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GASOLINE VEHICLE EXHAUST PARTICLE SAMPLING STUDY

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ABSTRACT

The University of Minnesota collaborated with the Paul Scherrer Institute, the University of Wisconsin (UWI) and Ricardo, Inc to physically and chemically characterize the exhaust plume from recruited gasoline spark ignition (SI) vehicles. The project objectives were:

- Measure representative particle size distributions from a set of on-road SI vehicles and compare these data to similar data collected on a small subset of light-duty gasoline vehicles tested on a chassis dynamometer with a dilution tunnel using the Unified Drive Cycle, at both room temperature (cold start) and 0 C (cold-cold start).
- Compare data collected from SI vehicles to similar data collected from Diesel engines during the Coordinating Research Council E-43 project.
- Characterize on-road aerosol during mixed midweek traffic and Sunday midday periods and determine fleet-specific emission rates.
- Characterize bulk- and size- segregated chemical composition of the particulate matter (PM) emitted in the exhaust from the gasoline vehicles.

Particle number concentrations and size distributions are strongly influenced by dilution and sampling conditions. Laboratory methods were evaluated to dilute SI exhaust in a way that would produce size distributions that were similar to those measured during laboratory experiments.

Size fractionated samples were collected for chemical analysis using a nano-microorifice uniform deposit impactor (nano-MOUDI). In addition, bulk samples were collected and analyzed.

A mixture of low, mid and high mileage vehicles were recruited for testing during the study. Under steady highway cruise conditions a significant particle signature above background was not measured, but during hard accelerations number size distributions for the test fleet were similar to modern heavy-duty Diesel vehicles. Number emissions were much higher at high speed and during cold-cold starts. Fuel specific number emissions range from 10^{12} to 3×10^{16} particles/kg fuel. A simple relationship between number and mass emissions was not observed.

Data were collected on-road to compare weekday with weekend air quality around the Twin Cities area. This portion of the study resulted in the development of a method to apportion the Diesel and SI contribution to on-road aerosol.

INTRODUCTION

Figure 1 illustrates relationships between idealized trimodal Diesel aerosol number, mass weighted size distributions [1], and an alveolar deposition curve [2]. A spark-ignition (SI) size distribution is similar, but has a mass median diameter that is typically smaller. For Diesel aerosol, the nuclei mode typically contains < 10% of the particle mass, but > 90% of the particle number. Most of the mass is composed of carbonaceous agglomerates and adsorbed materials; it is found in the accumulation mode. The coarse mode contains 5-20% of the mass. Nuclei mode particles emitted by either Diesel or SI engines are usually composed of nearly all-volatile material [3- 6].

Epidemiological and laboratory studies have linked environmental exposure to particles that are less than 2.5 µm in size with adverse health effects [7- 11]. These studies have elucidated a range of causal mechanisms, but have not developed a quantitative understanding of their relative importance. These studies have not, for the most part, focused on determining the physical characteristics of the aerosol to which people are exposed. Regulatory agencies, such as the U.S. Environmental Protection Agency (EPA), have adopted mass-based air pollution regulations for particulate matter, but other metrics, such as particle number or surface area, may also be important in characterizing the physical properties of aerosol related to health effects [12].

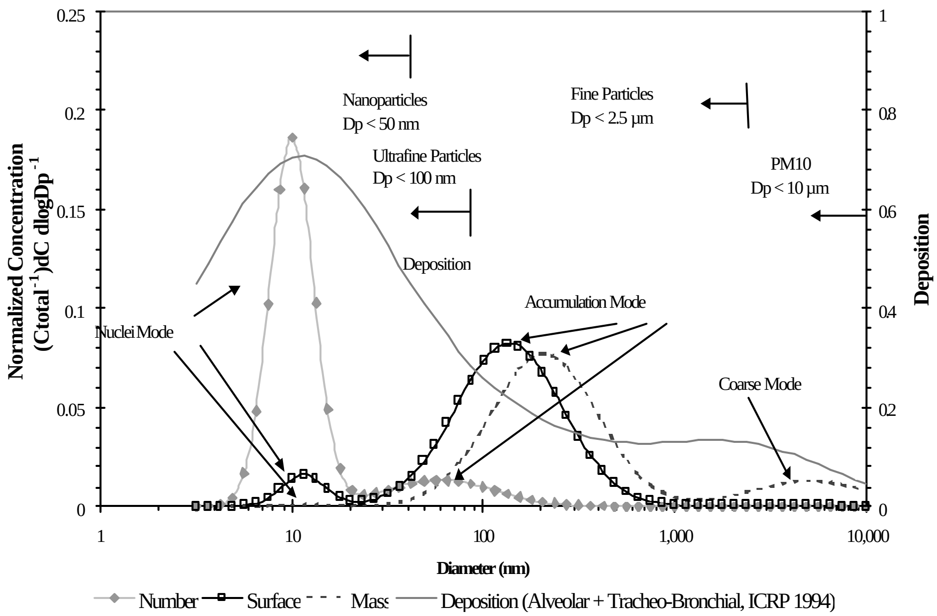

Figure 1. Typical Diesel mass and number weighted size distributions shown with alveolar deposition

Laboratory studies [4, 13-20] have shown that nanoparticle emissions from SI engines are much more speed- and load-dependent than Diesel engines, and that SI engines emit a higher proportion of smaller particles than do Diesels. Measurements from recent model SI vehicles, over the FTP and ECE driving cycles, showed tailpipe particulate matter emissions to be very low [19]. Mass emission rates from a light duty truck ranged from 4.3 mg/km, during the cold-start phase of the FTP drive cycle, to < 0.062 mg/km, during phase 2, for nearly half of the vehicles reported. All of the conventional gasoline vehicles in this study emitted < 1.24 mg/km particulate matter, compared to the current 50 mg/km particulate matter standard. The mean particle size for all vehicles tested ranged from 35 - 65 nm; by mass, it ranged from 100 - 300 nm. Minor changes in distribution were observed between vehicles. The number of particles emitted over the transient test conditions ranged from 3×10^{10} - 7.9×10^{13} particles/phase. Peak particle emissions occurred principally during short periods of heavy acceleration and coincide with peak hydrocarbon, CO, and NOx emissions.

