Effect of wind direction and speed on the dispersion of nucleation and accumulation mode particles in an urban street canyon

Prashant Kumar^{a,*}, Paul Fennell^b, Rex Britter^a

^aDepartment of Engineering, University of Cambridge, CB2 1PZ Cambridge, UK

^bDepartment of Chemical Engineering and Chemical Technology, Imperial College London, SW72AZ, UK

Abstract

There have been many studies concerning dispersion of gaseous pollutants from vehicles within street canyons; fewer address the dispersion of particulate matter, particularly particle number concentrations separated into the nucleation (10-30 nm or N₁₀₋₃₀) or accumulation (30-300 nm or N₃₀₋₃₀₀) modes either separately or together (N₁₀₋₃₀₀).

This study aimed to determine the effect of wind direction and speed on particle dispersion in the above size ranges. Particle number distributions (PNDs) and concentrations (PNCs) were measured in the 5-2738 nm range continuously (and in real-time) for 17 days between 7th and 23rd March 2007 in a regular (aspect ratio ~ unity) street canyon in Cambridge (UK), using a newly developed fast response differential mobility spectrometer (sampling frequency 0.5 Hz), at 1.60 m above the road level. The PNCs in each size range, during all wind directions, were better described by a proposed two regime model (traffic dependent and wind dependent mixing) than by simply assuming that the PNC was inversely proportional to the wind speed or by fitting the data with a best fit single power law. The critical cut-off wind speed ($U_{r,crit}$) for each size range of particles, distinguishing the boundary between these mixing regimes was also investigated. In the traffic dependent PNC region ($U_r \ll U_{r,crit}$), concentrations in each size range were approximately constant and independent of wind speed and direction. In the wind speed dependent PNC region ($U_r \gg U_{r,crit}$), concentrations were inversely proportional to U_r irrespective of any particle size range and wind directions. The wind speed demarcating the two regimes ($U_{r,crit}$) was $1.23 \pm 0.55 \text{ m s}^{-1}$ for N₁₀₋₃₀₀, ($1.47 \pm 0.72 \text{ m s}^{-1}$) for N₁₀₋₃₀ but smaller ($0.78 \pm 0.29 \text{ m s}^{-1}$) for N₃₀₋₃₀₀.

Key words: Particle number distribution; Nucleation and accumulation mode particles; Traffic-produced turbulence; Street canyon; Particle dispersion; Wind-produced turbulence

* Corresponding author: Hopkinson Laboratory, Department of Engineering, University of Cambridge Trumpington Street, CB2 1PZ, Cambridge, UK. Tel.: +44 1223 332681; Fax: +44 1223 765311, +44 1223 332662. E-mail addresses: pp286@cam.ac.uk (P. Kumar), rb11@eng.cam.ac.uk (R. Britter).

1. Introduction

The impacts of ambient particulate pollution on public health have been longstanding concerns for the air quality management community and regulatory authorities (Pope III, 2000; Seaton et al., 1995). Regulations controlling the emission of ambient particulate matter (PM) have been based on limits for PM₁₀ ($D_p \leq 10 \mu\text{m}$) and PM_{2.5} ($D_p \leq 2.5 \mu\text{m}$); these use particle mass concentrations, not particle number concentrations (PNC). Recent toxicological studies have suggested that the ultrafine fraction ($D_p \leq 100 \text{ nm}$), which is the main component of ambient particles by number, are more toxic than coarser particles, per unit mass (Oberdorster, 2000). Furthermore, epidemiological studies suggest correlation between exposure to ambient ultrafine particles at high number concentration, and adverse health effects (Davidson et al., 2005; Peters and Wichmann, 2001). The lack of standard methods and instrumentation for particle number measurements, and detailed understanding of the influence of ambient meteorology and traffic flows on particle dispersion have been major concern to design effective mitigation strategies for particulate pollution in urban areas.

Vehicles are the major source of ultrafine particles in urban areas (Fenger, 1999; Schauer et al., 1996). Particles between 10 and 300 nm diameter are those considered in detail here, since the majority (> 99%) of the total number of particles in this study were found to be in this range. These particles were investigated as a whole (N₁₀₋₃₀₀) and further separated into two broad size ranges, as nucleation (10 - 30 nm or N₁₀₋₃₀) and accumulation (30 - 300 nm or N₃₀₋₃₀₀) mode particles (Gourio et al., 2004; Kumar et al., 2008a; Roth et al., 2008). The nucleation mode particles are not present in primary exhaust emissions, but are thought to be due to condensation of the vapor phase present in the exhaust gases (Charron and Harrison, 2003; Kittelson et al., 2006); these particles are formed through nucleation (gas-to-particle conversion) in the atmosphere after the rapid cooling and dilution of exhaust emissions, when the saturation ratio of gaseous compounds of low volatility (e.g., sulphuric acid) reaches a

maximum (Rickeard et al., 1996; Charron and Harrison, 2003). Accumulation mode particles are formed in the combustion chamber, with associated condensed organic matter; they are composed of carbonaceous agglomerates (soot particles) and ash. They are produced mainly by diesel-engined or direct injection gasoline engined vehicles (Graskow et al., 1998).

Over the past two decades, several groups have studied the dispersion of vehicular emissions (gaseous pollutants and particulates) in urban street canyons (Boddy et al., 2005; Kastner-Klein et al., 2004; Kim and Baik, 2004; Kumar et al., 2008; Kumar et al., 2008a; Li et al., 2007; Weber et al., 2006; Wehner and Weidensohler, 2003), but the need for measurements of fine particulates (those below 1000 nm) to aid the production and evaluation of dispersion models for regulatory purposes, is acute. For micro-scale numerical modelling of street canyon air pollution, the traffic-related component of ambient pollutant concentration is generally assumed to be inversely dependent on above-roof wind speed, in particular when solar radiation is weak, stratification is neutral, and traffic-induced turbulence is ignored (Berkowicz, 2000). The direction of the wind (cross-canyon or along canyon) is also important in determining the flow and mixing processes in the street canyon and the consequent pollutant concentrations (Ketzel et al., 2002). At low wind speeds traffic-produced turbulence and thermal effects become important. Investigation of the dependence of particle number concentrations (PNCs) on wind speed and wind direction is vital for particulate dispersion models. Unfortunately, no studies could be located in the literature which enabled the comprehensive testing of dispersion models applied to particulates.

