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Comparing ELPI and SMPS Measurements of Motor Vehicle Exhaust PM

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Introduction

The past few years has witnessed increased interest in the measurement of particle size and number to characterize the particulate emissions of motor vehicles. This interest is motivated by an epidemiological correlation noted between episodes of elevated ambient particle concentrations and adverse health effects and by existing or anticipated regulations of PM emissions. In this contribution we compare the performance of two of the leading instruments used to measure particle size distributions, both under steady state and transient vehicle operation. The principal questions addressed are: 1) What physical particle properties are actually measured? 2) How do the size distributions compare? 3) What is the effect of particle density? 4) What is observed in transient measurements? and 5) What are the trade-offs between the two instruments?

The examples of particle size measurement presented in this work are taken from our past three years experiences using the ELPI (electrical low pressure impactor) and SMPS (scanning mobility particle sizer) to make PM emissions measurements. This includes vehicles with conventional gasoline engines, direct injection gasoline engines, and diesel engines. It includes measurements made using both a conventional chassis dynamometer and dilution tunnel system, as well as PM measurements made directly from the tailpipe, or from an engine dynamometer, using ejector pump sampling. From this work, we have chosen examples to illustrate the performance, and to enhance the comparison, between the two instruments.

Principles of operation

The ELPI (Figure 1) consists of a corona discharge, cascade low pressure impactor and electrometers. Particles in the sample aerosol are charged, and then separated according to aerodynamic size in the cascade impactor. The current deposited on each stage is recorded by the electrometers and divided by the corresponding charging efficiency, which has a power law dependence on particle diameter, in order to derive a particle number distribution.

Using the SMPS, the sample aerosol first passes through a bipolar charger in order to provide a Boltzmann distribution of charges on the particles. This aerosol enters the outer periphery of the electrostatic classifier (see Figure 1), through which a particle free sheath gas flows. A potential between the outer wall and the central electrode accelerates the positively charged particles towards the center of the cylinder. The particles reach a terminal velocity dependent on their electrical mobility. Those having the correct mobility enter the exit aperture and are counted with a condensation particle counter. Counting the particles at the exit during a scan of the high voltage leads to a mobility size distribution.

Initial comparison

Figures 2 and 3 superimpose number and volume weighted particle size distributions obtained simultaneously with the ELPI (symbols) and SMPS (lines). The horizontal axis represents aerodynamic diameter for the ELPI data and mobility diameter for the SMPS data. Note that only data from the lowest 5 - 6 stages of the ELPI is relevant to this comparison, since the SMPS does not cover the entire range of the ELPI, which is up to 10 μm . It is evident that the overall agreement between the ELPI and SMPS data is quite good. For the number weighted distributions, the ELPI overestimates particle concentrations on the lowest 1-2 stages by up to perhaps a factor of 2. For the volume weighted distributions, it underestimates

