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Corresponding Author: Mr Prashant Kumar, M.Tech, PhD Research Scholar

Corresponding Author's Institution: University of Cambridge

First Author: Prashant Kumar, M.Tech. PhD student

Order of Authors: Prashant Kumar, M.Tech. PhD student; Paul Fennell, PhD; Rex Britter, PhD, Professor

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turbulence. The PNCs at 2.60 m were lower by 10-40 % than those at 0.20 m and 1.0 m, suggesting a possible concentration gradient in the upper part of the canyon. The PNFs were estimated using an idealised and an operational approach; they were directly proportional to the traffic volume confirming the traffic to be the main source of particles. The PNEF were estimated using an inverse modelling technique; the reported values were within a factor of 3 of those published in similar studies.

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Authors

Prashant Kumar[a, *], Paul Fennell[b], Rex Britter[a]

Affiliation

[a]*Department of Engineering, University of Cambridge, CB2 1PZ Cambridge, UK*

[b]*Department of Chemical Engineering, University of Cambridge, CB2 3RA Cambridge, UK*

[*]**Corresponding author**

Hopkinson Laboratory

Department of Engineering

University of Cambridge

Trumpington Street, CB2 1PZ, Cambridge, UK

Tel.: +44-(0)1223-332681;

Fax: +44-(0)1223-765311, +44-(0) 1223-332662 (Attention: Prashant Kumar)

E-mail address: pp286@cam.ac.uk (Prashant Kumar), rb11@eng.cam.ac.uk (Rex Britter)

Statement

The work described has not been submitted elsewhere for publication, in whole or in part, and all the authors listed have approved the manuscript that is enclosed.

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[a]*Department of Engineering, University of Cambridge, CB2 1PZ Cambridge, UK*

[b]*Department of Chemical Engineering, University of Cambridge, CB2 3RA Cambridge, UK*

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Abstract

A newly developed instrument, the 'fast response differential mobility spectrometer (DMS500)', was deployed to measure the particles in the 5-1000 nm range in a Cambridge (UK) street canyon. Measurements were taken for 7 weekdays (from 09:00 to 19:00h) between 8 and 21 June 2006 at three heights close to the road level (i.e. 0.20 m, 1.0 m and 2.60 m). The main aims of the measurements were to investigate the dependence of particle number distributions (PNDs) and concentrations (PNCs) and their vertical variations on wind speed, wind direction, traffic volume, and to estimate the particle number flux (PNF) and the particle number emission factors (PNEF) for typical urban streets and driving conditions. Traffic was the main source of particles at the measurement site. Measured PNCs were inversely proportional to the reference wind speed and directly proportional to the traffic volume. During the periods of cross-canyon flow the PNCs were larger on the leeward side than the windward side of the street canyon showing a possible effect of the vortex circulation. The largest PNCs were unsurprisingly near to road level and the pollution sources. The PNCs measured at 0.20 m and 1.0 m were the same to within 0.5-12.5 % indicating a well-mixed region and this was presumably due to the enhanced mixing from traffic produced turbulence. The PNCs at 2.60 m were lower by 10-40 % than those at 0.20 m and 1.0 m, suggesting a possible concentration gradient in the upper part of the canyon. The PNFs were estimated using an idealised and an operational approach; they were directly proportional to the traffic volume confirming the traffic to be the main source of particles. The PNEF were estimated using an inverse modelling technique; the reported values were within a factor of 3 of those published in similar studies.

Keywords: Street canyon; Fine particles; Particle number flux; Dispersion; Particle number emission factor

1 Introduction

Particulate pollution and its impact on public health in urban areas (Seaton et al., 1995; Pope III et al., 1995), the global climate and local visibility (Hovarth, 1994; Anderson et al., 2003) have been longstanding concerns of the air quality management community and regulatory authorities. Vehicle emissions are clearly a major primary source of fine particles (those below 1000 nm) in urban areas (Shi et al., 1999; Longley et al., 2003; AQEG, 2005). Ultrafine or nucleation mode particles (those below 100 nm) are formed in combustion processes or formed from the homogeneous nucleation of supersaturated vapours. Accumulation mode particles (those between 100 nm and 1000 nm) are formed by coagulation of ultrafine particles and the condensation of gases on to pre-existing particles of both modes (AQEG, 2005). Ultrafine particles contribute very little to the total mass concentration of particles (Kittelson, 1998) but are the main component, by number concentration, of particulate pollution. Currently particulate emissions are regulated by the various authorities (i.e., European Union, United State Environmental Protection Agency and many others) using PM_{10} and $PM_{2.5}$ (PM stands for particulate matter, the subscript indicates the maximum aerodynamic diameter included in the standard, in μm) mass concentration rather than number concentration (QUARG, 1996; AQEG, 2005). The case for using number concentration of fine particles as markers of potential health hazards has been made by several researchers (QUARG, 1996; Donaldson et al., 1998; Pope III, 2000) since recent epidemiological studies suggest a correlation between exposure to ambient ultrafine particles with higher number concentration and adverse health effects (Peters and Wichmann, 2001).

City street canyons are the focus of discussion as they act as a trap for vehicle-sourced pollutants. Pollutant concentrations can be several times higher than those in unobstructed locations with well mixed-air depending on traffic characteristics, street canyon geometry and turbulence induced by wind, atmospheric instability, prevailing winds and the entrainment of emissions from adjacent streets etc., making pollutant dispersion in urban street canyons a complex problem. Understanding of the nature and impact of particulate pollution is inevitably limited by the availability of reliable technology to monitor the particles and by the complexity of urban pollution dispersion. It is clearly important to advance the understanding of the measurements and the dispersion behaviour of fine particles in urban street canyons. This would be helpful to develop new or improve existing fine particulate dispersion models that will enable regulatory authorities to make better predictions of human exposure, and to design mitigation strategies in urban areas.

Several groups (Shi et al., 1999; Colls and Micallef, 1999; Vardoulakis et al., 2002; Wehner and Weidensohler, 2003; Longley et al., 2003, 2004a; Weber et al., 2006) have examined the number concentration of fine particles in urban street canyons of large cities using a scanning mobility particle sizer, electrical low pressure impactor, ultrafine particle condensation counter alone or in a combination. Our study is somewhat different: Firstly, a newly developed instrument, the 'fast response differential mobility spectrometer DMS500', was used to measure the particle number concentrations in a broad range (5-1000 nm) with a high frequency (10 Hz output data rate) and this provided near real-time continuous measurements, unlike most other studies. Secondly, the

study is of a street canyon typical of many of Britain's towns and smaller cities and unlike the street canyons studied in larger cities. Finally, the particle number concentrations (PNCs) were measured close to the road level at three different heights (i.e. 0.20 m, 1.0 m, and 2.60 m), in order to show the dispersion behaviour of particles at these heights near where people may actually inhale particles. The main aims of the measurements were the investigation of the dependence of particle number distributions (PNDs) and concentrations (PNCs) and their vertical variations on wind speed, wind direction, and the dependence of particle number fluxes on traffic volume, and finally to estimate the particle number emission factors (PNEFs) for typical urban streets and driving conditions.

2 Experimental

2.1 Site Description

Measurements were carried out on a small section of the Fen Causeway street canyon, adjacent to the Department of Engineering in Cambridge. The chosen street section is one of the busiest roads in Cambridge. This section is approximately 200 m long and 20 m wide, runs in east-west direction, and carries two way traffic on a 10 m wide road with one lane in each direction. The heights and frontage of the buildings on either side of the road are not perfectly symmetric, but they are continuous and broadly follow the east-west line of the road. Measurements were taken at three different heights (i.e. 0.20 m, 1.0 m, and 2.60 m; hereafter called A, B and C respectively). The sampling points were on the north side of the road, 0.3 m away from the wall of Department of

Engineering building, 3.05 m away from the kerb, and approximately half-way through the section length. There is a range of building heights on both sides of the roads; on the south side from 18 to 22 m; on the north side from 15 to 22 m. The distance between the buildings on either side of the road is approximately 20 m. This section of road has an aspect ratio (height to width ratio, *H/W*) of about unity and has length to height ratio (*L/H*) about 5, making it of medium length (Vardoulakis et al., 2003). The roofs of the buildings along the south side are sloped parallel to the road while the geometries of those on the north side are more complex. Traffic flow is regulated by signals at both ends of the selected section; there are pedestrian crossings at both the eastern and western ends of the road section. The average traffic speed on the selected section was estimated to be about 30 km h^{-1}, by measuring the length of time 150 vehicles took to traverse the entire length of the section.

2.2 Instrumentation

A particle spectrometer (DMS500) was used in this study. Detailed description of the working principle and the application of the DMS500 can be seen in Collings et al. (2003), Biskos et al. (2005) and Symonds et al. (2007). It is capable of measuring the particle number distribution (PND) at a frequency of 10 Hz. However, our experiments recorded the average of 10 measurements to improve the signal/noise ratio. The instrument was calibrated by Cambustion Ltd. in September 2005 and the experimental duration was within the calibration validity period of 12 months. Generally, the instrument was calibrated in two ways, by using polystyrene spheres of a known diameter

(traceable), and by comparison to a scanning mobility particle sizer. The calibration error in particle diameter measurements and sample flow rate were about 4.3 % and 2 % respectively. When compared (private communication, Cambustion) with a Scanning Mobility Particle Sizer (SMPS) during calibration the DMS500 read 3.6% higher in number for a broadband salt aerosol at 24 nm, and 20% higher for an 8 nm H_2SO_4 monodisperse aerosol. Of course the SMPS has its own limitations. The particle number measurements with the DMS500 have been found to be consistent with those from commonly deployed instruments (i.e., SMPS and Electrostatic Low Pressure Impactor) during the road side measurements of Collings et al. (2003).