The effect of the above-roof wind speed and direction on the particles in the nucleation and accumulation modes was determined by measuring the particle number distributions (PNDs) in the 5-2738 nm size range, at 1.60 m above the road level of an 11.60 m deep (H) street canyon in Cambridge (UK), between the 7th and 23rd of March 2007 for 17 days. The measurement height (z) of 1.60 m (i.e., $z/H = 0.14$) was selected with the intention

that the mixing effects of both traffic and wind-produced turbulence in the street canyon could be observed (De Paul and Sheih, 1986; Di Sabatino et al., 2003; Solazzo et al., 2007). In this study, a recently developed instrument, the ‘fast response differential mobility spectrometer (DMS500)’ measured the PNDs in a broad range (5-2738 nm) with a high frequency (up to 10 Hz, though we used 0.5 Hz for our measurements), providing near real-time continuous measurements, unlike most other studies.

The main aims of this study were to determine the relative effects of the above-roof wind speed and wind direction on the dispersion of particles in the N_{10-300} , N_{10-30} and N_{30-300} size ranges, and to estimate the critical cut-off wind speed ($U_{r,crit}$) for these particles, which distinguishes the boundary between the traffic-dependent and the wind-dependent regimes. Ignoring the traffic dependent PNC regime may often lead to over prediction of concentrations. Therefore, a model providing information on $U_{r,crit}$, and reflecting the role of both traffic-produced (the PNCs that are independent of above-roof wind speed up to $U_{r,crit}$) and wind produced turbulence (the PNCs are inversely dependent on the wind speed above $U_{r,crit}$) was proposed and validated.

2. Methodology

2.1 Site description

Measurements were carried out in Pembroke Street (Cambridge, UK; $52^{\circ}12' N$ and $0^{\circ}10' E$), just outside the Chemical Engineering Department building. The studied section of street canyon (Fig. 1) is 167 m long, and runs approximately northeast to southwest. The Chemical Engineering Department is on the northwest (NW) side of the street and Pembroke College on the southeast (SE), with mean building heights (H) of about 11.6 m on both sides. The street canyon is nearly symmetrical, with pitched roofed (sloped parallel to the street) buildings on either side of the street. The street canyon is ~ 11.8 m wide (W_s) with one lane (6.65 m wide) travelling towards the northeast (NE). The studied section has an aspect ratio

1 (H/W_s) about unity and has a length to height (L/H) about 14, making it a long length street
2 canyon (Vardoulakis et al., 2003). The sampling was carried out 66 m from the SW end of the
3 street canyon, 0.40 m from the wall of the Chemical Engineering Department building and set
4 back 2.20 m from the kerb. Pembroke Street is close to a car park, which was closed during
5 the studied period, and the city centre. Distinct peaks in traffic occurred during morning
6 (07:00-09:00 h) and evening (18:00-20:00 h) office hours. Traffic flow at the NE end of the
7 street was regulated by traffic signals while the traffic flow was free at the southwest (SW)
8 end.

9 **2.2 Instrumentation and data acquisition**

10 A particle spectrometer (DMS500) measured the PNDs in the 5-2738 nm size range at
11 1.60 m. A sampling frequency of 0.5 Hz, rather than the maximal frequency of 10 Hz, was
12 used to improve the signal/noise ratio, and measurements were made continuously for 24
13 hours a day, for 17 days between 7 and 23 March 2007. The data from 20 March (16:00h) to
14 21 March (16:00 h) are not included in this analysis since pseudo-simultaneous measurements
15 at four different heights were made to assess the vertical variation of PNCs during this time;
16 results of these experiments are presented elsewhere (Kumar et al., 2008a). The DMS500 was
17 calibrated by the manufacturer (Cambustion Ltd.), immediately before the study, by using
18 polystyrene spheres of known diameter and also by comparing the results from sampling an
19 aerosol with those from a scanning mobility particle sizer. The calibration errors in particle
20 diameter measurements and sample flow rates were 3.4% and 2.3% respectively. A detailed
21 description of the working principle (Biskos et al., 2005) of the DMS500, and its application
22 in different scenarios and comparison with commonly deployed instruments (i.e., SMPS and
23 Electrostatic Low Pressure Impactor) during road side measurements can be found in Collings
24 et al. (2003) and Symonds et al.(2007). A cyclone, with a steel restrictor that has a 0.52 mm
25 diameter hole, was placed at the head of the sampling tube to maintain a sample flow rate of

2.5 l min⁻¹, and to reduce the pressure within the sampling tube to 0.16 bar in order to improve the time response of the instrument and to reduce particle agglomeration (Biskos et al., 2005).

An automatic, pole mounted, 3-cup vortex anemometer (Windware, UK) was used to record the above-roof (i.e., 16.60 m or $z/H = 1.43$) wind speed (hereafter called as U_r). A wireless weather station (Thermor, UK) was installed at 4.62 m (i.e., $z/H = 0.40$) and recorded ambient temperature, humidity, atmospheric pressure, wind speed and direction. The wind speed was recorded every minute during the entire sampling period by the anemometer which was set up above the roof ($z/H = 1.40$). The wind speed was also measured on an approximately half-hourly basis at the street level ($z/H = 0.40$), during working hours (0800:1900 hrs). Ambient temperature, relative humidity and pressure were also recorded with the same frequency. The half-hourly averaged reading from the Cambridge University operated AT&T weather station, which was approximately 500 m away from the sampling site (see Kumar et al. 2008 for details), were also collected and correlated with the local observations, which were found to be in reasonable (within 3%) agreement; these readings were used to determine the wind direction above the rooftop.

Traffic volumes were sampled through the measurement period by a movement sensitive CCTV camera. Manual traffic counts were also made for a few hours a day to ensure that the sampling was reliable. The traffic speed through the test site was manually measured to be about 30 ± 7 km h⁻¹. Traffic volume was consistent through different hours of the day, for example, this was 229 ± 92 , 1142 ± 71 , 705 ± 177 , 1147 ± 45 and 471 ± 120 veh h⁻¹ during 00:00-07:00, 07:00-09:00, 09:00-18:00, 18:00-20:00 and 20:00-24:00 h, respectively.

