

Submicrometer and Supermicrometer Particulate Emission from Spark Ignition Vehicles

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Particulate emissions from 11 gasoline-powered and 2 liquefied petroleum (LPG)-powered passenger vehicles were characterized during the Accelerated Simulation Mode driving cycles on a chassis dynamometer. The test fleet consisted of 10 catalyst-equipped vehicles operated with unleaded gasoline (5 Ford Falcons and 5 Holden Commodores), 2 LPG-powered vehicles (both Ford Falcons), and 1 older type nongasoline vehicle operated with leaded gasoline. Particulate characterization included determination of total particulate number concentration and size distribution using the scanning mobility particle sizer (SMPS) and the aerodynamic particle sizer (APS). The average particle number concentrations in the SMPS range for all modes was lower for Ford Falcons and somewhat higher for Commodores, with values of 1.5×10^4 and $4.1 \times 10^4 \text{ cm}^{-3}$, respectively. This difference is significant and was observed for all modes. The number concentration levels were higher for the LPG-fueled cars ($8.4 \times 10^4 \text{ cm}^{-3}$) and for the leaded gasoline-powered vehicle ($7.9 \times 10^5 \text{ cm}^{-3}$). There was not a significant variation in particle count median diameter in the SMPS and the APS ranges, either for different operating conditions of the vehicles investigated or between different vehicle groups. The observed size distributions were bimodal with average values of CMD ranging from 39.1 to 60.2 nm in the SMPS range and from 0.9 to 1.4 μm in the APS range. The results obtained from this study can be used as a first-order estimation toward emission inventories for vehicle groups included in the investigations.

Introduction

Exhaust emissions from spark ignition engine consist of gaseous and particulate phases. The main gaseous emissions include hydrocarbons (HC), CO, NO_x, CO₂, SO₂, and water vapor. Gasoline particulate matter is formed as a result of incomplete combustion of gasoline. The particles are mostly carbonaceous spherical submicron agglomerates ranging from 10 to 80 nm, consisting of a carbon core with various associated organic compounds. The main components of particulate phase include soot; ash; trace elements such as lead, iron, chlorine, and bromine; organic compounds; and low to medium boiling fractions of engine oil (1). Lubricating

oil and other fuel hydrocarbons may also contribute. The sulfate particles present in gasoline engine emission are mainly from catalyst-equipped vehicles utilizing unleaded gasoline (2). Common organic compounds are polycyclic aromatic hydrocarbons (PAHs), such as pyrene, chrysene, benzo[a]pyrene, and BaP.

Particle emissions from vehicles are currently under close scrutiny with respect to their contribution to ambient particles relative to other sources. The PM₁₀ fraction of the particles (mass concentration of particles smaller than 10 μm) has been linked to various health effects (3–7). The size distribution of these particles plays an important role as it influences the depth of penetration and deposition in the lung and hence the toxicological effects. The lack of knowledge of the size distribution makes it difficult to explain the toxicological mechanism related to the reported health effects. However, there is evidence of association between concentrations of particles and changes in a number of respiratory health indicators ranging from changes in lung function to hospital admissions and death.

There is significantly less information available on particulates from gasoline spark ignition emissions than from diesel emissions. Gasoline particulate matter is emitted at levels lower than a milligram per kilometer. Because it is emitted at such low levels, it is difficult to measure accurately. Total particulate mass emissions from spark ignition vehicles are significantly lower than diesel emissions and are usually well below any emission standards (1, 8–10). It has been noted, however, that while particulate emissions from individual spark ignition vehicles are lower than individual emissions from diesel vehicles, the total contribution of both types of vehicles to air emissions could be similar due to the exceedingly higher number of spark ignition vehicles than diesel-fueled vehicles (11). The low mass concentration of emissions from spark ignition engines is related to a lower number concentration in the emissions and also to the fact that the emitted particles are smaller than particles from diesel emissions (12).

Both air quality standards and current vehicle emissions legislation are based on mass measurements: the former on mass per unit volume (PM₁₀ and PM_{2.5}), and the latter on the total mass of particles emitted per kilometer. In both, there is no reference either to the size of the particles or to the number concentration of emitted particles. The available emissions data are limited and scattered. Most of the available data are on particle mass emission (8–11, 13), and there is only limited data on the size distribution of these particles or on the particle number emissions (9, 10), either in the submicrometer or supermicrometer region. Insufficient data on critical factors related to spark ignition emissions, including particulate emissions, have been identified as the most important issue in developing emission inventories (14, 15).

An important factor requiring further investigations and consideration in developing in-service vehicle testing procedures is documented test-to-test variability in emissions (16). The author showed that malfunctioning vehicles, which are among the highest emitters, often show very inconsistent emissions and are likely to escape emission testing if these are not frequent enough. Those vehicles are most often modern computer-controlled vehicles that have malfunctioning emission control systems that have not been tampered with.