A thermally and electrically conductive sampling tube, made of silicon rubber to which carbon has been added, 5.85 mm internal diameter and 5 m length, was used to obtain the air samples from each sampling points. A cyclone, with a 100 μm steel restrictor, was placed at the head of sampling tube to maintain a sample flow rate at 8 l min^{-1}, and to reduce the pressure within the sampling tube to 0.25 bar, improving the response time of the instrument. The sampling head also prevented particles larger than 1000 nm from entering the sampling tube. The residence time of the sample in this tube was estimated to be about 0.3 seconds. Hinds (1999) and Friedlander (2000) have studied particle losses in such scenarios. Of all the potential losses (i.e., sedimentation, inertial impaction, and thermophoretic and diffusion losses), those due to diffusion and inertial impaction are the most important for particles below 15 nm when using a long sampling tube such as the one used in our experiments. Theoretical estimates have shown that penetration (fraction of the entering particles that exit the tube) was 92-97 % for

particles between 5-10 nm, 97-99 % for particles between 10-15 nm and greater than 99-99.99 % for particles between 15-1000 nm in the system used for this study. Calculated particle losses were modest and are therefore not considered further.

2.3 Data acquisition

Particle measurements were taken at a frequency of one Hz, every second continuously for 10 hours between 09:00 and 19:00 h (BST), for 7 week-days on 8, 9, 12, 13, 16, 19 and 21 June 2006. To acquire a representative data set at each sampling height, the samples were taken for 20 minutes in an hour at each height, on two different occasions (i.e. 2 samples per hour, 10 minutes per sample) by manually re-positioning the sampling point every 10 minutes. Simultaneous measurements at each sampling height could not be performed due to the availability of only a single instrument; however, the fact that, sampling was done in 60 separate time periods in each day and 420 separate time periods in total whilst the PNC changed in an essentially random manner with respect to time, meant that sufficient measurements were made to draw tentative conclusions regarding the variation in PNC with height.

Meteorological data (wind speed hereafter called as reference wind speed, wind direction, temperature, and relative humidity) were obtained from a weather station operated by the University's AT&T Laboratories on the roof top of the Department of Engineering, on the north side of the road. The facility was about 40 m above road level at a point some 100 m from the sampling site. This location is above the average height for Cambridge city centre buildings and is not overlooked.

Visual traffic counts were taken throughout each period of measurement, allocating each vehicle into one of six categories i.e., cars and vans (gasoline), cars and vans (diesel), buses, light duty vehicles (LDV), heavy duty vehicles (HDV), and motorcycles.

3 Results and discussions

3.1 Particle number distributions and particle number concentrations analysed on a daily basis

The results are analysed on a daily basis and also on hourly and a half-hourly basis for some purposes in this paper; finer-scale analysis of the results will be presented in a later article. The daily average of the PND on each sampling day is shown in Fig. 1 (a-g). The PND at all the three sampling heights were found to be similar on each day. The PND on each day showed bi-modal PNDs with one peak at about 30 nm and other peak at about 100 nm. The peak at about 30 nm is attribute to particles formed by nucleation and condensation during the rapid cooling and dilution of semi-volatile species from the exhaust gases with ambient air whilst the peak at about 100 nm is attributed to particles formed in the combustion chamber with associated condensed organic matter. However, the PNDs varied from day to day depending presumably on the traffic volume, ambient meteorology (notably reference wind speed, wind direction), and possibly the presence, strength and sense of rotation of any street canyon vortex. In general, the PNDs were largest at the lowest sampling point and then decreased with increased sampling height. The only exception to this was on the 13 June where the PNDs at the two lowest sampling points were in the reverse order; a day on which the wind was generally from

the Northerly direction rather than from the Southerly direction.

3.1.1 Reference wind speed

Some of the factors influencing the PND may be more important than others in producing the day to day variation. To analyse the relative impact of these factors, the particle number concentrations (PNC) were obtained by integrating the PND profiles over the 5-1000 nm range. The daily average value of the PNC varied with the sampling height in the same way as the PNDs. Day to day variation of the PNC was quite marked as shown in Fig. 2. It should be noted that the daily averaged PNCs at each sampling height refer to the average of the hourly averages of the PNCs over all sampling hours on each day; and the hourly average of the PNCs are the average of two 10 minute samples within each hour but 20 minutes apart. The PNCs were strongly (and inversely) correlated with the reference wind speed (Fig. 2); for example the largest PNC and the smallest reference wind speed occurred on the 13 June. This dependence on the reference wind speed was clearly of prime importance with traffic volume as the next most important factor.

3.1.2 Traffic volume

The traffic volumes were counted continuously throughout the measurement period in six different categories which were identified visually, and are summarised in Table 1. The hourly traffic volume averaged over the whole sampling duration in both lanes were found to be 1566 vehicles h^{-1} with a standard deviation of 232 vehicles h^{-1}. This

comprised gasoline cars and vans (about 75 %), diesel cars and vans (19 %), buses (1 %), LDVs (3 %), HDVs (1 %) and motorcycles (1 %). The gasoline and diesel engined cars and vans were separated on the basis of sample survey performed on the measurement site where 20.4 % cars and vans were diesel engined. This local statistic compared well with the national statistic where at the end of 2005 the diesel share was little over 20.5 %, as shown by JD Power and Associates Automotive Forecasting. The deviation of the hourly traffic counts on each sampling day in all traffic categories was less than 20 % of the average value taken over all sampling days.

The correlation between the day to day variation of traffic volume and the PNCs was poor (see Fig. 2 for PNCs and Fig. 3 for traffic volume). In order to remove the prime dependence of the PNCs on the reference wind speed, the product of the PNCs and the reference wind speed was used as a primary variable and the day to day variation of this product was plotted against the traffic volume in Fig. 3. This clearly reveals that the products of the PNCs and the reference wind speed follow the traffic volume and appear to be directly proportional to it. The next important parameter was the wind direction.

3.1.3 Wind direction

The wind direction influences the flow in the street canyon. A vortex can form in the street canyon when the wind is across the canyon; this is less evident when the wind direction is parallel to the canyon. The flow can be a combination of an along street flow and a recirculating vortex flow (Belcher, 2005). Generally, in our experiments the wind direction was across the canyon; from a Northerly or from a Southerly direction. For the

9, 12, 16 and 21 June the wind was from the Southeast (SE) or the Southwest (SW). For the 8 and 19 June the wind was from the SE or SW for about 50 % and 75 % of the total sampling time respectively; otherwise the wind was from the West (W). On the 13 June the wind was from the Northeast (NE) or Northwest (NW). For the daily averaged data the PNCs decreased with the increased wind speed, showing no effect of wind direction. However more detailed half-hourly averaged data did show a slight effect of wind direction on the PNCs and this is discussed in section 3.2.

In general if the Reynolds number of the flow is large enough, so that the viscosity is no longer important and we do not consider any thermal influences or traffic generated turbulence, dimensional arguments require that the concentrations of a passive scalar must depend inversely on a reference wind speed and directly on the source release rate for any particular wind direction. Our observations are consistent with this requirement, though we have not specifically shown that the particle number behaves as a passive scalar.

The flow within the street canyon may also be affected by traffic produced turbulence (Eskridge and Rao, 1986), urban roughness elements within the canyon (Theurer, 1999), atmospheric stability and thermal effects produced by the differential heating of the walls and road within the canyon (Kim and Baik, 2001). The effects of these factors are not significant in our case except the traffic produced turbulence which may be important near the lowest level of the canyon, since the reference wind speed was always well in excess of 1.5 m s^{-1} during our entire sampling duration and there was the possibility of vortex formation (DePaul and Sheih, 1986) particularly as the wind direction was

typically at an angle of more than 30^0 to the street axis (Oke, 1988). Additionally at the wind speeds experienced during the experiments it was expected that the exchange of particles from the canyon was dominated by wind-produced turbulence rather than traffic produced turbulence (Vardoulakis et al., 2003).

3.1.4 Temperature and humidity

The day to day variations in temperature were very small during the entire sampling duration therefore the influence of temperature on the PNCs could not be distinguished. The humidity also had little variation, except for 13 June, but the large PNC observed on that day was principally due to the low wind speed.

3.2 Dependence of particle number concentrations on wind speed and wind direction based on half-hourly averaged data

The AT&T weather station provided a categorisation of the wind directions on a half-hourly averaged basis. These half-hourly averaged measurements were found to be suitable to study the effect of the reference wind speed and wind direction on the PNCs. The selected canyon runs in an east-west direction. We can broadly categorize the wind flows on a daily basis as being Southerly on all days (sampling points being situated on the windward side of the canyon) except on 13 June when it was Northerly (sampling points being situated on the leeward side of the canyon). Because we had half hour averaged wind directions it was possible to categorise the directions more finely into three groups; from the (S, SE, SW), from the (NE, NW), or from the (W).

To analyse the effect of wind speed and wind direction on the PNCs based on the half-hourly averaged data, the PNCs were averaged over the three sampling positions and plotted in Fig. 4 against the reference wind speed and wind directions for the entire sampling duration. For all wind directions the PNCs were clearly found to decrease with increasing wind speed. Only on 13 June were the sampling points on the leeward side of the canyon and those measurements were generally larger than for the other days at similar wind speeds. These observations indicate a vortex in the street canyon; a vortex that would transport pollutants away from the windward side of the canyon and towards the leeward side of the canyon producing higher concentrations on the leeward side (DePaul and Sheih, 1986; Hunter et al., 1992; Boddy et al., 2005). Somewhat surprisingly, the data for the wind from the West were much the same as that from the (S, SE, SW); possibly reflecting the small angle from an along-street wind required to produce a vortex structure.

3.3 Vertical variation of total particle number concentration

The PNCs on each sampling day at A, B and C were found to be similar but showed a discernible decrease with height (Fig. 2). Closer inspection indicated that the concentrations differences between the two lower positions were always significantly smaller (between 0.5-12.5 %) than the concentration differences between the two upper positions (between 10-40 %). The higher PNCs at the lower levels can be attributed to the presence of the points of emission close to the road level; and the smaller concentration difference between the two lower positions is indicative of a well-mixed region close to

the road level caused by enhanced mixing from traffic produced turbulence (Di Sabatino et al., 2003; Kastner-Klein et al., 2003). These results are in agreement with some street canyon models, such as the operation street dispersion model (OSPM), which assumes a uniformly mixed region close to the road level (Berkowicz, 2000). However, a consistent decrease of PNCs from the two lowest positions to the upper most position indicates a concentration gradient in the street canyon. This observation is supported by many street canyon studies (Zoumakis, 1995; Vakeva et al., 1999; Vardoulakis et al., 2002; Murena and Vorraro, 2003) for the measurement of particulates and gaseous pollutants where they reported the maximum concentration close to the canyon bottom and found an exponential decreasing concentration with the increasing height. To test whether a similar variation occurs for fine particles we tried to fit an exponential variation to the daily averaged data for each day. The PNCs on each day at A, B and C were normalised and plotted against the dimensionless height. The relationship is expressed as,

$$(C_Z - C_b)/(C_0 - C_b) = \exp[-k(z/H)] \quad (1)$$

where C_z and C_b are the PNCs at any height z and background respectively, C_0 is the PNC at road level which is assumed equal to the PNC at 0.20 m, H is the canyon height, k/H ($=k_1$) is the exponential decay coefficient in m^{-1}. The inverse of k_1 indicates the characteristic dispersion height which corresponds to the height above the road level at which the dimensionless concentration is $e^{-1}=0.37$.

