
<td>8-300 nm</td>
<td>6.6 ± 1.0x10¹⁴</td>
</tr>
<tr>
<td>Imhoff et al. (2005) Birrhard Location</td>
<td>CPC</td>
<td>&gt;7 nm</td>
<td>7.3x10¹⁵</td>
</tr>
<tr>
<td>Imhoff et al. (2005) Humlikon Location</td>
<td>CPC</td>
<td>&gt;7 nm</td>
<td>6.9x10¹⁵</td>
</tr>
<tr>
<td>Imhoff et al. (2005) Weststrasse Location</td>
<td>CPC</td>
<td>&gt;7 nm</td>
<td>5.5x10¹⁵</td>
</tr>
<tr>
<td>Abu Allaban et al. (2002)</td>
<td>SMPS</td>
<td>approx 10 to 400 nm</td>
<td>5.16 to 21.0x10¹³</td>
</tr>
<tr>
<td>Gidhagen et al. (2003)</td>
<td>DMPS</td>
<td>&gt;10nm</td>
<td>5.88x10¹⁵</td>
</tr>
<tr>
<td>Gidhagen et al. (2003)</td>
<td>DMPS</td>
<td>Nuc. mode + &gt;10nm</td>
<td>7.33x10¹⁵</td>
</tr>
<tr>
<td>Jamriska and Morawska (2001)</td>
<td>SMPS</td>
<td>17 to 890 nm</td>
<td>1.75 ± 1.18x10¹⁴</td>
</tr>
<tr>
<td>Ketzel et al (2003)</td>
<td>DMPS</td>
<td>10-700 nm</td>
<td>2.8 ± 0.5x10¹⁴</td>
</tr>
<tr>
<td>Kirchstetter et al. (1999)</td>
<td>CNC</td>
<td>&gt;10nm</td>
<td>2.49x10¹⁵</td>
</tr>
<tr>
<td>Kittelson et al. (2004)</td>
<td>CPC</td>
<td>&gt;3nm</td>
<td>1.9 to 9.9x10¹⁴</td>
</tr>
<tr>
<td>Kittelson et al. (2004)</td>
<td>SMPS</td>
<td>&gt;8nm</td>
<td>8.7 to 22.4x10¹³</td>
</tr>
<tr>
<td>Kristensson et al. (2004)</td>
<td>DMPS</td>
<td>3-900 nm</td>
<td>4.6 ± 1.9x10¹⁴</td>
</tr>
</tbody>
</table>

*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)
- Dr. Nick Barsic, John Deere
- Minnesota Department of Transportation (MNDOT)
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

Apportioned Size Distributions (per unit traffic volume)

- Heavy Duty 2006
- Heavy Duty 2007

Large reduction in nanoparticle range

Only accumulation mode remains.
References