2.3 Particle Losses in sampling tube

A thermally and electrically conductive sampling tube, made of silicon rubber to which carbon has been added, 7.85 mm internal diameter and 5.17 m length (L_I), was

used to obtain the air samples. To quantify the particle losses in this tube, particle measurements were made with the same sampling frequency from a stationary diesel engine car (approximately 500 mm from the exhaust), and compared with separately taken measurements using a reference tube of much shorter length ($L_{ref} = 1.0$ m). We assumed that losses in L_{ref} would be equivalent to the losses in the first meter of the sampling tubes which were used in this, and the previously mentioned study (Kumar et al., 2008a), so that approximately the same number and distribution of particles entered the 2nd meter of the sampling tube for each of sampling tubes, which was the size and number distribution measured for L_{ref} . Next, we correlated the size-dependent penetration through the “corrected” length of sampling tube (i.e. the total length of the tube minus L_{ref}). This enabled us to correlate the losses in a sampling tube as a function of its actual length. Figure 2 shows this correlation. Comparison of the experimental results with laminar and turbulent flow regime models (Hinds, 1999) were also made; the results were better described by the turbulent flow model, even though the Reynolds number in the sample line lengths was within the laminar regime. It is clear from Figure 2 that particle losses below 10 nm seem to be highly significant, showing the maximum losses as high as ~ 90% for 5 nm particles. Therefore the data below 10 nm are not considered in the analysis below, with particle losses between 10 and 20 nm particles being corrected using the results from Fig. 2. Further details of the correction methods are given in Kumar et al. (2008a).

3. Results and Discussions

3.1 Above-roof wind speed

The dispersion of pollutants in a street canyon is closely related to the mixing mechanisms within it. Wind-produced and traffic-produced turbulence are considered to be the dominant mixing mechanisms of the particles in this study. Earlier studies have shown that when the wind blows across a regular street canyon with U_r greater than around 1.2 m s^{-1} ,

the mixing of gaseous pollutants is dominated by wind-produced turbulence, and below this wind speed the mixing of the pollutants in the lower part of the street canyon (up to $z/H = 0.20$) is dominated by the traffic-produced turbulence (De Paul and Sheih, 1986; Di Sabatino et al., 2003; Kastner-Klein et al., 2003; Solazzo et al., 2007). For about 71% of the total duration of sampling, U_r was greater than 1.2 m s^{-1} ; otherwise it was $\leq 1.2 \text{ m s}^{-1}$ (Fig. 3).

Mixing effects produced by differential heating of the walls and road within the canyon, are considered to be negligible (Kim and Baik, 2001), especially since changes in temperature were modest (average $7.4 \text{ }^{\circ}\text{C}$, standard deviation $4.6 \text{ }^{\circ}\text{C}$) over the entire measurements and the fact that thermal effects are mainly from variations in solar heating of the street walls and the ground during the day (Kovar-Panskus et al., 2002). In our experiments, solar radiation was weak (as evidenced by the low temperatures) throughout the entire sampling period.

3.2 Wind direction

The flow within a street canyon can be characterized by the roof geometry, roughness elements, street canyon geometry such as aspect ratio and street orientation and with the synoptic (above-roof) wind conditions (Kastner-Klein et al., 2004). The most important factor influencing the flow in the street canyon is wind direction. A single vortex can form in a regular (aspect ratio ~ 1) street canyon when the wind is across the canyon (i.e., wind direction to the street axis exceeds 30°) and U_r is greater than 1.5 m s^{-1} (De Paul and Sheih, 1986). However, such vortices are less evident when the wind direction is more parallel to the canyon. The flow can also be a combination of an along-street flow and a re-circulating flow (Belcher, 2005). Our study covered wind flow from most directions (see Fig. 3), though the majority of the wind flow was from the SW.

Experiments were undertaken during different wind directions. The entire data set was half hourly averaged and was divided into eight categories of wind directions. These wind

directions were northwest (NW), north (N), northeast (NE), east (E), southeast (SE), south (S), southwest (SW) and west (W), which represent the wind angles 292.5° - 337.5° , 337.5° - 22.5° , 22.5° - 67.5° , 67.5° - 112.5° , 112.5° - 157.5° , 157.5° - 202.5° , 202.5° - 247.5° and 247.5° - 292.5° respectively. As shown in Fig. 3, NW and SE represent cross-canyon flow for the leeward and windward situations respectively whilst SW and NE represent respectively along canyon flow with and against the direction of traffic, with the other directions representing the conditions between cross and along canyon flow. The frequency of winds from NW, SE, NE, SW, S and W were 16, 5, 3, 38, 23 and 16% respectively; indicating that only a small data set was available for SE and NE, and no data was obtained for E and N winds (Fig. 3).

3.3 Particle number distributions and concentrations

The PNDs can be described as consisting of different populations in different size-modes, and further quantified by the total particle number concentrations in these modes. The modes were categorized as nucleation mode (N_{10-30}), accumulation mode (N_{30-300}) and coarse mode (those between 300 and 2738 nm or $N_{300-2738}$). The average PNDs for each wind direction are shown in Fig. 4 (a-f). The PNDs were representative of a pollution originating from typical urban traffic (Jones and Harrison, 2006; Roth et al., 2008), exhibiting a strong peaks at ~ 15 nm and another peak at ~ 87 nm. The peak at ~ 15 nm was attributed to the 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 ~ 87 nm was attributed to particles formed in the combustion chamber, with associated condensed organic matter. However, the magnitude of PNDs varied with wind direction (Fig. 4 a-f). This variability is presumably due to the different building geometries seen by the wind, the traffic volume, the ambient meteorology (notably U_r and wind direction), and possibly the presence, strength and sense of rotation of any street canyon vortex. What is most striking about these

1 plots is that the magnitudes of the local maxima at 15 nm and 87 nm do not move in
2 sympathy when considering the various geometrical situations.

