1 **Street versus rooftop level concentrations of**
2 **fine particles in a Cambridge street canyon**

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15 **STATEMENT**

16 The work described has not been submitted elsewhere for publication, in whole or
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3 **ABSTRACT**

4 Dispersion of particles, as evidenced by changes in their number distributions (PNDs) and
5 concentrations (PNCs), in urban street canyons, is still not well understood. This study compares
6 measurements by a fast-response particle spectrometer (DMS500) of the PNDs and the PNCs (5–
7 1000 nm, sampled at 1 Hz) at street and rooftop levels in a Cambridge (UK) street canyon, and
8 examines mixing, physical and chemical conversion processes, and the competing influences of
9 traffic volume and rooftop wind speed on the PNDs and the PNCs in various size ranges. PNCs at
10 street level were ≈ 6.5 times higher than at rooftop. Street-level PNCs followed the traffic volume
11 and decreased with increasing wind speed, showing a larger influence of wind speed on 30–300
12 nm particles than on 5–30 nm particles. Conversely, rooftop PNCs in the 5–30 nm size range
13 increased with wind speed, whereas those for particles between 30–300 nm did not vary with wind
14 speed.

15 **Keywords:** Dispersion; Fine particles; Number concentration; Particle number
16 distribution; Street canyon

1. INTRODUCTION

Current regulations address amount of the ambient particulate matter (PM) as PM_{10} ($D_p \leq 10 \mu m$) and $PM_{2.5}$ ($D_p \leq 2.5 \mu m$); these regulations use mass concentrations of particles, not particle number concentrations (QUARG 1996). Fine particles (below 1000 nm, which is within PM_{10}) are not included in the regulatory limits because they contribute very little to the particle mass. Recent toxicological studies have suggested that ultrafine particles, i.e., the fraction of fine particles below 100 nm and the main component of ambient particles by number, are more toxic than coarser particles, per unit mass (Oberdorster 2000). Furthermore, epidemiological studies suggest a correlation between exposure to ambient ultrafine particles at high number concentrations, and adverse health effects (Peters and Wichmann 2001; Davidson et al. 2005). The case for using the particle number concentration (PNC – the total number of particles, of all sizes, per unit volume, whereas PND is the distribution function of the total number of particles per unit volume against particle diameter) as a marker of potential health hazards has been made by several researchers (Pope III 2000; AQEG 2005), but progress has been hampered by a lack of standard methods and instrumentation to measure the particle number concentrations. Furthermore, a lack of a detailed understanding of the influence of ambient meteorology and traffic flows on particle dispersion has been the major problem for the design of effective mitigation strategies for particulate pollution in urban areas.

Vehicles are the main source of ultrafine particles in urban street canyons (Schauer et al. 1996). Zones of high PNCs can be commonly seen in urban street canyons, because of the limited dispersion of exhaust emissions due to the surrounding built-up environment (Van Dingenen et al. 2004). Over the past two decades several groups (Shi et al. 1999; Vardoulakis et al. 2002; Longley et al. 2003; Li et al. 2007) have studied the mass and number concentrations of particles at different heights in urban street canyons, but the effects of mixing and physical and chemical conversion of particles on particle numbers and size distributions are still a matter of discussion.

Concentrations at street level can be much higher than those in unobstructed locations with well-mixed air. The differences depend on traffic characteristics, the geometry of the street canyon, advection of emissions from

1 adjacent streets and turbulence produced by wind, atmospheric instability and
2 traffic, all making pollutant dispersion in urban streets a complex problem
3 (Bauman et al. 1982). There are several studies related to the spatial variation of
4 *gaseous* pollutants in urban street canyons. Thus Qin and Kot (1993) reported an
5 average vertical difference of approximately a factor of two for both CO and NO_x
6 between road level and the 25 m rooftop level. Zoumakis (1995) measured a drop
7 by a factor of 3 to 4 in CO concentrations at 29 m rooftop from road level.
8 According to Bauman et al. (1982) CO concentrations are higher by a factor of 4
9 at road level than at 60 m rooftop, while Vakeva et al. (1999) found a factor of 5
10 difference in the concentrations of CO, NO_x and O₃ between street level (3 m) and
11 rooftop (25 m). All the above studies show a high correlation between levels of
12 primary (gaseous) pollutants and traffic volume, as well as strong concentration
13 gradients in a street canyon.

