Submicrometer and Supermicrometer Particles from Diesel Vehicle Emissions

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There is growing evidence that fine airborne particulates could play the most important role in determining health effects. The aim of this work was to investigate the number concentration and size distributions of particulates in the exhausts of diesel vehicles (mainly buses) of different ages and make, operating under different loads. Particle-sizing instruments used were the Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS). The average particle number concentration of the exhausts was in the range $(0.7-3.9) \times 10^7$ cm^{-3} in the SMPS range (0.0075−0.304 μm) and $(0.3-32) \times 10^3$ cm^{-3} in the APS range (0.5−30 μm). In most cases, particle number concentrations increased with the increased power output from the engine and, in both SMPS and APS ranges, varied significantly within each group of vehicles, but the differences between the groups were small. For individual vehicles, there was no relation between emissions in the smaller and larger particle ranges. Emission characteristics did not appear to be correlated with engine model or age. The implications of these findings to particle emission testing and control as well as to exposure and risk analysis are discussed.

Introduction

Airborne particulate pollution has been emerging as a global problem with growing scientific evidence linking increased health risk to exposure to particulates in general and specifically to fine particulates (*1*, *2*). The size of airborne particles determines in which parts of the respiratory tract the particles are deposited (*3*, *4*); small airborne particles less than 1 μm in diameter (submicrometer particles) have a high probability of deposition deeper in the respiratory tract and are likely to trigger or exacerbate respiratory diseases. Small particles have also higher burdens of toxins, which when absorbed in the body can result in health consequences other than respiratory health effects.

In the urban environment, motor vehicle emissions are often the main anthropogenic source of air pollution, significantly contributing to the deterioration of urban air quality (*5*, *6*). Most of the research for motor vehicle particulate emissions has focused on mass concentrations in the exhaust, either as total mass concentration (*7*), or PM_{10} (mass concentration of particles smaller than 10 μm). The mass of submicrometer particles is often insignificant in comparison with the total mass of emitted particles due to the fact that a small number of large particles contribute most of the mass; submicrometer number concentrations can change by orders of magnitude without consequent changes in the total mass concentration measurements. From this approach, no information can be obtained on fine particle characteristics or on changes to fine particle emissions due to differences in vehicle operation, maintenance, or fuel characteristics (*8*, *9*).

Current emissions and air quality standards are based on the assumption that mass concentration is the critical indicator, but this assumption is no longer definitive. An example of the inadequacy of current technological and management solutions is that, even as the levels of total particulate mass concentrations are decreasing in most of the cities in the western world, concentration levels of fine particulates remain unchanged or increase. Technical solutions aimed at reductions in total particle emissions do not necessarily result in concomitant reductions in fine particle emissions; in certain cases, the opposite effect is observed, resulting in increased emissions of fine particles. An example of this is the reduction of the total mass emissions of motor vehicles in the U.S. by a factor of about 10, accompanied at the same time by an increase in the of number of fine particles emitted by a factor of about 20 (*10*).

Characteristics of Diesel Particulates. Diesel exhaust particles are mostly submicrometer agglomerates of carbonaceous spherical particles ranging from 10 to 80 nm. Larger particles contain up to 4000 individual spherical particles clustered as agglomerates up to 30 μm (*3*, *11*−*13*). A significant proportion of diesel emission particulates have aerodynamic diameters smaller than 1 μm. These particles are primarily elemental carbon, but contain also adsorbed or condensed hydrocarbons, hydrocarbon derivatives, sulfur compounds, and other materials (*3*, *9*, *13*). Solvent extractable organic components of diesel aerosols represent 5−40% of the particle mass depending on the fuel and the operating conditions (engine speed, power, torque, and temperature) of the vehicle (*9*, *14*, *15*).

Experimental Techniques for Measurements of Particle Size Spectra. The experimental techniques for measurement of particle size spectra are complicated and expensive. Such measurements must cover a broad range of particle sizes from several nanometers to at least several micrometers. Due to the differing physical characteristics, different methods have to be applied for measurements of size spectra of small particles and large particles.

Measurement of the total particulate concentration and particle size distribution of the diesel exhaust normally requires dilution of a measured proportion of the diesel exhaust flow with a measured flow of clean dry air. However, because diesel aerosols are highly dynamic and reactive nonlinear systems, their properties will be dependent on the parameters of the dilution process, namely the dilution ratio and the residence time in the dilution tunnel. In roadway situations, dilution of the diesel exhaust occurs within time periods less than 1 s, and from experimental studies, the dilution ratio is estimated to approach 1000 (*13*). In contrast, the first sampling stages of laboratory test rigs have dilution ratios in the range 5−20 and have residence times 1−5 s (*16*). The diluted exhaust is then sampled for instrument analysis

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S0013-936X(97)00826-2 CCC: $15.00
Published on Web 05/30/1998

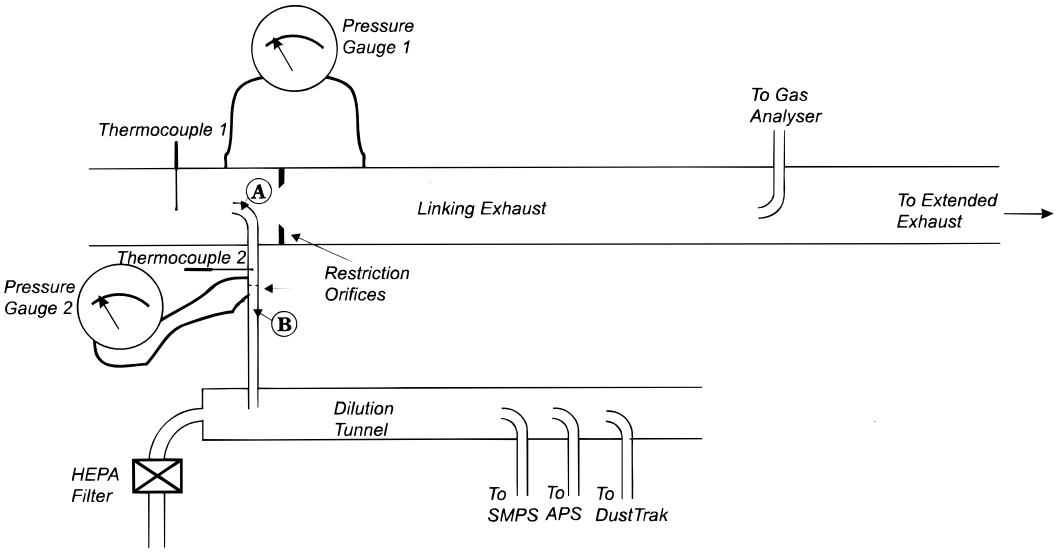

FIGURE 1. Exhaust sampling and dilution system that was built and used in measurements.