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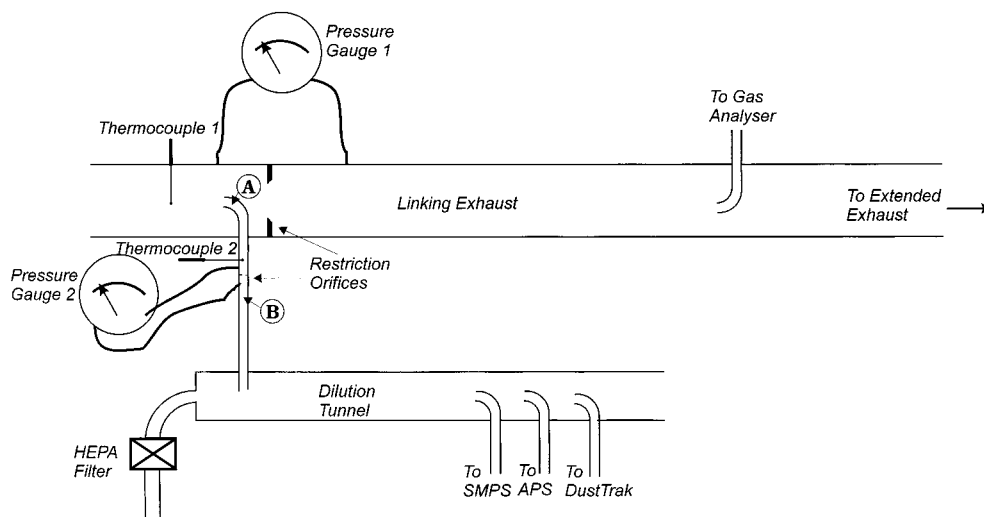


FIGURE 1. Exhaust sampling and dilution system.

An additional support for the importance of the focus on individual vehicles was provided by Small and Kazimi (17), who conducted cost estimates of various aspects of pollution control and showed that emissions from individual vehicles should be the prime target.

The aim of this work was to characterize particle emissions from a range of spark ignition vehicles, fueled with different fuels (unleaded and leaded gasoline and LPG), using a portable sampling line that allowed sampling for particulate sizing instruments. The ultimate objective of the investigation was to provide information on the concentration and size distributions of fine particulates in the exhausts of spark ignition vehicles operating under different load conditions as well as to provide indications as to the main factors affecting particle emissions (such as vehicle make, age, or maintenance record). The size distributions were measured in a broad range of particle sizes from about 0.01 to 30 μm . The comprehensive knowledge of spectral characteristics of petrol particle emissions is essential for conducting emission inventories as well as for developing better technologies for lowering of the emissions. The literature data on this topic are still very limited. With the small number of vehicles tested within the framework of this study, the outcome of the study can only be used as an estimate toward emission inventories and cannot be used for drawing global conclusions on emission trends. The main significance of the study is the identification of relationships between emissions in the different size ranges and the variation of the relationship between different vehicles. This information can be directly used for optimization of particle emission testing procedures as well as for developing strategies toward lowering the emissions of fine particles.

The full scope of investigations included measurements of a number of other emission characteristics including gaseous emissions (CO_2 , CO, HC, and excess O_2). Due to the large volume of information collected, this paper will focus only on particle size distribution characteristics, while relationships between other emission parameters measured as well as between different methods of particle measurements will be reported separately.

Exhaust Measurement and Sampling

The system used for measurement of petrol exhaust emission is a modification of a design used previously for measurements of diesel exhaust emissions (18). The main differences that had to be accommodated in the modified design were significantly lower flow rates and lower particle concentrations for petrol exhaust emission measurements as compared with diesel emission measurements.

A diagram of the sampling system developed for these studies is presented in Figure 1. The vehicle exhaust was connected to the primary sampling segment where the primary sample was taken and temperature and other measurements were made. It also contained a restriction orifice. The primary sampling segment was a stainless steel tube of 100 mm diameter connected to the vehicle exhaust by a flexible tube of similar diameter so that different vehicle heights could be accommodated. The distance between the car exhaust and the sampling port was of the order of 1 m. The primary sampling segment continued to the extended exhaust, a tube of larger diameter to convey exhaust gases away from the work area without introducing backpressure. The volumetric flow rate of the total exhaust gas was measured by the pressure difference across the restriction orifice in the primary sampling segment and the flow of the exhaust sample by the pressure difference across the restriction orifice in the primary sample line. The sample flow was introduced to the dilution tunnel where it mixed with a constant flow of ambient air filtered through HEPA filters.

The experimental system design was based on variable dilution sampling, where the clean air flow rate in the dilution tunnel was held constant while the sample flow rate varied with different vehicle operating conditions, resulting in varying dilution ratios. Changes in dilution ratio can potentially affect the coagulation rate and hence the particle concentration and count median diameter. It was considered, and later confirmed experimentally, that the concentration levels in the dilution tunnel would not exceed 10^6 cm^{-3} . It was estimated that for such concentrations and for residence times of less than 10 s, the effect of changes in dilution ratio on number concentrations should not exceed 1% (19). The variable dilution sampling system has an advantage of being relatively cheap to construct, but the disadvantage is that it requires adjustments for different vehicles sampled and different sampling conditions.

