Fast response measurements of the dispersion of nanoparticles in a vehicle wake and a street canyon

Prashant Kumar^{*,a}, Alan Robins^a and Rex Britter^b

^a*Faculty of Engineering and Physical Sciences, University of Surrey, Guildford GU2 7XH, UK*

^b*Senseable City Laboratory, Massachusetts Institute of Technology, Boston, USA*

Abstract

The distributions of nanoparticles (below 300 nm in diameter) change rapidly after emission from the tail pipe of a moving vehicle due to the influence of transformation processes. Information on this time scale is important for modelling of nanoparticle dispersion but is unknown because the sampling frequencies of available instruments are unable to capture these rapid processes. In this study, a fast response differential mobility spectrometer (Cambustion Instruments DMS500), originally designed to measure particle number distributions (PNDs) and concentrations in engine exhaust emissions, was deployed to measure particles in the 5–1000 nm size range at a sampling frequency of 10 Hz. This article presents results of two separate studies; one, measurements along the roadside in a Cambridge (UK) street canyon and, two, measurements at a fixed position (20 cm above road level), centrally, in the wake of a single moving diesel-engined car. The aims of the first measurements were to test the suitability and recommend optimum operating conditions of the DMS500 for ambient measurements. The aim of the second study was to investigate the time scale over which competing influences of dilution and transformation processes (nucleation, condensation and coagulation) affect the PNDs in the wake of a moving car. Results suggested that the effect of transformation processes was nearly complete within about 1 s after emission due to rapid dilution in the vehicle wake. Furthermore, roadside measurements in a street canyon showed that the time for traffic emissions to reach the roadside in calm wind conditions was about 45 ± 6 s. These observations suggest the hypothesis that the effects of transformation processes are generally complete by the time particles are observed at roadside and the total particle numbers can then be assumed as conserved. A corollary of this hypothesis is that complex transformation processes can be ignored when modelling the behaviour of nanoparticles in street canyons once the very near-exhaust processes are complete.

Keywords: Dispersion; Fast response particle measurements; Nanoparticles, Particle number distribution; Street canyon; Vehicle wake

*Corresponding author. Division of Civil, Chemical and Environmental Engineering, University of Surrey, Guildford GU2 7XH, UK; Tel.: +44 1483 682762; Fax: +44 1483 682135. Email addresses: p.kumar@surrey.ac.uk, prashant.kumar@cantab.net (P.Kumar)

1. Introduction

Recent studies have indicated that ultrafine particles (those below 100 nm in diameter) may have greater impacts than coarse particles (PM_{10} or $PM_{2.5}$) on human health, atmospheric visibility and the global climate (Donaldson et al., 2005; Horvath, 1995; IPCC, 2007). These particles possess negligible mass, but contribute significantly to particle number concentrations (PNCs). Measurements of ultrafine particles (including their size distributions) are still rare, because of the lack of standard methods and instrumentation to measure them, the limited knowledge of their dispersion and the complex particle dynamics. This article describes the use of a fast response particle spectrometer (Cambustion DMS500) to measure particle number distributions (PNDs) and concentrations in the 5–1000 nm size range. This instrument is capable of measurements over two size ranges (i.e. 5–1000 nm and 5–2500 nm) but our interest was confined to the former. Measurements were taken in the open-air in a vehicle's wake and at the roadside. This is believed to be the first time a DMS500 was deployed for such experiments, because this instrument was originally designed for real-time measurements of engine exhaust emissions where the levels of PNCs are well-above (i.e. many times larger than those in urban environments) its noise level.

PNDs change rapidly in the wake of a moving vehicle. This change is mainly governed by the competing influences of transformation processes (i.e. coagulation, dilution, condensation and nucleation) and dilution (Kittelson, 1998; Zhang and Wexler, 2004a, b; Zhang et al., 2005). Information on such time scales was not available previously because most of the widely used instruments (e.g. the Scanning Mobility Particle Sizer, the Electrical Low Pressure Impactor or

the Aerosol Particle Sizer) have slower sampling frequencies than required for useful experiments.