or for further dilution, since certain instruments require very low input particle concentrations. Processes taking place in the relatively concentrated streams of laboratory dilution tunnels can lead to particle size distribution and chemical compositions that are different from those produced under roadway dilution conditions.

Techniques for exhaust sampling can be divided into three major groups including (i) dilution of a significant portion of the full exhaust, (ii) dilution of small portion of exhaust sample (mini dilution tunnels), and (iii) microsampling (microdilution particulate measurement systems). While the quality of the data obtained with all methods is comparable, portability and the price of the methods vary. In general, the cost of the diesel exhaust dilution and sampling apparatus decreases with decreasing complexity and size.

Current Work. The aim of this work was to conduct measurements of size distributions and concentrations of particle emissions from a range of diesel powered buses, as well as from other diesel vehicles for comparison, using a portable sampling line designed and constructed for this study that allowed sampling for particulate sizing instruments. The size distributions were measured in a broad range of particle sizes from about 0.01 to 30 μm for the vehicles operating under differing, steady-state load conditions. The comprehensive knowledge of spectral characteristics of diesel particle emissions is essential for conducting emission inventories as well as for developing better technologies for lowering of the emissions, and the literature on this topic is still very limited. With the small number of vehicles tested within the framework of this study, the outcomes 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 relations between emissions in the different size ranges and the variation of the relations between different vehicles. This information can be directly used for optimization of particle emission testing procedures as well as for developing strategies toward lowering of emissions of fine particles.

In addition to the determination of particle number concentrations, the full scope of investigations included measurements of a number of other emission characteristics including gaseous emissions (NO_x, CO_2, CO, and excess O_2). Smoke was measured using an opacimeter and particle mass concentration using the Dust Trak, a simple instrument based on the scattering of laser light which gives a direct reading of mass concentration on the basis of the manufacturer's calibration from a test aerosol. Due to the large volume of information collected, this paper will focus only on particle size distribution characteristics, while relations between other emission parameters measured, as well as between different methods of particle measurements, will be reported separately.

Exhaust Measurement and Sampling

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 made and which contained a restriction orifice. The primary sampling segment was a stainless steel tube of diameter 100 mm, connected to the bus exhaust by a flexible tube of similar diameter so that different bus heights could be accommodated. The distance between the bus exhaust and the sampling port was on 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 10^7 cm^{-3}. It was estimated that, for such concentrations and for residence times of less than 5 s, the effect of changes in dilution ratio on number concentrations should not exceed 1% (*16*). 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.

A critical requirement in the design of the primary sample line was to maintain isokineticity. This requirement applies

to measurements of larger particles with diameter over 4 μm (*16*). While from the preliminary investigations, it was expected that the majority of emitted particles would have diameters smaller than this value, care was taken to ensure isokinetic sampling. In the design shown in Figure 1, isokinetic flow was achieved by small adjustments of back-pressure in the primary sample line and the pressure in the primary sample segment of the exhaust at the sampling orifice, by empirical adjustment of design specifications (mainly the size of the orifices). However, once the dimensions were set, they did not need further adjustment, and the sampling system was shown to be isokinetic, or nearly so, for a range of the exhaust flows.

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 1.8×10^3 and 2.2×10^4 L min^{-1}. As the distance between the bus 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 sampling ports for gas analysis and opacity were located downstream of the restriction orifice in the primary sampling segment. 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 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) and the Dust Trak aerosol mass monitor. The velocity and temperature of the diluted exhaust were measured with a TSI velocity meter. The method of the exhaust flow measurement was verified by comparison with measurements of the engine air intake flow. The differences between the exhaust flow rates and the corresponding air intake flow rates were on average in the range of 13% and were not large considering the multiplicity of factors that can affect them.

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 20 L min^{-1}, the total flow rate of the mixture in the dilution tunnel was about 200 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. Due to the different physical characteristics of the particulates in different size ranges, different methods have to be applied for measurements of size spectra of small particles and large particles. The TSI model 3394 Scanning Mobility Particle Sizer (SMPS) and the TSI model 3310A Aerodynamic Particle Sizer (APS) were used in this study to measure the size distribution of a sampled aerosol. 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 which 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 is ramped. 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. 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 0.0075−0.304 μm was selected as the optimum to cover the number distribution spectra for most of the vehicle tested and for most of the operation modes. The APS counts particles in the size range 0.5−30 μm; the underlying principle 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 accelerating flow field is dependent on its inertia and 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 which appears when the particle crosses the laser beams.

It must be noted that each of the two instruments is based on a different principle, supplies data based on different assumptions, and it 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, mass concentration, and distributions may be computed from the original number distributions.

Measurement Procedure

Definition of Test Cycles. The measurements were performed for each of a set of steady-state operating modes defined by engine power and road speed. Two mode sets were used—the 13 steady-state mode cycle based on standard SAE tests for heavy duty diesel engines and its modification, the 15 mode cycle, incorporating some features of the standard European legislative emission cycle (ECE-15). The 15 mode steady-state cycle consist of idling modes, a group of modes at intermediate road speed (corresponding to maximum torque) and a group of modes at rated road speed (corresponding to maximum power). Within the intermediate and rated speed groups, individual modes corresponded to power settings of 2, 10, 25, 50, 75, and 100% of maximum power. In the 13 mode steady-state cycle, the modes for 10% of the intermediate and rated power were omitted.