The volumetric flow rate of the total exhaust was determined from the pressure difference across the restriction orifice in the primary sampling segment using the magnehelic gauge 1. The temperature of the exhaust tube was measured with thermocouple 1. The flow rates through the primary sampling line varied between 1 and $3 \times 10^3 \text{ L min}^{-1}$. As the distance between the car exhaust and the sampling port was about 1 m, the residence time in this section was less than 1 s at the smallest flow rate. The temperature and flow rate of the exhaust sample were measured with thermocouple 2 and magnehelic gauge 2 connected across the restriction orifice in the exhaust primary sampling segment. Clean air was supplied to the dilution tunnel by pump 1 and filtered

TABLE 1. Specifications of the Vehicles Tested

| vehicle type | vehicle code | year of manufacture | odometer reading (km) | fuel | capacity/configuration |
|---------------------------------|--------------|---------------------|-----------------------|----------|------------------------|
| Ford Falcon, station wagon | 1, 10 | 24/7/95 | 45 019 | unleaded | 4, fuel injected |
| | 2 | 30/6/95 | 41 053 | unleaded | |
| | 3 | 23/6/95 | 62 901 | unleaded | |
| | 4 | Aug 1995 | 37 999 | unleaded | |
| | 13 | 27/6/97 | 4 403 | unleaded | |
| Holden Commodore, station wagon | 5 | 22/2/95 | 24 042 | unleaded | 3.8, v6, fuel injected |
| | 8 | 7/11/95 | 25 482 | unleaded | |
| | 9 | 27/3/97 | 3 913 | unleaded | |
| | 12 | 25/7/95 | 25 388 | unleaded | |
| Holden Acclaim, sedan | 6 | 1995 | 34 000 | unleaded | |
| Ford Falcon, sedan | 11 | pre 1992 | | LPG | 4, carburetor |
| | 15 | pre 1992 | | LPG | |
| Ford, utility | 14 | 1976 | | leaded | 5.2, carburetor |

with HEPA filters. The mixture of the sample and clean air was passed via the dilution tunnel to the sampling ports for the particle sizing instruments (scanning mobility particle sizer and aerodynamic particle sizer). The velocity and temperature of the diluted exhaust were measured with a TSI velocity meter.

The flow rate of the clean air in the dilution tunnel was 180 L min⁻¹. Considering that the flow rate of an average exhaust sample was usually around 5 L min⁻¹, the total flow rate of the mixture in the dilution tunnel was about 185 L min⁻¹. Small changes in the flow rate of the exhaust in the primary sample line would make little difference to the total flow rate and hence to the linear velocity in the dilution tunnel. Once isokinetic conditions were established, the sampling was approximately isokinetic over the whole sample range. The linear flow rate in the dilution tunnel also matched the flow in the sampling lines to the particle sizing instruments.

Particle Size Distribution Instruments. *Scanning Mobility Particle Sizer.* The scanning mobility particle sizer (SMPS) and the aerodynamic particle sizer (APS) are instruments that measure the size distribution of a sampled aerosol. The SMPS operates within a window in the range 0.0075–1 μm, the window size depending on the value of the sample and sheath air flow rates that are selected. For these measurements, a window of 0.015–0.7 μm was selected as the optimum to cover the number distribution spectra for most of the vehicle tested in most of the operation modes. The SMPS consists of the 3071A TSI electrostatic classifier and the 3010 condensation particle counter. Before entering the electrostatic classifier, the sample passes through an inertial impactor in order to remove larger particles. In the electrostatic classifier, the sample passes through a charger that imparts a bipolar equilibrium charge level to the particles. The aerosol charged in this way enters the tube of the differential mobility analyzer where a fraction of the particles is selected according to its electrical mobility. The selected fraction of the particles is counted in the condensation particle counter and expressed as particle concentration. Throughout the sampling process the voltage on the electrostatic classifier ramps. This corresponds to a continuous scan of mobility and therefore particle size. The process of measurement is controlled with an interfacing computer that does the necessary calculations, controls the classifier, and stores the data supplied by the counter.

Aerodynamic Particle Sizer. The aerodynamic particle sizer (APS) counts particles in the size range 0.5–30 μm. The principle of operation of this instrument involves measuring the acceleration of aerosol particles in response to the accelerating flow field created by the inlet nozzle. The particle response to the flow accelerating field is dependent on its

inertia and the aerodynamic size. As the particle leaves the nozzle its time-of-flight over a given distance depends on its aerodynamic size. The time-of-flight of the particle is found from the scattered light that appears when the particle crosses the laser beams.

It must be noted that each of the two instruments (SMPS and APS) is based on a different principle, supplies data based on different assumptions, and does so for a different size range. Both instruments essentially produce particle number concentration. If assumptions are made concerning the sphericity of the particles and the particle density, surface area, volume and mass concentration, and distributions may be computed from the original number distributions. It should be noted that this has to be done with some caution as some chain aggregates may be formed.