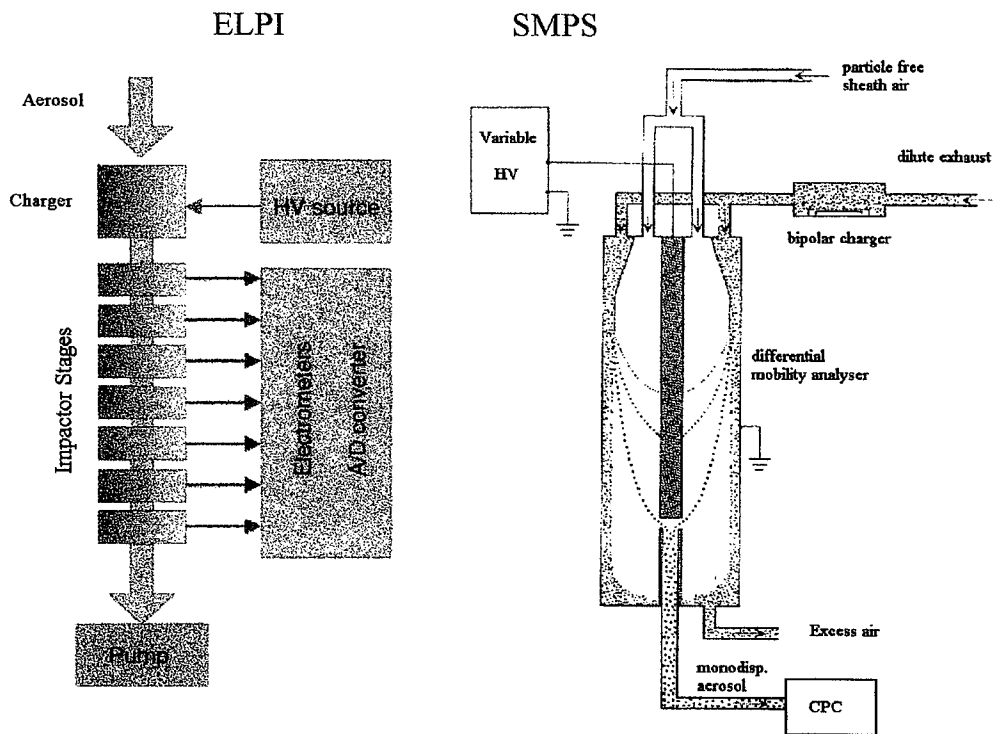


Figure 1. Schematic representations of the ELPI and SMPS.

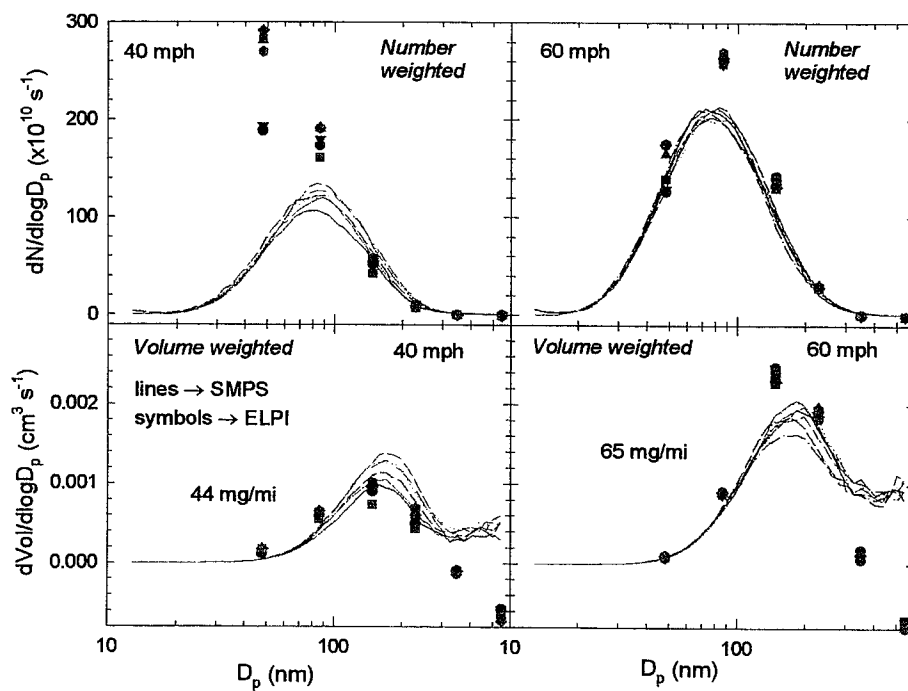


Figure 2. Comparison of ELPI and SMPS measurements of steady state diesel PM emissions.

the concentrations for stages 5 - 6, with negative values sometimes present. The overestimation of the number concentrations is likely due to a combination of effects. On one hand, electrometer noise and offsets are exacerbated by the low charging efficiency of particles in the ~ 40 nm range. On the other hand, the comparison in these figures is between aerodynamic and mobility diameter, which are not the same. The underestimation of the particle volume for stages 5 and 6 is a combination of electrometer offsets and an overcorrection for small particle loss within the cascade impactor, that is exacerbated by volume weighting.