Vehicle Tests. Eleven in-service buses, selected from the Brisbane Transport Fleet, and one near-new Caterpillar-engined bus were tested on the chassis dynamometer of Hastings Deering in Brisbane, Q, Australia, using the 15 mode cycle. Two other vehicles (a four-wheel drive diesel vehicle and a medium truck) were tested on the chassis dynamometer of TAFE in Wetherill Park, Sydney, NSW, Australia, using the 13 mode cycle. All vehicles were supplied with diesel fuel from the same production batch. The vehicles from the Brisbane Transport Fleet comprised four types of engine: Volvo Mark II, III, and IV and MAN. The buses with the same type of engine were of similar age; there were, however, significant differences in age of buses equipped with different types of engines. The MAN bus was the oldest (over 13 years), and Volvo Mark IV the youngest (over 2 years). The characteristics of the vehicles tested are summarized in Table 1. It should be noted that buses 6 and 10 refer to the same bus, tested at different times, as are 13 and 14.

The measurements were conducted according to the following procedure. Each vehicle was subjected to a power test so that maximum power, maximum torque, and intermediate and rated road speeds were identified. The vehicles

TABLE 1. Specifications of the Vehicles Tested

engine type	vehicle ID	vehicle age (month)	engine specification	power specification
Volvo	buses: 2	30	engine: THD 103 KF	rated power: 180 kW/2000 rpm
Mark IV	4	26	cylinders: 6	max torque: 1050 N m/1230 rpm
	12	29	displacement: 9.6 L	
Volvo	bus 1	45	engine: THD 101 GC	rated power: 178 kW/2200 rpm
Mark III	3	44	cylinders: 6	max torque: 890 N m/1400 rpm
	13 (14)	44	displacement: 9.6 L	
Volvo	bus 5	113	engine: THD 101 GC	rated power: 180 kW/2000 rpm
Mark II	6 (10)	113	cylinders: 6	max torque: 1050 N m/1400 rpm
	7	113	displacement: 9.6 L	
	8			
MAN	bus 9	159	engine: D 2566MUH	rated power: 145 kW/2200 rpm
			cylinders: 6	max torque: 775 N m/1400 rpm
			displacement: 11.4 L	
Caterpillar	bus 11		engine: Cat 3126	rated power: 202 kW/2200 rpm
			cylinders: 6	max torque: 1165 N m/1440 rpm
(CAT)			displacement: 7.2 L	
Toyota Landcruiser	vehicle 1		Engine: 1 HZ	
			cylinders: 6 in line	
			displacement: 4.2 L	
RTA truck	vehicle 2			

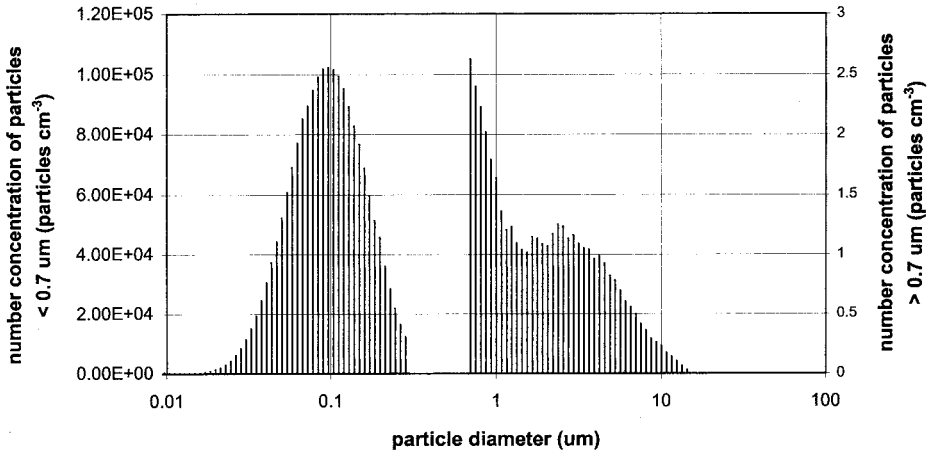

FIGURE 2. A typical size distribution of particles from a diesel bus running on low power, measured independently by the SMPS and APS.

were then tested in each mode of the appropriate cycle in sequence. After attaining the required conditions of power and speed for the mode, steady conditions were maintained for 5 min. The sampling instruments were active only during the last minute of the mode. Readings from the magnehelic gauges, from which sample flows rates were calculated, were averaged over the last minute. The temperature of the exhaust, the temperature of the exhaust primary sample, and the temperature of the diluted exhaust were taken at the beginning and at the end of the last minute of the mode and the readings averaged. At the end of the 15 mode cycle some of the modes were repeated in order to assess reproducibility of the measurements.

Results and Discussion

Figure 2 presents a typical size distribution of particle emissions from a diesel bus running on low power mode (bus 4: Volvo Mark IV, 2% power, rated speed mode). The results of measurements conducted independently by the SMPS and APS instruments are presented on one diagram. Note that the number concentration scales for SMPS and APS size ranges differ by orders of magnitude.

Particulate Data by SMPS. The SMPS data refer to a particle size range from 0.0075 to 0.305 μm. The data collected with the SMPS have been presented in terms of particle count median diameter (CMD) and total concentration. Particulate mass concentration was calculated from the particle number concentration for the undiluted exhaust and temperature 25 °C and assumed particle density of 1 g cm^{-3}.

Figure 3 presents typical relations between particle number concentration, CMD, and mass and the power setting for intermediate and rated speeds in the SMPS range (bus 4). Average particle number concentrations in the SMPS range in the undiluted exhaust range from 7.2×10^6 cm^{-3} for idle modes to 3.9×10^7 cm^{-3} for rated speed modes. A summary of particle number concentration, CMD, and mass concentration in the SMPS range for all buses is provided for all vehicles in Table 2.