The Measurement Procedure

Vehicles. Ten in-service Falcon and Commodore cars from the Queensland University of Technology fleet, two LPG Falcon taxis, and one privately owned, older Falcon Utility were selected for testing. The tests were conducted on the chassis dynamometer of Zillmere Dyno Tuning in Brisbane. The characteristics of the vehicles tested are summarized in Table 1. Cars 1 and 10 denote the same vehicle.

The unleaded vehicles were all run for the tests on the same unleaded test fuel supplied by Unleaded Fuel AMPOL Ltd. LPG vehicles were run on fuel from the same batch, while the older Falcon Utility was run on commercially available leaded petrol fuel.

Definition of Test Cycles. The only available test cycle for passenger vehicles in Australia is the ADR 37. This is a transient cycle that was developed for gas emissions and not for particulates. As measurement of particulates requires certain finite time, a steady test cycle is preferred to a transient one. The only available steady test cycle for passenger vehicles was the five-mode steady cycle available from the U.S. EPA Final ASM Test Procedure Documentation (U.S. EPA, 1996), which was adopted and modified for the purpose of this study. Modifications were needed in the length of the test (which was 3 min) at each mode, as the minimum time required for a measurement at each mode was 5 min.

The measurements were performed for each of a set of operating modes defined by engine power and road speed. The vehicle operation modes are presented in Table 2.

Measurements of the exhaust volume and sampling for particulates and gas analysis were performed as shown in the modal sequence from 1 to 5. Having attained the mode conditions of power and speed, the sampling tunnel (dilution tunnel) was flushed with clean air for 30 s. This was considered to be sufficient time to clean the tunnel after the previous tests and prepare it for the new test. Repeat

TABLE 2. Definition of Modes

| mode | dynamometer load (kW) | dynamometer speed (km h ⁻¹) |
|-------------------------------|---|---|
| ASM 5015 | 10.6 (6 cylinder station wagon) 10.7 (6 cylinder sedans) | 25 |
| ASM 2525 | 9.1 (6 cylinder station wagon) 9.2 (6 cylinder sedan) | 40 |
| steady mode at road load | depending on the vehicle mass | 80 |
| idle mode in drive (break on) | 0 | 0 |
| idle mode in neutral | 0 | 0 |

measurements at a single mode point were conducted. Between three and five samples were collected with the SMPS, and 10 samples were collected with the APS. Readings from the magnehelic gauges, from which sample flow rates were calculated, were averaged over the last minute. The average duration of each mode was about 5 min.

The temperature of the exhaust, the temperature of the exhaust primary sample, and the temperature in the sampling line were taken at the beginning and at the end of each mode, and the readings were averaged.

Results and Discussion

All tested cars were ordered into three groups according to the type of the car/engine. The first group represents the Ford Falcons with a 4.0-L fuel-injected engine, the second group is the Holden Commodores with a 3.8-L fuel-injected engine, and the last group represents the two Ford Falcons running on LPG. In addition to the three groups, one old Falcon Utility running on leaded petrol fuel was tested for comparison. The first two groups of cars, Ford Falcons and Holden Commodores, were fitted with a catalytic converter and ran on unleaded gasoline.

The data collected with both instruments (SMPS and APS) have been analyzed in terms of particle count medium diameter (CMD), total number concentration, and particulate mass concentration, calculated from particle number concentration assuming particle density of 1 g cm⁻³.

Measurements with the SMPS. Size Distribution. A typical size distribution spectrum measured by the SMPS, in the range of 15–750 nm, for a unleaded vehicle (no. 8) is presented in Figure 2a. The spectrum was measured at 40 km h⁻¹ steady-state conditions. The horizontal axis presents particle size on a logarithmic scale, while the vertical axis represents the number of particles per cm³ of exhaust gas. A full line presents a fit to a log-normal distribution with a peak close to 50 nm. As can be seen, the correspondence between the fitted and measured values is excellent. The fitting procedure was done in order to estimate the amount of particles smaller than the lower limit of the SMPS range, 15 nm.

Figure 2b shows a typical spectrum from the SMPS for vehicle 14, which ran on leaded gasoline fuel. Sampling conditions were same as for the measurements presented in Figure 2a (ASM2525). It is important to notice that the size distribution is almost identical with a CMD of 51 and 49 nm for unleaded and leaded fueled cars, respectively. The main difference is in the total concentration, which was 7.9 × 10⁴ cm⁻³ for the unleaded vehicle and 1.79 × 10⁶ cm⁻³ for the vehicle that ran on leaded fuel, more than 200 times higher.