In spite of these discrepancies both instruments provide similar values for the particle number emissions and show diesel particles to have a number weighted mean of about 100 nm. The ELPI and SMPS both show in Figure 3 that the addition of dimethoxy methane (DMM) to diesel fuel does not alter significantly the number of particles emitted; rather it reduces their average size by about 20 - 30 nm. Both instruments show that this size reduction leads to a lowering of the PM volume (also mass) emissions by roughly a factor of 2.

Steady state measurement of diesel exhaust PM

Apparent agreement is good except for:

- particle #, lowest 1-2 stages
- particle vol., stages 5-6.

Both methods show shift to smaller particle size when adding DMM to the fuel.

Both show decrease in particle volume when DMM is added.

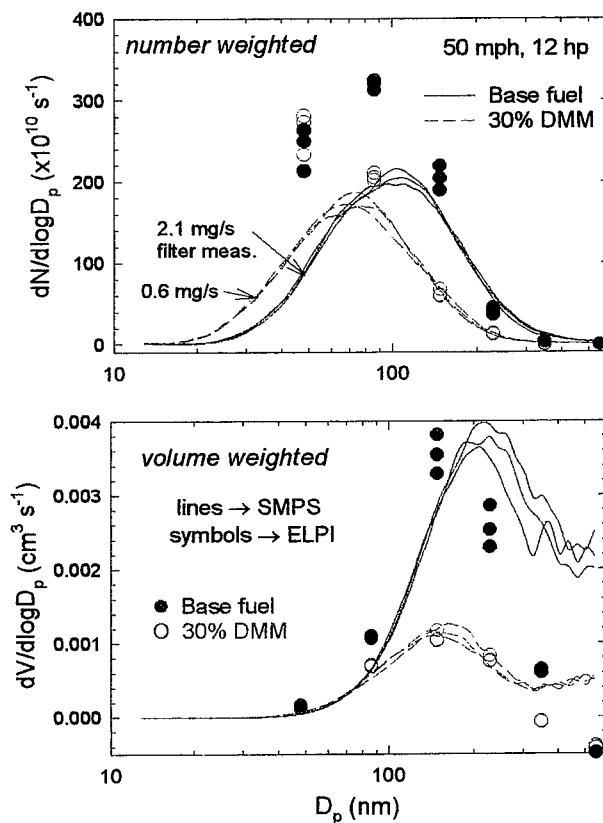


Figure 3.

What is really measured?

The above comparisons are made ignoring the distinction between aerodynamic and mobility diameter. To measure particle size with the SMPS, the sample aerosol is first brought to a Boltzmann charge distribution using bipolar charging. The positively charged particles of the correct mobility exit the classifier and are counted, resulting in a mobility distribution. For the ELPI, unipolar charging is used, the efficiency of which depends on particle mobility. However, particles are separated according to aerodynamic diameter. Since the mobility diameter is not measured, the ELPI uses the aerodynamic diameter to calculate charger efficiency, and thereby to convert the measured currents to a number distribution. To place the comparison on equal footing, we can following Ahlvik et al., (*SAE Technical paper No. 980410, 1998*) assume particle density do decrease from about $1.5 g/cm^3$ at 40 nm to