3 In further analysis we assume that the dependence of the PNDs on traffic volume and
4 wind speed is as commonly observed; it increases (linearly) with increasing traffic volume
5 and decreases inversely with increasing wind speed. Furthermore, if we assume that the
6 number of particles at each peak diameter (15 and 87 nm) is proportional to the total number
7 of particles in the N_{10-30} and N_{30-300} ranges, and normalise them for each wind direction by
8 dividing by the traffic volume (T) and multiplying by U_r (assuming that the number count is
9 proportional to the traffic volume and the inverse wind speed law holds), before finally
10 dividing through by the minimum value for the cross-canyon wind direction (from the SE) the
11 differences in the normalised number of particles should indicate the effect of the various
12 wind directions. Comparison of normalised PNDs in the N_{30-300} range for different wind
13 directions showed only modest (0.94 ± 0.23 times the SE) variations. However, the
14 normalised PNDs in the N_{10-30} range were larger by a factor of 5, 4, 3, 2 and 2 during NE,
15 NW, SW, S and W winds respectively than those during the SE winds. This variation is itself
16 of interest but, possibly of more interest is why such a variation exists for the N_{10-30} range but
17 not for the N_{30-300} range.

18 The reason for this could be that the PNCs in the nucleation mode (N_{10-30}) are affected
19 differently by increased dilution than the PNCs in the accumulation mode (N_{30-300}). Kittleson
20 et al. (2006) reported that nucleation mode particles are not present in the tailpipe and their
21 formation is driven by the concentration of nucleating species (mainly sulphuric acid and
22 hydrocarbons) and its degree of super-saturation; dilution conditions such as temperature,
23 residence time in the tail pipe, dilution ratio and dilution rate may change the number
24 concentrations of these particles by an order of magnitude or more. Conversely, accumulation
25 mode particles are composed of primarily of carbonaceous agglomerates and ash, and are

1 formed inside the engines of the vehicles during combustion or thereafter; these are less
2 influenced by sampling and dilution conditions (Kittelson et al., 2006). Our previous studies
3 (Kumar et al., 2008; Kumar et al., 2007a; Kumar et al., 2007b) for street canyon
4 measurements also showed that transformation processes for particles in the accumulation
5 mode were generally complete by the time particles were measured, and their total number
6 can be assumed to be conserved (i.e., their concentration only changes when the air in which
7 they are suspended in is diluted by fresh, uncontaminated, air). This is discussed further in
8 Section 3.6.

9 The PNCs were obtained in selected size ranges by integrating the areas under PND
10 curves over a given size range. The average PNCs over the entire measurements in the N_{10-30} ,
11 N_{30-300} and $N_{300-2738}$ range were about 66 ± 5 , 32.5 ± 5 and $0.5 \pm 0.03\%$ of the total ($N_{10-2738}$)
12 PNCs, respectively. Similar results were reported by Tuch et al. (1997) for European cities
13 where they found that the PNCs in the N_{10-30} range were dominant and that there was
14 negligible particle number concentration of particles above 500 nm during measurements of
15 particles between 10 and 10000 nm. These observations were later confirmed by Wehner and
16 Wiednesohler (2003) in their long term study (over 4 years) in Leipzig (Germany). As
17 expected, the PNCs in the $N_{300-2738}$ were found to be negligible in this study. Therefore the
18 overall range (N_{10-300}) and the split of this into N_{10-30} (nucleation mode) and N_{30-300}
19 (accumulation mode) only are considered in subsequent analysis.

20 **3.4 Traffic dependent and wind speed dependent particle number** 21 **concentrations**

22 There are two limiting cases for the dilution of the PNCs. *Firstly*, the traffic dependent
23 case, where dilution is dominated by traffic-produced turbulence occurring at smaller wind
24 speeds. *Secondly*, the wind dependent case, where dilution is dominated by wind-produced

turbulence occurring at higher wind speeds. For both cases, normalised number concentration of the traffic component of total concentrations can be expressed as (Ketzel et al., 2002);

$$\frac{N_{i-j} - C_{b,i-j}}{T^m E_{i-j}} = a U_r^{-n} \quad \text{Or} \quad \frac{N_{i-j} - C_{b,i-j}}{T^m} = a_w U_r^{-n} \quad (1)$$

where N_{i-j} is the PNC in any size range, $C_{b,i-j}$ is the background PNC in any size range, m and n are the exponents of T and U_r respectively, a is a constant, E_{i-j} (taken to be constant in this study) is the average particle number emission factor ($\# \text{ veh}^{-1} \text{ km}^{-1}$) in any particle size range for all vehicles in the fleet, and a_w is the product of a and E_{i-j} .

For the first case n must be zero. For the second case, n is often taken to be unity (the inverse wind speed law holds). The inverse wind speed law arises if dilution of vehicle-produced particles is assumed to be proportional to above-roof wind speed (i.e., to the ventilation rate of the canyon). As noted earlier, for this assumption to hold, it is also important that stratifications are neutral, solar radiation is weak, and traffic produced turbulence is ignored. For both cases it is assumed that $m = 1$, that is the particulate emission is assumed to be proportional to the traffic volume. Considering these assumptions, a model with two distinct regimes, reflecting the role of traffic-produced and wind-produced turbulence, was proposed by modifying Eq. (1) thus:

$$\frac{N_{i-j}}{T^m} = \left(\frac{N_{i-j}}{T^m} \right)_{crit} \quad \text{For } U_r \ll U_{r,crit} \quad (n = 0) \quad (2)$$

$$\frac{N_{i-j}}{T^m} = \left(\frac{N_{i-j}}{T^m} \right)_{crit} U_{r,crit} U_r^{-n} \quad \text{For } U_r \gg U_{r,crit} \quad (n = 1) \quad (3)$$

where $U_{r,crit}$ is the critical cut-off wind speed at which the gradient (n) of the best-fit line changes, and $(N_{i-j}/T^m)_{crit}$ is the traffic-normalised PNC in any size range below $U_{r,crit}$. Equation (2) represents the flat regions (i.e., $n = 0$) whereas Eq. (3) represents the regions where the inverse wind speed law holds (i.e., $n = 1$). Equations (2) and (3) were combined to give a continuous function spanning all experimental measurements. The fit of the function to the experimental data was optimised by varying the value of $U_{r,crit}$ to achieve the lowest sum of

squared errors between model and experiment. Results are shown in Figs 5-10. This appears to be the first time such a model has been applied to dispersion of fine particles. The values of $U_{r,crit}$ for all size ranges during different wind directions are presented in Fig. 11, which are discussed in detail in Section 3.6. It should be noted that the above formulations (Eqs. 2 and 3) do not include the background PNCs (probably small) (Kumar et al., 2008) because these were not directly measured. Therefore, this was removed from Eq. (1) and the PNCs in each size range were simply divided through by the traffic volume, to obtain a traffic-normalised PNC in each size range, and plotted logarithmically against U_r for all wind directions, as shown in Figs. 5-10.