This article presents results of studies which were taken separately in two different environments. Firstly, sample measurements (10 Hz) for a short duration were made along the roadside in Pembroke Street (Cambridge, UK). The aim was to test the suitability and to suggest the best operating conditions of the DMS500 for *ambient* measurements. Secondly, fast response measurements (10 Hz) of PNDs were made in the *wake* of a moving car. The aim was to investigate the *time* scale over which transformation processes and dilution affect PNDs. The *time* taken for vehicle emissions to reach the roadside was also measured in Pembroke Street. It was compared with the time scales of the transformation processes responsible for changing PNDs after emissions in the wake of a moving vehicle. This comparison allowed us to formulate a hypothesis on the importance of particle dynamics during ambient measurements. The proposed hypothesis could be useful for developing models of nanoparticle dispersion.

2. Application of a DMS500 for ambient measurements

Before using the DMS500 for ambient measurements, it was necessary to: (i) check the sensitivity level of the instrument, and (ii) identify suitable operating conditions (mainly sampling frequency) of the instrument that optimises its utility.

2.1 Noise and sensitivity of the DMS500

The sensitivity is defined as the smallest detectable signal that exceeds the peak-to-peak noise or, if peak-to-peak noise is not defined, as the smallest signal that can be identified from the noise with a 99% confidence (Cambustion, 2008). Fig. 1 shows the typical noise is greatest at the highest sampling frequency (10 Hz) and reduces with increasing sample averaging time (Cambustion, 2008). Typical street background and roadside PNDs at Pembroke Street have also

been plotted in Fig. 1 (see Kumar et al., 2008b, c for the site description and experimental methodology). Street background refers to the PNDs measured at a time when there had been no movement of traffic in the street for about 10 min (considered as the maximum ventilation time of the street) prior and subsequent to measurements. The reason for plotting these PNDs was to compare the noise level of the instrument with the minimum level of PNDs in the street. All measurements were taken at the highest sampling frequency of 10 Hz, where the instrument signal noise is also greatest. Comparison of the measured PNDs with the maximal noise (i.e. at 10 Hz) indicate that both the roadside and background PNDs are far above the noise level of the instrument for all diameters except particles below about 7 nm. It should be noted that our later studies showed that there were substantial losses of particles less than 10 nm in the sampling tubes, a factor that was not considered in these initial experiments due to small length of sampling tubes (Kumar et al., 2008d). Considering these observations, it was decided that a relatively low sampling rate (i.e. 1 Hz or lower) rather than maximum (10 Hz) was more appropriate for ambient measurements, unless the experiments relied critically upon fast response data. This sampling rate was used in our later street canyon measurements (Kumar et al., 2008a–d, 2009a, b) and not only helped maintain the measured PNDs well above the instrument’s noise level, but also reduced the size of data files to more manageable proportions during continuous measurements over many days.

2.2 Time between vehicle emissions and measurements at roadside

A sample survey was performed in Pembroke Street to estimate the lapse time of emissions for various types of vehicles. Lapse time is defined as the time for the emissions of a moving vehicle to reach the roadside (i.e. to the DMS500 which was placed ≈ 2 m away from the kerb and operated with a 2.5 m long sampling tube (see Kumar et al., 2008b, c for details). Sampling