14 Conversely, there is very little (Bauman et al. 1982; Vakeva et al. 1999)
15 information on similar comparisons for fine particles. Vakeva et al. (1999) found
16 the number concentration of particles to be 5 times higher at street (1.5 m) level
17 than that at rooftop (25 m); also Bauman et al. (1982) reported a factor of 2 to 4
18 difference for fine particles between road level and 60 m rooftop. However, no
19 information on particle number distributions (PND) was presented in the above
20 two studies.

21 In this work PNDs were continuously measured in 5–1000 nm size range
22 at street and subsequently at rooftop level, in Cambridge (UK). This study is
23 novel in two respects; firstly, we used a recently developed instrument, the ‘fast
24 response differential mobility spectrometer DMS500’, to measure the PNDs,
25 providing near real-time continuous measurements, unlike other studies, which
26 used, for example, a scanning mobility particle sizer (SMPS), a electrical low-
27 pressure impactor (ELPI), an ultra particle condensation particle counter or optical
28 particle counters, either alone or in combination. Secondly, we measured a broad
29 range (5–1000 nm) of particles at a high sampling frequency (10 Hz). However, to
30 ensure maximal data quality, our experiments recorded the ten point rolling
31 average of the PNDs with a 1 Hz sampling frequency. The main aims of these
32 measurements were to determine the effects of mixing and physical and chemical
33 conversion processes, as well as the competing influences of rooftop wind speed

and traffic volume on both the PNDs and particle number concentrations (PNCs) in various size ranges, along with the production of new particles at both levels.

2. EXPERIMENTAL

2.1 Description of Site

Measurements were carried out on a small section of the street, Fen Causeway, adjacent to the university's Department of Engineering in Cambridge (UK). The section of street canyon studied is 200 m long, and runs approximately east to west, as shown in Fig. 1. The road carries two-way traffic, and is 10 m wide, with one lane in each direction. The heights and frontages of the buildings are not perfectly symmetrical, but they are continuous and broadly follow the east-west line of the road. The roofs of the school buildings along the south side are sloped and parallel to the road, whereas the geometries of those on the north side are more complex. The buildings vary in height on both sides of the road: on the south side from 18 to 22 m and on the north side from 15 to 22 m. The average height (H) and the width (W) (distance between buildings on opposite sides of the road) of the street canyon at the measurement location are approximately 18 m and 20 m, respectively, giving an aspect ratio (H/W) near unity (Vardoulakis et al. 2003). Traffic is regulated by signals at both ends of the selected section through pedestrian crossings. The street level 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 along the length of the section; altogether three different heights from 0.2 to 2.6 m were sampled. Rooftop sampling was also performed on the north side of the road above the roof (20 m) of Department building and set back 3.35 m from the kerb. During the study winds were from the southwest, with few sources of pollution upwind in the countryside. Therefore, it was considered unlikely that there was a change in rooftop PNCs due to a change in background PNCs on the two different days with similar weather.

2.2 Instrumentation

A fast response differential mobility spectrometer (DMS500) was used to measure PNDs. A detailed description of the DMS500, its working principles, applications in different areas and a comparison with other instruments (i.e.,

1 SMPS and ELPI) for road-side measurements can be seen elsewhere (Collings et
2 al. 2003; Biskos et al. 2005; Symonds et al. 2007). The DMS500 was calibrated
3 by using polystyrene spheres of known diameter (traceable) and also by
4 comparing the results from sampling an aerosol with those from a SMPS. The
5 calibration errors in the particle measurements and sample flow rate were about
6 4.3% and 2%, respectively.