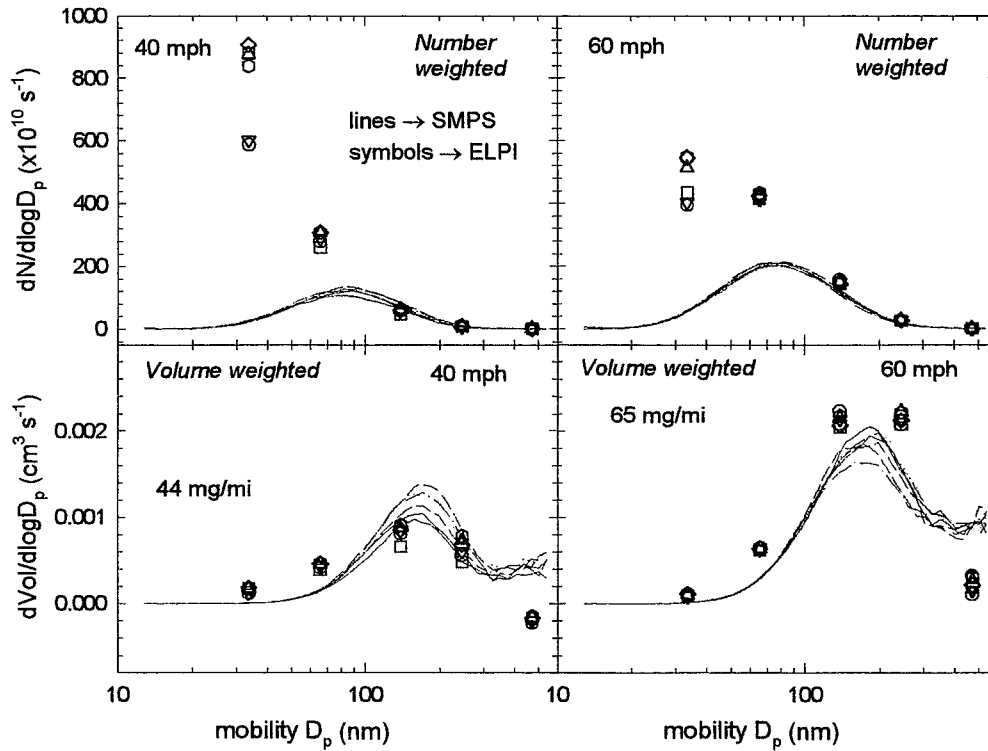


Figure 4. Same comparison as in Figure 2, but with ELPI data converted to mobility diameter.

Tandem SMPS - ELPI size distributions diesel exhaust

Use SMPS to create mono-disperse exhaust aerosol at mobility diameters corresponding to ELPI stages 1-5.

Sample mono-disperse aerosol with ELPI and determine aerodynamic size distribution.