Figure 2c shows a size distribution spectrum for the emission from a vehicle run on LPG. The conditions were the same as for the previous two vehicles. In the vehicle that ran on LPG, the CMD is 79 nm and is higher than for the previous two vehicles. The total number concentration was higher than for the unleaded vehicle but still lower than for the leaded vehicle. It should be noted that the last two

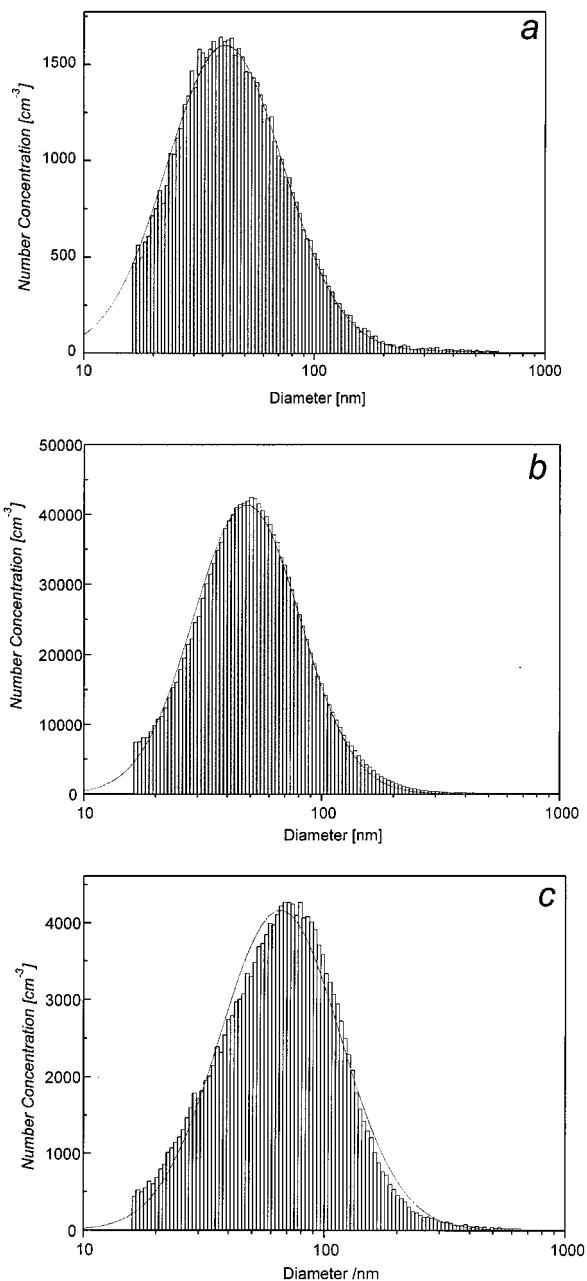


FIGURE 2. Typical size distribution spectra measured by the SMPS in the range of 15–750 nm at 40 km h⁻¹, mode 2, for a vehicle run on (a) unleaded gasoline (no. 8) (b) leaded gasoline, and (c) LPG. Vehicles, leaded and LPG, were significantly older and with a higher mileage than the unleaded vehicles. Also the leaded vehicle had a carburettor and a higher capacity engine, hence comparison should be made with caution. The plotted size distributions are generally typical for the group they present.

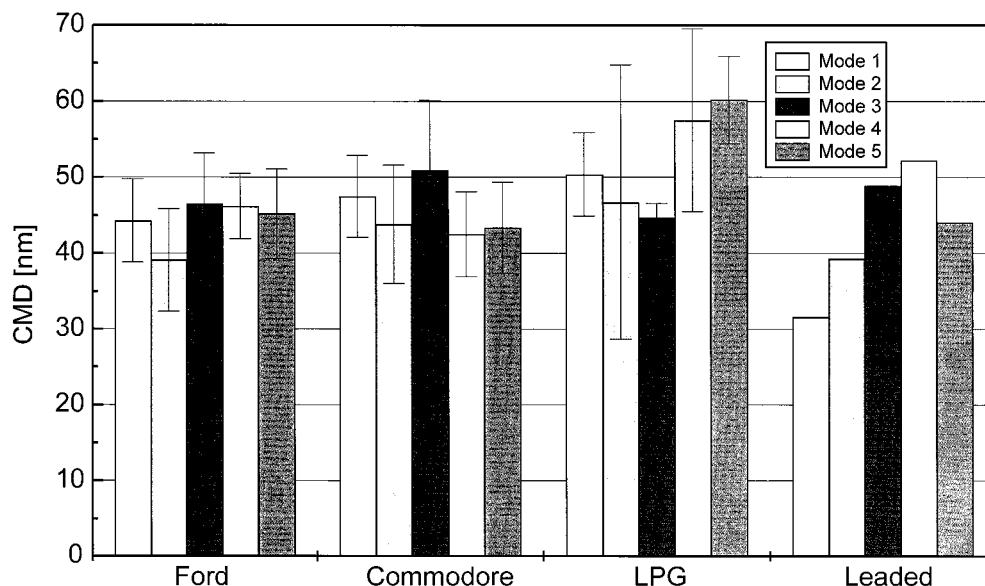


FIGURE 3. Dependence of the CMD, measured by the SMPS in the range from 15 to 700 nm, on the mode for all three groups of vehicles (Ford Falcons, Holden Commodores, and LPG) together with the leaded vehicle.