The Northern Front Range Air Quality Study [21, 22] found that vehicle exhaust was the largest PM2.5 total carbon (TC) contributor, constituting about 85% of PM2.5 carbon at sites in the Denver metropolitan area. Sources with emissions similar to light-duty-gasoline vehicles contributed about 60% of PM2.5 TC at urban Denver sites; these contributions were 2.5-3 times the Diesel exhaust contributions. Emissions were particularly high during cold, cold starts and from vehicles identified as "smokers." Still, Diesels contributed the largest share of the "elemental" carbon (EC) at these locations.

Uncertainty exists regarding the relative contribution of atmospheric aerosol from SI- and Diesel-exhaust in real-world conditions. Diesels emit more particulate matter per vehicle; but, because SI vehicles, at least in the U.S., account for most of the vehicles operating on-road, the direct PM emissions from SI engines may be more important [22]. More than three times as much gasoline is consumed in the U.S. compared to Diesel fuel for transportation purposes.

The project, which is described in detail elsewhere [23], was carried out over a 31-month period, beginning in December 2000, and had the following objectives:

- Measure representative particle size distributions from a set of on-road SI vehicles and compare these data to similar data collected on a small subset of light-duty gasoline vehicles tested on a chassis dynamometer with a dilution tunnel using the Unified Drive Cycle (UDC), at both room temperature (cold start) and 0 C (cold-cold start).
- Compare data collected from SI vehicles to similar data collected from Diesel engines during the Coordinating Research Council E-43 project [6].
- Characterize on-road aerosol during mixed mid-week traffic and Sunday mid-day periods and determine fleet-specific emission rates.
- Characterize bulk- and size- segregated chemical composition of the particulate matter (PM) emitted in the exhaust from the gasoline vehicles.

This paper gives an overview of the project and summarizes the major findings.

MOBILE EMISSIONS LABORATORY AND INSTRUMENTATION

The University of Minnesota mobile emissions laboratory (MEL) was used to collect on-road air quality data. Data were collected while driving on Minnesota highways under normal traffic conditions and during chase experiments in which the MEL followed test vehicles. The sample air intake was located 69 cm above the highway and 43 cm in front of the MEL. Total sample-line length was less than 11 m, with a flow rate of 400 L/min through a 10.2 cm diameter tube; the calculated penetration efficiency for 10 nm size particles was greater than 95%. A manifold distributed the sample air to the instruments.

The TSI 3934 scanning mobility particle sizer (SMPS) was used to determine the number, surface area, and volume size distributions. The SMPS consists of a TSI 3071A electrostatic classifier and a TSI 3025A condensation particle counter (CPC). It was used to classify particles by an electrical mobility equivalent diameter. The SMPS was configured to cover the size range of 8 to 283 nm in the high flow mode (10 L/min sheath air flow and 1.5 L/min aerosol flow). The high-flow mode was used to reduce internal particle losses.

A standalone TSI 3025A CPC was used to determine the total number concentration for particles ranging in size from about 3 to 1,000 nm. TSI reported a counting efficiency of the 3025A to be 50 % for a particle size of 3 nm, which has been confirmed by at least two studies [24, 25]. The 3025A CPC measures a maximum concentration of 9.99×10^4 particles/cm^3, so it was used with a leaky-filter dilutor. The leaky filter dilutor is a passive dilutor and is dependent upon the instrument flow rate. A glass capillary tube was placed inside an absolute capsule filter to create a leak through the filter. The diameter of the capillary tube determined the dilution ratio. Several capillaries, with different diameters, were made to attain different dilution ratios ranging between 16 and 600:1. To insure proper mixing after the aerosol passed through the leaky filter and before passing to the CPC, a mixing orifice was introduced into the sampling line. The dilution ratio was established using a polydisperse ammonium sulfate aerosol, both with and without the leaky filter dilutor in line. A nebulizer was used to spray the polydisperse ammonium sulfate aerosol to the particle instruments on a daily basis to check the consistency of the size distributions and number concentrations.

The total particle number concentrations measured by the 3025A CPC and calculated from the SMPS were different because the 3025A CPC counts particles as small as 3 nm, while the SMPS counts (or measures) down to only 8 nm. This situation is analogous to PM10 and PM2.5, where the metric being measured is the same but the cutoff size is different.

A nano-MOUDI [26] was used to determine the aerosol mass size distribution and to collect size fractionated samples for chemical analysis during the laboratory phases of the project. The nano-MOUDI is a modification of the microorifice uniform deposit impactor (MOUDI) developed previously [27]. The nano-MOUDI had the following impactor plate cut-sizes: 18, 10, 5.62, 3.20, 1.80, 1.00, 0.56, 0.32, 0.18, 0.10, 0.056 µm, plus three additional low-pressure impactor stages with cut-sizes at 0.032, 0.018, and 0.010 µm, followed by an after filter. The MOUDI stages were rotated using a MOUDI turner. The nano-MOUDI stages were not rotated. The 10 upper stages of the nano-MOUDI were operated at a flow rate of 30 L/min, while the lower 4 stages had a flow rate of 10 L/min. Two pumps were required to operate the nano-MOUDI.

Different types of substrates and after filters were used in the project to collect samples for chemical analysis. These included 37 mm ungreased Al foil or Teflon filter substrates and 47 mm ultra pure quartz fiber or Teflon filters. The array of substrates and filters in a nano-MOUDI sample was pre-determined based upon the method of chemical analysis.

Additional aerosol instruments were used in the project. These included the photoelectric aerosol sensor (PAS), diffusion charger (DC), and electrical low pressure impactor (ELPI). Details of these instruments are presented elsewhere [23].