Figs. 5-10 clearly reveal that the normalised PNCs in all size ranges are approximately independent of wind speed, up to a critical cut-off value ($U_{r,crit}$). Above $U_{r,crit}$, there is an approximately inversely proportional decrease in normalised PNC with increasing wind speed. In this latter region, the normalised PNC data above $U_{r,crit}$ was used to test whether n is really unity for all wind directions in each size range. The best-fit lines were drawn to this data (shown in Figs. 5-10), and comparisons were made between the obtained values of n and an assumed $n = 1$. The average values of n over all wind directions for particles in the N_{10-300} , N_{10-30} and N_{30-300} range were 1.00 ± 0.25 , 0.98 ± 0.36 and 0.94 ± 0.14 (see Table 2 or Figs. 5-10); these were close to the assumed value (unity), confirming an inverse wind speed law in each size range. Moreover, these observations also confirm that the proposed model (Eqs. 2 and 3), which provides simultaneous information on $U_{r,crit}$ by using two distinct regimes ($n = 0$ and 1), better fits the entire PNC data (shown in Figs. 5-10) than the other model simply fitting the entire PNC data with a best fit single power law (not shown in Figs. 5-10). The overall performance of these models are shown in Table 1 by comparing the commonly used following statistical indicators (Yadav and Sharan, 1996).

- Correlation coefficient (R) - this describes the degree of association between the predicted and the observed values; its value lies between 0 and 1, and ideal value is 1.
- Mean fractional bias (FB) - this describes the tendency of the model to overestimate (FB<0) or underestimate (FB>0) the observed values; its value lies between -2 and +2, and desired value is zero.
- The fraction of predictions within a factor of 2 (FAC2) - this describes the fraction of the data for which $0.5 \leq (\text{predicted concentration}/\text{observed concentration}) \leq 2$; ideal value 100%.

Table 1 clearly reveals that using a single power law fit rather than a proposed two regime model on the PNC data in each size range over the entire U_r range lead to significant over prediction (see difference in the values of FB) of concentrations.

After confirming from the above discussions that the normalised PNCs are inversely proportional to the U_r in the region where $U_r \gg U_{r,crit}$, and that proposed model fit the entire data set well for all wind directions, the next interesting aspect is to show the effect of wind directions on normalised PNCs in both regions.

3.5 Role of traffic and wind produced turbulence

Both traffic-produced and wind-produced turbulence influence the normalised particle number concentrations within the street canyon. More precisely it is both the turbulence and any mean flow that might be set up by the traffic and the wind that will influence the magnitude and the spatial distribution of the normalised PNC. In this paper we will not consider any thermal effects both for simplicity and because they were unlikely to be of significance over the measurement period (see Section 3.1).

Under low wind speed conditions the traffic-produced turbulence is the dominant process in the dilution of particles emitted at street level (Di Sabatino et al., 2003; Solazzo et al., 2007; Vachon et al., 2002). This is the case for the left hand side of the plots (Figs. 5-10)

where the normalised PNCs are independent of the wind speed ($n = 0$). This is the expected behaviour for $U_r \ll U_{r,crit}$ but we will interpret this behaviour to be valid up to $U_r = U_{r,crit}$. In this case we expect the same values of normalised concentrations (y-intercepts of Figs. 5-10) in each size range irrespective of wind direction. As expected, the normalised PNCs in the N_{10-300} range were similar, with a mean of 161 and a standard deviation of 68 (Table 2). Similarly, the normalised PNCs in N_{10-30} and N_{30-300} ranges were 90 ± 25 and 87 ± 24 respectively (Table 2). Interestingly, the normalised PNCs in each size range were the largest for the winds from the S, indicating a relatively smaller effect of traffic-produced turbulence or possibly a relatively larger emission rate per vehicle. We could not see any particular reason why these observations should be correlated with winds from the S. But overall our observations are generally as expected, that is the PNCs in each size range in the low wind speed regimes are independent of wind speed. Under higher wind speed conditions the wind-produced turbulence is the dominant process in the dilution of particles emitted at street level (Britter and Hanna, 2003; Kastner-Klein et al., 2004).

For the right hand side of the plots (Figs. 5-10) where $U_r > U_{r,crit}$ the normalised PNCs decreased with increased wind speed as is expected. There are two interesting aspects to address in this region. Firstly, is there any deviation in the values of n from that expected of about unity for $U_r > U_{r,crit}$ for each wind direction? Secondly, is there any effect of wind direction on the normalised PNC in each size range? These are discussed below.

- The fitting of the data with a negative unity exponent is seen to be not unreasonable. For N_{10-300} , the best fit values of n were 0.64, 0.85, 1.15, 1.10 and 1.27 for winds from the NW, SE, SW, S and W respectively. Given the scatter of the original data it is argued here that these results are consistent with a negative unity exponent that is required by dimensional arguments (Table 2). It is probably fortuitous that the average of the 5 calculated exponents (omitting that for winds from the NE where very little data was

available) was 1.00 ± 0.25 . Similarly, the average values of n over all wind directions for particles in the N_{10-30} and N_{30-300} range were 0.98 ± 0.36 and 0.94 ± 0.14 , respectively (Table 2). This is particularly interesting given the findings in Section 3.6 (Fig. 11) that the $U_{r,crit}$ is affected by the relative orientation of the canyon and the wind. However, there were exceptions for N_{10-30} during the winds from the NW and the SW where n was the smallest (0.4) and the largest (1.35), respectively. Similarly, for N_{30-300} , n was smallest (0.69) during winds from the SW. The reason for the smallest n for N_{10-30} during NW can be that the data was very sparse (Fig. 5b); no clear explanation for the remaining variation was found. However, the different flow conditions and levels of wind-produced turbulence during different wind directions, as described below, could be a possible reason.