The estimation of k_1 excluding sources other than traffic, required the subtraction of any background concentration. Daily background concentrations could not be directly measured during the experiments for logistical reasons. However, an estimate of

the background concentration was made using rooftop measurements that were taken on 22 June; these are not included in this paper but are presented elsewhere (Kumar et al., 2007). On this date, continuous measurements were taken between 09:00 and 19:00 h at the rooftop of Department of Engineering at about 20 m height and about 2 m away from the sampling position. These measurements should represent the background concentration on 22 June and will be similar to those of 16 June since the wind speed, wind direction, temperature, relative humidity and traffic volume were similar on both days. The value of rooftop PNCs were about 15 % of the in-canyon PNCs (average of A, B and C). If we assume the same proportion of background for each day the best fit exponential produced coefficient k_1 is 0.10 m^{-1} (see Fig. 5).

Since there are no fine particle studies available in the literature for the direct comparison of k_1 we compared our results with some street canyon studies performed for gaseous pollutants. In spite of the sparseness of data, our value of k_1 for particles in the 5-1000 nm range were close to those obtained (between 0.08-0.15 m^{-1}) by Murena and Vorraro (2003) for benzene and at the upper end of those obtained (between 0.04 and 0.07 m^{-1}) by Zoumakis (1995) for CO. Of course, further measurements with a greater range of heights in the canyon are necessary to confirm this tentative conclusion.

3.4 Dependence of particle number fluxes on traffic volume

The net particle number fluxes (PNF) out of the street canyon (i.e. the net number of particles passing through unit upper surface area in unit time) depend on the particle production rate within the canyon and any conversion or similar processes. The PNFs

were estimated in two ways; one by using an idealized approach (Caton et al., 2003) and the other using an operational approach such as that used in the OSPM model (Berkowicz, 2000). In the first approach, the PNFs were estimated using the measured PNC which was averaged over A, B and C and an estimated exchange velocity that depends directly on the reference wind velocity. Caton et al. (2003) showed that in a regular (H/W≈1) street canyon for cross canyon flow when the shear layer drives the flow and creates the turbulence the particle number flux (PNF) will vary in proportion to the external velocity (our reference velocity) (Caton et al., 2003) as,

$$PNF = C\frac{U_r}{4\sigma_0\sqrt{\pi}} \tag{2}$$

where C is the concentration inside the street canyon in # cm^{-3}, PNF is in # cm^{-2} s^{-1}, U_r is the reference wind speed in cm s^{-1} and σ_0=11 is a dimensionless parameter (Rajaratnam, 1976). In order to make an estimate of PNFs using the second approach, the exchange wind velocity between the rooftop and street level winds near the rooftop was used as 0.10 U_r (for U_r greater than 1.5 m s^{-1}) (Berkowicz, 2000), and the PNCs near the top of the canyon are predicted by using Eq. (1) with k_1 = 0.10 m^{-1}. Interestingly, the differences among the estimated PNFs from both the approaches on each day were less than 10%. This was because the exchange velocity and the PNCs used in the first approach are about 7.5 times smaller and about 7 times larger respectively than those used in the second approach.

The estimated daily average of the total PNF using Eq. (2) varied from a minimum value (2.36 × 10^5 # cm^{-2} s^{-1}) on 8 June to a maximum value (6.1 × 10^5 # cm^{-}

^{2} s^{-1}) on 21 June (Fig. 6) with a mean over the entire sampling period of 4.1×10^5 # cm^{-2} s^{-1} and a standard deviation value of 1.8×10^5 # cm^{-2} s^{-1}. Estimated values of the PNFs are similar to those directly measured by Dorsey et al. (2002) above the City of Edinburgh and Longley et al. (2004b) in a busy street canyon in Manchester, UK. Dorsey et al. (2002) measured the average PNFs in the 11 nm to 3000 nm range between 9×10^3 cm^{-2} s^{-1} to 9×10^4 # cm^{-2} s^{-1} and a value as high as 1.5×10^5 # cm^{-2} s^{-1} on some occasions. Our values of the PNFs were about 2-6 times higher than those directly measured by Dorsey et al. (2002). There could be various reasons for the higher PNFs in our case; an important difference is that our PNFs reflect the flux out of the street canyon rather than the flux coming out over the whole city, and the other reason is that the average traffic was up to 3 times larger in our experiments than in those of Dorsey et al. (2002). Longley et al. (2004b) reported the PNFs as 3.7×10^4 # cm^{-2} s^{-1} in the 100-500 nm range which was measured at 3.5 m height in a busy asymmetric street canyon between 09:00-19:00 h; these PNFs are about 10 times lower than those reported in this study. There are two reasons for these differences: Firstly, Longley et al. (2004b) only measured particles in the 100-500 nm range. Our measurements show that particles between 5 nm and 100 nm comprise about more than 50 % of the total number of particles, meaning that this previous study may have underestimated the PNFs. Secondly, average traffic volume was up to a factor of 3 larger in our experiments than this study.

The daily averaged data of estimated PNFs and traffic activities on each sampling day is plotted in Fig. 7 in order to analyse their relationship. The best fit lines were drawn for two cases (i.e. including and excluding the estimated background PNFs). The

regression coefficients obtained from both the best fit lines were close to each other showing little effect of the background and the PNFs to be directly proportional to the traffic volume.

3.5 Estimation of particle number emission factors

Modelling of urban air quality relies on having comprehensive data on the emission factors for the various vehicles under a range of driving situations. Less information is available on a particle number basis (as distinct from particle mass), and particularly for fine particles under typical urban driving conditions. However, an inverse modelling technique (Palmgren et al., 1999) can be used to estimate the particle number emission factors (PNEF) from our measurements. We assume that the selected stretch of the road is longitudinally homogeneous and that the production of the PNF due to traffic emissions within the canyon and the removal of PNF due to exchange with background from the canyon top must be equal apart from any deposition and gravitational settling losses, though these are considered to be negligible (Jamriska and Morawska, 2001). Under these conditions, the PNEF can be estimated from,

$$PNEF \approx \frac{(10^5)(PNF)(W)}{T} \tag{3}$$

where PNEF is in # veh^{-1} km^{-1}, W is the width of the canyon in cm, PNF is in # cm^{-2} s^{-1} as described in Eq. (2), and the T is the traffic volume in veh s^{-1}. But we should note that the PNF includes the contribution both from the background and traffic.

The estimated values of daily averaged PNEFs including and excluding the background were in the range of 1.43-2.63 $\times 10^{14}$ # veh^{-1} km^{-1} and 1.21-2.23$\times 10^{14}$ #

veh^{-1} km^{-1}, respectively over the entire sampling period for any average traffic speed about 30 km h^{-1}, which of course has a significant effect on the PNEFs, but it did change significantly depending on the time of the day. The background PNCs were very low (less than 15%) compared to the traffic produced PNCs, so did not significantly affect the value of PNEFs.

There are several studies in which the PNEFs were measured either in the laboratory (Rickeard et al., 1996; Kirchstetter et al., 2002; Graskow et al., 1998; Farnlund et al., 2001; Kristensson et al., 2004; Geller et al., 2005), estimated using models (Jamriska and Morawska, 2001; Gramotnev et al., 2003) or estimated in the field for highway/rural motorway conditions i.e., constant speed (Kittelson et al., 2001; Abu-Allaban et al. 2002; Kittelson et al., 2004; Corsmeier et al., 2005; Imhof et al., 2005; Zhang et al., 2005). All these studies measured or estimated the emission factors in the range of 0.4-9.9 × 10^{14} # veh^{-1} km^{-1} depending on the traffic fleet, traffic speed, measured particle size range and measurement conditions. Jones and Harrison (2006) review these studies. Only a few studies (Ketzel et al., 2003; Morawska et al., 2005; Jones and Harrison, 2006) could be located in the literature for direct comparison with our results that represent typical urban driving conditions in the street canyons.

In a Copenhagen street canyon study (Ketzel et al., 2003), for a mixed traffic fleet (6-8 % HDVs) and traffic speed about 40-50 km h^{-1}, the PNEFs in the 10-700 nm particle size range were estimated in the range of 2.8 ± 0.5 ×10^{14} # veh^{-1} km^{-1}. In another street canyon study (Morawska et al., 2005) the emission factors in the 18-880 nm size range were reported as 2.18 ± 0.57 ×10^{13} # veh^{-1} km^{-1} and 2.04 ± 0.24 ×10^{14} # veh^{-1} km^{-1} for

petrol and diesel engined vehicles respectively. In a recent study (Jones and Harrison, 2006) in London street canyon conditions, PNEFs in the 11-450 nm size range were estimated as 1.22 $\times 10^{13}$ # veh^{-1} km^{-1} and 6.36 $\times 10^{14}$ # veh^{-1} km^{-1} for LDVs and HDVs respectively, for vehicle speeds less than 50 km h^{-1}.