A variety of ambient and exhaust gas analyzers were also used throughout the project. These included non-dispersive infrared (NDIR) CO_2 and CO analyzers, and a chemiluminescence NOx analyzer. These instruments have a response time of about 1 s, and were used to determine background concentrations and the exhaust dilution ratio. Each instrument was zeroed and spanned daily according to manufacturers' instructions and 10 point calibrations were done every six months. The quality assurance protocol was developed during the Coordinating Research Council E-43 project and is reported elsewhere [28, 29].

ON-ROAD CHASE EXPERIMENTS

The MEL was used to capture plumes from five of the test vehicles shown in Table 1. These included the Windstar, F-150, Tracer, Voyager, and Caprice. The Voyager and Caprice were high mileage cars and emitted visible emissions under some driving conditions. An automotive repair shop conducted a safety inspection and determined that the vehicles were safe to drive. However, neither the Voyager nor the Caprice was in a "well-maintained" condition. For comparison, Table 1 also shows information for vehicles tested at the chassis dynamometer.

Table 1. Test vehicle data

Car	Class	Model year	Engine displacement, L	Mileage, mi	Transmission	Estimated weight, kg	Tested[1]
F-150	Truck	1999	4.6	29,500	Automatic	2273	OR,CD
Prism	Compact	1999	1.8	56,600	Automatic	1250	CD
Windstar	Van	1998	3.8	46,805	Automatic	1932	OR,CD
Tracer	Compact	1995	1.9	104,000	Manual	1307	OR
Escort	Compact	1995	1.9	>140000	Manual	1307	CD
Voyager	Van	1989	3.0	157,933	Automatic	1704	OR
Caprice	Full-size	1984	5.0	>119,000	Automatic	1932	OR

[1] OR = On-road, CD = chassis dynamometer

The test conditions included highway cruise (approximately 88 km/hr), hard acceleration (80 to 120 km/hr) and various idle conditions. The SI tests were conducted on flat roads with two people in the front seat and 68 kg of sand in the trunk. To be included in the analysis plume samples had to meet three criteria.

- The ratio of the plume total integrated SMPS particle number concentration to the background total integrated SMPS number concentration had to be greater than 2:1.
- The plume dilution ratio had to be $\geq$ 100:1 and $\leq$ 5,000:1.
- The data set had to be sufficiently complete to allow data analysis.

It should be emphasized that our tests were conducted on a limited fleet of vehicles under warm weather conditions. The influence of cold weather and unusually high emitting vehicles was not evaluated in this project. For the chase tests, the average daily temperature ranged from 20 to 30^{o}C, the dew point ranged from 7 to 24^{o}C, and wind speed ranged from 10 to 23 km/h.

Under steady highway cruise conditions, we were not able to measure a significant particle signature above background for any of the cars in the test fleet. However, during hard accelerations, our test fleet produced size distributions (Figure 2) that were surprisingly similar to modern heavy-duty Diesel vehicles (Figure 3). Generally, gasoline vehicles have somewhat lower particle number concentrations in the upper end of the accumulation mode where most of the particle mass is found, thus, they have lower mass emissions.

Figure 4 shows fuel specific number emissions for the five test vehicles that ranged from 2×10^{14} to 3×10^{16}. It is evident from the chart that many particles were below the level of detection of the SMPS but were counted by the CPC. This indicates that emissions of extremely small particles, with particle diameters >3 nm and < 10 nm, were often more than an order of magnitude higher than emissions of particles with diameters > 10 nm.

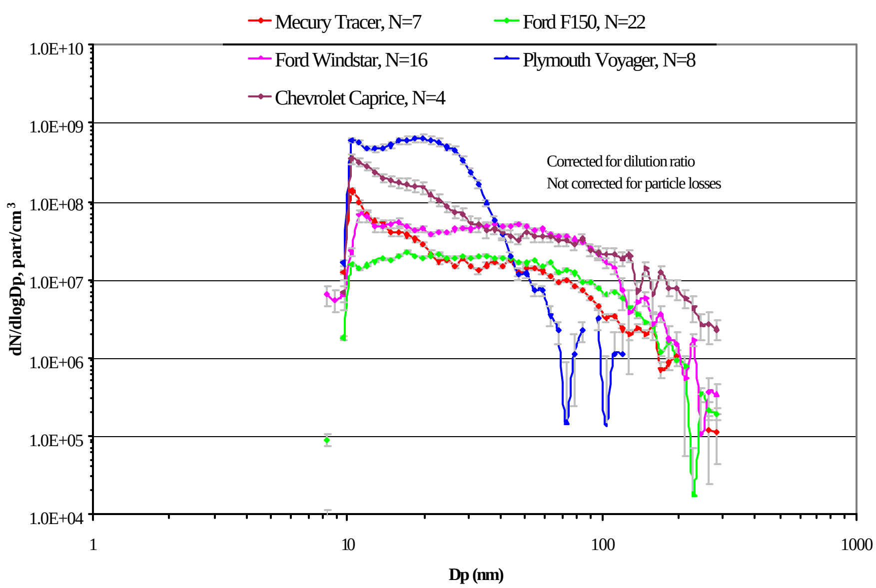

Figure 2. Average SI acceleration size distribution with the standard deviation of the mean (SDOM) and sample size (N) by car corrected for background and dilution ratio