1 tubes were thermally and electrically conductive; they were made of silicon rubber to which
2 carbon had been added to reduce particle diffusion losses (see Kumar et al., 2008b, d for further
3 details). The DMS500 was calibrated by the manufacturer and all measurements were made
4 within the validity period of certified calibration. Wind conditions during the survey period were
5 light (i.e. less than 1.5 m s^{-1}); these were recorded above the roof (i.e. 16.60 m above road level)
6 of the Chemical Engineering Department, which is on the northwest (NW) side of Pembroke
7 Street (refer Kumar et al., 2008c for a detailed site description). The sample survey was
8 conducted over 17 days during the street canyon measurement campaign; other results of this
9 study are presented in Kumar et al. (2008c). Average traffic volumes were 229 ± 92 , 1142 ± 71 ,
10 705 ± 177 , 1147 ± 45 and $471 \pm 120 \text{ veh h}^{-1}$ during the periods 00:00–07:00, 07:00–09:00, 09:00–
11 18:00, 18:00–20:00 and 20:00–24:00 h, respectively. As seen in Fig. 2, about 50 cars and vans
12 and a similar number of other vehicles (buses, trucks etc.), moving at an average measured speed
13 $30 \pm 7 \text{ km h}^{-1}$, were sampled. Data were selected for the periods when winds were near-calm
14 ($\leq 1.5 \text{ m s}^{-1}$) and PND signatures of passing vehicles could clearly be observed by the DMS500
15 for a period of a few seconds. In these circumstances, the average lapse time for all vehicles
16 surveyed was found to be about $45 \pm 6 \text{ s}$ (Fig. 2).

17 In a similar study, Minoura et al. (2009) used an engine exhaust particle sizer for roadside
18 nanoparticle measurements in Tokyo (Japan) and observed a cyclic change in number
19 concentrations over about 2 s when a single vehicle passed the sampling location. In such cases,
20 when winds are generally calm, the dominant mixing mechanism is expected to be traffic-
21 produced turbulence (Sollazo et. al., 2007), there being no organised canyon vortex to carry
22 emissions towards the road side (De Paul and Sheih, 1986). In practice, the lapse time may vary

considerably, depending on the local canyon geometry, sampling location, traffic conditions and the above-roof wind speed and direction.

This time scale derived from the data obtained in Pembroke Street describes the transport of vehicle emissions to the roadside in a particular street canyon during calm wind conditions and is used for comparison with the PND evolution time scale in vehicle wakes (see Section 4.2 for details). Note that even if the lapse time scale is smaller than the observed value of ~ 45 s under other street canyon geometry, traffic and meteorological conditions, it will in nearly all circumstances remain far larger than the evolution time observed in the vehicle wake study presented in Section 4.2. Therefore, its precise value does not affect our comparison to any significant degree.

3. Measurements in the wake of a moving diesel car

3.1 Site description

Measurements were performed at the New Museum Site of Cambridge University, just in front of the Chemical Engineering Department building (see Fig. 3). The entrance of the site has a 15 m long, 5 m wide and 4.5 m deep covered passage, which is followed by a 10 m wide and 60 m long, straight and open road leading to other departments of the University. Sampling was carried out on the straight section of this open road (i.e. about 10 m away from the end of the passage). This road runs approximately northwest to southeast. Buildings on either side of the road are about 15 m high; 3 m away from the road on the northeast side and about 20 m away on the southwest side.

3.2 Instrumentation and data acquisition

A DMS500, with sampling tube 2.5 m long and 7.85 mm internal diameter, was used to measure PNDs in the 5–1000 nm size range at a sampling frequency of 10 Hz (see Section 2.2 for further details). Measurements were taken at a single point, 20 cm above the road level, centrally, in the wake of a single diesel-engined car that was moving at a speed of $\approx 15 \text{ km h}^{-1}$ (as illustrated in Fig. 3). In fact, several sets of measurements were taken for a variety of vehicles, but in most cases there was insufficient time before or after the observations in which the background was established and could be characterised. Therefore, results for a particular diesel-engined car were selected as an example for discussion in this study. In the selected case there was no movement of traffic for about 10 min (i.e. assumed as the clearing time of emissions) prior and subsequent to the vehicle passing over the sample point.