From the SMPS data, the general conclusion can be drawn that for both intermediate and rated speed settings the number concentration of submicrometer particles increases significantly with load. The ratio of number concentrations in the maximum power modes to idle modes, for intermediate speeds, ranges from 1 to 80 with an average of 13 and for rated speed modes (except for vehicle 1) from 3 to 40 with an average of 15. The differences between the vehicles in terms of the number concentration in the SMPS range are seen in Figure 4 for idling, intermediate, and rated modes, respectively. For the idling modes, Volvo Mark II and MAN buses had higher levels of particulate emissions than the other vehicles. For the intermediate and rated speed setting,

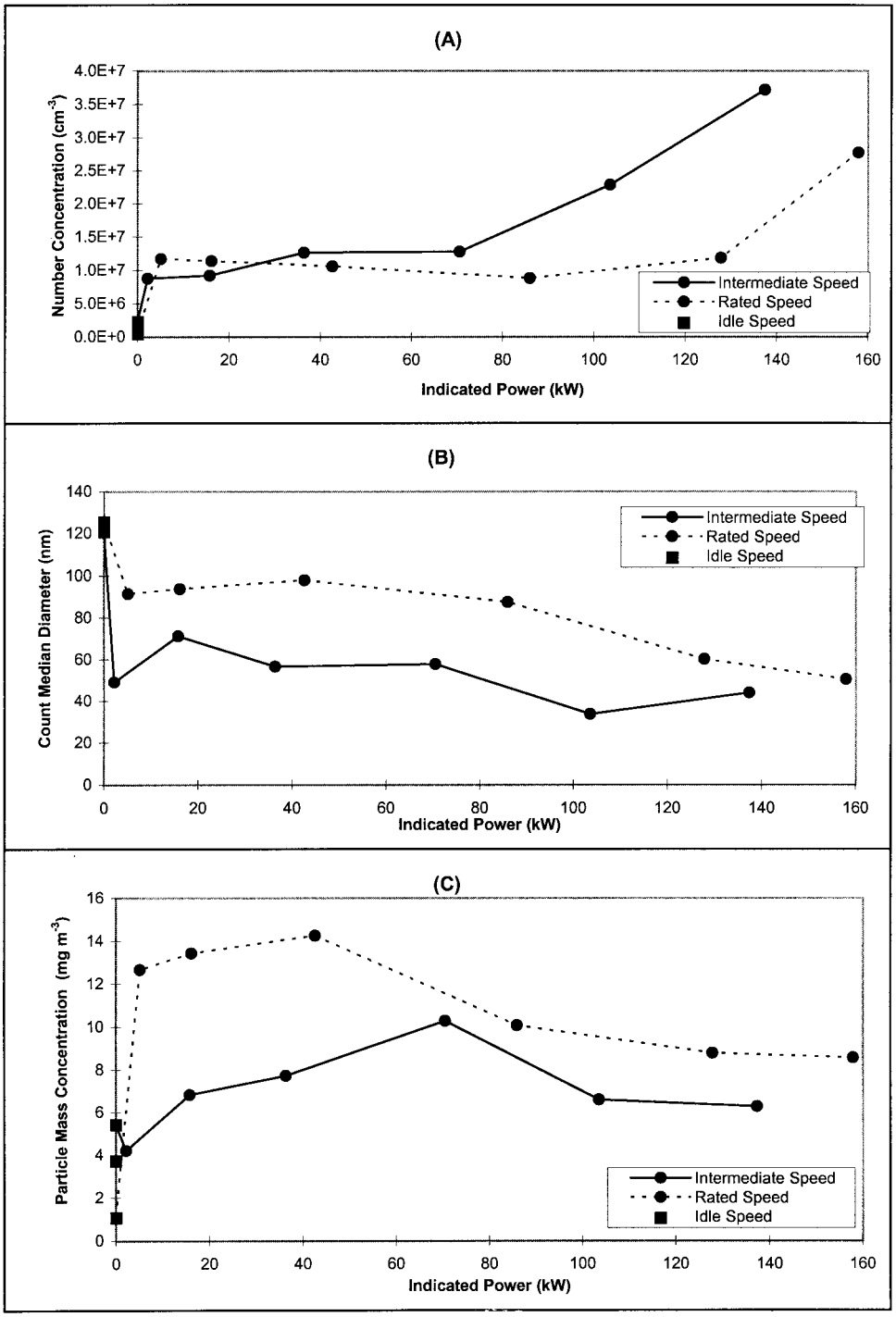

FIGURE 3. Typical shapes of the relations between (a) particle number concentration, (b) CMD, and (c) mass and the power setting for intermediate and rated speeds in the SMPS range (bus 4).

TABLE 2. Summary for Buses Only of Particle Number Concentration, CMD, and Mass Concentration in SMPS Ranges

	particle concn (particles cm^{-3})		CMD (nm)		calcd mass concn (mg m^{-3})	
modes	range	avg	range	avg	range	avg
idle	6.1×10^4–1.8×10^8	7.2×10^6	18–137	63	0.12–9.09	3.1
intermediate[a]	4×10^5–5.6×10^7	2.8×10^7	17–211	67	2.09–95	24.4
rated[a]	2.9×10^5–8.7×10^7	3.9×10^7	18–187	68	1.24–96	22.0

[a] At maximum power.

the number concentration in the SMPS range varies significantly within each of the groups, but the differences between the groups in terms of average number concentrations are small.