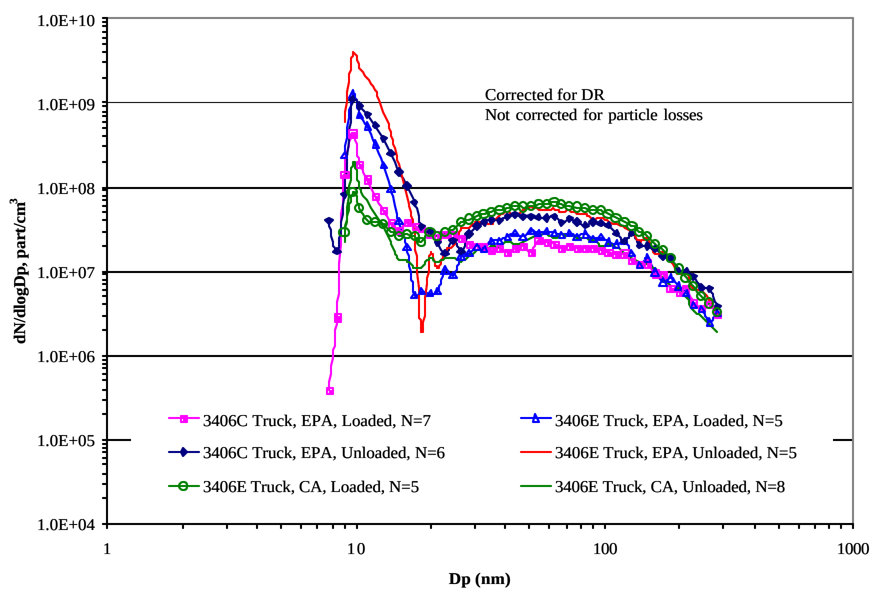

Figure 3. Average Diesel acceleration size distribution with the sample size (N) by truck (3406E electronically controlled, 3406C mechanically controlled) corrected for background and dilution ratio. Tests were conducted using two fuels EPA certification fuel and California Diesel fuel. (Data from [6])

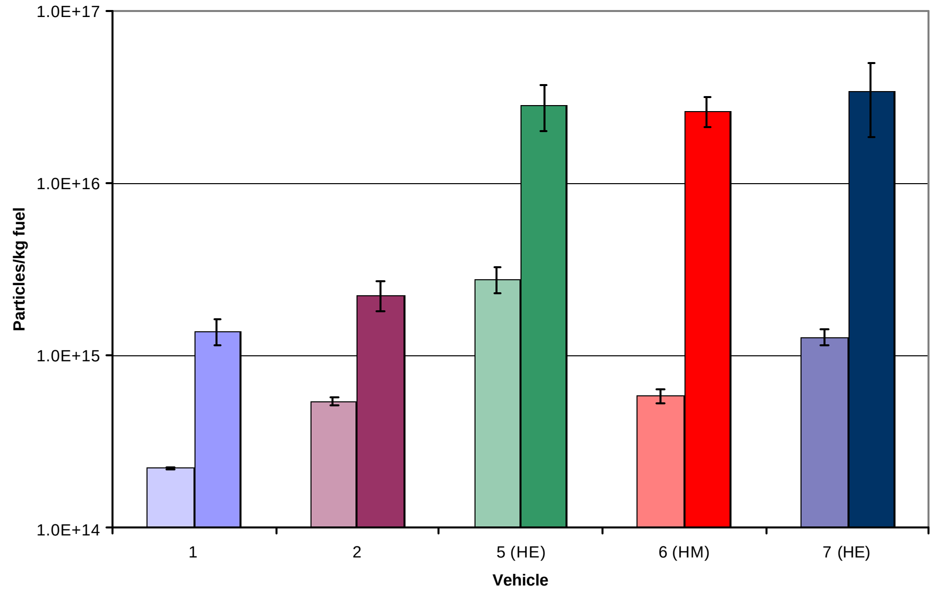

Figure 4. Number emissions measured during hard acceleration. CPC solid dark fill and SMPS lighter hatched fill. Cars 5 and 7 were heavy emitters (HE) and car 6 had high mileage (HM). Error bars are SDOM.

CHASSIS DYNAMOMETER EXPERIMENTS

As stated previously, the objective of the chassis dynamometer experiments was to compare information obtained from the on-road chase tests to laboratory data on a subset of vehicles. Both steady-state and Unified Drive Cycle (UDC) data were obtained. The UDC data were obtained under cold-cold start (0^o C) and hot start conditions. The same instruments were used as in the on-road tests, plus additional gas analyzers were used to measure raw and diluted concentrations in the CVS tunnel. A specially designed dilutor was used for these tests with CVS dilution occurring at the tailpipe to minimize loss of particle precursors.

Approximately four hours of steady-state operation followed during which filter samples and nano-MOUDI samples were collected for chemical analysis. Two more cold-cold start UDCs were run on the second day with a cold soak period of about 5 hr between tests. Then, another car was installed on the chassis dynamometer. Standard protocols were followed for the conduct of each test.

Figure 5 shows that number emissions were much higher for cold-cold UDC tests, particularly with low mileage vehicles. The fuel specific emissions ranged from $6x10^{13}$ to $3x10^{15}$ part/kg fuel.

Calculated size distributions for the hot and cold-cold UDC are shown in Figure 6. The distributions were obtained by running the

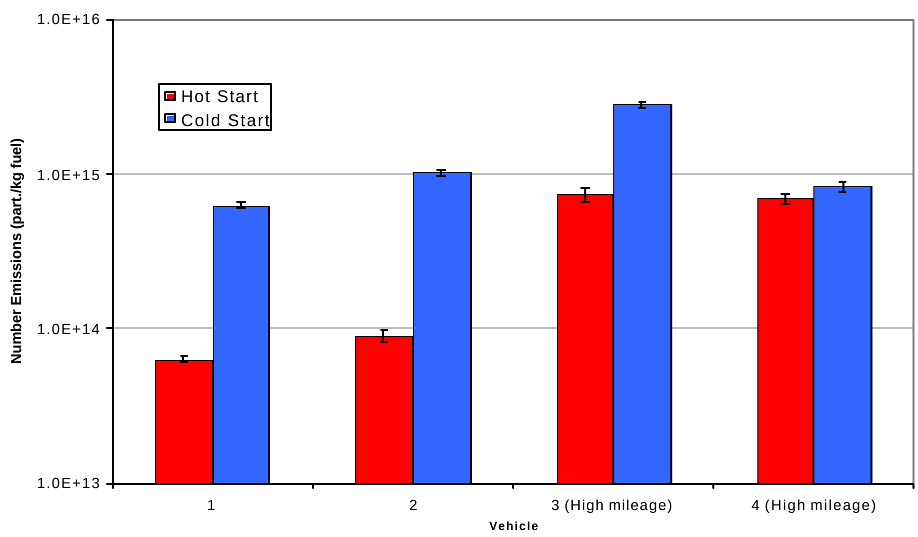

Figure 5. Chassis dynamometer average CPC number emissions for cold-cold UDC test

