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Evaluating the Health Impacts of Ethanol Blend Petrol

Final Report KW48A/17/F3.3

June 2008

Department of the Environment, Water, Heritage and the Arts

Appendix B5

Treatment of Particle Size Distributions

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Important Disclaimer

1. TREATMENT OF PARTICLE SIZE DISTRIBUTIONS

Newer engine technology is leading to smaller emissions from petrol fuelled vehicles (EMEP/CORINAIR, 2007, Maricq et al., 1999). A consequence of this is that there has been increasing attention devoted to the use of real time particle counters to estimate the mass of emissions. However, this has proven to be far from straight forward as complications arise due to the measurement technique as well as the physical and chemical characteristics of the particles (Mohr et al., 2000, Kittelson et al, 2004, 2006).

In the current project two real-time particle analysers are employed, namely the EEPS (Engine Exhaust Particle Sizer) and the ELPI (Electrical Low Pressure Impactor). Both impart charge to the aerosol using a corona discharge and measure the current as particles reach specific collectors thereby counting the number of particles in a particular size bin. Due allowance is made for the degree of charging as a function of particle size. Plots of the number distribution as a function of particle diameter as determined by the EEPS or the ELPI nearly always show the ELPI distribution shifted to larger diameters and with higher counts in the large particle sizes than might be expected by extrapolating the EEPS distribution (see eg Zervas E and Dorlhene P, 2006.; Kittelson et al, 2004). This is also the case in this study as shown in Figure B5.1 for E10, other fuels exhibit similar distributions

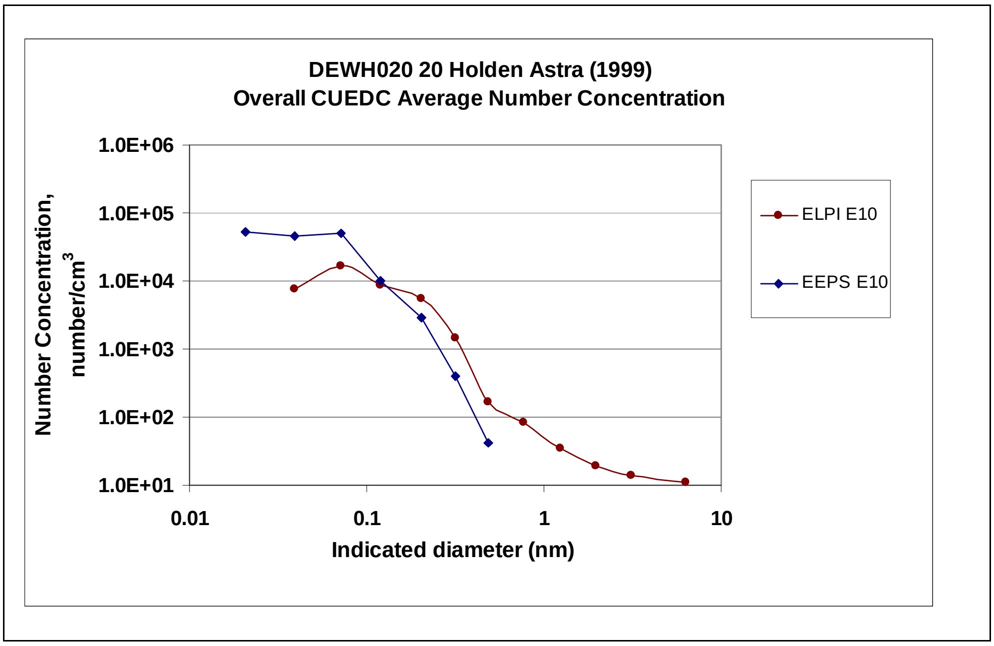

Figure B5.1 – Plot of particle number distribution with particle

Part of the reason, at least, lies with the degree of particle charging in respect of sooty agglomerates which are characteristic of vehicle exhaust.