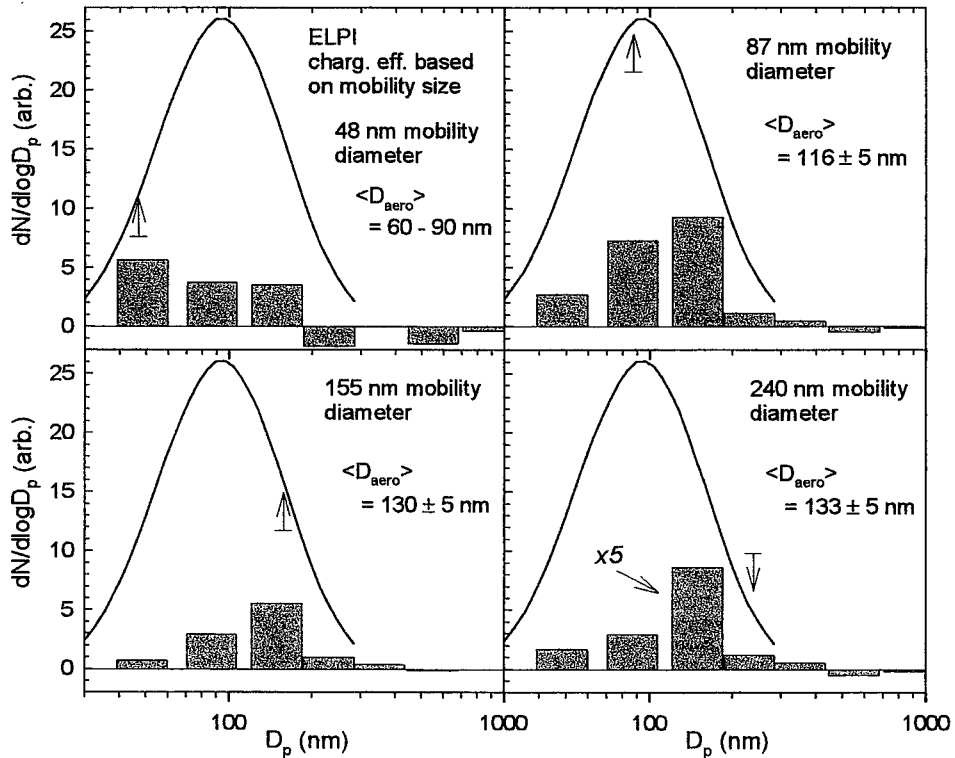


Figure 5.

$\sim 0.5 \text{ g/cm}^3$ at 400 nm. The larger than unit density for the lowest two ELPI stages causes a decrease in particle diameter when converting from aerodynamic to mobility units, and therefore a decrease in charging efficiency. This leads to an increase in the discrepancy between ELPI and SMPS data for these two stages, as evident in Figure 4. Stages 3 - 5 are not much affected, since the lower than unit density implies an increase in diameter when converting from aerodynamic to mobility units, and an increase in charging efficiency, but this just follows the shape of the size distribution.

Alternatively, a tandem experiment can be performed, as also done by Ahlvik et al., in which the SMPS is used to select particles having a well specified mobility diameter and the ELPI utilized to examine their aerodynamic diameter. As Figure 5 illustrates, a well defined mobility does not lead to a specific aerodynamic diameter. In contrast, the mobility selected diesel soot particles exhibit a wide range of aerodynamic diameters. Figure 6 shows the average "effective density" of these particles, along with the standard deviation of the density (given by the vertical bars).

- ✓ Average diesel soot density decreases as particle diam. increases.
- ✓ At a given mobility diameter diesel soot has a wide distribution of aerodynamic diam.
- ✓ Close agreement between ELPI and SMPS somewhat fortuitous.
- ✓ Cannot simply "correct" ELPI by converting to mobility diameter.

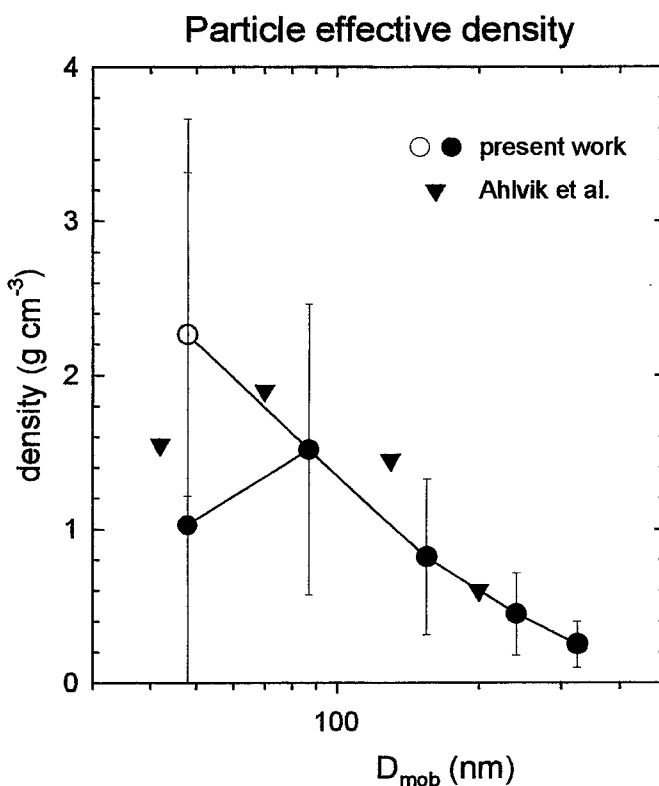


Figure 6.