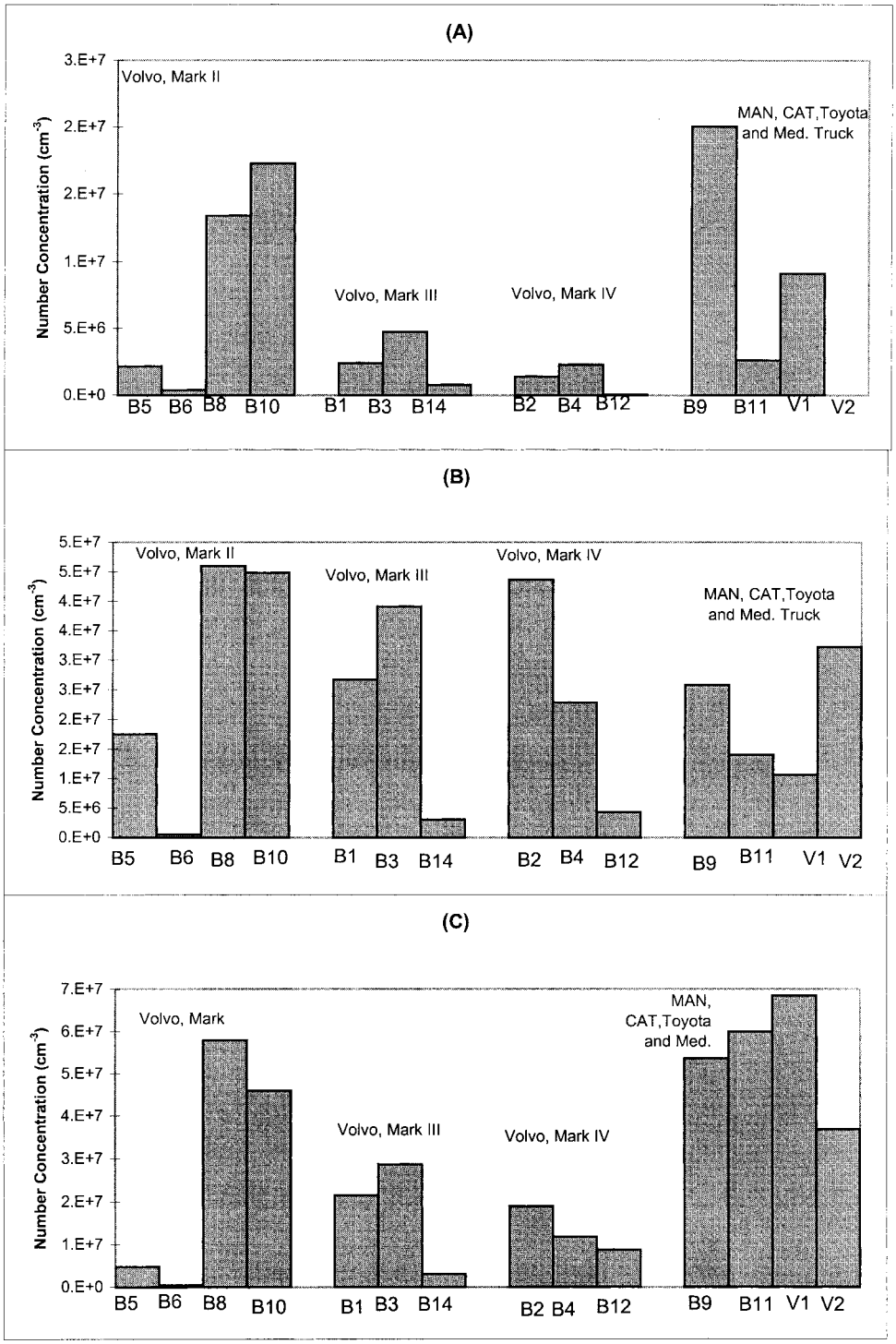

FIGURE 4. The difference between the tested vehicles in terms of particle number concentration in the SMPS range for (a) idling, (b) intermediate. and (c) rated modes.

The Count Medium Diameter (CMD) for the SMPS range fluctuates moderately but it does seem to depend on vehicle operating conditions. Figure 5 shows that there are large differences in CMD for different buses. The differences between the CMD of buses of the same type are quite significant, and it is difficult to relate the value of the CMD to the type/age of the engine.

The calculated mass concentration in the SMPS range increases with increasing power for all the vehicles investigated except for vehicle 1 at rated speed. The ratio of the mass concentration at maximum power to idle for intermediate speed modes ranges from over 1 to 24 with an average of 8.5, and for rated speed modes from over 1 to 28 with an average of 7.6. The trends in the mass concentration in the SMPS range cannot be related to the age or the type of the bus in a simple manner.

Particulate Data by APS. Average particle number concentrations in the APS range in the undiluted exhaust from 3.3×10^4 cm^{-3} for idle modes to 2.4×10^4 cm^{-3} for rated speed modes. A summary of the particle number concentration and CMD in the APS range is presented for buses only in Table 3. PM_{10} mass concentration obtained by summing the calculated mass concentration in the SMPS range and in the APS range below 10 μm is presented