4. Results and discussion

4.1 Temporal Variation of Particle Number and Mass Distributions

The PNDs and PMDs are plotted as a function of particle diameter in Fig. 4 on the primary and secondary y-axes, respectively. The PMDs ($dM/d\log D_p$) were obtained by multiplying the PNDs ($dN/d\log D_p$) by the mass per particle $M(D_p)$ (Park et al., 2003), i.e.

$$\frac{dM}{d\log D_p} = M(D_p) \frac{dN}{d\log D_p} \quad (1)$$

$$M(D_p) = C' \rho_p D_p^{D_f} \quad (2)$$

For $dM/d\log D_p$ in $\mu\text{gm cm}^{-3}$, $M(D_p)$ is in Kg, D_p is in nm, $C' (= 6.95 \times 10^{-24})$ is a constant, $\rho_p (= 1 \text{ g cm}^{-3})$ and $D_f (= 3)$ are the density and the fractal dimension, respectively, of assumed spherical shape particles (Park et al. 2003). The fractal dimension is a measure of the

“stringiness” of the agglomerate. Long chain-like structures have fractal dimensions close to 1 while compact spherical shapes have fractal dimensions that approach to 3 (Friedlander, 2000).

Fig. 4 includes 28 sub-figures, each incremented from the previous by 0.1 s, starting from time $t = -0.8$ and running to $t = 21.7$ s; where $t = -0.8$ s is the time when the tail pipe of the vehicle passed over the sampling point. The origin $t = 0.0$ s is the time when a first indication of exhaust emissions was observed. These figures cover three stages: (i) pre-evolution, (ii) evolution, and (iii) post-evolution. Changes in PNCs and PMCs (total and without site-background) during these stages are shown in Figs. 5 and 6, respectively. These figures provide a clear summary of the observations, each showing a similar pattern (i.e. almost negligible change in concentrations during pre-evolution stage, transient increase in concentrations during evolution stage and then decreasing concentrations in post-evolution stage).

4.1.1 Pre-evolution stage

The first eight sub-figures between $t = -0.8$ and -0.1 s (Figs. 4a–h) show almost identical PNDs and PMDs, representing the “pre-evolution” stage. The distributions in this stage can be considered as site background (i.e. the presence of suspended particles in the air at the sampling site without any local emission source). The PNDs illustrate a large nucleation mode (assumed below 30 nm) peak at 15.4 nm and a small accumulation mode (assumed between 30 and 300 nm) peak at 86.6 nm (Fig. 7). The latter is not clear in the plots but can be identified by plotting them on a reduced concentration scales on y-axis. These distributions also indicate that the majority of particles, by number, are below 100 nm in diameter. As expected, the PMDs are near-zero below 30 nm, but show a small amount of mass between 30 and 300 nm. These characteristics are in accordance with the typical ambient distributions of particles, as shown in

Kumar et al. (2008a–d). Given the scales used, the PMDs are almost inconsequential in many of the pre–evolution plots; however, they become substantial during the later phase.

4.1.2 Evolution stage

The next ten sub–figures between $t = 0.0$ and 0.9 s (Figs. 4i–r) correspond to the “evolution” stage, ending when the changes in bimodal forms are no longer apparent. Our results suggest that the overall time–scale for this stage to be of order 1 s. The first evidence of exhaust emissions was seen at $t = 0.0$ s (Fig. 4i) where a small *transient* peak appeared at about 10 nm due to the injection of fresh emissions; this was no longer evident after $t = 0.1$ s. The first clear bimodal distribution (with peaks at about 16.6 and 86.6 nm) appeared at 0.4 s (Fig. 4m). The maximum number and mass distributions occurred between $t = 0.4$ s (Fig. 4m) and 0.5 s (Fig. 4n). After $t = 0.5$ s, the PNDs decrease rapidly with time (Figs. 4n–r), mainly by the mixing of emissions with ambient air due to traffic–produced turbulence (Solazzo et al., 2007; Zhang and Wexler, 2004a).