Transient PM emissions

The ability to measure transient PM emissions is important for a number of reasons: emissions regulations are based on transient vehicle tests, and engine performance and calibration information can be learned from transient operation. The SMPS performs size scans too slowly (minimum 30 s) to measure size distributions directly for transient tests. Instead, it is fixed to transmit particles at a set mobility diameter, and the transient emissions at this diameter are monitored as a function of time. Size distributions are subsequently constructed from repeated transient tests conducted for a series of mobility diameters. The ELPI is able to record size distributions on a second by second basis (400 pA scale). However, its response depends on the sensitivity scale that is chosen, and decreases substantially to > 20 s if the 10 pA scale is employed.

Transient electrometer response to burst of particles

- ✓ “AC” response on upper stages due to passage of charged particles.
- ✓ “peak” in current on lower stages due to particle impaction.
- ✓ time delay due to transit time of aerosol sample.

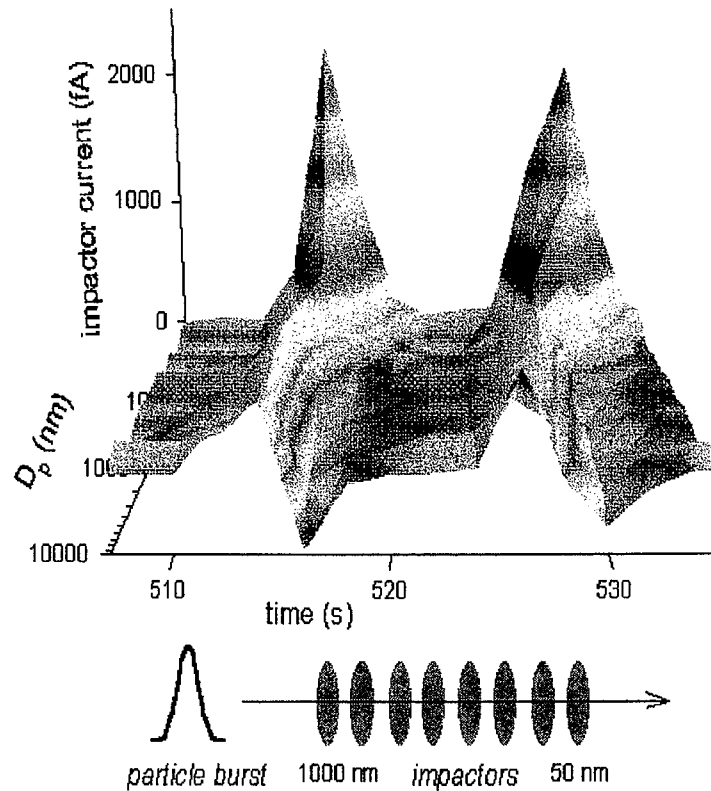
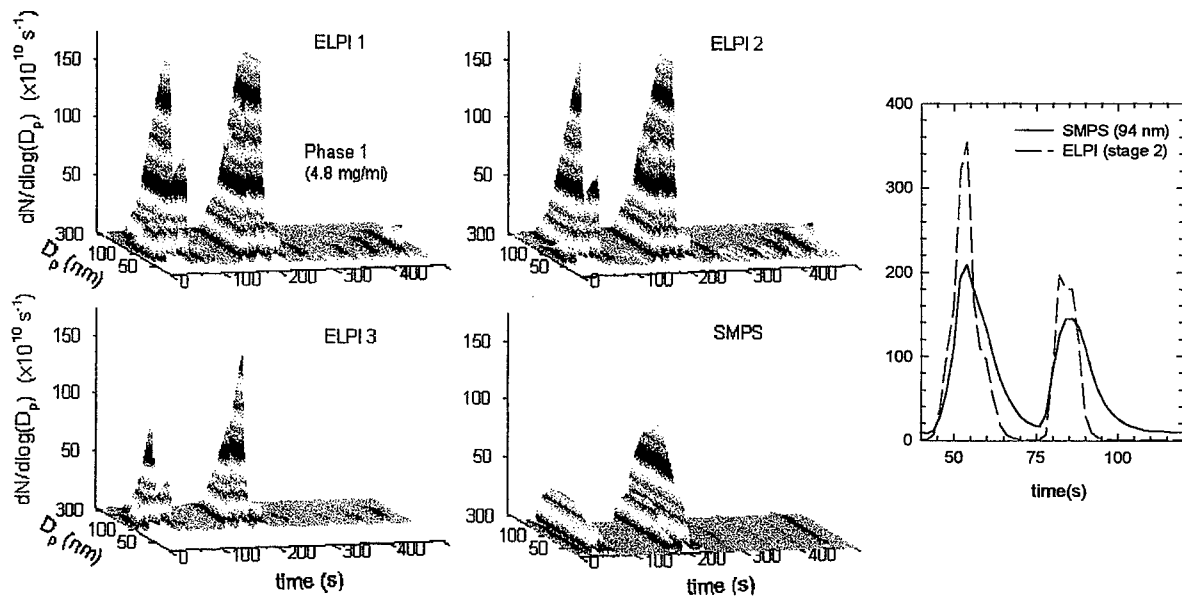