On the first day of testing for each car, a cold-cold start UDC cycle was run after soaking the vehicle at 0 C until the oil temperature was 0^o C $\pm$ 2 C. Immediately following the completion of the first cold-cold start UDC, three hot start UDCs were conducted.

SMPS in the single channel mode for 10, 32 and 100 nm size particles and fitting the size distributions using a standard routine developed at the UMN. The measured points are shown as symbols in Figure 6. These distributions are not corrected for particle losses, dilution ratio, or the background concentration. The highest concentrations, both in the nuclei and accumulation modes, are for the cold-cold start UDC. Higher accumulation mode concentrations are consistent with higher mass emissions that were expected during a cold-cold start test. The hot start emissions were much lower and, based upon the CPC data, decreased from test to test; this is reasonable since the engine and exhaust system would be warmer for each subsequent test. Figure 7 shows SMPS data from one set of three cold-cold start UDC tests conducted on the Escort. Note that the highest 10 nm particle emissions occured at the highest vehicle speed, which was about 113 km/hr and that the largest particles were emitted early in the test cycle. Particle number emissions for SI vehicles are very dependent upon engine load conditions.

The steady-state conditions were composed of about 2 hrs at 105 km/hr, 1 hr at 56 km/hr, and a one hour period composed of 15 min periods of idle, 113, 32, and 113 km/hr segments. Figure 8 shows that number emissions ranged from 10^{12} to $3x10^{16}$ particles/kg fuel and that number emissions were higher at high speed. Data not shown, but included in the final report for this project [23], suggest that the highest number emissions are associated with storage and release of volatile material from the exhaust system. The CPC consistently measured more particles than the SMPS since the CPC counted particles down to 3 nm, compared to 8 nm for the SMPS.