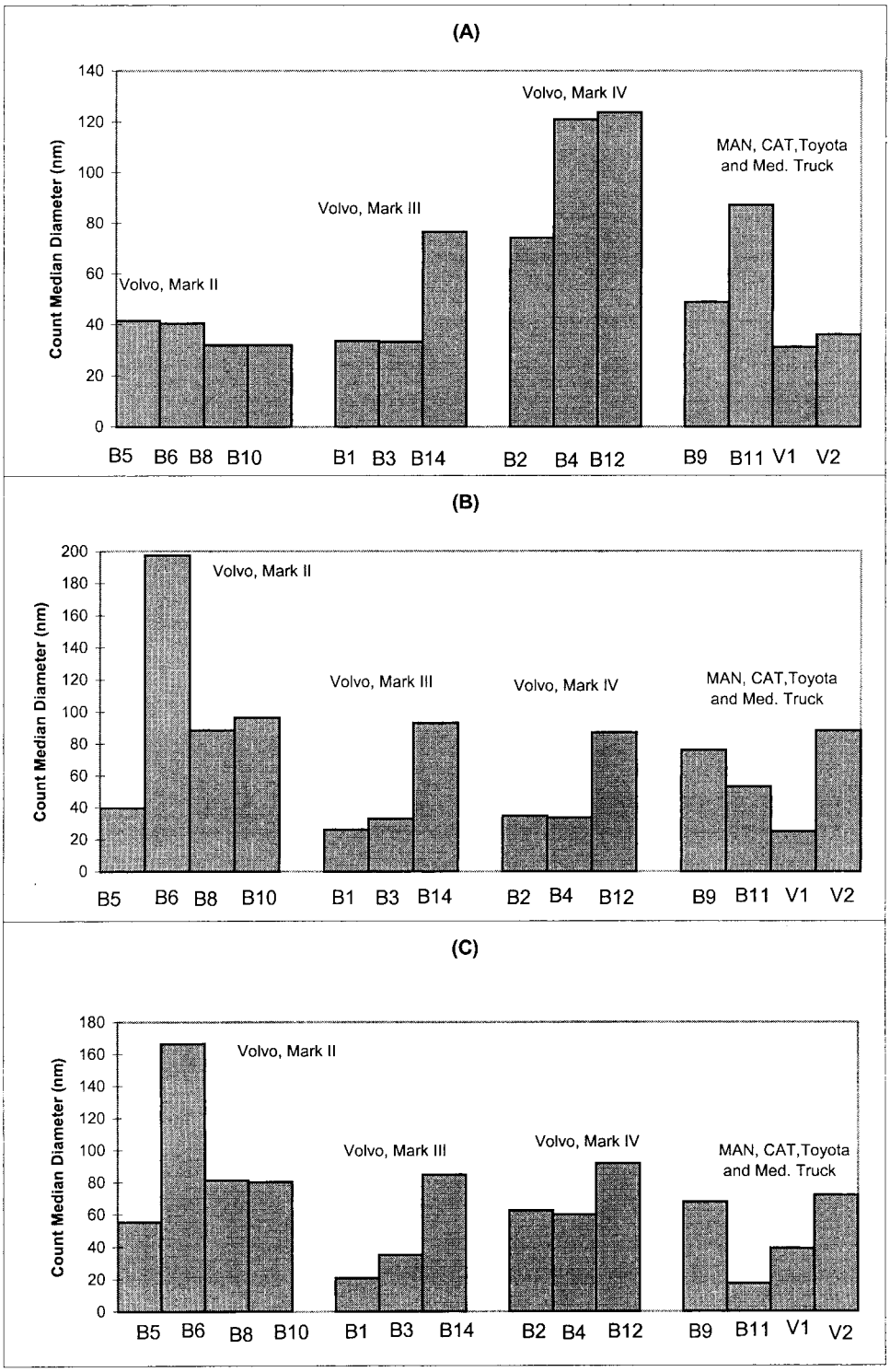

FIGURE 5. The difference between the tested vehicles in terms of particle count median diameter in the SMPS range for (a) idling, (b) intermediate, and (c) rated modes.

in the last column of Table 3. The choice of calculated PM_{10} fraction rather than the total particle mass in the APS range for this presentation was made to enable comparison with the existing standards or guidelines that are expressed in terms of PM_{10} and not PM_{30} (the upper range of the APS).

The particulate data in the APS range display relatively less variation than those in the SMPS range. The number concentration in the APS range increases with increasing power for all the buses and both the intermediate and the rated speed settings. The increase in the number concentration with power is moderate at first, but at a power output greater than 80−110 kW it is more rapid. It should be noted here that the light duty vehicles (1 and 2) emit significantly higher particulate concentrations in the larger size range than do the buses. The ratio of the number concentrations at maximum power to idle for intermediate speed modes ranges from over 1 to 1.4×10^3 with an average of 130 (the ratio would be 52 if bus 12 of exceptionally high ratio was excluded from the calculations),

TABLE 3. Summary for Buses Only of Particle Number Concentration and CMD in APS Range and Calculated PM_{10} Mass Concentration

	particle concn (particles cm^{-3})		CMD (mm)		calculated PM10 mass concn (mg m^{-3})	
modes	range	avg	range	avg	range	avg
idle	1.0×10^1–3.8×10^5	2.8×10^2	0.61–2.10	1.06	0.03–58	5.8
intermediate[a]	$(6.2–2.5) \times 10^5$	3.2×10^4	0.63–2.38	1.26	$(25–1.3) \times 10^4$	413
rated[a]	3.0×10^2–7.4×10^4	2.4×10^4	0.69–1.63	1.04	$(62–1.1) \times 10^4$	434

[a] At maximum power.