TABLE 3. Average and Standard Deviation of Total Number Concentration for Different Groups of Vehicle for Each Mode Together with Average of All Modes and Average for a Certain Mode and All Tested Vehicles

| vehicle type | total concentration (cm ⁻³) | | | | | all modes |
|-------------------|---|----------|----------|----------|----------|-----------|
| | mode 1 | mode 2 | mode 3 | mode 4 | mode 5 | |
| Ford average | 1.51E+04 | 2.28E+04 | 1.85E+04 | 9.86E+03 | 1.06E+04 | 1.54E+04 |
| SD ^a | 9.27E+03 | 9.77E+03 | 9.75E+03 | 2.95E+03 | 3.65E+03 | 4.34E+03 |
| Commodore average | 6.70E+04 | 9.03E+04 | 2.20E+04 | 1.35E+04 | 1.28E+04 | 4.11E+04 |
| SD | 2.43E+04 | 8.44E+04 | 5.45E+03 | 4.12E+03 | 3.64E+03 | 1.92E+04 |
| LPG average | 2.21E+04 | 9.32E+04 | 2.64E+05 | 2.25E+04 | 1.81E+04 | 8.40E+04 |
| SD | 1.81E+04 | 1.00E+05 | 2.04E+05 | 1.65E+03 | 1.29E+03 | 1.25E+05 |
| old Ford Ute | 2.37E+05 | 1.79E+06 | 1.90E+06 | 2.25E+04 | 5.92E+03 | 7.90E+05 |
| total average | 5.05E+04 | 1.83E+05 | 1.89E+05 | 1.39E+04 | 1.21E+04 | 8.97E+04 |

^a SD, standard deviation.

Count Median Diameters. Since the distributions generally follow a log-normal pattern, the CMD presents a better index of the size than the mean. While the CMD is not a perfect measure for the distributions with more than one peak, which is not the case in our measurements, it does provide a simple measure to detect any large shifts in the size distribution. Figure 3 illustrates the dependence of the CMD on the mode for all three groups of vehicles (Ford Falcons, Holden Commodores, and LPG) together with the leaded vehicle. There was a certain degree of variation in terms of CMD between the three groups of vehicles tested, but more variation was between individual modes and for the same mode between vehicles belonging to the same group. The average for all modes for the Commodores was 43 nm (range from 35 to 66 nm), the average for the Falcons was 45 (range from 30 to 55 nm), and the average for the LPG cars was 60 (range from 34 to 66 nm). Overall, the average values for all three groups are very close. The leaded gasoline car had a similar value of the CMD with the range from 32 to 52 nm and with the average for all modes of 44 nm. It is obvious that there is no clear trend in the CMD of emitted particles.

Number Concentration. The number concentration was measured both by the SMPS in the range from 0.015 to 0.7 μm and by the APS in the range from 0.5 to 30 μm . Total number concentration emitted in the APS range (0.5–30 μm) was smaller than the concentration emitted in the SMPS range by several orders of magnitude; therefore, the re-

mainder of the discussion focuses on the SMPS data, which covers the size range of emitted particles.

In Table 3, the average and standard deviation (SD) of the total number concentration for different groups of vehicle for each mode together with the average of all modes and the average for a certain mode and all tested vehicles are presented.

Figure 4 presents the total number concentration together with the standard deviation for each group of vehicles. On the same figure, the values for the vehicle using leaded gasoline is also presented. It is important to notice that the scaling for the leaded vehicle (right y-axis) is an order of magnitude higher than for other three groups of vehicles. The first two groups of vehicles (unleaded petrol) show the same trend with maximum emissions at 40 km h⁻¹. LPG vehicles show a similar trend to the unleaded fuel vehicles, with maximum emission for the highest speed. It is useful to notice that the second mode at 40 km h⁻¹ had a maximum load for the first two groups of vehicles.

The most consistent results in terms of the total number emissions exist for all three groups of vehicles for both drive and neutral idle modes (modes 4 and 5). There is little variation for those modes for vehicles belonging to the same group and for repeat measurements for the same vehicle. The average number concentration for the Commodores for these two modes were 1.35×10^4 and 1.28×10^4 cm⁻³, for the Falcons was 9.86×10^3 and 1.06×10^4 cm⁻³, and for the LPG-fueled cars was 2.25×10^4 cm⁻³ and 1.81×10^4 cm⁻³.