Our range of estimated PNEFs compare well (within a factor of 3) with the street canyon studies representing the typical urban driving conditions but overall are at the lower end of those reported in the literature. The significant reasons for this difference could be the dominance of the gasoline engined vehicles and the lower vehicle speeds measured. The emissions for the gasoline engined vehicles are much more engine-load and speed dependent than those for diesel engined vehicles (Kittelson et al., 2004) and the PNEFs for the gasoline engined vehicles can be as low as 3.7 $\times 10^{11}$ # veh^{-1} km^{-1} (Farnlund et al., 2001) and as high as 5 $\times 10^{13}$ # veh^{-1} km^{-1} at 50 km h^{-1} and 1.2 $\times 10^{14}$ # veh^{-1} km^{-1} at 120 km h^{-1} (Rickeard et al., 1996). Our PNEF estimates are smaller than those of the most comparable other study Ketzel et al. (2003); 1.21 - 2.23 x 10^{14} # veh^{-1} km^{-1} compared with 2.8 ± 0.5 x 10^{14} # veh^{-1} $km^{-1.}$ This difference may be due to the different percentages of heavy duty diesel engine vehicles in the two studies; 2 % compared with 6-8 %. Assuming that the PNEF for heavy duty diesel engine vehicles are roughly an order of magnitude larger than those for light duty gasoline engine vehicles our results can be modified to mimic their study. This produced PNEFs of our experiments of 1.7 - 3.1 x 10^{14} # veh^{-1} km^{-1} to be compared with 2.8 ± 0.5 x 10^{14} # veh^{-1} km^{-1} from Ketzel et al. (2003); as good an agreement as might be expected from the experiment and the modeling. It was also found that when the vehicle speed fell by a

factor of about two from its average speed, the PNEFs fell by a factor of about 1.5 from their average values.

4 Summary and conclusions

A newly developed instrument was used to measure the real time particle number distributions (PND) in the 5-1000 nm range at three different heights close to the road level in a Cambridge (UK) street canyon. The PNDs were found to be similar at each sampling height and showed a consistent and discernible decrease with the sampling height. Largest particle number concentrations (PNCs) were closest to the road level due to the presence of points of emissions. These observations were in agreement with most street canyon studies but in contrast to the findings of Weber et al. (2006). The PNCs at the two lowest sampling positions were very close to each other indicating a well-mixed region close to the road level, presumably due to the enhanced mixing by the traffic produced turbulence. Such observations have not been previously reported for fine particles. However these results are in agreement with the street canyon dispersion models for gaseous pollutants such as the OSPM model which assume a well-mixed region close to the road level.

The measured PNCs in the street canyon were found to be inversely dependent on the reference wind speed. The effect of wind direction on PNCs during cross canyon flow could not be confirmed due to the limited data set; however the results support the commonly held view that, due to a vortex like flow in the street canyon the PNCs were larger on the leeward side than the windward side of the street for the same wind speeds. The trend of decreased PNCs with increased wind speed was also observed on

the days when the flow was along the canyon. Such dependence, because of the fine-scale details of air flow within the canyon, was also reported by Longley et al. (2003) for fine particles and Kukkonen et al. (2001) for gaseous traffic pollutants.

Many street canyon studies for gaseous and particulate pollutants report an exponentially decreasing concentration with increasing canyon height. In our study, a consistent decrease of PNCs from the two lowest positions to the upper most position also indicated a concentration gradient. Due to sparseness of our PNC data at the upper canyon height, this trend could not confirmed. However, we tested our data set assuming similar variations; the exponential decay coefficient produced by the best fit line was similar in magnitude to those of obtained for gaseous pollutants (Zoumakis, 1995; Murena and Vorraro, 2003).

The particle number fluxes (PNF) were estimated using an idealized and an operational approach. Both approaches complemented each other, with a less than 10% difference in PNF values. Moreover, direct proportionality of the PNFs with the traffic volume confirmed the traffic volume to be the main source of particles at the measurement site.

The particle number emission factors (PNEF) were estimated using an inverse modelling technique for typical British urban streets and driving conditions. There is limited literature available on PNEFs in our considered size range for these typical conditions. The estimated PNEFs were in the range of 1.21-2.23 $\times 10^{14}$ # veh^{-1} km^{-1} with an average value of 1.57 ± 0.76 $\times 10^{14}$ # veh^{-1} km^{-1} which were within a factor of 3 than those published in similar studies (Jones and Harrison, 2006). It should be noted

that our reported PNEFs are for gasoline engined vehicles dominated traffic fleet, with a low proportion of HDVs (1 %) and buses (1 %) in the total traffic fleet, and an estimated average speed of the mixed traffic fleet about 30 km h^{-1}.

Since measurements were made only in the lowest 2.6 m of the 20 m high street canyon, this limited the scope for analysing of the vertical variations of particles across the whole height of the canyon. Meteorological data (wind speed and direction, temperature and humidity) was available only on a half hourly basis. This limited the finer-scale detailed analysis of PNCs, based on the meteorology. More detailed experiments are in progress for the study of the vertical profiles and dispersion of fine particles in typical urban streets and driving conditions at a finer scale.

5 Acknowledgements

Prashant Kumar thanks the Cambridge Commonwealth Trust for a Cambridge-Nehru Scholarship and the Higher Education Funding Council for England for an Overseas Research Scholarship (ORS) Award. The authors thank Prof. A.N. Hayhurst and Dr. J.S. Dennis for lending the DMS500 for the study. They also thank Prof. Nick Collings and Dr. Kingsley Reavell for their support and technical discussions during the study and the preparation of this manuscript.

6 References

Abu-Allaban M, Coulomb W, Gertler AW, Gillies J, Poerson WR, Rogers CF, Sagebiel JC, Tarnay L. Exhaust particle size distribution measurements at the Tuscarora Mountain Tunnel. Aerosol Sci Technol 2002; 36: 771–789.

Anderson TL, Charlson RJ, Schwartz SE, Knutti R, Boucher O, Rodhe H, Heintzenberg J. Climate forcing by aerosols-a hazy picture. Sci 2003; 300: 1103-1104.

AQEG (Air Quality Expert Group). Particulate matter in the United Kingdom. Defra, London;2005. http://www.defra.gov.uk/environment/airquality/aqeg (last accessed: 22 November 2006).

Belcher SE. Mixing and transport in urban areas. Phil Trans R Soc 2005; 363: 2947-2968.

Berkowicz R. Operational street pollution model-a parameterized street pollution model. Environ Monit Assess 2000; 65: 323-331.

Biskos G, Reavell K, Collings N. Description and theoretical analysis of a Differential Mobility Spectrometer. Aerosol Sci Technol 2005; 39(6): 527-541.

Boddy JWD, Smalley RJ, Dixon NS, Tate JE, Tomlin AS. The spatial variability in concentrations of a traffic-related pollutant in two street canyons in York, UK - Part I: the influence of background winds. Atmos Environ 2005; 39: 3147–3161.

Corsmeier U, Imhof D, Kohler M, Kuhlwein J, Kurtenbach R, Petrea M, Rosenbohm E, Vogel B, Vogt U. Comparison of measured and model-calculated real-world traffic emissions. Atmos Environ 2005; 39: 5760–5775.

Caton F, Britter RE, Dalziel S. Dispersion mechanisms in a street canyon. Atmos Environ 2003; 37: 693-702.

Collings N, Reavell K, Hands T, Tate J. 194 Roadside aerosol measurements with a fast particle spectrometer. Soc Auto Engg 2003; 20035407.

Colls JJ, Micallef A. Measured and modelled concentrations and vertical profiles of airborne particulate matter within the boundary layer of a street canyon. Sci

Total Environ 1999; 235: 221-233.

DePaul FT, Sheih CM. Measurements of wind velocities in a street canyon. Atmos Environ 1986; 20(3): 455-459.

Di Sabatino S, Kastner-Klein P, Berkowicz R, Britter RE, Fedorovich E. The modelling of turbulence from traffic in urban dispersion models - part I: theoretical considerations. Environ Fluid Mech 2003; 3: 129–143.

Donaldson K, Li XY, MacNee W. Ultrafine (nanometer) particle mediated lung injury. J Aerosol Sci 1998; 29: 553-560.

Dorsey JR, Nemitz E, Gallagher MW, Fowler D, Williams PI, Bower KN. Direct measurements and parameterisation of aerosol flux, concentration and emission velocity above a city. Atmos Environ 2002; 36: 791-800.

Eskridge RE, Rao ST. Turbulent diffusion behind vehicles: experimentally determined turbulence mixing parameters. Atmos Environ 1986; 20: 851-860.

Farnlund J, Homan C, Kageson P. Emisions of Ultrafine particles from different types of light duty vehicles, Swedish National Road Administration Publication 2001, 10: 16.

Friedlander SK. Smoke, Dust and Haze: Fundamentals of Aerosol Dynamics. Oxford University Press, UK, 2000.

Geller MD, Sardar SB, Phuleria H, Fine PM, Sioutas C. Measurement of particle number and mass concentrations and size distributions in a tunnel environment. Environ Sci Technol 2005; 39: 8653–8663.

Gramotnev G, Brown R, Ristovski Z, Hitchins J, Morawska L. Determination of

average emission factors for vehicles on a busy road. Atmos Environ 2003; 37: 465–474.

Graskow BR, Kittelson DB, Abdul-Khaleek IS, Ahmadi MR, Morris JE. Characterization of exhaust particulate emissions from a spark ignition engine. SAE Technical papers 1998; 980528.

Hinds WC. Aerosol technology: Properties, behaviour and measurement of airborne particles. 2nd edition, John Wiley & Sons, UK, 1999.

Hovarth H. Atmospheric aerosols, atmospheric optics visibility. J Aerosol Sci 1994; 25: S23-S24.

Hunter LJ, Johnson GT, Watson ID. An investigation of three-dimensional characteristics of flow regimes within the urban canyon. Atmos Environ 1992; 26B(4): 425-432.

Imhof D, Weingartner ED, Ordonez C, Gehrig R, Hill M, Buchmann B, Baltensperger U. Real-world emission factors of fine and ultrafine aerosol particles for different traffic situations in Switzerland. Environ Sci Tech 2005; 39: 8341-8350.

Jamriska M, Morawska L. A model for determination of motor vehicle emission factors from on-road measurements with a focus on sub micrometer particles. Sci Total Environ 2001; 264: 241–255.

Jones AM, Harrison RM. Estimation of the emission factors of particle number and mass fractions from traffic at a site where mean vehicle speeds vary over short distances. Atmos Environ 2006; 40: 7125-7137.

Kastner-Klein P, Fedorovich E, Ketzel M, Berkowicz R, Britter RE. The modelling of

turbulence from traffic in urban dispersion models - Part II: evaluation against laboratory and full-scale concentration measurements in street canyons. Environ Fluid Mech 2003; 3: 145–172.