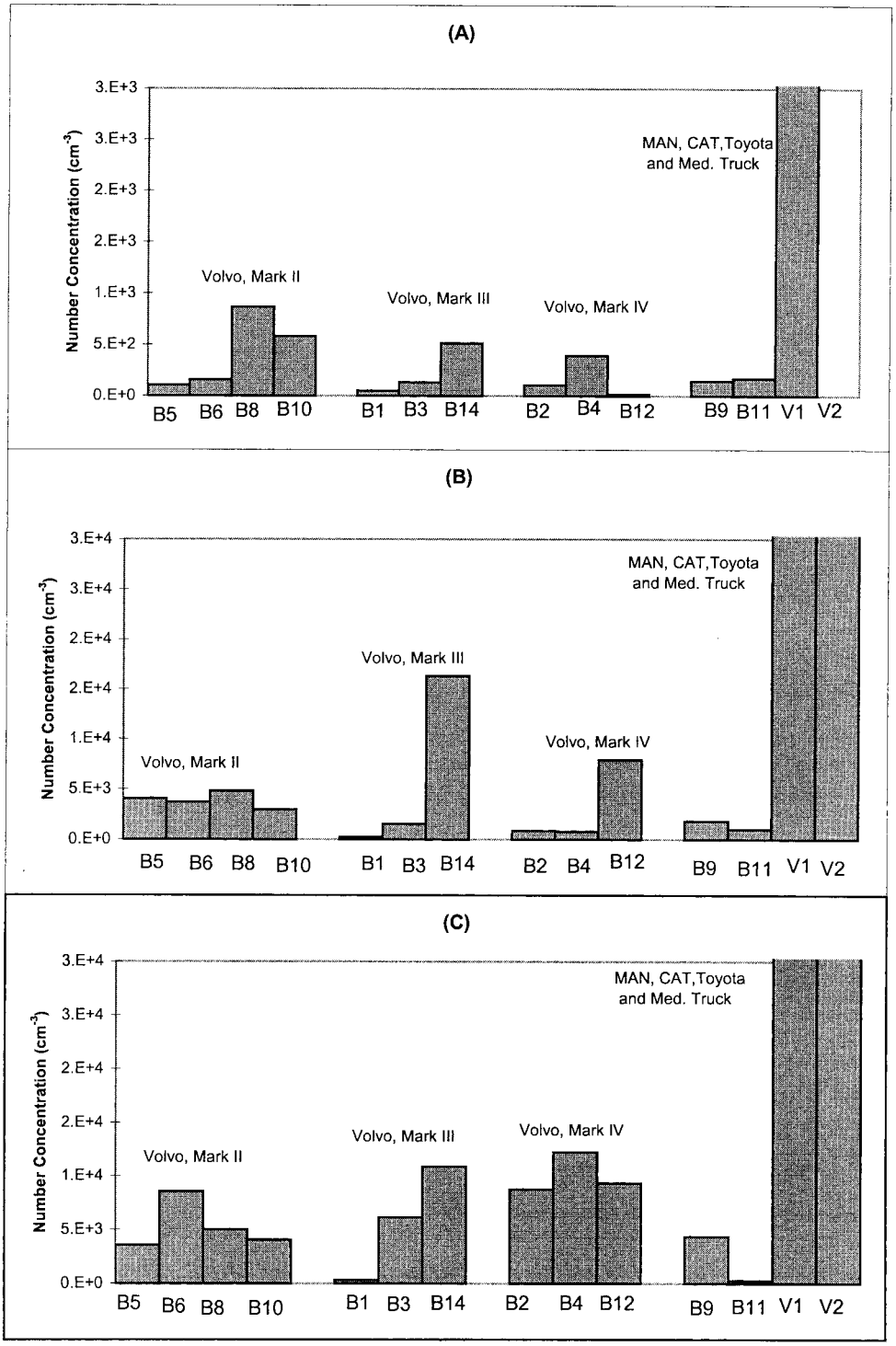

FIGURE 6. The difference between the tested vehicles in terms of particle number concentration in the APS range for (a) idling, (b) intermediate, and (c) rated modes.

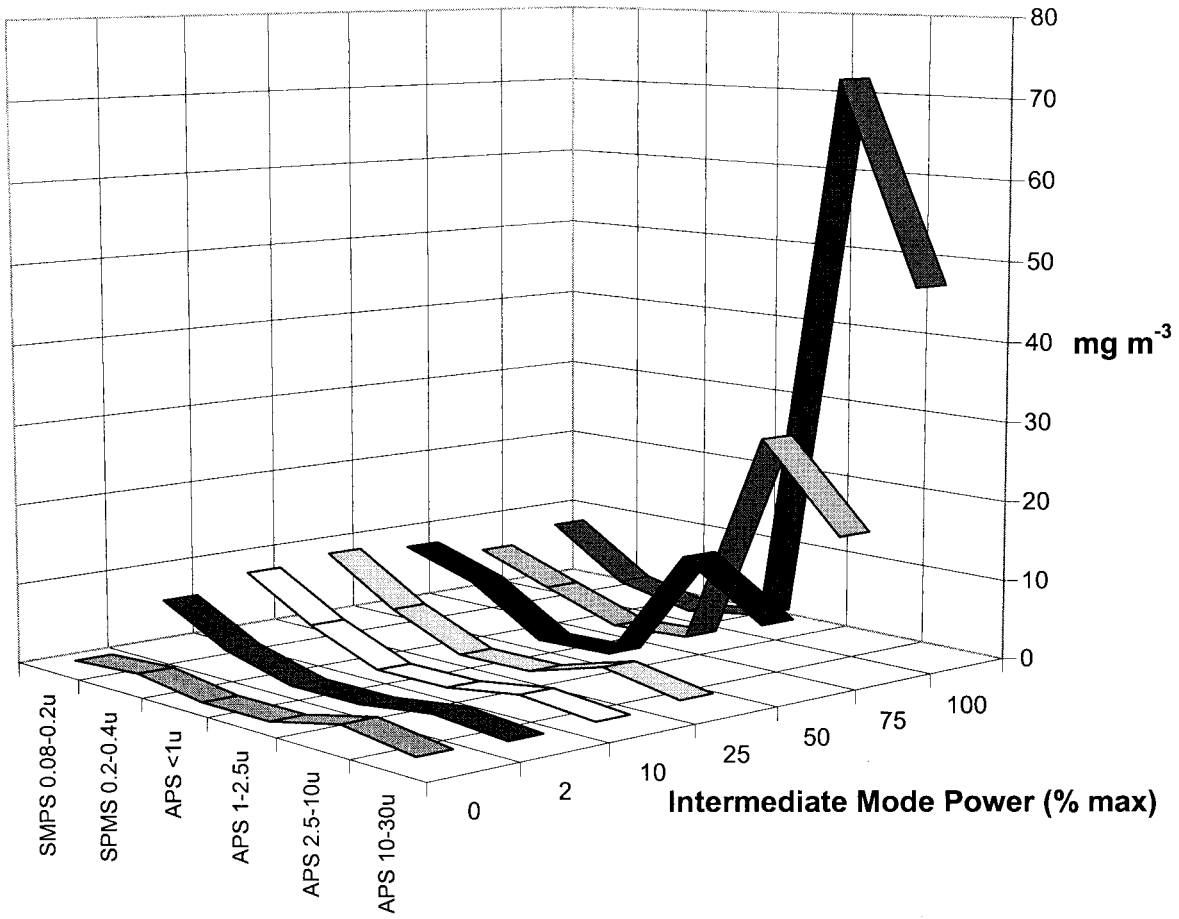

FIGURE 7. Comparison of particle emissions in the SMPS and APS ranges for bus 2 for different intermediate power setting.

and for rated speed modes from 1.5 to 134 with an average of 62. The comparison of the particulate emissions for all vehicles in terms of number concentration in the APS range is presented in Figure 6. The CMD in the APS range exhibits some variation for buses 2−7, but for other buses it remains almost constant. The variation in the CMD within any of the groups appears to be larger than between the CMD group averages.