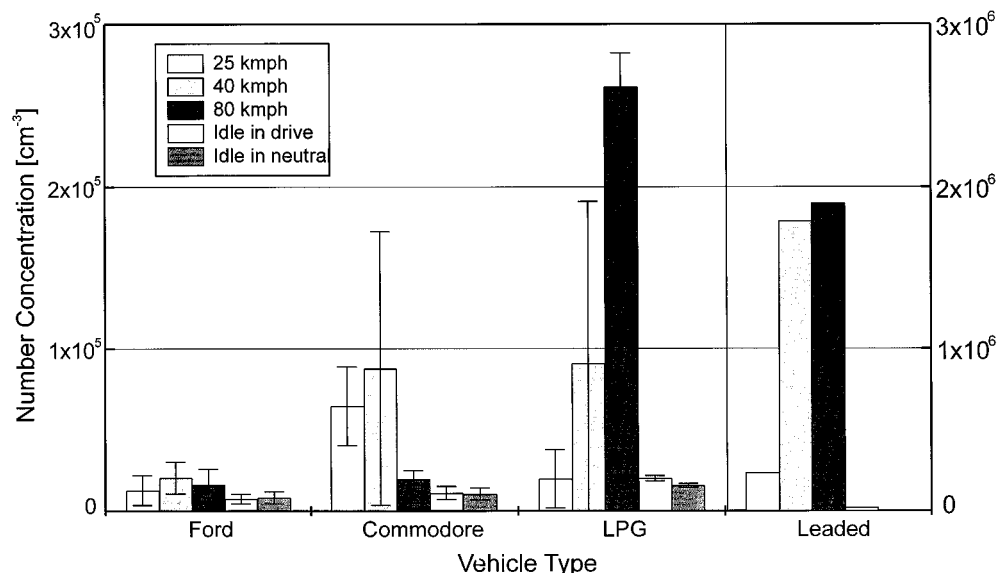


FIGURE 4. Total number concentration together with the standard deviation for each group of vehicles and for each mode. Right y-axis is for the vehicle ran on leaded gasoline.

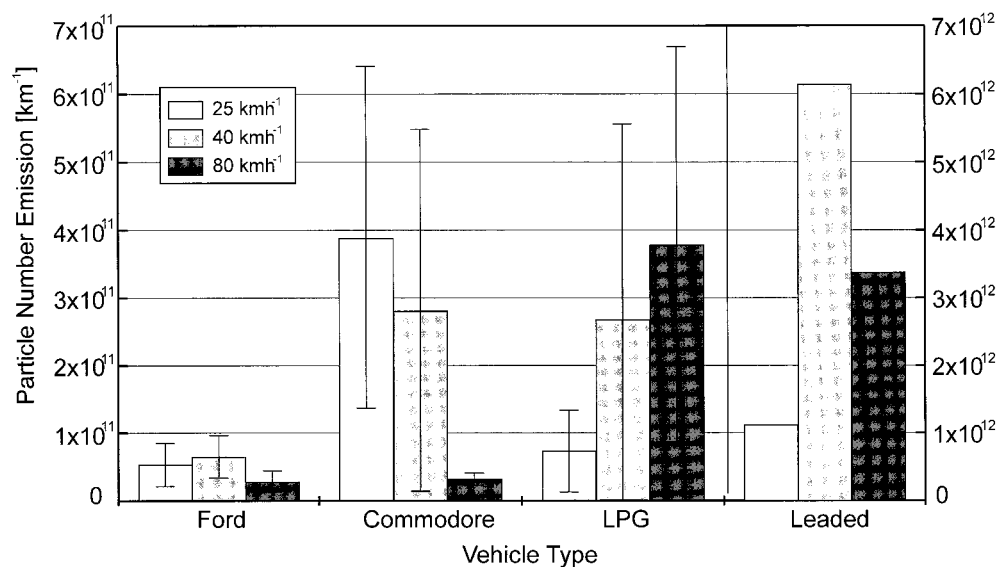


FIGURE 5. Total number of particles emitted at the tailpipe in the size range detected by the SMPS per kilometer.

Very consistent emission values were measured also for mode 3 for the Commodores and Falcons. The values of $2.2 \times 10^4 \text{ cm}^{-3}$ for the Commodores and $1.85 \times 10^4 \text{ cm}^{-3}$ for the Falcons for mode 3 are elevated in comparison with the emissions at the idle modes for these vehicles. There was slightly more variation at mode 3 between the two LPG-fueled cars, but the main feature was that emissions at this mode were significantly higher than at idle modes, and in fact the highest for these vehicles.

For the Commodores and Falcons, the highest levels of emissions, the highest variability between individual vehicles, and the lowest repeatability for the same vehicle were displayed for mode 2. The average emissions for this mode for the Commodores was $9.03 \times 10^4 \text{ cm}^{-3}$ with the range from 2.3×10^4 to $2.25 \times 10^5 \text{ cm}^{-3}$, which is a variation for an order of magnitude. The average emission for the Falcons was $2.28 \times 10^4 \text{ cm}^{-3}$ with the range from 1.24 to $3.58 \times 10^4 \text{ cm}^{-3}$. For the LPG cars, the emissions for this mode were lower than for mode 3; however, the emission values for the two vehicles tested differed by close to an order of magnitude.

The number concentration in the SMPS range for the older type vehicle using leaded fuel was an order of magnitude

higher than the average emissions from the previous three group of vehicles. The highest number concentration was achieved for mode 3, at $1.9 \times 10^6 \text{ cm}^{-3}$, and was 2 orders of magnitude higher than the total number concentration obtained with the two groups of vehicles that ran on unleaded gasoline.

Particle Emissions per Kilometer. There are no vehicle emission legislations for the particle number emissions. The only legislations that exist are for particle mass expressed in terms of the mass emitted per kilometer. The emissions of submicrometer particles can be expressed as the number of particles emitted per kilometer. Figure 5 shows the total number of particles emitted at the tailpipe, in the SMPS size range, per kilometer. Emissions per kilometer are presented only for the first three modes as modes 4 and 5 are idle with zero speed. In Figure 5, the averages together with standard deviations for each of the three groups of vehicles are presented.