Ketzel M, Wahlin P, Berkowicz R, Palmgren F. Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations. Atmos Environ 2003; 37: 2735-2749.

Kim JJ, Baik JJ. Urban street canyon flows with bottom heating. Atmos Environ 2001; 35: 3395-3404.

Kirchstetter TW, Harley RA, Kriesberg NM, Stolzenburg MR, Hering SV. Corrigendum to - On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles. Atmos Environ 2002; 36: 6059.

Kittleson DB. Engines and nano-particles: a review. J Aerosol Sci 1998; 29: 575-588.

Kittelson DB, Watts WF, Johnson JP. Fine particle (nanoparticle) emissions on Minnesota highways. Minnesota Department of Transportation, St. Paul, MN, Final Report, May 2001.

Kittelson DB, Watts WF, Johnson JP. Nanoparticle emissions on Minnesota highways. Atmos Environ 2004; 38: 9-19.

Kristensson A, Johansson C, Westerholm R, Swietlicki E, Gidhagen L, Wideqvist U, Vesely V. Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden. Atmos Environ 2004; 38: 657-673.

Kumar P, Britter R, Langley D. 2007. Street versus rooftop level concentrations of fine particles in a Cambridge street canyon. Proceedings of the 6th International

Conference on Urban Air Quality Limassol, Cyprus, 27-29 March 2007, ISBN: 978-1-905313-46-4; 147: 35-38.

Longley ID, Gallagher MW, Dorsey JR, Flynn M, Allan JD, Alfarra D, Inglish D. A case study of aerosol (4.6nm<Dp<10μm) number and mass size distribution measurements in a busy street canyon in Manchester, U.K. Atmos Environ 2003; 37: 1563-1571.

Longley ID, Gallagher MW, Dorsey JR, Flynn M, Bower KN, Allan JD. 2004a. Street canyon aerosol pollutant transport measurements. Sci Total Environ 2004a; 334-335: 327-336.

Longley ID, Gallagher MW, Dorsey JR, Flynn M. A case-study of fine particle concentrations and fluxes measured in a busy street canyon in Manchester, UK. Atmos Environ 2004b; 38: 3595-3603.

Morawska L, Jamriska M, Thomas S, Ferreira L, Mengersen K, Wraith D, McGregor F. 2005. Quantification of particle number emission factors for motor vehicles from on road measurements. Environ Sci Technol 2005; 39: 9130–9139.

Murena F, Vorraro F. Vertical gradients of benzene concentration in a deep street canyon in the urban area of Naples. Atmos Environ 2003; 37: 4853-4859.

Oke TR. Street design and urban canopy layer climate. Energy and Buildings 1988; 11: 103 -113.

Palmgren F, Berkowicz R, Ziv A, Hertel O. Actual car fleet emissions estimated from urban air quality measurements and street pollution models. Sci Total Environ 1999; 235: 101-109.

1 Peters A, Wichmann HE. Epidemiological evidence on health effects of ultrafine
2 particles. Epidemiology 2001; 12:544.
3 PopeIII CA. Review: Epidemiological basis for particulate air pollution health standards.
4 Aerosol Sci Technol 2000; 32: 4-14.
5 PopeIII CA, Dockery DW, Schwart J. Review of epidemiological evidence of health
6 effects of particulate air pollution. Inhalation Toxicology 1995; 7: 1-18.
7 QUARG (Quality of the Urban Air Review Group). Airborne particulate matter in the
8 United Kingdom. The third report of the Quality of the Urban Air Review
9 Group. Technical report, Department of Environment, London, UK, 1996.
10 Rajaratnam N. Turbulent Jets. Elsevier, Amsterdam, New York, 1976.
11 Rickeard DJ, Bateman JR, Kwon YK, McAughey JJ, Dickens CJ. Exhaust particulate
12 size distribution: vehicle and fuel influences in light duty vehicles. Society of
13 Automotive Engineers, Warrendale 1996; PA: 961980.
14 Seaton A, MacNee N, Donaldson K, Godden D. Particulate air pollution and acute health
15 effects. Lancet 1995; 345: 176-178.
16 Shi PJ, Khan AA, Harrison RM. Measurements of ultra fine particle concentration and
17 size distribution in the urban atmosphere. Sci Total Environ 1999; 235: 51-64.
18 Symonds JPR, Reavell JS, Olfert JS, Campbell BW, Swift SJ. Diesel soot mass
19 calculation in real-time with a differential mobility spectrometer. J Aerosol Sci
20 2007; 38: 52-68.
21 Theurer W. Typical building arrangements for urban air pollution modeling. Atmos
22 Environ 1999; 33: 4057-4066.

Vakeva M, Hameri K, Kulmala M, Lahdes R, Ruuskanen J, Laitinen T. Street level versus rooftop concentrations of submicron aerosol particles and gaseous pollutants in an urban street canyon. Atmos Environ 1999; 33: 1385-1397.

Vardoulakis S, Gonzalez-Flesca N, Fisher BEA. Assessment of traffic-related air pollution in two street canyons in Paris: implications for exposure studies. Atmos Environ 2002; 36: 1025-1039.

Vardoulakis S, Fisher BRA, Pericleous K, Gonzalez-Flesca N. Modelling air quality in street canyons: a review. Atmos Environ 2003; 37: 155-182.

Weber S, Kuttler W, Weber K. Flow characteristics and particle mass and number concentration variability within a busy street canyon. Atmos Environ 2006; 40: 7565-7578.

Wehner B, Weidensohler A. Long term measurements of submicrometer urban aerosols: statistical analysis for correlations with meteorological conditions and trace gases. Atmos Chem Phys 2003; 3: 867-879.

Zhang KM, Wexler AS, Niemeier DA, Zhu YF, Hinds WC, Sioutas C. Evolution of particle number distribution near roadways. Part III: Traffic analysis and on-road size resolved particulate emission factors. Atmos Environ 2005; 39: 4155-4166.

Zoumakis NM. A note on average vertical profiles of vehicular pollutant concentrations in urban Street Canyons. Atmos Environ 1995; 29: 3719-3725.

Table 1: The daily average hourly traffic counts on both lanes in various categories

Date	Cars and Vans (gasoline) (count h^{-1})	Cars and Vans (diesel) (count h^{-1})	Buses (count h^{-1})	LDVs (count h^{-1})	HDVs (count h^{-1})	Two Wheelers (count h^{-1})	Total	
							Count h^{-1}	Standard Deviation
08 June 2006	846	285	12	27	11	9	1189	125
09 June 2006	1388	466	10	39	13	19	1936	381
12 June 2006	1185	399	8	67	17	13	1688	277
13 June 2006	1153	388	11	44	15	18	1629	303
16 June 2006	1148	386	12	48	8	21	1623	165
19 June 2006	984	330	11	62	19	16	1423	134
21 June 2006	1039	348	11	46	16	17	1478	278
Average	1106	372	11	48	14	16	1566	--
Standard Deviation	172	57	1	14	4	4	234	--

Figure Captions

Fig. 1. Particle number distribution on; (a) 8 June 2006; PWD: SE (50%), W (50%) (b) 9 June 2006; PWD: SE (c) 12 June 2006; PWD: SE (d) 13 June 2006; PWD: NE (55%), NW (45%) (e) 16 June 2006: PWD: SW (f) 19 June 2006; PWD: SW (75%), W (25%) (g) 21 June 2006; PWD: SW. Acronyms WS, T, RH and PWD represent the daily average, reference wind speed, temperature, relative humidity and predominant wind direction respectively. The lines joining the triangles, circles and squares represent the PNDs at 0.20 m, 1.0 m and 2.60 m respectively.

Fig. 2. Day to day variation of PNCs at each sampling height with reference wind speed. Error bars represent the standard deviation of the hourly averaged data. The dotted lines are as aid to the eye only since the measurements were not continuous.

Fig. 3. Day to day variation of product of the PNCs and the reference wind speed at each sampling height with the traffic volume. Error bars represent the standard deviation of the hourly averaged data. The dotted lines are as aid to the eye.

Fig. 4. Effect of half-hourly averaged wind speed and direction on the half-hourly averaged PNCs during the entire sampling period. The half-hourly averaged PNCs shown here are the averages of A, B and C; and each height (A, B and C) contain 10 minutes sampling in every half-hour.

Fig. 5. Normalised vertical profiles of particle number concentrations over the whole

sampling period.

Fig. 6. Day to day variation of estimated PNFs with the traffic volume. Error bars represent the standard deviation of the hourly averaged data. The dotted lines are as aid to the eye.

Fig. 7. Relationship between the particle number flux and the traffic volume. Solid and dotted line represents the case including and excluding the background PNFs respectively. The best fit solid line is forced to pass through the background PNF values (which is the intercept of the best fit line on the y-axis) while the dotted line is forced to pass through zero on the y-axis assuming because of the absence of traffic. Error bars represent the standard deviation of the hourly averaged data.