7 Samples of air from both street and rooftop levels were obtained via a
8 thermally and electrically conductive sampling tube (5.85 mm i.d.; length 5 m)
9 with a residence time of ≈ 0.3 s. A cyclone, with a steel restrictor (with a hole of
10 diameter 1.0 mm), was placed at the head of the sampling tube to maintain a flow
11 rate of $8 \times 10^{-3} \text{ m}^3 \text{ min}^{-1}$ for the sample, and to reduce the pressure inside the
12 sampling tube to 0.25×10^5 Pa to improve the time response of the instrument and
13 to reduce agglomeration of particles (Biskos et al. 2005).

14 Theoretical estimates of particle losses in the sampling tube show that
15 diffusion and inertial impaction were the most important mechanisms for particles
16 below 15 nm (Hinds 1999), since the penetration (the fraction of the entering
17 particles leaving the tube) was 92–97% for particles between 5 nm and 10 nm,
18 97–99% for particles between 10 nm and 15 nm and 99–99.99% for particles
19 between 15 nm and 1000 nm. These penetrations indicate losses sufficiently small
20 to be ignored.

21 **2.3 Measurements**

22 The DMS500 can record the PNDs at a frequency of 10 Hz. However, our
23 experiments recorded the average of 10 measurements with a sampling frequency
24 of 1 Hz at both street and rooftop levels to improve the signal/noise ratio. The
25 measurements at street level were taken between 8 and 21 June 2006 (from 09:00
26 to 19:00 h) for 7 weekdays at three different heights (i.e., 0.20 m, 1.0 m, and 2.60
27 m above road level; hereafter called A, B and C, respectively). Further
28 measurements were taken on another weekday (22 June 2006) at rooftop level
29 (i.e., about 20 m above road level; hereafter called R). It should be noted that only
30 the street level measurements for 16 June have been compared with the rooftop
31 measurements, since the meteorology (wind speed, wind direction, temperature
32 and relative humidity) and the traffic volumes were similar on both days (see

1 Table 1). Other results from this study are reported elsewhere (Kumar et al.
2 2008a).

3 To acquire a representative data set at each street level (i.e., at points A, B
4 and C), the samples were taken for 20 min in an hour at each sampling point, on
5 two different occasions (i.e., 2 samples per hour, 10 min per sample) by manually
6 re-positioning the height of the probe every 10 min. Rooftop measurements were
7 taken continuously between 09:00 and 19:00 h on 22 June 2006. Simultaneous
8 measurements at both levels (i.e., each sampling height) could not be performed,
9 due to the availability of only one instrument. With regards to street-level
10 sampling, since this was done in 60 separate time periods in the day, whilst the
11 PNC changed in an essentially random manner with respect to time, sufficient
12 measurements were made to draw tentative conclusions about the average PNCs
13 at street level.

14 Meteorological data (rooftop wind speed, hereafter called wind speed U_r ,
15 wind direction, temperature, and relative humidity) were obtained from a weather
16 station, operated by the University's AT&T laboratories on the roof of the
17 Department of Engineering building. The facility was about 40 m above the road
18 level at a point some 100 m from the sampling site. This location is above the
19 average height for Cambridge buildings and is not overlooked.

20 Traffic volumes were counted manually throughout each period of particle
21 measurements. The average traffic volumes during 16 June and 22 June were
22 1623 ± 165 vehicle h^{-1} and 1629 ± 125 vehicle h^{-1} , respectively, of which cars and
23 vans (gasoline), cars and vans (diesel), buses, light duty vehicles (LDV), heavy
24 duty vehicles (HDV) and motorcycles were about 74.85%, 19.15%, 1%, 3%, 1%
25 and 1%, respectively. The traffic speed through the test site was manually
26 measured to be about 30 km h^{-1} . Kumar et al. (2008a) give further details of the site,
27 the instrumentation, data acquisition and other street level results of this study.