The EEPS classifies the size of the particles according to their electrical mobility. The mobility is proportional to the charge on the particle and inversely proportional to the diameter. Charged particles pass down an annular tube made up of a series of insulated electrical collectors. The more mobile particles find their way across the gas stream to the topmost collectors whilst the larger ones take longer and arrive lower down. Measurement of the current generated in each of the collectors provided estimates of particle numbers. The EEPS has a sizing range from 10-600 nm, smaller than the ELPI which goes from 50 – 10,000nm.

By contrast, the ELPI classifies the particles according to their aerodynamic mass. The rate of charge arriving on each of the series of electrically insulated impactor plates provides a count of the particle numbers for sizes bins ranging from 50 – 10,000 nm. The charge on a given particle is proportional to its diameter over the ELPI size range but increases with the square of the diameter as the particles get bigger due to the onset of a different charging mechanism (field charging). For example, for a given charging situation, particles of 40 nm diameter are estimated to have an average of 0.26 charges per particles whilst 4000 nm particles average 2580 charges/particle (Hinds, 1999). A further complication that affects the ELPI is that different particles of similar aerodynamic size can have different physical diameters and thus deposit different amounts of charge on the same collector. Particles of diameter ten times that expected aerodynamically are not uncommon each of which would be recorded as 100 particles for that impactor. At the large end of the size spectrum, a few agglomerated particles can readily distort the mass distribution. The presence of such agglomerates is obvious in Figure 2 which was observed for impactor plate 9 (2.32 µm) with an optical microscope. If the particles were solid then all should appear similar to the smallest in the photograph ie ~ 1/5th the 10 µm scale bar. Lower plates are likely to have greater proportions of sooty material.

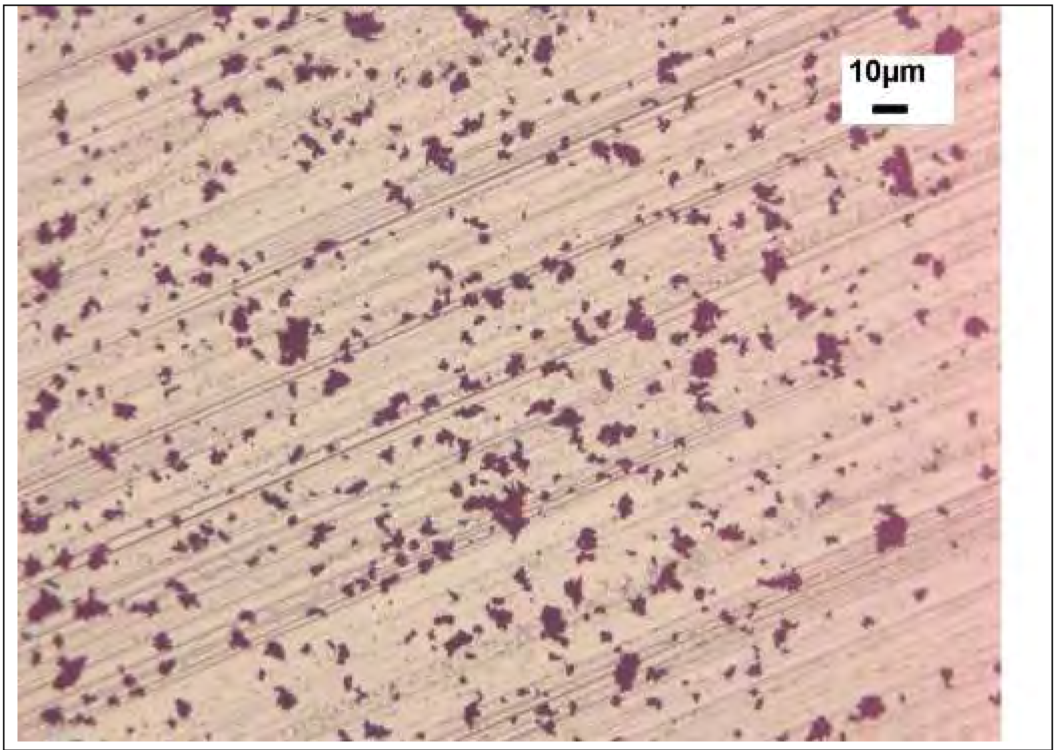

Figure B5.2 – Sooty agglomerates on ELPI plate 9 (aerodynamic dia. 2.32 µm). Note some particles exceed 10 µm in diameter

Typically the ELPI plates are used over a test period spanning a batch of three vehicles each performing multiple tests. In order to better understand the ELPI's particle behaviour a separate experiment was performed whereby the ELPI plates were used for only a single CUEDC test, thereby providing a comparator to the multiple test observations. This single test was performed for DEWH20 Astra (~1999).