Important points to note at this stage are the changes in the diameters at which the peaks occur in the PNDs (simply referred as peak diameters) in both the nucleation (<30 nm) and accumulation (30–300 nm) modes. While the peak diameters in the nucleation mode changed from 15.4 to 17.8 nm between $t = 0.4$ and 0.9 s, those in the accumulation mode were nearly unchanged (Fig. 7). Here, both dilution and transformation processes could play an important role. The change in the peak diameters shows an influence of transformation processes, because dilution would decrease the PNDs proportionally and not change the peak diameters. Moreover, the constant peak diameter in the accumulation mode suggests that dilution is the dominant process for reducing the PNDs in this size range. However, the changes in peak diameter in the nucleation mode could possibly be due to condensation of semi–volatile vapours onto the larger

1 concentrations of pre-existing (coarser) particles (Kerminen et al., 2001; Kulmala et al., 2004).
2 Other processes such as nucleation could be important, but these would appear to be too rapid to
3 be captured by our measurements.

4 Note that lateral drift of the vehicle wake during this period ($t = 0$ to 0.9 s) due to any mean
5 flow was small, as the prevailing wind conditions were calm, $< 1.5 \text{ m s}^{-1}$ above roof level. This
6 point is reinforced by the observed time (~ 45 s) taken for emissions to reach the kerb-side. Thus
7 the changes in PNDs and PNCs were not influenced by wake drift but were a result of the causes
8 ascribed above.

9 **4.1.3 Post-evolution stage**

10 Figs. 4s-ab (from $t = 1.0$ to 21.7 s) correspond to the “post-evolution” stage. The peak
11 diameters in both modes were found to be similar (at 15.4 and 86.6 nm) to those found during the
12 pre-evolution stage. However, the particle mass and number distributions were slightly greater in
13 the accumulation mode than during the pre-evolution stage (Fig. 4a-h). These additional
14 concentrations reduced over periods of the order of 10 s before the distributions approached
15 those seen in the pre-evolution stage (see Figs. 4a-h and Figs. 4aa-ab). However, this time scale
16 is variable and may change depending on the ventilation due to the above-roof wind speed and
17 direction.

18 The aim of these experiments was to determine the time scales in various stages of the
19 “evolution” of the PNDs. The constant peak diameters in the nucleation modes in the pre- and
20 post-evolution stages and in the accumulation modes throughout all the three stages strongly
21 indicate that transformation processes were very fast and their effects very nearly complete
22 within a second of release due to rapid dilution. A more detailed study of transformation
23 processes was beyond the scope of the work presented here.

4.2 Hypothesis

The vehicle wake study showed that the effects of transformation processes were generally over within about 1 s after emission, due to rapid dilution in the near-wake region (Section 3.4.1). Furthermore, the roadside study showed that the typical time for the traffic emissions to reach the roadside was about 45 ± 6 s (Section 2.2) in near-calm wind conditions. These observations led to the hypothesis that “the effect of transformation processes on the particles are effectively complete by the time these particles are measured at roadside and total particle numbers can then be assumed to be conserved”. The measurements presented in this article are consistent with this hypothesis. The hypothesis was further tested during our recent street canyon measurements (Kumar et al., 2008b–d, 2009a, b), the results again being consistent with the hypothesis, allowing us to ignore particle dynamics during analysis of the street canyon measurements.

It is interesting to note that the evolution time of 1 s is similar to the wake residence time, which is likely to be in the range from 1–3 s in the case studied (Fackrell, 1984). This suggests that evolution, will in general, be complete by the time particles escape from the near-wake of a vehicle. In all but the strongest of cross-winds, this time scale will be considerably less than the advection time to kerb-side.

5. Summary and conclusions

This article presents the preliminary results of PND measurements in a vehicle’s wake and at the roadside using a Cambustion Instruments DMS500. Application of the DMS500 was found to be satisfactory because both the roadside and site-background PNDs were well above the instrument’s noise level. The measurements suggested that a sampling frequency of 1 Hz or lower would be acceptable. This is well below the maximum available (10 Hz) with a DMS500

1 and reduces the effects of noise considerably as well as results in more manageable data file
2 sizes. A sample survey to estimate the time for the emissions to reach the roadside in calm roof-
3 level wind conditions gave 45 ± 6 s. Furthermore, the results of vehicle wake experiments
4 indicated that dilution was an important process influencing PNDs and PNCs, and that the effects
5 of transformation processes such as nucleation and condensation were generally complete within
6 about 1 s after emission. Comparison of time scales for PNDs to stabilise with the time for
7 sampling suggested a possibility to neglect particle dynamics during roadside measurements in
8 street canyons and total particle numbers can be assumed to be conserved. Transformation
9 processes have not been discussed in detail in this work because the focus was the characteristics
10 of particles that had been within the ambient atmosphere for much longer duration than the time
11 span for rapid change in PNDs after emission.