28 **3. RESULTS AND DISCUSSION**

29 **3.1 Particle number distributions at street level and rooftop** 30 **level**

31 The PNDs, hourly averaged over the entire measurement period, at street
32 and rooftop levels are shown in Fig. 2a. The shape of street level PNDs at all three

1 heights were found to be similar. The maxima of the PND profiles at three heights
2 were close to each other, showing a small, but consistent decrease with height.
3 However, significant differences can be seen between the PNDs at street and
4 rooftop levels. These observations depend on the traffic volume, ambient
5 meteorology (notably rooftop wind speed and wind direction), strength and sense
6 of rotation of any street canyon vortex. The PNDs at both street and rooftop levels
7 showed a typical bi-modal distribution, as seen in Fig. 2a. At street level the
8 PNDs showed peaks at about 30 nm and 100 nm, whereas the rooftop PNDs
9 showed peaks at about 20 nm and 100 nm. The peaks at smaller diameters (i.e., 20
10 nm and 30 nm) can be attributed to particles formed by nucleation and
11 condensation during the rapid cooling and dilution of semi-volatile species from
12 the exhaust gases with ambient air. This contrasts with the peak at about 100 nm,
13 corresponding to larger particles formed in a vehicle's combustion chamber by
14 condensation of organic matter (Kittelson et al. 2004). Apart from exhaust
15 emissions, there may be a contribution of nucleating species (typically sulphates
16 and nitrates) from the upwind countryside at all levels. However, measurements at
17 minimum traffic levels and different wind speeds indicated that such particles
18 from the regional background made a minimal contribution.

19 If we treat the rooftop PNDs as background and subtract them from street
20 level PNDs, the resulting PNDs, shown in Fig. 2b, should reflect local traffic.
21 These resulting PNDs show an additional peak at about 10 nm, which probably
22 reflected fresh emissions from traffic and/or tiny particles formed in the
23 atmosphere from various hydrocarbons precursors, because the background PNCs
24 has been shown to be relatively small and $\approx 15\%$ of the total PNC (Kumar et al.
25 2008a). Interestingly, the street level PNDs are not similar to those obtained in
26 another recent study in a different Cambridge (UK) street canyon (Kumar et al.
27 2008b), where a significantly larger peak was observed at ≈ 13 nm and a smaller
28 peak at ≈ 87 nm, in contrast to the large peak in Fig. 2b at ≈ 30 nm and a smaller
29 peak at ≈ 100 nm. This change in PNDs can be attributed to a type and volume of
30 traffic, driving conditions (more congested in this study) and the effect of the
31 surrounding built-up environment on the dispersion of exhaust emissions in the
32 street canyons. Apart from the difference in canyon geometry, the proportions of
33 gasoline and diesel engined vehicles were different in this study, being about 76%
34 and 24%, respectively, compared with 68% and 32%, respectively, in the

1 previous study on several (8–10% of total traffic volume) bus routes (Kumar et al.
2 2008b). The present study comprised diesel cars and vans (19%), buses (1%),
3 light duty commercial vehicles (3%) and heavy duty commercial vehicles (1%)
4 (Kumar et al. 2008a). The relatively high proportion of buses in the previous
5 study would have had a disproportionate effect on the modal particle diameter,
6 because of the large number of particles emitted by these vehicles.

7 Since the three sampling points (A, B and C) recorded similar PNCs and
8 PNDs (differences in PNC ranged between 13% and 21%) for the entire period of
9 measurements, for simplicity the PNDs at A, B and C were averaged and used as
10 “street level” PNDs in the analysis below.