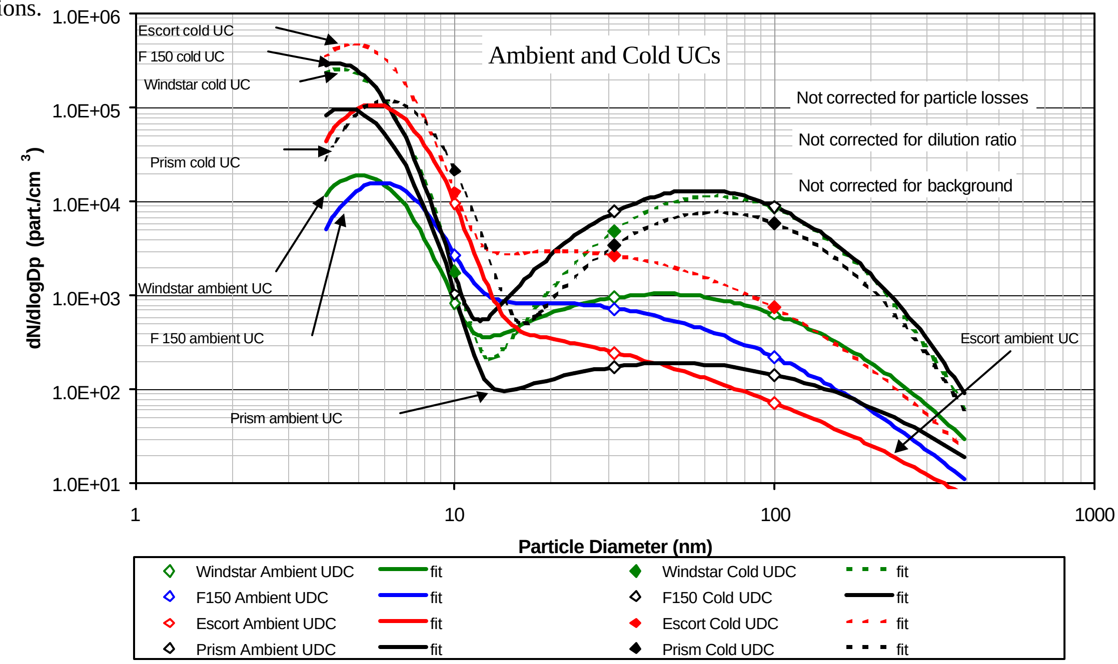

Figure 6. Calculated SMPS size distributions for the UDC tests

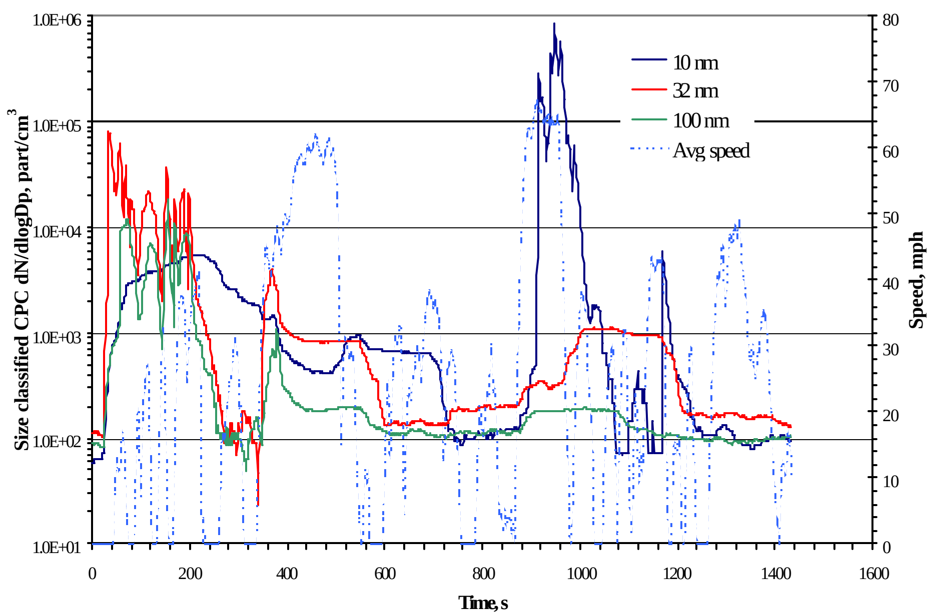

Figure 7. Size classified UDC number emissions obtained from the SPMS operated in the single channel mode

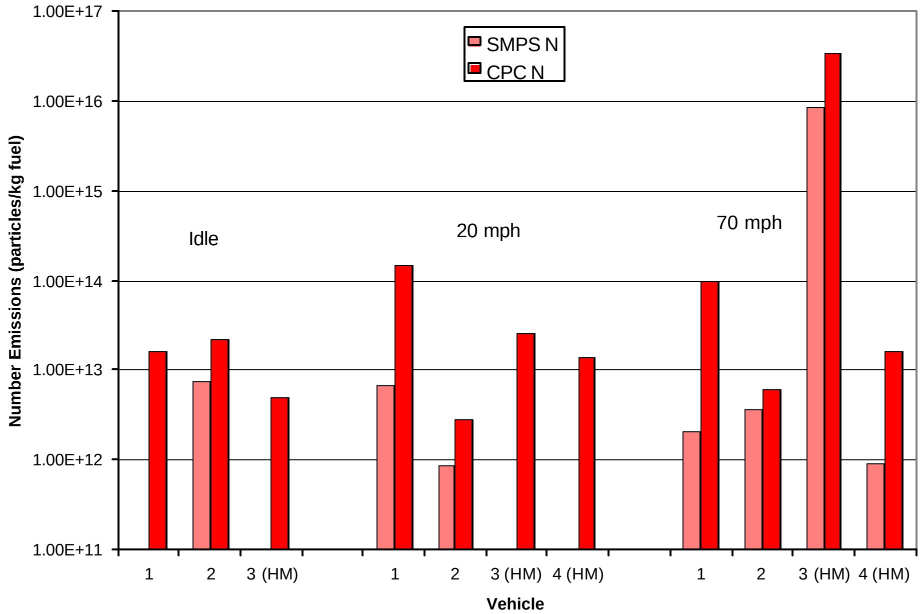

Figure 8. Steady-state number emissions for four test vehicles including high mileage (HM) vehicles

Filter and size selective impactor samples were collected for chemical analysis by the University of Wisconsin. Complete details of the analysis and results from these samples are found elsewhere [30]. Mass emissions from the three composited cold-cold UDC tests ranged from 1 to 7 mg/km for the test fleet. The elemental carbon fraction varied from 30 to 60% of the total mass in these tests. One vehicle was an unusual emitter. It produced the lowest mass on the cold-cold start UDC tests, but was the highest number emitter (Figure 5). However, it produced higher mass emissions during the steady-state test periods. Data reported elsewhere [30] show that the bulk of the emitted mass was composed of ammonium and sulfate ions in roughly stoichiometric proportions. This illustrates that there is no simple relationship between mass and number emissions.

ON-ROAD CHARACTERIZATION OF REAL WORLD FLEETS

The MEL collected gaseous and particulate matter air quality data while driving on an interstate highway route in the Minneapolis and Saint Paul metropolitan area. Measurements were made for CO_2, CO, NOx, particle number, surface area, and aerosol number size distribution. Measurements were made on both weekdays (Tuesdays and Thursdays) and on weekends (Sundays). By exploiting the difference in the relative volumes of heavy duty (mostly Diesel vehicles) and light-duty vehicles (mostly SI vehicles) over the roadway from weekdays to weekends, we were able to apportion gas and particle production to its source.

The results of this apportionment show that on a weekday, weekly weighted basis, the majority of particle number can be attributed to heavy-duty Diesel traffic. Weekend production of particles can be attributed to light-duty SI sources. On a per vehicle basis, heavy-duty Diesel vehicles produced substantially greater numbers of particles. When compared on a per kg of fuel burned basis, heavy-duty vehicle particle production was higher than that of

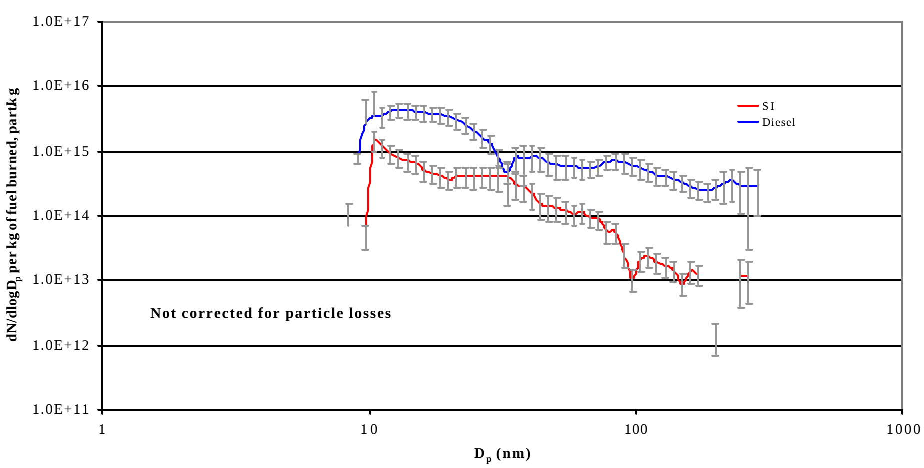

Figure 9. Diesel and SI apportioned size distributions on a fuel specific basis

light-duty vehicles.