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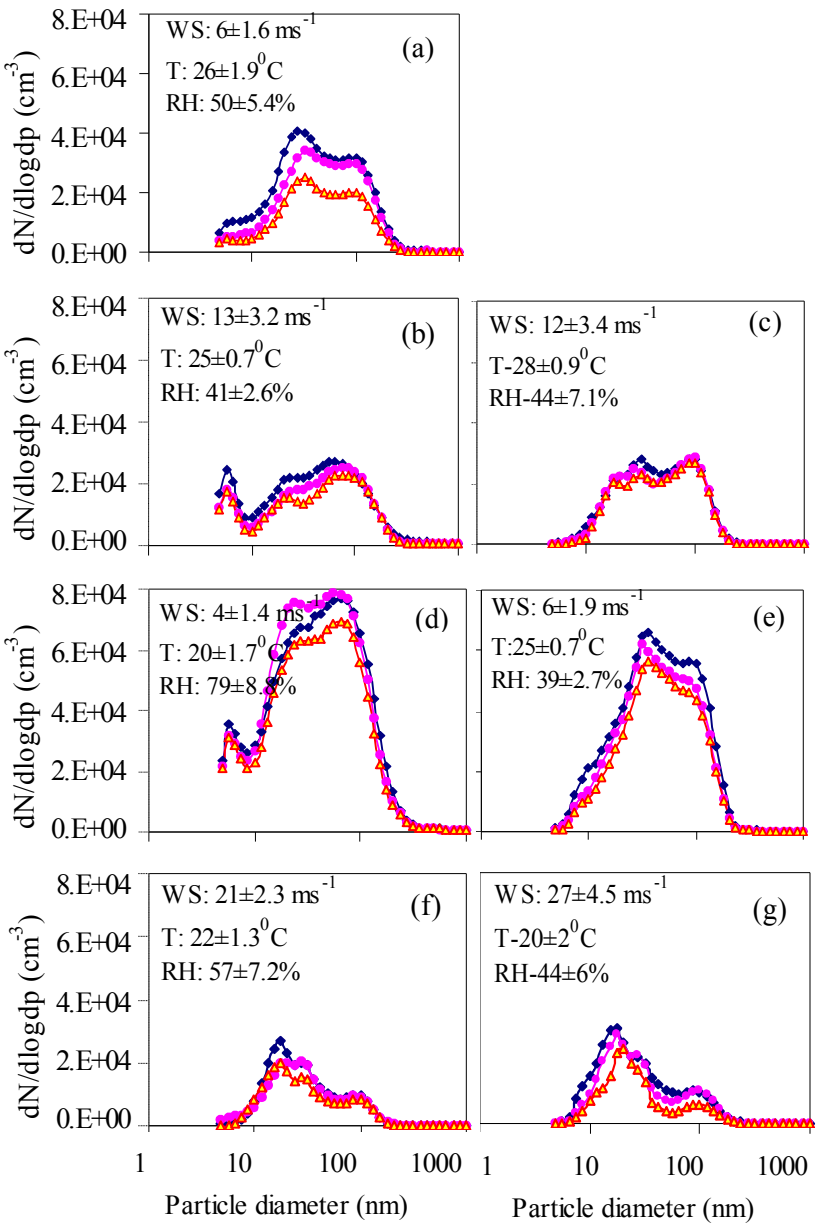

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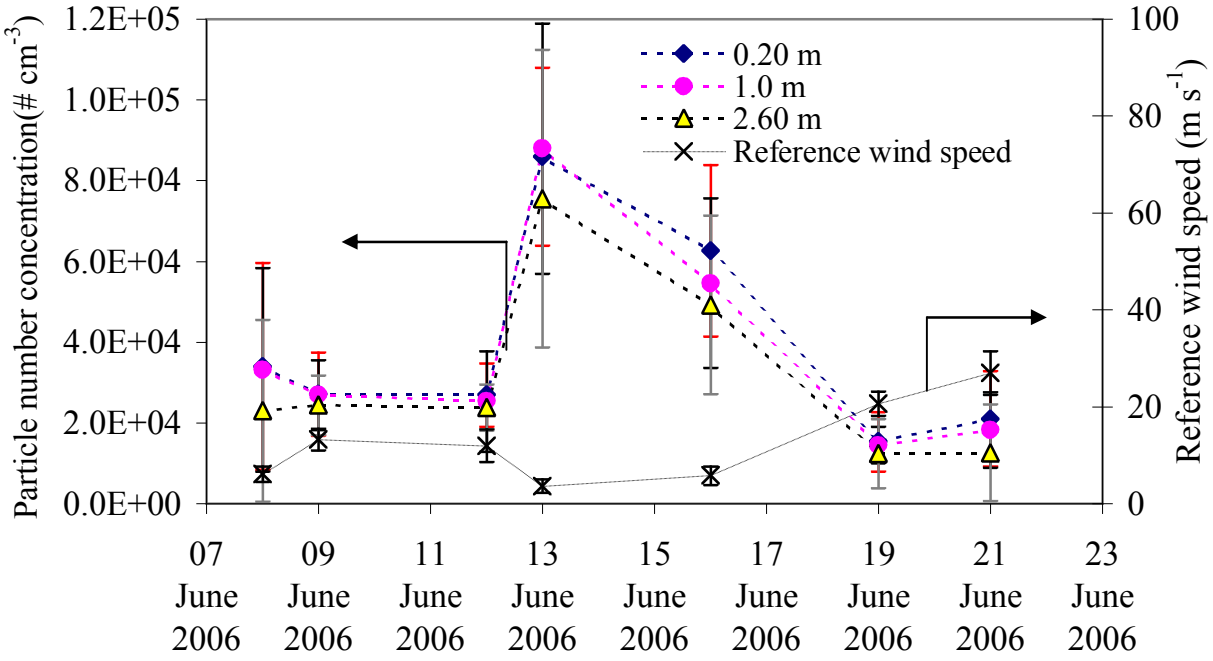

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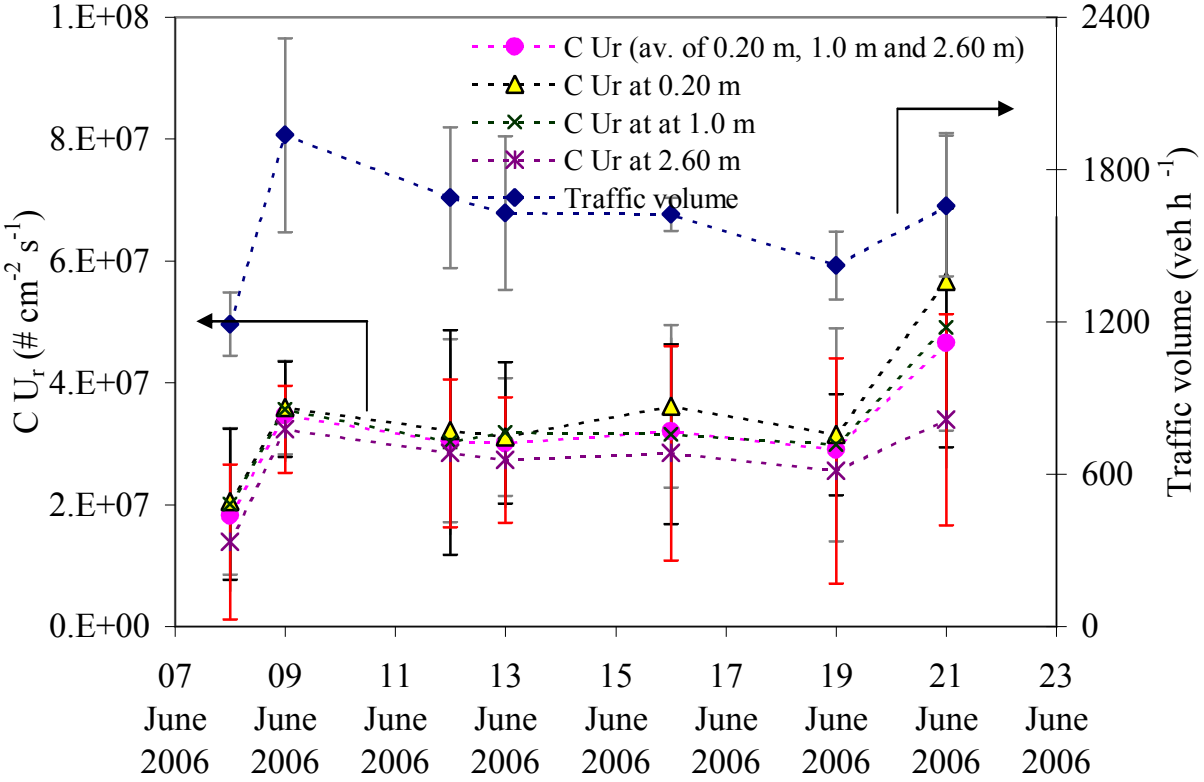

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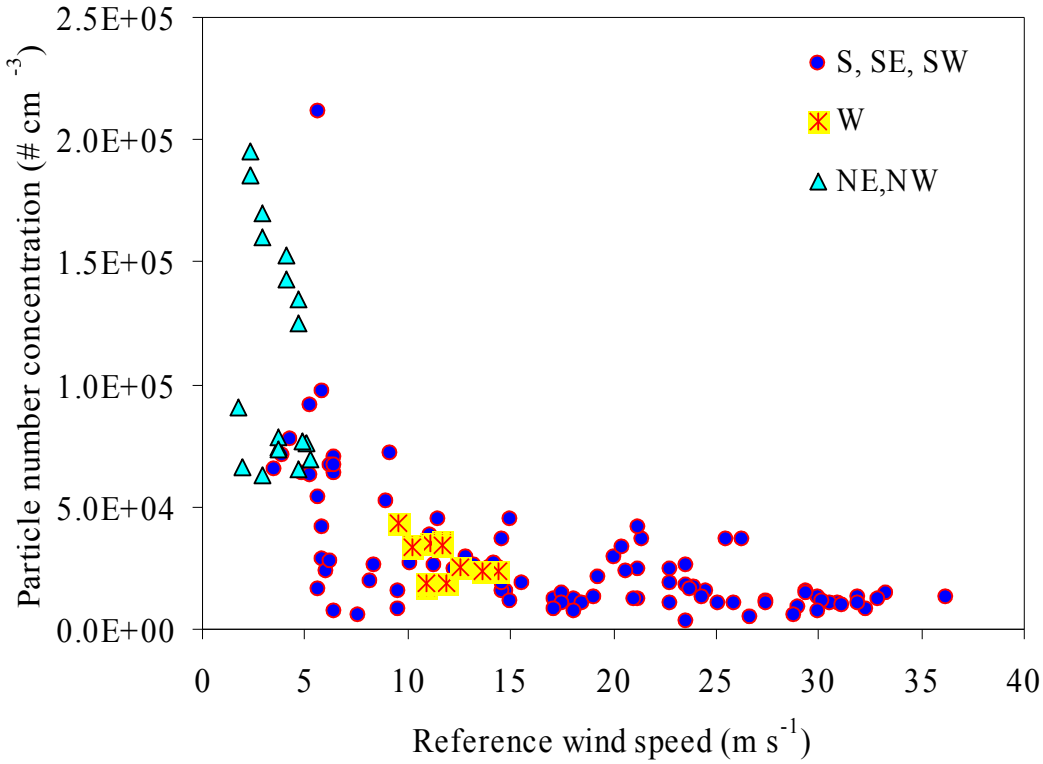