11 **3.2 Effect of wind speed on PNDs at street and rooftop** 12 **levels**

13 The half-hourly averaged wind speed and wind direction are used below to show
14 their effect on PNDs at street and rooftop levels. The wind speeds were
15 categorized in various ranges and the average PND for all measurements in each
16 category of wind speed is plotted in Fig. 3. The winds were always above 2.5 m s^{-1}
17 during the study. The frequencies of winds in the designated categories of wind
18 speed $>2.5 \leq 3.5$, $>3.5 \leq 4.5$, $>4.5 \leq 5.5$, $>5.5 \leq 6.5$, $>6.5 \leq 7.5$, $>7.5 \leq 8.5$ and $>8.5 \leq 9.5$
19 m s^{-1} were, respectively, 18, 18, 12, 35, 0, 0 and 17% during measurements at
20 street level, but 22, 20, 13, 12, 27, 6 and 0%, respectively, during measurements
21 at rooftop level. *Street* level PNDs are plotted in Fig. 3a and seen to be acutely
22 dependent on the wind speed; the largest PNDs were found at the time of the
23 smallest wind speed and vice-versa. However, this effect was not consistent
24 across the entire range of particle sizes; for example, the variations in PNDs for
25 particles between 5 and 30 nm were significantly smaller than for particles
26 between 30 and 300 nm. This shows a greater influence of wind speed on the
27 larger particles with diameters of 30–300 nm, than on smaller particles (5–30 nm).
28 This contrasts with *rooftop* PNDs, which are presented in Fig. 3b. They were
29 much more affected by changes in wind speed for smaller particles of 5 to 30 nm
30 than those between 30 nm and 300 nm, whose abundances were independent of
31 wind speed. The larger variations in the PNDs between 30 nm and 300 nm at
32 street level seems mainly due to transport of particles out of the canyon (exchange
33 between the street canyon and the wind above, or *ventilation*), presumably due to

1 dilution by turbulent mixing produced by the wind. Moreover, the influence of
 2 rooftop PND_s on street level PND_s seems to be insignificant, since changes in
 3 rooftop PND_s in the 30–300 nm size range are nearly negligible, despite the
 4 change in wind speed (Fig. 3b), throughout the measurements. Therefore their
 5 transport into the canyon should not affect the street level PND_s. During the entire
 6 measurement period, conditions were found to be favourable for transporting
 7 particles out of the street canyon through a street canyon vortex (see Fig. 1), since
 8 the wind direction was nearly across the canyon (i.e., SW or about 22.5°–67.5° to
 9 the street) and the wind speeds were always in excess of 1.5 m s⁻¹ throughout the
 10 measurements. The work of De Paul and Sheih (1986) showed that for wind
 11 speeds above 1.5 m s⁻¹ at an angle of more than 30° to the street's axis (Oke
 12 1988), a vortex is likely to form, as shown in Fig. 1. Furthermore, dilution of
 13 particles at street level by clean air from above the canyon was most likely to be
 14 dominated by mixing due to wind-produced turbulence, which dominates any
 15 turbulence produced by traffic, when the wind blows across a regular street
 16 canyon with a speed greater than 1.2 m s⁻¹ (Kastner-Klein et al. 2003; Solazzo et
 17 al. 2007).

18 **3.3 Particle number concentrations in various size ranges** 19 **at street and rooftop levels**

20 The PND_s can be described as consisting of distinct populations in
 21 different size-modes and further quantified by their total particle number
 22 concentrations (PNC_s) in these size ranges. Consequently PNC_s were obtained in
 23 various size ranges by integrating the areas under PND curves over a given size
 24 range; these size ranges were 5–10 nm, 10–30 nm, 30–300 nm and 300–1000 nm
 25 and the number of particles within each range is referred to as N_{5–10}, N_{10–30}, N_{30–}
 26 ₃₀₀ and N_{300–1000}, respectively. Particles within the first two size ranges (i.e., N_{5–30})
 27 can be referred to as the nucleation mode, whereas those in N_{30–300} were referred
 28 to as the accumulation mode.