Trends for particle emissions per kilometer are similar to the total concentrations, shown in Figure 4. The main difference is that in this case the emission of the Commodores is similar to the emission of the LPG cars but with opposite

TABLE 4. Number Concentrations of Emission in the APS Range for All Tested Vehicles and All Modes

| vehicle type | vehicle no. | total concentration (cm ⁻³) | | | | | all modes |
|-------------------|-------------|---|--------|--------|--------|--------|-----------|
| | | mode 1 | mode 2 | mode 3 | mode 4 | mode 5 | |
| Ford | 1 | 1.07 | 1.34 | 0.04 | 1.96 | 0.00 | 0.88 |
| Falcon | 2 | 0.60 | 0.68 | 1.28 | 1.02 | 0.84 | 0.88 |
| | 3 | 1.11 | 0.91 | 0.60 | 0.38 | 0.48 | 0.70 |
| | 4 | 0.92 | 1.30 | 2.28 | 1.37 | 0.94 | 1.36 |
| | 10 | 2.49 | 1.38 | 2.40 | 2.49 | 1.56 | 2.06 |
| | 13 | 2.33 | 1.42 | 2.24 | 1.77 | 0.67 | 1.69 |
| Ford average | | 1.42 | 1.17 | 1.47 | 1.50 | 0.75 | 1.26 |
| Commodore | 5 | 0.50 | 0.51 | 0.34 | 0.36 | 0.27 | 0.40 |
| | 6 | 0.40 | 0.32 | 0.33 | 0.32 | 0.65 | 0.40 |
| | 7 | 2.22 | 1.34 | 1.38 | 1.26 | 0.88 | 1.41 |
| | 8 | 2.82 | 2.09 | 2.93 | 1.61 | 39.70 | 9.83 |
| | 9 | 1.69 | 1.40 | 11.72 | 1.65 | 75.86 | 18.46 |
| Commodore average | | 1.52 | 1.13 | 3.34 | 1.04 | 23.47 | 0.78 |
| LPG | 11 | 1.56 | 1.83 | 2.55 | 0.98 | 0.56 | 1.49 |
| | 15 | 2.42 | 2.58 | 3.85 | 0.93 | 0.74 | 2.10 |
| LPG average | | 1.99 | 2.20 | 3.20 | 0.95 | 0.65 | 1.80 |
| leaded car | 14 | 16.96 | 4.23 | 15.82 | 4.50 | 7.00 | 9.70 |
| total average | | 2.65 | 1.52 | 3.41 | 1.47 | 9.30 | 3.67 |

trends. The highest emission for the Commodores is for mode 1, 25 km h⁻¹, while the LPG cars have the highest emission for the maximum speed of 80 km h⁻¹, mode 3. The older leaded fuel vehicle had again emissions of particles that were an order of magnitude higher than the other three groups of vehicles.

The results of the study imply that there could be a strong relationship between the level of emissions and age/generation of the vehicles. The lowest emissions were related to the youngest, new generation engines, and the highest emissions were related to the oldest vehicle investigated. While this is only a hypothesis that would need to be tested by investigations of larger sample of vehicles, including vehicles of all ages, the difference in level of emissions identified here was of an order of magnitude.

It would be also interesting to note that emissions from LPG-fueled cars, in the submicrometer range, were not lower but in fact higher than the emissions from the modern gasoline-fueled cars. It could then be a concern that the emissions from LPG fuel (which is considered a clean fuel) are not necessarily lower than gasoline emissions. Again, this hypothesis would require further investigation as both the LPG-fueled cars investigated had considerable mileage and age.

Measurements in the APS Range. The APS data refer to particle size in the range from 0.5 to 30 μm. The total number concentration averaged for all vehicles and all modes in a certain group of vehicles is shown in Table 4. The average concentrations are of the same order of magnitude as background outdoor concentrations and do not present a significant source of particles. Only 2 vehicles (nos. 8 and 9) in mode 5, which is idle in drive, showed significantly higher concentrations than usual. These two points are assumed to be outliers and were not used in the calculation of the mean value for the Commodores. Even the significantly higher emitter in the SMPS range (vehicle 11 using LPG) did not show significantly higher emissions in the APS range. Vehicle 11 had an average number concentration for all modes of 1.49 cm⁻³, which is comparable with the values for the Fords of 1.26 cm⁻³. The older vehicle that ran on leaded gasoline did not have that high emission in the APS range, about 7 times higher than the Fords, as compared to its emission in the SMPS range, where it emitted more than 50 times more than the Fords.

There was not a big variation in the CMD between all five modes or between the three groups of vehicles. The average

CMD for all modes was 1.3, 1.4, and 1.1 for the Fords, Commodores, and LPG vehicles, respectively. Only the leaded gasoline-fueled vehicle had an average CMD smaller than 1 μm.

Of importance for modeling of emission contributions from different types of vehicles is that different groups of vehicles have different contributions under different driving conditions. For example, identified in this study was a higher contribution from the Commodores and Falcons at lower speeds and LPG-fueled cars at higher speeds.

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