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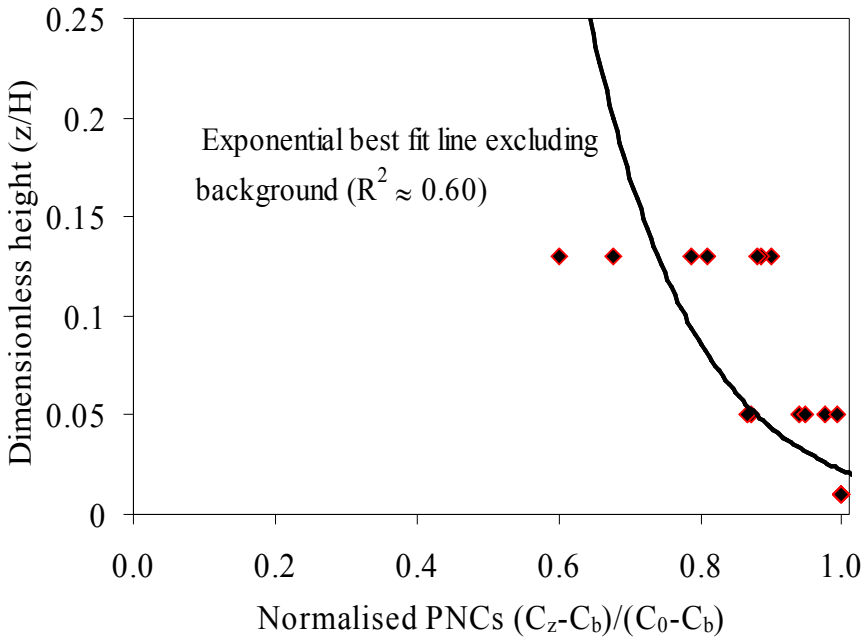

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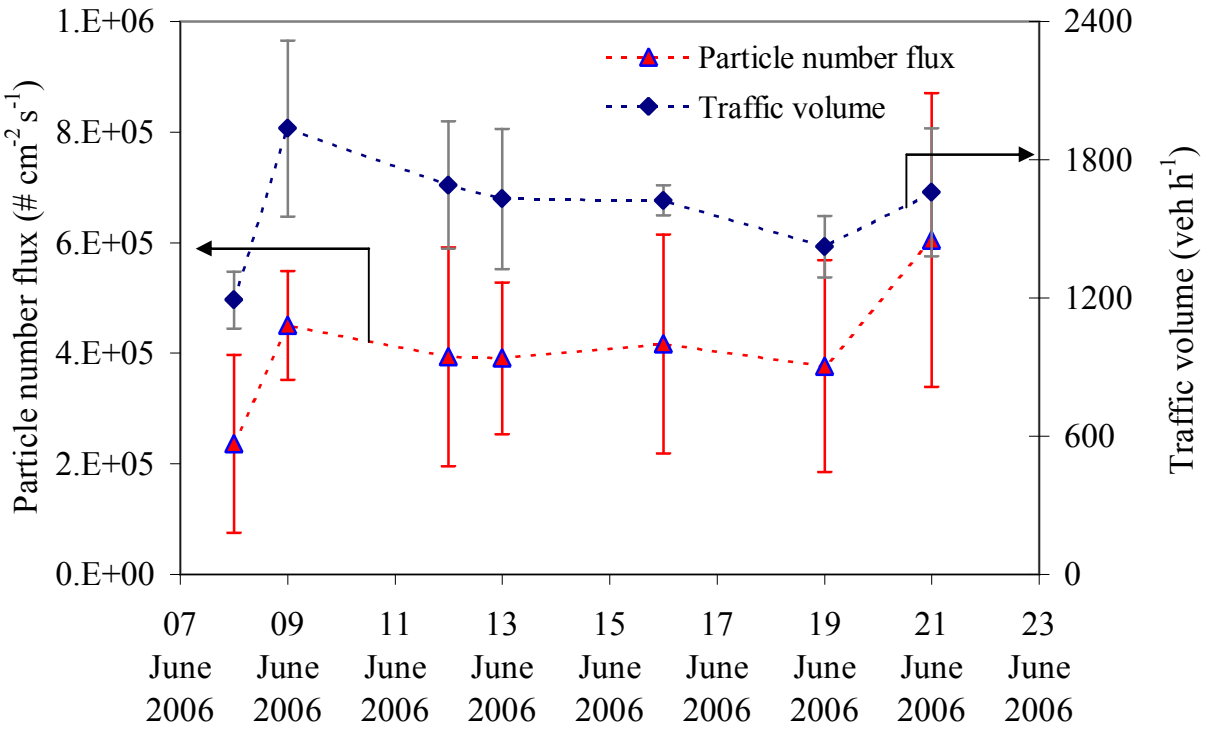

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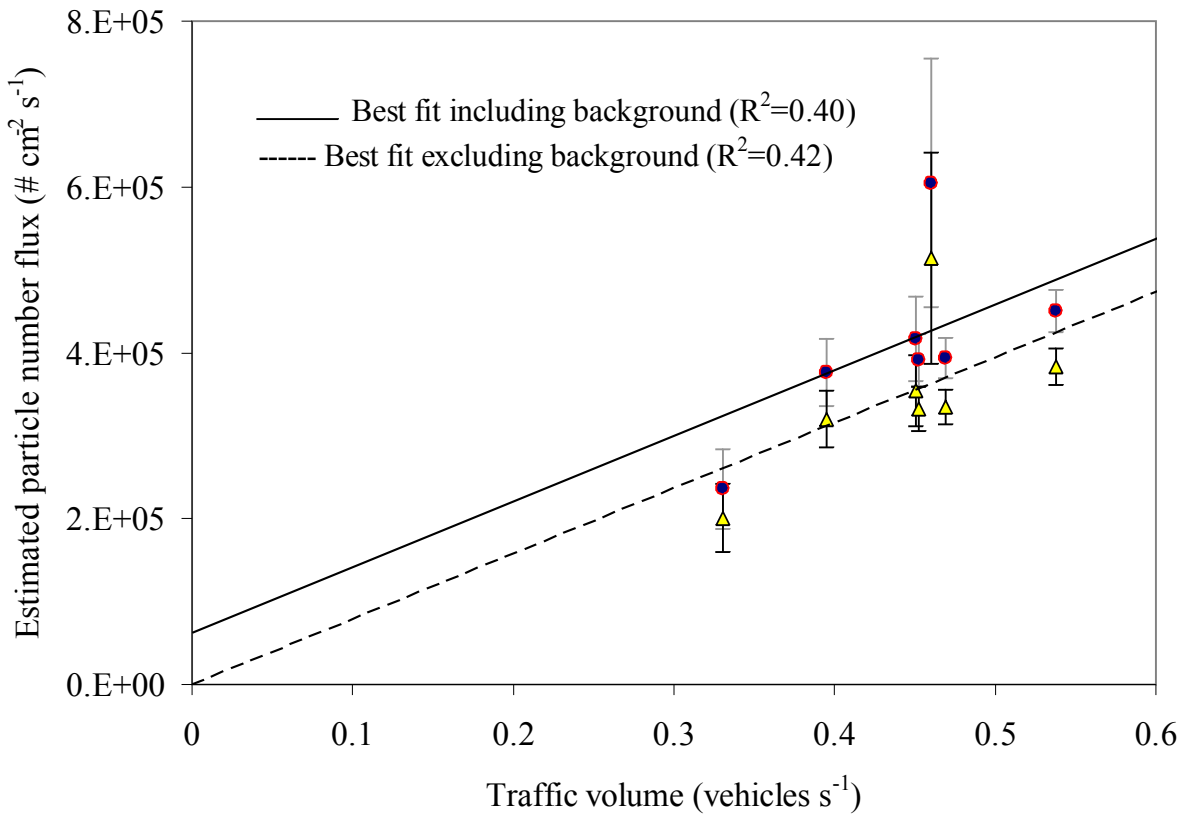


Response to Reviewer's Comments – STOTEN-D-07-00639

The authors fully accept the comments made by Reviewer #1 and 2. Detailed responses to Reviewer's comments are given below;

Reviewer #1

(a) Page 3, line 11: Changed the word 'Fine particles ... to.. *ultrafine particles'*.

(b) Page 3, lines 20-21 (now lines 20-22): Changed the sentence 'since health effects are correlated more closely with the number concentration of particles than their mass concentration (Donaldson et al., 1998).. to.. *since recent epidemiological studies suggest a correlation between exposure to ambient ultrafine particles with higher number concentration and adverse health effects (Peters and Wichmann, 2001)'*

Reference (Peters and Wichmann, 2001) included on Page 30, lines 1-2.

(c) Page 6, Section 2.2: A few recent references (Collings et al., 2003; Biskos et al., 2005; Symonds et al., 2007) have been added in Section 2 (Page 6, lines 14-16) which explains the working principle and the application areas of the DMS500.

In terms of number concentration, the calibration of the instrument is based entirely upon (a) the correct calibration of the sensitivity detection ring amplifiers based upon a known current source and (b) the computer model. There is currently no available absolute standard which could be used to make a reliable empirical calibration. As mentioned by the reviewer a Condensation Particle Counter is perhaps the closest available technology, but even it can only be described as an absolute standard when PNCs are $<10^4$, as above this value the device switches from *count mode* to *photometric mode,* the latter being based upon bulk optical properties of the aerosol and is dependent on manufacturer. An SMPS system (consisting of DMA and CPC) does allow number comparison to be made more within the working dynamic range of the DMS500, as band-pass action of the DMA reduces the concentration. Good number agreement between an SMPS and the DMS500 is obtained when the CPC is kept in count mode.

However, an SMPS is a complex system, reliant upon its own calibration, so is only used as a check.

The instrument read 3.6% higher in number than SMPS for a broadband salt at 24 nm and 20% higher for an 8nm H_2S0_4 monodisperse aerosol. But please note, there is diffusion correction in the DMS model and not in the case of SMPS model (and the DMS500 inlet flow is much higher) so more losses would be expected (especially for the long DMA used) in the case of the SMPS.