Figure 9 shows the Diesel and SI apportioned size distributions on a fuel specific basis. Diesels emitted more particles per mass of fuel burned than SI vehicles. This method of comparison allows the results to be compared with other data that we have collected, as shown in Figure 10. This method reduces the impact of any error in traffic counts in the apportionment method. Complete details of the apportionment method and data collection and analysis process are reported elsewhere [23].

The heavy-duty Diesel-apportioned size distribution is remarkably similar to size distributions measured in real world and laboratory studies (Figure 10). The light-duty apportioned size distribution is bounded by laboratory and real world studies. This result seems to indicate that the apportioned size distribution is a combination of size distributions produced during steady-state and transient testing.

In Figure 11, the SI apportionment distribution has a nuclei mode below the acceleration, 65 and 70 mph cruise conditions and an accumulation mode that lies between the acceleration and 65 and 70 mph cruise conditions. The shape of the SI apportionment distribution is qualitatively similar to that of the chase acceleration. The apportioned SI distribution also falls in the middle of the chase and lab conditions for the continuous instruments as shown by data in Table 2. It is notable that the apportioned SI production of small particles, N30/N = 0.769, where N30 is the number of particles $\leq$ 30 nm in diameter and N is the total number of particles, was significantly smaller than the laboratory, where N30/N equaled 0.9995, 0.998, respectively. The high speed chassis dynamometer tests showed a large nuclei mode during the first few minutes of operation, which gradually disappeared over time leaving only the accumulation mode. It may be the case that the preceding low speed engine conditions deposited volatile material on the vehicle's exhaust system that was

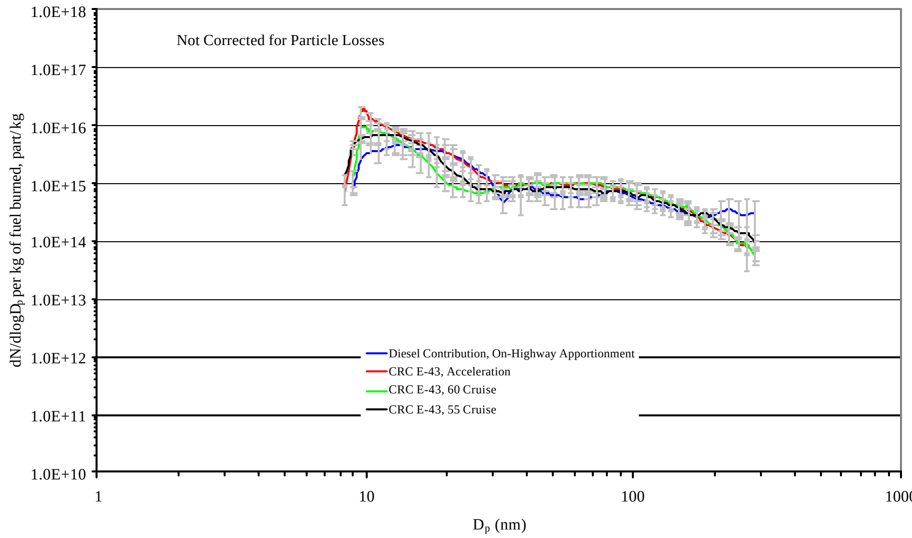

Figure 10. Comparison of on-highway fuel-specific Diesel apportionment results to comparable fleet average conditions from CRC E-43 Diesel testing

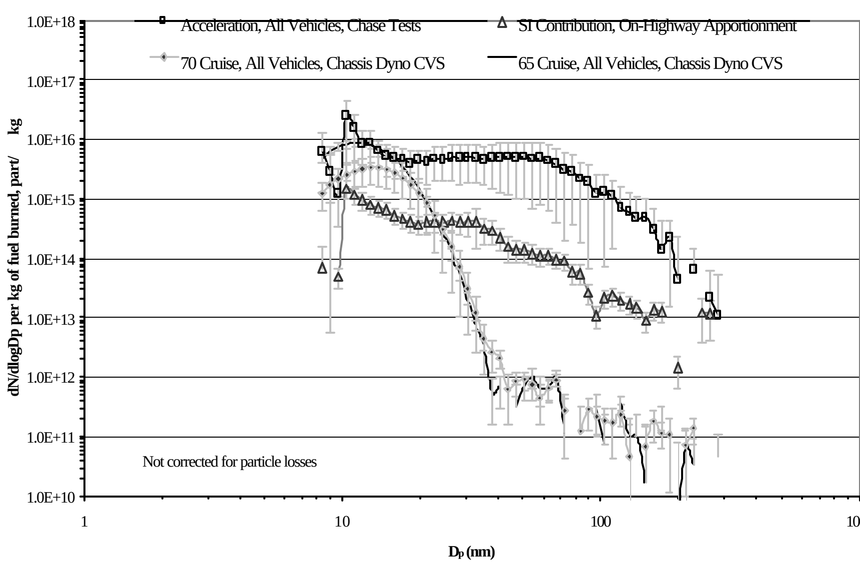

Figure 11. Comparison of on-highway fuel-specific SI apportionment results to comparable fleet average conditions from chase and chassis dynamometer testing

Table 2. Fuel-specific statistics for the size distributions shown in Figure 10 and the corresponding continuous instrument data. Statistics in gray box are not statistically significant to a 95% confidence level.