The mass concentration in the APS range increases with increasing power for both intermediate and rated speed settings. This increase is similar to that observed for the number concentration in the APS range. In the APS range, the ratio of the mass concentrations at maximum power to those at idle for intermediate modes ranges from (2.5 to 6.3) $\times 10^3$ with an average of 516; the ratio would be 125 if bus 12 of exceptionally high ratio was excluded from calculations. For rated speed modes, the ratio varies from 2.5 to 442 with an average of 114 (vehicles 1 and 2 are not included in the average). Similarly, as it was in the SMPS range, the mass concentration of the particulates emitted by a vehicle does not correlate with the age or model of the engines investigated. Hence, the individual characteristics and history of an engine appear to play a more important role than the age, model, or the type of engine. The smallest mass concentration in the APS range is that for CAT diesel engine.

A comparison of particulate mass emissions in SMPS and APS ranges for one of the vehicles investigated, bus 2, is presented in Figure 7 and illustrates how mass emissions vary with the different operating conditions of the vehicle. For lower power modes, emissions in the submicrometer range are dominant, while for high power modes, emissions in the supermicrometer range are dominant.

Reproducibility of Particulate Data. The reproducibility of number concentration and CMD is taken as the difference between two determinations relative to the mean value for a particular mode setting. The reproducibility of the CMD for the SMPS range is better than 10% (and in most of the cases better than 3%) and of the number concentration in the SMPS range varies between 0.5 and 50%; for few cases, it is even larger. The reproducibility of the CMD for the APS range is better than 6% and, in most cases, is better than 2%. The reproducibility of the number concentration in the APS range varies between 0.5 and 30%. The lower level of reproducibility of the number concentration for the SMPS and APS ranges is caused by the fact that the repeat tests for specific modes were never conducted for the same condition as the initial measurements. For example, if mode 6 was repeated after completion of the whole tests, the engine conditions were different from those that pertained after completion of mode 5, which preceded mode 6 in the initial tests. The variability of conditions for repeated modes and hence the variation in emission characteristics was also seen in the gas emissions.

Inter- and Intragroup Variation for the Vehicles Investigated. Particle number concentrations in both SMPS and APS ranges vary significantly within each group of vehicles, but the differences between the groups are small. Emission characteristics do not appear to correlate with age, model, or type of the engine, and there are large variations in particle emissions from vehicles powered by the same type engines, suggesting that significant reduction of emissions could possibly be related to the tuning of the vehicles. With the focus on fine particles, in-service vehicles would have to be tested for emissions of fine particles, which would require simplification of testing techniques for fine particles. The currently available techniques for measurements of fine particles, such as these used in this study, are costly and

complicated, and cannot be easily employed for in-service vehicle testing.

Relation between Particle Emissions in Different Size Ranges. Analysis of particle emission levels in different size ranges reveals that for individual vehicles there is no relation between emissions in the smaller and larger particle ranges. The vehicles that emit the highest levels of particles in the SMPS range do not necessarily emit the highest levels in the APS range. This finding has a significant implication for vehicle testing for particle emissions and indicates that emissions in small and large particle ranges should be tested independently to provide a comprehensive characterization of the vehicle particle emissions. This indicates further that mass emission concentration measurements, whether total mass or PM_{10} fraction, will not help to identify vehicles which are the worst emitters of fine particles. For management and control of fine particle emissions, particle number or particle mass in the ultra fine range should be used as an indicator.

Implications for Exposure or Risk Analysis. The current air quality emission standards and guidelines are based on particle mass concentrations, which means that there is a bias toward larger particles whose contribution to the total mass is often considerably higher than the mass of smaller particles. This applies to all mass-based standards, regardless of the cut-off point for particle diameter. Emission testing and air quality monitoring data are similarly also usually available in relation to particle mass concentrations. If this data set is used for health risk analysis, there could be a significant misinterpretation of exposures and thus risks, as particle deposition in deeper parts of respiratory tract is much greater for smaller particles than for larger, and should be considered in terms of size related to number, rather than mass. Comparisons of particle emissions in the SMPS and APS ranges measured in this study show (see Tables 2 and 3) that there are 2−3 orders of magnitude difference in particle numbers emitted in these ranges, but only over an order of magnitude difference in the calculated mass of particles in the SMPS range and calculated PM_{10} fraction. Thus, if risk is related to very fine particles and particle number, correlating it with particle mass may result in underestimation of risk, unless relation between particle number and mass is known and taken into account.

Size distributions for the purpose of exposure assessment would also require measurements that were conducted under environmental conditions, where environmental dilution and long-term particle aging effects could be taken into account. Direct exhaust sampling results are applicable for vehicle intercomparison, but there are limitations in using them for exposure assessment where changes in particle size distribution are of importance. These changes, however, do not affect the total mass balance.

The data on particle emission levels in different size ranges and variation of the emissions for different bus operating conditions can be used for assessing emissions from a bus fleet; however, with the small number of vehicles tested within the framework of this study, the outcomes of the study can only be used as estimates in emission inventories.

Acknowledgments

Environment Australia provided funding for this project. The authors would like to acknowledge the technical support of Jane Hitchins, Nick Giannakis, Wendy Barron, and Marcin Metter. Brisbane City Council provided vehicles for the measurements and invaluable support for this project from Mr. Jacek Kaluza and Mr. Ian Whitelaw; the authors are also indebted to Mr. Matthew C. Armstrong from Hastings Deering whose dedication, technical expertise, and friendly participation in the dynamometer tests made them possible.

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Received for review September 16, 1997. Revised manuscript received March 19, 1998. Accepted April 16, 1998.

ES970826+