We have included the following sentences on page 7, lines 3-9 *'When compared (private communication, Cambustion) with an SMPS during calibration the DMS500 read 3.6% higher in number for a broadband salt at 24 nm and 20% higher in number for an 8nm* H_2S0_4 *monodisperse aerosol. Of course the SMPS has its own limitations. The particle number measurements with the DMS500 have been found to be consistent with those from commonly deployed instruments (i.e., Scanning Mobility Particle Sizer and Electrostatic Low Pressure Impactor) during the road side measurements of Collings et al. (2003)'.*

Given the capabilities of the DMS500 (e.g., real-time continuous measurements, fast response and broader size range) and its previous application to different areas references (Collings et al., 2003; Biskos et al., 2005; Symonds et al., 2007) the authors could not find any reason not to use this instrument for ambient measurements.

(d) Page 7, line 1 (now lines 10-11): To reduce static buildup whilst particles passing through the tube, carbon is added to make the *silicon rubber tube* thermally and electrically conductive.

The point raised by the reviewer is correct because the sampling tube is made of *silicon rubber* not of *simple rubber*. Reworded the sentence as *'A thermally and electrically conductive sampling tube, made of silicon rubber to which carbon has been added, 5.85 mm internal ....'*

These tubes are commercially available, and the manufacturers can be contacted at http://www.siliconex.com/.

(e) Page 9, line 3 (now line 12): Authors agree the reviewer's view and the latter part of the sentence *'an evidence of gasoline and diesel engine vehicles'* is removed.

However, as suggested by the reviewer, the explanation for this bimodality may be of interest to the readers which is added on page 9, lines 12-15 as *'The peak at about 30 nm is*

attributed to particles formed by nucleation and condensation during the rapid cooling and dilution of semi-volatile species from the exhaust gases with ambient air whilst the peak at about 100 nm is attributed to particles formed in the combustion chamber with associated condensed organic matter.'

(f) Page 16, line 12 (now page 17, line 4): Authors support the reviewer's view that the idealised approach should refer to a reference height.

Our results have been arbitrarily based on the concentrations averaged over the three measurement heights. An alternative approach based on a specific reference height of 1 metre was considered. The two methods produced results that were very similar (typically less than 5%).

(g) Page 21, lines 1-4 (now lines 13-22): As suggested we have included the following sentences

'Our PNEF estimates are smaller than those of the most comparable other study Ketzel et al. (2003); 1.21 - 2.23 x 10^{14} # veh^{-1} km^{-1} compared with 2.8 ± 0.5 x 10^{14} # veh^{-1} km^{-1}. This difference may be due to the different percentages of heavy duty diesel engine vehicles in the two studies; 2% compared with 6-8%. Assuming that the PNEF for heavy duty diesel engine vehicles are roughly an order of magnitude larger than those for light duty gasoline engine vehicles our results can be modified to mimic their study. This produced PNEFs of our experiments of 1.7 - 3.1 x 10^{14} # veh^{-1} km^{-1} to be compared with 2.8 ± 0.5 x 10^{14} # veh^{-1} km^{-1} from Ketzel et al. (2003); as good an agreement as might be expected from the experiment and the modeling.'

Page 23, line 4 (now page 24 line 2): To make the percentage of heavy duty engine vehicles (2%) consistent with above included sentence, the words '….*and buses (1%)'* are included.

(h) Page 21, lines 20-22 (now page 22, lines 18-21): Authors agree with the reviewer's view that this is an artefact of the small data set collected since there were only very few periods (13 June only described in section 3.1.3) throughout the sampling period when the winds were northerly and these were always below 6 m s^{-1}.

The sentence has been reworded as *'The effect of wind direction on PNCs during cross canyon flow could not be confirmed due to the limited data set; however the results support the commonly held view that, due to a vortex like flow in the street canyon the PNCs were larger on the leeward side than the windward side of the street for the same wind speeds.*

Reviewer #2

Page 3, line 21 (now line 20): This is 1998. Corrected '1988 to *1998 in Reference list on page 26, line 8 '*.

Page 4, line 13 (now line 14): Removed '*reference Harrison et al., 1998 from text* since there are enough other references quoted in the sentence to support the statement'; Corrected 'Cools to *Colls*'

Page 5, line 1 (now lines 2-3): Defined 'PNCs as *particle number concentrations*'

Section 2: The schematic diagram of street canyon is not included to save the space and reduce the length of the paper since detailed explanation of sampling positions and street canyon geometry are given in this section.

Page 6, line 13 (now line 14): As suggested by the reviewer some recent references (Collings et al., 2003; Biskos et al., 2005; Symonds et al., 2007) explaining the working principle and the application in different areas of the DMS500 are included.

We have included the following sentence on Page 6, lines 14-16 as *'Detailed description of the working principle and the application of the DMS500 can be seen in Collings et al. (2003), Biskos et al. (2005) and Symonds et al. (2007).'*

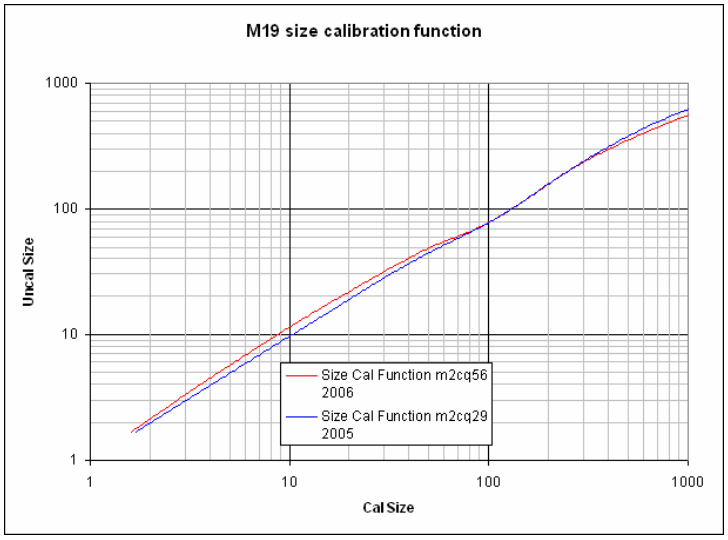
We have included the following references:

Biskos et al. (2005) on Page 25, lines 9-10.

Collings et al. (2003) on Page 25, lines 19-20.

Symonds et al. (2007) on Page 30 lines 18-20.

Page 6, line 16 (now lines 19-20): Calibration drifts (2005-2006); shows the relationship between base computer model and empirical calibration, as shown in Fig. below. Infact, the majority of the apparent difference between the two calibrations (as shown in Fig. below) is due [Cambustion, private communication] to a change in the charging model, which was improved in 2006.



As seen in the figure this small calibration drift is due to the improved charging model; however it is recommended by the manufacturer (http://www.cambustion.com/instruments/dms500/index.html) to get the DMS500 calibrated every after 12 months.

Non spherical particles: These achieve progressively higher charge levels than spherical particles as particles get larger. This manifests itself as a reduction in size and an increase in gain; however this only begins to affect particles larger than 100 nm. Since the size is determined by inertial means; it is similar to the mechanical mobility equivalent diameter, as sized by mobility spectrometers such as SMPS. The DMS instrument effectively classify by charge to drag ratio, or by electrical mobility which is the diameter of a spherical particle with the same charge:drag ratio as particle being measured. Symonds et al., 2007 can be referred for further detailed discussion on this topic.

Page 8, line 10 (now line 18): The wind directions were measured at the rooftop as this was representative of flow above the urban canopy. No measurements were made within the street canyon for wind speed/direction.

Page 9, line 3 (now line 12): deleted '*an*'

Page 10, line 5 (line 17): Changed the sentence 'The traffic volumes were manually counted in six…..to… *the traffic volumes were counted continuously throughout the sampling period in six*….'.

Yes, the overall numbers were compared with those monitored by Cambridge City Council, and were found to be similar (variation within 10%). However, we used our monitored traffic data for analysis since the traffic volumes were counted continuously throughout the sampling period.

Page 10, line 18 (now Page 11, line 10): Traffic volume is plotted in Fig. 3 as stated in the text ' … was poor (see Fig. 2 for PNCs and *Fig. 3 for traffic volume*)'. This is not plotted in Fig. 2 because the wind speed is plotted on secondary y-axis, and to avoid the repetition (as already plotted in Fig. 3).

Page 12, line 5 (now line 9): There may be effects of urban roughness (trees and other vegetation, parked vehicles and street furniture etc.) however these would be generally similar throughout the sampling period..

The measurements were made during day time (between 09:00 and 19:00 h, BST) in June 2006, and the changes in temperature were small (average 23.5^{0}C, standard deviation 2.9^{0}C), and the wind speeds were high, averaging around 5 m s^{-1} throughout the sampling period. Therefore, the effect of atmospheric stability was assumed to be negligible.

Thermal effects are mainly from the variation of solar heating of the street walls and ground during the day. It is expected that the thermal effects are greatest under low wind conditions and wind perpendicular to the street axis. These are important in a narrow region close to the windward heated wall. As the flow in the wall boundaries carries air from the street level upwards, where normally cleaner air is transported from above. Since there were always the periods of high winds (always greater than 1.5 ms^{-1}) throughout the experiments, and the solar radiation was weak during the experiments, therefore thermal effects are assumed to be negligible. The relevant Froude number for flow would be around 10, well above the value of near unity, see Kovar-Panskus et al. (2002), required for thermal effects within the street canyon to be of consequence.

Kovar-Panskus A, Moulinneuf L, Savory E, Abdelqari A, Sini J-F, Rosant J-M, Robins A, Toy N. A wind tunnel investigation of the influences of solar-induced wall-heating on the flow regime within a simulated urban street canyon. Water, Air, and Soil Pollution 2002: 2: 555-571.

Page 13, line 6 (now line 16): deleted '*from the*' from lines 16 and 17.

Page 13, line 19 (now page 14, line 10): Changed 'Hunter, 1992 to *Hunter et al., 1992'*.

Page 14, line 4-5 (lines 18-19): 'significantly smaller' already quantified in the sentence as '…always significantly smaller *(between 0.5-12.5 %)* than the concentration differences between the two upper positions *(between 10-40 %)*.'

Page 19, line 20 (now page 20, line 11): Imhof reference added on '*Page 27, lines 13-15'* in Reference list.

Page 28, line 19 (now page 30, line 3): Park reference '*deleted from reference list'*.

Supplementary Material
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