Phase	Name	CPC (part/kg)		SMPS Number (part/kg)		DC (μm^2/kg)		SMPS Active Surface (μm^2/kg)	
		Mean	SDOM	Mean	SDOM	Mean	SDOM	Mean	SDOM
On-Highway	Diesel Contribution	1.34E+16	1.86E+15	2.13E+15	3.04E+14	3.15E+12	6.15E+11	1.45E+13	2.15E+12
CRC E-43	55 Cruise, All Vehicles	1.04E+16	2.64E+15	2.69E+15	1.42E+14	2.39E+13	5.13E+12	1.28E+13	4.80E+11
CRC E-43	60 Cruise, All Vehicles	7.20E+15	1.88E+15	2.48E+15	2.57E+14	1.43E+13	2.51E+12	1.29E+13	7.06E+11
CRC E-43	Acceleration, All Vehicles	2.04E+16	5.28E+15	3.88E+15	2.58E+14	2.70E+13	6.03E+12	1.35E+13	8.48E+11
On-Highway	SI Contribution	7.10E+15	1.55E+15	3.88E+14	6.11E+13	1.77E+11	3.91E+10	9.41E+11	1.52E+11
Chase	Acceleration, All Vehicles	1.04E+17	5.05E+16	6.02E+15	9.32E+14	2.84E+13	1.89E+13	2.60E+13	4.43E+12
CD	65 Cruise, All Vehicles, CVS	9.27E+15	1.25E+15	2.47E+15	1.10E+14	3.08E+11	5.42E+10	1.26E+12	5.69E+10
CD	70 Cruise, All Vehicles, CVS	4.19E+15	1.74E+15	1.04E+15	1.47E+14	2.50E+11	1.39E+11	6.48E+11	9.74E+10

Phase	Name	PAS (fA/kg)		SMPS Volume (μm^3/kg)		DGN (nm)		DGV (nm)	
		Mean	SDOM	Mean	SDOM	Mean	SDOM	Mean	SDOM
On-Highway	Diesel Contribution	2.91E+12	4.04E+11	6.21E+11	1.40E+11	22.7	1.7	195.7	12.1
CRC E-43	55 Cruise, All Vehicles	2.89E+12	7.87E+11	4.30E+11	2.39E+10	18.7	0.4	166.1	3.5
CRC E-43	60 Cruise, All Vehicles	2.67E+12	7.57E+11	3.93E+11	2.60E+10	19.6	1.1	152.3	3.8
CRC E-43	Acceleration, All Vehicles	1.48E+12	2.35E+11	3.75E+11	2.75E+10	17.0	0.5	148.3	3.9
On-Highway	SI Contribution	5.69E+11	1.26E+11	1.44E+10	4.44E+09	19.3	0.7	104.0	24.3
Chase	Acceleration, All Vehicles	8.57E+12	3.72E+12	4.35E+11	9.82E+10	23.2	2.8	95.3	10.9
CD	65 Cruise, All Vehicles, CVS	6.88E+09	1.53E+09	3.16E+09	1.54E+08	12.2	0.1	15.1	0.7
CD	70 Cruise, All Vehicles, CVS	1.13E+10	1.60E+09	1.84E+09	2.94E+08	13.4	0.4	16.9	1.3

Phase	Name	N/V Ratio		N_{30}/N Ratio		V_{30}/V Ratio	
		Mean	SDOM	Mean	SDOM	Mean	SDOM
On-Highway	Diesel Contribution	3.44E+03	7.76E+02	0.760	0.125	0.008	0.002
CRC E-43	55 Cruise, All Vehicles	6.25E+03	4.79E+02	0.794	0.067	0.009	0.001
CRC E-43	60 Cruise, All Vehicles	6.32E+03	7.76E+02	0.743	0.128	0.007	0.001
CRC E-43	Acceleration, All Vehicles	1.03E+04	1.02E+03	0.834	0.086	0.015	0.002
On-Highway	SI Contribution	2.70E+04	7.69E+03	0.769	0.086	0.059	0.017
Chase	Acceleration, All Vehicles	1.38E+04	3.78E+03	0.620	0.168	0.022	0.007
CD	65 Cruise, All Vehicles, CVS	7.81E+05	5.17E+04	0.9995	0.063	0.986	0.067
CD	70 Cruise, All Vehicles, CVS	5.66E+05	1.21E+05	0.998	0.200	0.967	0.221

burned off during the high-speed conditions. The first few minutes of high particle production may have skewed the fleet average distribution. Alternatively, the CVS (laboratory) testing, itself, may have overestimated the production of the smallest particles, when compared to the real world where we were unable to capture a plume under steady-state conditions. Also, shown in Table 2, are the number to volume ratio (N/V) and the V30/V. The V30/V ratio is the volume of particles $\leq$ 30 nm in diameter to total volume and DGN and DGV, which are the geometric mean number and volume diameters, respectively. An N/V ratio > 10^4 indicates the presence of a nuclei mode [31]. The Diesel/SI ratios for the CPC, SMPS active surface and SMPS volume data in Table 2 show that, proportionately, Diesels contribute more large particles, consequently more mass, whereas SI engines contribute a greater number of small particles.

The principal uncertainties in these estimates are attributable to traffic counts and background correction. Further, the measurements were made during the summer months and would not account for higher emissions that would be expected during colder periods. We view these results as preliminary, but think that an expansion of this method could be useful in apportioning real-world mobile source pollution, without the need for traffic controls or the artificial confinement of the study to roadway tunnels.

CONCLUSIONS

- Modern in-use SI vehicles have significant mass emissions (1-7 mg/km) in the cold-cold start (0 C) UDC.
    - o EC emissions in these test cycles were surprisingly high, typically 30-60% of PM2.5 mass
- Emissions of extremely small particles (Dp < 10 nm) at high road speed may be very high (10^{14} - 10^{16} part/kg fuel) even for nominal low emitters. Storage and release play an important role in the formation of these particles.
- Normal emitting SI engines under high speed and load conditions may emit number emissions that equal or exceed Diesel levels
- On-road source apportionment shows promise in determining emission factors for Diesel and SI engines in on-road, real-world fleets.

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