29 The proportion of PNC_s in each size range at both street and rooftop levels
 30 are shown in, respectively, in Fig. 4 (a) and (b). At both levels, particles in the N_{5–}
 31 ₃₀₀ size range accounted for most (≈ 99.9%) of the PNC_s, showing negligible
 32 quantities of particles in the larger (N_{300–1000}) size range. These results in Fig. 4
 33 are in accordance with Tuch et al. (1997), who reported that the contribution of

1 particles larger than 500 nm to the overall PNC was negligible in the European
2 urban environment. Since the PNC in the $N_{300-1000}$ size range at both street and
3 roof levels has been shown here to be almost negligible, this size range is not
4 considered separately in the subsequent analysis. Interestingly, particles in the
5 N_{30-300} size range dominated ($\approx 68\%$ of N_{5-1000}) the PNCs at street level, whereas
6 particles in the N_{5-30} size range dominated ($\approx 66\%$ of N_{5-1000}) the PNCs observed
7 at rooftop level. Our results for street level measurements also agree with those
8 measured by Longley et al. (2003) in a street canyon in Manchester, UK, where
9 particles in the N_{30-300} size range dominated the range 4.6 to 10000 nm.

10 Figure 5 shows clear differences in the PNCs in each size range between
11 street and rooftop levels. A ratio, averaged over the entire sampling period, of
12 about 6.5 was observed between PNCs at street and rooftop levels in the N_{5-1000}
13 range. This ratio changed significantly when the comparison was made for various
14 size ranges; it was respectively, 2.4, 3.2, 12.8 and 6.6 for particles in the N_{5-10} ,
15 N_{10-30} , N_{30-300} and $N_{300-1000}$ size ranges. The highest ratio (≈ 13) was observed for
16 the accumulation mode (N_{30-300}) particles. This was expected because of the larger
17 emissions (including particles formed during the cooling of exhaust gases) of
18 primary particles by the traffic at street level, and also the greater dilution by
19 stronger winds at rooftop level. Furthermore, this ratio N_{30-300} at street and rooftop
20 levels is plotted in Fig. 6, where it is seen to decrease with increasing wind speed.
21 This supports the statement above that the particles in this size range are largely
22 influenced by the wind speed. Interestingly, variations in this ratio were largely
23 dominated by the changes in street level PNCs (N_{30-300}) due to ventilation (Fig.
24 3a), since the PNCs in the N_{30-300} size range at rooftop level were nearly
25 unchanged, despite the change in wind speed, throughout the measurements (Fig.
26 3b).

27 Our previous analysis (Kumar et al 2007; Kumar et al. 2008a–c) showed
28 that the particles in the accumulation mode were nearly unaffected by the
29 transformations undergone by particles, so that their concentration only changes
30 when the air which they are suspended in is diluted by fresh, uncontaminated, air.
31 Assuming that particle numbers in the accumulation mode are, in fact, conserved,
32 the ratio ($N_{5-30}:N_{30-300}$) at any level should indicate the rate of production of new
33 (nucleation mode) particles; this ratio, averaged over the entire sampling period,
34 was 0.52 at street level, compared with 2.49 at rooftop level. Interestingly, this

1 ratio ($N_{5-30}:N_{30-300}$) was reasonably constant at street level (0.52 ± 0.21)
2 throughout the measurements, despite changes in wind speed (Fig. 6). However,
3 this ratio showed much higher values and variations (2.49 ± 1.01) at rooftop level
4 (Fig. 6), indicating the possibility of a much higher production of nucleation mode
5 particles at this level than at street level.

6 Vakeva et al. (1999) reported two important factors which can favour a
7 much greater production of particles at rooftop level than at street level: firstly,
8 the higher concentration of condensable gases and secondly, the smaller
9 concentrations of pre-existing particles. These arguments were later supported by
10 Kulmala et al. (2000), who concluded that small pre-existing aerosol
11 concentrations favour both the production of new particles and their growth to
12 measurable sizes in the atmosphere. Subsequently, Kerminen et al. (2001) found
13 that high concentrations of pre-existing particles both promote the condensation
14 of the semi-volatile vapours and disfavour both the growth of fresh nuclei and
15 their survival from high coagulation scavenging.