12 This preliminary study showed the rapid evolution of particle number and mass
13 distributions in the wake of a diesel-engined car. The next step should be to perform similar
14 experiments to estimate the evolution time of particle number and mass distributions for various
15 types of vehicles running at a range of speeds in different urban settings, meteorological
16 conditions and busy routes where background PNCs differ considerably. Additionally, it would
17 be of considerable value for the development of particle dispersion models if detailed
18 investigations of the transformation processes were also carried out to help in addressing
19 important questions about the role of fine scale particle dynamics.

20 **6. Acknowledgements**

21 Authors thank Dr. Paul Fennell from Imperial College London for helping with the
22 measurements during the field study. Prof. A.N. Hayhurst and Dr. J.S. Dennis, both from
23 University of Cambridge, are thanked for the loan of a DMS500.

7. References

- Cambustion, 2008. Cambustion DMS500 Fast Particulate Size Spectrometer. Cambustion Ltd. Cambridge, UK, Downloadable from:
<http://www.cambustion.com/instruments/dms500/dmsprinciple.html>.
- Donaldson, K., Tran, L., Albert Jimenez, L.A., Duffin, R., Newby, D.E., Mills, N., MacNee, W., Stone, V., 2005. Combustion-derived nanoparticles: A review of their toxicology following inhalation exposure. *Particle & Fibre Toxicology* 5/6, 553-560.
- De Paul, F.T., Sheih, C.M., 1986. A tracer study of dispersion in an urban street canyon. *Atmospheric Environment* 20, 455-459
- Fackrell, J.E., 1984. Parameters characterising dispersion in the near wake of a building. *Journal of Wind Engineering and Industrial Aerodynamics* 16, 97-118.
- Friedlander, S.K., 2000. *Smoke, Dust and Haze: Fundamentals of Aerosol Dynamics*. Oxford University Press, UK, pp. 397.
- Horvath, H., 1995. Estimation of the average visibility in central Europe. *Atmospheric Environment* 29, 241-246.
- IPCC, 2007. *Climate change 2007: The Physical Science Basis*. Contribution of working group I to the fourth assessment report of the intergovernmental panel on climate change. S. Solomon, D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor and H.L. Miller. Cambridge University Press, Cambridge, United Kingdom.
- Kerminen, V.-M., Pijrola, L., Kulmala, M., 2001. How significantly does coagulation limit atmospheric particle production? *Journal of Geophysical Research* 106, 24119-24125.
- Kittelson, D.B., 1998. Engines and nanoparticles: a review. *Journal of Aerosol Science* 29, 575-588.
- Kulmala, M., Vehkamäki, H., Petaja, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., McMurry, P.H., 2004. Formation and growth rates of ultrafine particles: a review of observations. *Journal of Aerosol Science* 35, 143-176.
- Kumar, P., Fennell, P., Britter, R., 2008a. Measurements of particles in the 5-1000 nm range close to road level in an urban street canyon. *Science of the Total Environment* 390, 437-447.
- Kumar, P., Fennell, P., Langley, D., Britter, R., 2008b. Pseudo-simultaneous measurements for the vertical variation of coarse, fine and ultra fine particles in an urban street canyon. *Atmospheric Environment* 42, 4304-4319.
- Kumar, P., Fennell, P., Britter, R., 2008c. Effect of wind direction and speed on the dispersion of nucleation and accumulation mode particles in an urban street canyon. *Science of the Total Environment* 402, 82-94.