Figure 7.

Transient measurements incur two aspects of ELPI operation that are not apparent during steady state applications. The first is that the usual picture of ELPI operation, based on the electrometers recording the charge deposited by the impacting particles, is an oversimplification. Actually, the impactors act like Faraday cages, which respond to the appearance and disappearance of charge as it passes through the impactor. Thus, in Figure 7, transient pulses of vehicle PM emissions cause AC responses on the electrometers of the upper ELPI stages as they pass through. On the lower stages, of appropriate aerodynamic diameter for combustion particles, the particles impact and produce positive going peaks in electrometer current. The second feature to note is the time delay incurred in the particles traveling between stages 1 and 12. This must be accounted for if one wants to examine properly the size distribution at a particular point in time.

Figure 8 provides a comparison between ELPI and SMPS transient size distributions for a gasoline vehicle driven over the FTP drive cycle (the third ELPI plot is given omitting the data from stage 1). The various ELPI plots show that good repeatability is achieved between individual FTP tests. The SMPS plot is constructed from 8 repeat drive cycles. It appears that the ELPI measurements consistently overestimate the SMPS data by about a factor of 2. However, in order to make a proper comparison, one must take into account the time responses of the instruments. As shown in the 2-d plot accompanying Figure 8, the ELPI has a faster time response than the SMPS, and will thus have a larger response to the narrow particle emissions peaks that characterize gasoline vehicle emissions.

ELPI vs SMPS transient size distributions



- ✓ PM emissions peaks occur during vehicle acceleration.
- ✓ ELPI distributions appear $\sim 2 \times$ more intense than SMPS.
- ✓ SMPS time response slower than ELPI.

Figure 8.