- In the wind produced turbulence regime, taken as $U_r > U_{r,crit}$, the magnitude of the normalised PNCs due to change in wind directions is directly measured by the coefficient (a_w). The estimated values of a_w are shown in Table 2 and these clearly change significantly with wind direction. The change in a_w during different wind directions can be due to two main reasons; change in flow conditions (which will lead to a change in the level of wind produced turbulence and to the transport of particles out of the canyon) and the presence of any organised vortex structure within the canyon (which will lead to a spatial variation of concentration within the canyon). Note that a smaller a_w represents smaller concentrations and larger dilution. For the N_{10-300} , the results for the NW and SE winds suggest the presence of an organised vortex giving larger concentrations when the measuring station is on the leeward side. Winds from the SW will be along the street and in the direction of traffic flow. This would seem to be a case where there is little turbulence generation due to flow separation from the canyon walls or from the co-flow of the wind and the traffic. Hence large normalised PNCs are

observed. A similar though weaker argument could be made for winds from the S. However we are unable to explain the small normalised PNCs for winds from the W. Interestingly, when looking at the split range of particles the a_w for N_{10-30} and N_{30-300} showed, in general, the same trend as explained for N_{10-300} (Table 2), with an exception for N_{30-300} during the winds from the NW and the SE where these were in contrast to the expected results. The main reason for this seems to be the small quantity of data set available for SE.

It can be concluded from this section that when $U_r < U_{r,crit}$ the normalised PNCs are nearly similar in each size range irrespective of any wind direction. Moreover, when $U_r > U_{r,crit}$ the particles are inversely proportional to the wind speed irrespective of any particle size range and wind directions, and that effect of wind directions seems to be similar on the dispersion of particles in each size range. However, the values of $U_{r,crit}$ are different for all three size ranges, and change with changes in wind direction, which is discussed in subsequent section.

3.6 Critical cut-off wind speed

Fig. 11 shows the values of $U_{r,crit}$ for each size range during different wind directions. These are obtained from Figs. 5-10. The value of $U_{r,crit}$ was significantly influenced by the relative orientation of the canyon and the wind. The value of $U_{r,crit}$ for particles in the N_{10-300} range ranged from 0.70 to 1.98 $m s^{-1}$, spanning the often quoted value of 1.2 $m s^{-1}$ (De Paul and Sheih, 1986; Di Sabatino et al., 2003; Kastner-Klein et al., 2003; Solazzo et al., 2007; Vachon et al., 2002) for *gaseous* pollutants, with a mean and standard deviation of 1.23 $m s^{-1}$ and 0.55 $m s^{-1}$ respectively. The derived $U_{r,crit}$ was always smaller (average 0.78 $m s^{-1}$ and standard deviation 0.29 $m s^{-1}$, range 0.45 - 1.13 $m s^{-1}$) for N_{30-300} than for N_{10-30} (average 1.47 $m s^{-1}$ and standard deviation 0.72 $m s^{-1}$, range 0.70 – 2.22 $m s^{-1}$) and the latter showed larger variations for all wind directions. These observations produced two interesting questions.

Why was the $U_{r,crit}$ for each size range different for different wind directions? Why was the $U_{r,crit}$ not the same for particles in N_{10-30} and N_{30-300} ranges? These are explained as below.

- From our analysis it is apparent that the $U_{r,crit}$ as defined here, is a reflection of the magnitude of the coefficient a_w (see Eq. 1 and Section 3.5 for details, and Table 2 for values). The $U_{r,crit}$ has been defined as the intersection of the traffic-related and wind-related correlations. The first of these has been assumed independent of wind speed and direction while the second varies with the “turbulence-generating capacity” of the mean wind and a particular geometry. Thus when a_w is large, $U_{r,crit}$ should also be, must depend upon the wind direction. Of course, it is also important to decide whether this difference is operationally important or not.
- Fig. 11 shows that the $U_{r,crit}$ is always larger for N_{10-30} than for N_{30-300} for all wind directions, with larger variations for N_{10-30} than N_{30-300} . These observations indicate two possibilities. The first is that particles in the N_{10-30} range (nucleation mode) are relatively more affected than the particles in N_{30-300} (accumulation mode) range for the same level of traffic-produced turbulence during any wind direction. The other is that particles in the N_{10-30} range are relatively less affected than the particles in N_{30-300} range for same level of wind-produced turbulence during any wind direction. The most probable reason seems to be the first, since the nucleation mode particles are formed within the turbulent wake of a vehicle, so that traffic-induced turbulence plays a much greater role in their measured number than does the wind. The gas-to-particle conversion leading to the nucleation mode mainly depends on dilution ratio and amount of surface area available for volatile organics to adsorb to (Kittelson et al., 1999). The dilution ratio due to traffic-produced turbulence in the wake of a moving vehicle can be as high as 1000 in the first 1-5 second, and can increase only a further factor of ~ 10 in

up to 10 minutes (Zhang and Wexler, 2004). However, information on dilution in the near-vehicle wake could not be obtained with the sampling arrangement used.

It is important to note that the estimations of the above discussed $U_{r,crit}$ did not include the background concentration ($C_{b,i-j}$). In order to show whether the inclusion of this parameter affect the $U_{r,crit}$ an approximate estimates of the $C_{b,i-j}$ in each size range during all wind directions were made by modifying Eq. (1) to:

$$N_{i-j} = a_w T^m U_r^{-n} + C_{b,i-j} \quad (4)$$

For all wind directions, the estimated $C_{b,i-j}$ were found to be relatively much smaller (<10%) than the total PNCs in any size range. The incorporation of the estimated background concentrations into Eq. (1) did not lead to any significant changes in $U_{r,crit}$ as shown in Fig. 11, except during NE winds (though the small quantity of data from which the results are derived means that this results should be treated with care).