1 Kumar, P., Fennell, P., Symonds, J., Britter, R., 2008d. Treatment of losses of ultrafine aerosol
2 particles in long cylindrical sampling tubes during ambient measurements. *Atmospheric*
3 *Environment* 42, 8831-8838.

4 Kumar, P., Fennell, P., Hayhurst, A., Britter, R.E., 2009a. Street versus rooftop level
5 concentrations of fine particles in a Cambridge street canyon. *Boundary-Layer Meteorology* 131,
6 3-18.

7 Kumar, P., Garmory, A., Ketzel, M., Berkowicz, R., Britter, R., 2009b. Comparative study of
8 measured and modelled number concentrations of nanoparticles in an urban street canyon.
9 *Atmospheric Environment* 43, 949-958.

10 Minoura, H., Takekawa, H., Terada, S., 2009. Roadside nanoparticles corresponding to vehicle
11 emissions during one signal cycle. *Atmospheric Environment* 43, 546-556.

12 Park, K., Cao, F., Kittelson, D.B., McMurray, P.H., 2003. Relationship between particle mass
13 and mobility for diesel exhaust particles. *Environmental Science and Technology* 37, 577-583.

14 Solazzo, E., Vardoulakis, S., Cai, X., 2007. Evaluation of traffic-producing turbulence schemes
15 within operating schemes within operational street pollution models using road side
16 measurements *Atmospheric Environment* 41, 5357-5370.

17 Zhang, K.M., Wexler, A.S., 2004a. Evolution of particle number distribution near roadways -
18 Part I: analysis of aerosol dynamics and its implications for engine emission measurement.
19 *Atmospheric Environment* 38, 6643-6653.

20 Zhang, K.M., Wexler, A.S., 2004b. Evolution of particle number distribution near roadways -
21 Part II: the 'Road-to-Ambient' process. *Atmospheric Environment* 38, 6655-6665.

22 Zhang, K.M., Wexler, A.S., Niemeier, D.A., Zhu, Y.F., Hinds, W.C., Sioutas, C., 2005.
23 Evolution of particle number distribution near roadways. Part III: Traffic analysis and on-road
24 size resolved particulate emission factors. *Atmospheric Environment* 39, 4155-4166.

25

Figure Captions

Fig. 1. Noise level of the DMS500 at 10, 1 and 0.1 Hz sampling frequencies, taken from Cambustion (2008). Typical roadside and background PNDs are also plotted; these were measured at the highest (10 Hz) sampling frequency. Error bars represent the standard deviation of 20 PND samples taken at every 0.1 s.

Fig. 2. Sample measurements showing the time for vehicle emissions to reach the kerb-side DMS500 in Pembroke Street. Winds were calm ($\leq 1.5 \text{ m s}^{-1}$) during measurements.

Fig. 3. Schematic diagram of sampling site showing the sampling arrangement and location at New Museum Site (NMS).

Fig. 4. Temporal changes at 0.1 s intervals in PNDs and PMDs due to emissions from the passage of a single diesel-engined car; a–h, the pre-evolution stage where all figures show almost identical distributions that are assumed as site background; i–j, the first evidence of fresh exhaust emissions (the start of the evolution stage); k–n, the development of bi-modal distributions; o–r, completion of the evolution stage; s–ab, the post-evolution stage where distributions return to the background forms (nearly similar to a–h).

Fig. 5. Temporal change in PNCs (total and without site-background) at a fixed point in the wake of a moving diesel car.

Fig. 6. Temporal change in PMCs (total and without site-background) at a fixed point in the wake of a moving diesel car.

Fig. 7. Temporal change in peak diameters of 0.1 s averaged PNDs in both the nucleation and accumulation modes. The peak diameters of the accumulation mode are not clearly visible in the plots until about 0.2 s, but can be identified in the raw data.

Fig. 1.ppt

Fig. 2.ppt

Fig. 3.ppt

Fig. 4a-h.ppt

Fig. 4i-p.ppt

Fig. 4q-x.ppt

Fig. 4y-ab

Fig. 5.ppt

Fig. 6.ppt

Fig. 7.ppt