16 Our results seem to be in accordance with the above two conditions
17 favouring much higher particle production at rooftop level than at street level.
18 Firstly, smaller concentrations of pre-existing particles at rooftop level can be
19 seen from the ratio (N_{30-300} at street level: N_{30-300} at rooftop level) of about 12.8.
20 Secondly, the possibly higher concentration of condensable gases at rooftop level
21 could be attributed in urban areas to atmospheric chemistry (including
22 photochemistry) producing sufficient amounts of condensable gases for gas-to-
23 particle conversion. The evidence for the above arguments is shown in Fig. 6 by
24 the ratio ($N_{5-30}:N_{30-300}$), which indicates that the relative rate of production of new
25 particles at *rooftop* level increases with the wind speed. Also, the ratio (N_{30-300} at
26 street level: N_{30-300} at rooftop level), which indicates the relative number of pre-
27 existing particles at each level, decreases with increased wind speed. Furthermore,
28 increasing the wind speed generates a shift of the particle number distributions, as
29 measured at the rooftop level, to finer diameters; this is shown in Fig. 7, which
30 plots the peak diameter in a number distribution against the wind speed. Also, the
31 increase of relative particle number concentrations in Fig. 6 indicates decreased
32 agglomeration and coalescence of particles at rooftop level. Similar evidence for
33 the formation of new particles and their growth in urban areas has been reported

in many experimental (Makela et al. 1998; Vakeva et al. 1999; Charron and Harrison 2003; Kulmala et al. 2004, Curtius 2006) studies.

Conversely, at street level the number and surface area of pre-existing particles emitted from vehicles can be so large that condensation of gases on to them outweighs any new particles being formed. This argument is supported by the ratios ($N_{5-30}:N_{30-300}$) in Fig. 6; they indicate the relative rate of production of new particles. Also, the peak diameters in the particle number distributions are seen in Fig. 7 to be nearly constant at street level, despite the change in wind speed, throughout the measurements.

A nucleation event was also observed during the midday hours (between 11:00 h and 13:00 h), when the weather was “bright sunny”. The ratios ($N_{5-30}:N_{30-300}$) during this time increased from an overall average of 2.49 to 3.59. This increase might well be due to photochemically induced nucleation, in addition to new particles being produced through gas-to-particle conversion. The relative contributions of photochemically induced nucleation and nucleation through gas-to-particle conversion could not be deduced because of the limited availability of measurements. However, Wehner and Weidensohler (2003) and Vakeva et al. (1999) have reported strong evidence for an association between the formation of new particles and nucleation, photochemically induced by solar radiation.

3.4 Correlation of PNCs with traffic volume and wind speed

Traffic volume and rooftop wind speed are generally the principal variables influencing PNCs at both street and rooftop levels of a canyon. In order to show the effect of rooftop wind speed on PNC, the PNCs in the N_{5-30} and N_{30-300} size ranges at both levels were correlated with wind speed using:

$$N_{i-j} = a U_r^{-n} \quad (1)$$

where N_{i-j} is the PNC in any given size range, a is constant and n is the exponent of wind speed, U_r . Table 2 shows the results. It is clear that the values of n are constant and positive at street level, but negative at rooftop level. This indicates that PNCs decrease with increased wind speed at street level, but the opposite trend holds at rooftop level. The values of n for each size range at street level were close to unity, as shown in Fig. 8a. This confirms the idealised inverse ($n \approx 1$) wind speed law (Ketzel et al. 2002). However, the values of R^2 were larger for N_{30-300} than for N_{5-30} : 0.48 compared to 0.28. By contrast, rooftop level PNCs in

1 the N_{5-30} size range correlated better with wind speed than those for PNCs in the
 2 N_{30-300} size range: R^2 was 0.80 compared with 0.58. Moreover, the negative values
 3 of n for both size ranges at rooftop level reflect that PNCs increase with wind
 4 speed, as seen in Fig. 8c. Also, there were much higher variations in the values of
 5 n at rooftop level than at street level. One of the highest values of R^2 and the value
 6 of n with largest magnitude is $n = -2.3$, indicating a greater concentration of
 7 particles with higher wind speed for N_{5-30} . This again suggests more formation of
 8 particles at rooftop level. Harrison et al. (2000) and Charron and Harrison (2003)
 9 also reported that “cleaner atmospheres” created by stronger winds favour the
 10 occurrence of large numbers of smaller particles. The dependence of the PNCs at
 11 both levels on wind speed is clearly of prime importance, with traffic volume as
 12 the next most important factor.