4. Summary and Conclusions

This paper presents the results of a study performed in a regular street canyon ($H/W \sim$ unity) in Cambridge (UK) continuously for 17 days between 7th and 23rd March 2007 at 1.60 m above the road level. Real-time continuous measurements of particle number distributions (PNDs) were made in 5-2738 nm size range using a fast-response particle spectrometer at a sampling frequency of 0.5 Hz. This study considered particles in the N_{10-300} range and split these into nucleation (N_{10-30}) and accumulation (N_{30-300}) mode particles to study the effect of above-roof wind speed and wind directions on the dispersion of these particles. The study tested the inverse wind speed law for wind-dependent dispersion, the constancy of the PNC's for the traffic-dependent dispersion and a model for distinguishing the boundary between these two processes

The average PNDs showed typical bi-modal distributions during each wind direction, with a strong nucleation mode peak at ~ 15 nm and an accumulation mode peak at ~ 87 nm.

The magnitude of the PNDs varied according to the wind direction, and showed much higher changes for the nucleation mode than for the accumulation mode. The main reasons for larger changes were attributed to the larger effect of increased dilution on particles in the N_{10-30} range than on particles in the N_{30-300} range. The average PNCs in the N_{10-30} range were the largest ($66 \pm 5\%$) fraction of the total ($N_{10-2738}$) PNCs. The PNCs in the N_{30-300} and $N_{300-2738}$ range were about 32.5 ± 5 and $0.5 \pm 0.03\%$, respectively of the total. Broadly speaking, these results were in line with the literature (Longley et al., 2003; Roth et al., 2008; Tuch et al., 1997; Wehner and Weidensohler, 2003).

When the rooftop wind speed was less than a critical value, $U_{r,crit}$, traffic-produced turbulence dominated the mixing in the lower part of the canyon and the dilution of normalised PNCs was independent of wind speed. However, when U_r was greater than $U_{r,crit}$, wind-produced turbulence dominated the mixing in the canyon and the concentration of the normalised PNCs was often found to be inversely proportional to U_r . This inverse dependence of concentrations on wind speed is required on dimensional grounds subject to some idealisations. Initially we tested this inverse ($n=1$) dependence of PNCs on U_r . The average values of n over all wind directions for particles in the N_{10-300} , N_{10-30} and N_{30-300} range were 1.00 ± 0.25 , 0.98 ± 0.36 and 0.94 ± 0.14 respectively, which were considered to be reasonably close to unity.

This two regime model was shown to statistically provide a better fit to the data than a single exponent power law model applied to the PNC data. In the wind speed dependent region, the magnitude of normalised (with respect to traffic volume) PNCs in each size range changed significantly with the change in wind directions (in fact, as did the value of $U_{r,crit}$). These changes were characterised by the coefficient (a_w) which quantified the “turbulence-generating capacity” of the mean wind and the particular geometry. In general, the trend of

1 results for all three size ranges in both traffic and wind speed dependent PNC regions were
2 almost similar for all wind directions, except the change in $U_{r,crit}$.

3 Changes in $U_{r,crit}$ with wind direction were because the normalised PNCs for the
4 traffic-produced turbulence case was approximately independent of wind direction and
5 because the normalised PNCs for the wind-produced turbulence case did depend upon wind
6 direction (all else held equal). Thus the intercept of these two cases (that is $U_{r,crit}$) must and
7 does depend upon the wind direction. Of course, it is also important to decide whether this
8 difference is operationally important or not. The value of $U_{r,crit}$ for N_{10-300} range was $1.23 \pm$
9 0.55 m s^{-1} , with a similar value ($1.47 \pm 0.72 \text{ m s}^{-1}$) for N_{10-30} but smaller ($0.78 \pm 0.29 \text{ m s}^{-1}$)
10 for N_{30-300} . Interestingly, $U_{r,crit}$ was always smaller for N_{30-300} than for N_{10-30} for all wind
11 directions. This was attributed to a possible greater effect of dilution due to traffic-produced
12 turbulence on particles in the nucleation mode than on particles in the accumulation mode
13 since the nucleation mode particles are formed within the turbulent wake of a vehicle, so that
14 traffic-induced turbulence may play a much greater role in their measured number than does
15 the wind.

16 Operational dispersion models which do not include the effects of traffic-produced
17 turbulence may often lead to over prediction of concentrations, as is also shown in Table 1.
18 While these results are preliminary, they clearly provide useful information on the dispersion
19 of particles within street canyons and on the $U_{r,crit}$ for particles in different size ranges which
20 could be useful for micro-scale numerical modelling of particles in urban street canyons. Of
21 course, our study is only for one canyon geometry, for a limited time period and with one
22 particular type of vehicle fleet. Clearly, the specific conditions within different canyons will
23 affect dispersion mechanisms, meaning that a great deal of more work is required in this area,
24 in street canyons of different geometrics and for different vehicle fleets.

5. Acknowledgements

Prashant Kumar acknowledges receipt of the Cambridge Nehru Scholarship and the Overseas Research Scholarship Award for his Ph.D. The authors thank Prof. A.N. Hayhurst and Dr. J.S. Dennis for lending the DMS500 for the study. Thanks also to Dr Jonathan Symonds from Cambustion Ltd. for lending the sampling heads and for the technical advice, and to Dr. David Langley for helping with some of the traffic measurements.

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Figure Captions

Fig. 1. Schematic diagram of Pembroke Street, showing the street dimensions, sampling point and traffic flow as described in the text.

Fig. 2. Size dependent penetration of particles in sampling tube. For comparison, results of laminar and turbulent regime models for particle penetration in sampling tube are also plotted with experimental results.

Fig. 3. Wind rose diagram for half-hourly averaged wind speed over the entire sampling duration. As classified in the text, wind directions cover different wind angles. The thick blue line represents the orientation of street canyon. Values shown against each wind directions in parenthesis are the total frequencies of winds.