Examining the situation in more detail, we find that, for DEWH20 particle numbers exceed $6x10^5$ at the 6.2 micron collection plate alone which translates to ~0.3 mg and that this accounts for ~80% of the total ELPI mass. Electron microscopic examination and manual counting of the material collected on the topmost impactor plates (10 and 11) from 30 CUEDC tests covering a range of vehicles demonstrates that the actual number of particles collected is much less than counted by the ELPI as is shown in the Table B5.1.

Table B5.1 – Comparison of the averaged ELPI count (over 30 vehicle tests) with manual count/30 from electron micrographs

D_{50} (um)	ELPI particle nos /run (ELPI Count)	Microscopic nos /run (Manual Count)
3.09 (plate 10)	839,000	10,114
6.29 (plate 11)	662,000	605

Particles from the single CUEDC test (performed with DEWH20) were also counted, this time by optical microscopy. The data for the same plates as in Table B5.1 are listed in Table B5.2.

Table B5.2 – Comparison of the ELPI count for DEWH20 (one vehicle test) with manual count from optical microscopy

D_{50} (um)	ELPI particle nos /run (ELPI Count)	Microscopic nos /run (Manual Count)
3.09 (plate 10)	950,000	13,272
6.29 (plate 11)	730,000	996

These results have been used to adjust the ELPI distribution to match the particle numbers determined by microscopy for plates 10 and 11. The attenuation factors have been extended to particle sizes down to 0.76 µm by extrapolation to provide a smooth union with the remaining ELPI data. The attenuation factors are given in Table B5.3.

Table B5.3 – Attenuation factors (AF) for ELPI stages 0.76 µm and above

D_{50}	AF
0.76 (plate 07)	5
1.23 (plate 08)	10
1.97 (plate 09)	23
3.09 (plate 10)	76
6.29 (plate 11)	900

Converting number distribution to volume or mass distribution entails a number of assumptions. The instrument calibration curves assume that the density is (ρ) is 1 g/cc and that the particle mass is $\pi/6D^3\ \rho$. For the ELPI, if the aerosol contains large sooty agglomerates, the particles carry much more charge than would be expected on the basis of their aerodynamic size and thus the ELPI records the arrival of many particles instead of just one. To correct for this, it is assumed that particles up to 0.76 µm are in the diffusion charging regime so that the acquired charge is proportional to the physical diameter. The diameter of soot agglomerates can be deduced from the literature values of soot density versus diameter (Maricq MM and Ning Xu, 2004). These authors show that a 1 µm diameter soot particle has a density of only ~ 0.1 g/cc. As the diameter is proportional to the cube root of the density, the changes are less dramatic. Such a correction cannot be readily applied to particle sizes above 0.76 µm for two reasons at least. The first is that the charging regime changes from diffusional to field in nature meaning that the extent of charge on a particle becomes proportional to the surface area of the particles rather than just the diameter. Secondly the electron micrographs indicate the presence of very significant proportion of non-agglomerated inorganic material on the largest impactor plate, more than might be deduced from the particle size distributions in Figure E.2. Thus correcting for excess charging would require knowledge of the particle composition. This reinforces the decision to match the actual particle counts to the ELPI response function.

All the ELPI data have been processed using the above attenuation factors. That is particles numbers with diameter 0.76 µm and above have been converted to mass using the factors in Table E.3 with finer-sized particles assumed to be made up of sooty agglomerates with measured numbers in need of correction for excess charge due to their diameters being larger than aerodynamic expectations. At the time of writing a uniform density across all particle sizes has been assumed to calculate the PM mass fractions. While a density correction based on the above discussion is currently being applied it is not anticipated that it will change the results, significantly.