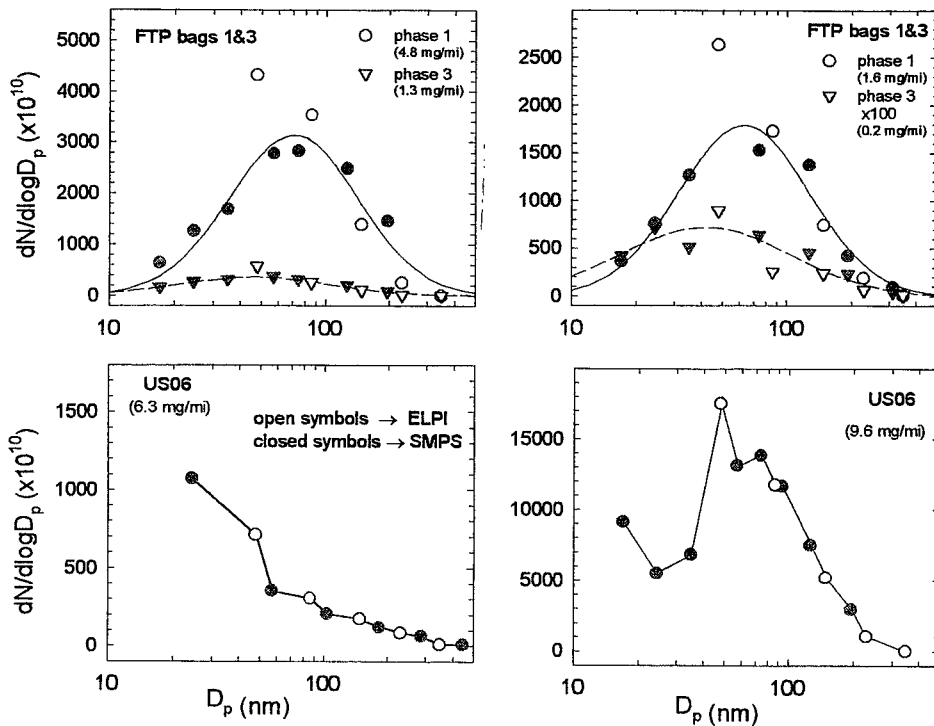


Figure 9. Time integrated ELPI and SMPS distributions for gasoline vehicles, FTP and US06 drive cycles.

The slower response of the SMPS implies less intense, but wider PM peaks recorded along the drive cycle. When the transient emissions are integrated over the drive cycle (or one phase of the cycle), this distinction cancels and, as shown in Figure 9, there is good quantitative agreement between particle size distributions recorded by the ELPI and SMPS (again noting that one is comparing aerodynamic to mobility diameter). The agreement is found to hold for phase 1, the cold start, of the FTP, phase 3 of the FTP, and the more aggressive US06 drive cycle. Both instruments show cold start PM emissions to outnumber those from the hot start (phase 1 versus phase 3). With gasoline vehicle PM emissions occurring primarily during vehicle acceleration, higher levels are recorded by both instruments for the US06 versus hot start FTP drive cycles.

Conclusions

Considering the different methods for particle sizing, cascade impactor versus electrostatic classifier, and considering the different physical properties that are measured, aerodynamic diameter versus mobility diameter, the vehicle emissions particle size distributions recorded by these two instruments are remarkably similar. When they are used to record trends in the size distributions, e.g. for different fuels or under different speed and load conditions, the two instruments register similar trends. This consistency is found for both steady state operation and for transient operation, as long as the transient tests are sufficiently repeatable so as to allow the construction of SMPS distributions from a sequence of tests.

Used in a tandem approach, recording aerodynamic size distributions of mobility selected diesel particulate matter allows calculation of an "effective" particle density. It is effective in the sense that it derives from a comparison between aerodynamic and mobility equivalent spheres to the actual fractal geometry soot particles. It is also "effective" in the sense that it is based on the assumptions underlying the calculations used to determine particle trajectories through the impactor stages. With these qualifications, the average "effective" particle density decreases from about 1.5 g/cm^3 at a mobility diameter of 48 nm to about 0.2 g/cm^3 at 340 nm. Yet, at each size, the particles have a wide range of densities.

For steady state applications, the SMPS has the advantage of narrow size resolution and the ability to record particles diameters to 10 nm or less. The ELPI lower limit of about 35 nm does not allow it to record ultrafine particles. However, the ELPI has the advantage that it can measure particle sizes up to $10 \mu\text{m}$ (though caution must be exercised before extending volume distributions to this size when many small, but no large particles, are expected). In transient tests, the ELPI has the advantage of being able to record size distributions on a second by second basis. Though it can only be used at a single size per test, the SMPS remains useful to examine the transient emissions of ultrafine particles.