Fig. 4. Half hourly averaged measured and corrected particle number distributions during winds from the (a) NW (b) SE (c) NE (d) SW (e) S and (f) W. Acronyms D_p , T , U_r , RH and T_a stands for particle diameter, traffic volume, above-roof wind speed, relative humidity and ambient temperature respectively, and numbers against them show half hourly average values and their standard deviations over the entire sampling duration. Bars show the standard deviation of the half hourly averaged PNDs.

Fig. 5. Half hourly averaged normalised particle number concentrations against wind speed during winds from NW (i.e., cross canyon winds, leeward situation) in (a) N_{10-300} , (b) N_{10-30} and (c) N_{30-300} size ranges. For both size ranges, fit results showing $n = 0$ and 1 represent the model Eqs. 2 and 3, and the U_r on x-axis at which n changes from 0 to 1 corresponds to $U_{r,crit}$. Furthermore, to test the inverse wind speed law in the wind speed dependent PNC region, other fit results are drawn from the PNC data above $U_{r,crit}$ which shows the deviations in n from assumed unity. Similar fit results are shown in Figs. 6-10.

1 **Fig. 6.** Half hourly averaged normalised particle number concentrations against wind speed
2 during winds from SE (i.e., cross canyon winds, windward situation) in (a) N_{10-300} , (b) N_{10-30}
3 and (c) N_{30-300} size ranges.

4 **Fig. 7.** Half hourly averaged normalised particle number concentrations against wind speed
5 during winds from NE (i.e., along canyon winds, and winds directed against the traffic
6 movement) in (a) N_{10-300} , (b) N_{10-30} and (c) N_{30-300} size ranges.

7 **Fig. 8.** Half hourly averaged normalised particle number concentrations against wind speed
8 during winds from SW (i.e., along canyon winds, and winds directed with the traffic
9 movement) in (a) N_{10-300} , (b) N_{10-30} and (c) N_{30-300} size ranges.

10 **Fig. 9.** Half hourly averaged normalised particle number concentrations against wind speed
11 during winds from S (i.e., between along and cross canyon winds, windward situation) in (a)
12 N_{10-300} , (b) N_{10-30} and (c) N_{30-300} size ranges.

13 **Fig. 10.** Half hourly averaged normalised particle number concentrations against wind speed
14 during winds from W (i.e., between along and cross canyon winds, leeward situation) in (a)
15 N_{10-300} , (b) N_{10-30} and (c) N_{30-300} size ranges.

16 **Fig. 11.** The $U_{r,crit}$ for particles in the N_{10-300} , N_{10-30} and N_{30-300} range (including and excluding
17 the background) during different wind directions.

1 Tables

2 **Table 1.** Overall performance of proposed model (Eqs. 2 and 3) fitted on entire PNC data
3 (shown in Figs. 5-10), and the other model (best fit single power law) fitted on entire PNC
4 data (best fit line not shown in Figs. 5-10). R is the regression coefficient, FAC2 is the
5 fraction of predictions within a factor of two and FB is the fractional bias.

Wind		Proposed model (Eqs. 2 and			Other model (best fit single		
directions		3) fitted on entire PND data			power law) fitted on entire PNC		
		data					
		N ₁₀₋₃₀₀	N ₁₀₋₃₀	N ₃₀₋₃₀₀	N ₁₀₋₃₀₀	N ₁₀₋₃₀	N ₃₀₋₃₀₀
NW	R	0.35	0.31	0.54	0.41	0.23	0.51
	FAC2	53%	61%	52%	48%	48%	40%
	FB	-0.02	-0.01	-0.04	-0.36	-0.46	-0.21
SE	R	0.48	0.52	0.42	0.44	0.49	0.38
	FAC2	77%	90%	70%	80%	87%	57%
	FB	0.01	0.01	0.01	-0.15	-0.11	-0.21
NE*	R*	0.42*	0.40*	0.42*	0.34*	0.31*	0.32*
	FAC2*	93%*	81%*	94%*	93%*	81%*	93%*
	FB*	-0.03*	-0.03*	-0.03*	-0.04*	-0.05*	-0.04*
SW	R	0.56	0.55	0.58	0.49	0.41	0.54
	FAC2	76%	74%	75%	75%	74%	75%
	FB	0.01	0.01	-0.05	-0.15	-0.18	-0.15
S	R	0.79	0.68	0.79	0.59	0.50	0.61
	FAC2	84%	80%	79%	77%	79%	67%
	FB	0.03	0.00	0.04	0.11	0.01	0.37
W	R	0.64	0.64	0.79	0.53	0.54	0.68
	FAC2	72%	73%	80%	66%	66%	78%
	FB	0.03	-0.01	-0.01	-0.19	-0.16	-0.27

6 * Based on very little available data, therefore these are not considered or estimated for analysis.

1 **Table 2.** Y-intercepts of the normalised PNCs for traffic dependent PNC case and the values
2 of n and a_w for wind dependent PNC case for particles in each size ranges; these are estimated
3 from Figs. 5-10.

Wind direc- tions	Wind Freq- ency (%)	Y-intercept of normalised PNCs for traffic dependent PNC case ($n =$ 0)			Values of n for wind dependent PNC case (obtained from PNC data above $U_{r,crit}$)			Coefficient (a_w) for wind dependent PNC case ($n = 1$)		
		N ₁₀₋₃₀₀	N ₁₀₋₃₀	N ₃₀₋₃₀₀	N ₁₀₋	N ₁₀₋₃₀	N ₃₀₋₃₀₀	N ₁₀₋₃₀₀	N ₁₀₋₃₀	N ₃₀₋₃₀₀
					300					
NW	16	162	98	71	0.64	0.40	0.98	256	217	54
SE	5	139	53	84	0.85	1.04	0.99	119	71	98
NE*	3*	281*	199*	82*	--	--	--	--	300*	127*
SW	38	119	76	57	1.15	1.35	0.69	232	166	58
S	23	277	116	158	1.10	0.92	1.03	195	115	93
W	16	107	108	67	1.27	1.19	1.03	80	75	29
Average	--	161	90	87	1.00	0.98	0.94	176	129	66
Stdev.	--	68	26	41	0.25	0.36	0.14	75	62	29

4 *Based on very little available data, therefore these are not considered or estimated in analysis.

Please cite this article as:

Kumar, P., Fennell, P., Britter, R., 2008. 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.