Kittelson et al, 2003 and Zielinska et al, found that more than 98% of PM from gasoline fuelled vehicles was $PM_{2.5}$ with more than 93% of the total being within the PM_1 size fraction. However Yang et al found 89% $PM_{2.5}$ and 83% PM_1 for motor cycle emissions

2. EXAMINATION OF PARTICLES ON THE ELPI COLLECTION PLATES

The ELPI selectively captures particle on a size basis with the particles being deposited on cleaned foil substrates.

Figure B5.2 displays a series of 12 ELPI impactor stages from this current work. The particle size fractions shown increase from 0.039µm up to the largest size fraction where particles with diameters greater than 10µm are collected. This data represents the particles deposited from over 20 CUEDC tests from a range of test vehicles. This is qualitatively consistent with the observation that the majority of PM is below 2.5µm.

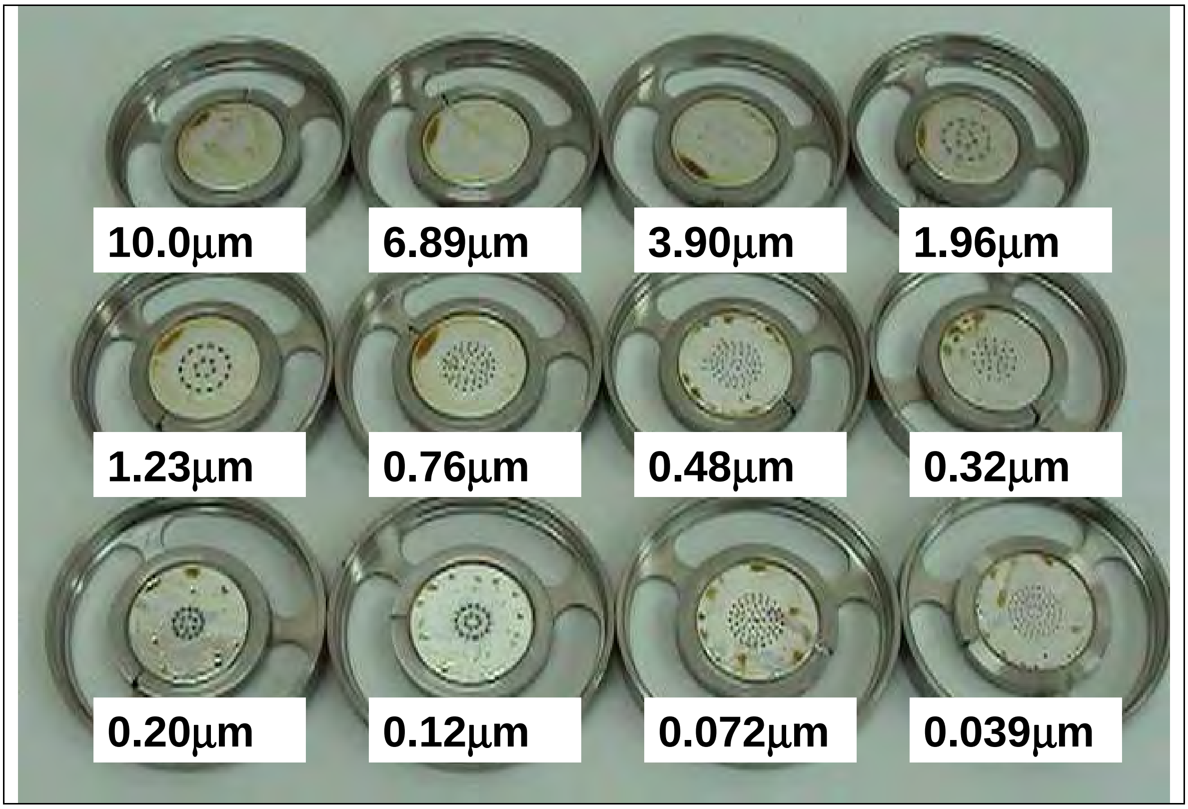

Figure B5.2 – ELPI Substrates showing Typical Particle Deposits

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