13 The correlations between the PNCs in each size range at both levels and
 14 the volume of traffic at both levels are shown in Table 3, for which it was
 15 assumed that N_{i-j} is linear in the traffic volume. As expected, these correlations
 16 were found to be better at street level than at rooftop level. Also worthy of note, is
 17 that the values of R^2 at street level are for correlations showing linearly *increasing*
 18 PNCs in each size range with increasing traffic volume, as shown in Fig 8b. On
 19 the other hand, at rooftop level, the correlations surprisingly show linearly
 20 *decreasing* PNCs in each size range with increasing traffic volume, as illustrated
 21 in Fig. 8d.

22 Finally, it is important to investigate the combined effect of both wind
 23 speed and traffic volume on the PNCs for each size range. In order to remove the
 24 prime dependence of the PNCs on traffic volume, each PNC was divided by the
 25 traffic volume and this was used as a primary variable and plotted against wind
 26 speed. Such a correlation can be represented by changing Eq. (1) to:

$$27 \quad N_{i-j}/T^m = aU_r^{-n} \quad (2)$$

28 where m is the exponent (assumed to be unity) of traffic volume (T). The results
 29 of correlating the normalised PNCs in each size range with Eq. (2) at *street* level
 30 are given in Table 4. The best fit values of n are similar to those presented in
 31 Table 2 (PNCs vs U_r) and Table 3 (PNCs vs T); this confirms the individual
 32 relations of the PNCs with wind speed (Table 2) and traffic volume (Table 3),
 33 showing an equal influence of wind speed and traffic volume on the PNCs.
 34 Furthermore, correlations for the normalised PNCs in each size range with Eq. (2)

1 at *rooftop* level (Table 4) were fairly similar to those presented in Table 2 (PNCs
2 vs U_r), but much better than those shown in Table 3 (PNCs vs T), showing a much
3 larger influence on the PNCs of wind speed than traffic volume.

4 **4. SUMMARY AND CONCLUSIONS**

5 Particle number distributions were measured at both street and rooftop
6 levels in a Cambridge (UK) street canyon. Measurements were taken at three
7 different heights (i.e., 0.20 m, 1.0 m and 2.60 m) close to the road, whereas at
8 rooftop level measurements were taken ≈ 20 m above the road. The PNDs showed
9 a typical bi-modal distribution at both levels, peaking at about 30 nm and 100 nm
10 at street level, and about 20 nm and 100 nm at rooftop level. Closer inspection of
11 the measurements indicated that street level PNDs depended more on wind speed
12 for particles in N_{30-300} size range than particles in N_{5-30} size range. Conversely,
13 rooftop PNDs depended more on wind speed for the particles in N_{5-30} size range
14 than did particles in N_{30-300} size range, whose PND was hardly altered by a
15 change in wind speed.

16 At both levels, the PNCs in N_{5-300} accounted for more than 99% of the
17 total (N_{5-1000}) PNCs. Street level PNCs were dominated by the particles in the N_{30-}
18 300 size range, whereas rooftop level PNCs were dominated by particles in the N_{5-}
19 30 size range. One interesting result of this study was the ratios of PNCs at rooftop
20 and street levels. A ratio of about 6.5 was observed between street and rooftop
21 level PNCs in the N_{5-1000} size range. However, this ratio changed significantly for
22 other size ranges, showing the highest ratio for particles in N_{30-300} size range. This
23 was attributed to the larger emissions of primary particles from traffic at the street
24 level and also more intense dilution by stronger winds at rooftop level. The
25 assumption was checked that particles in the N_{30-300} size range (accumulation
26 mode) were largely unaffected by the transformation processes, i.e., changes in
27 concentration were only due to dilution of traffic-produced particles by “clean”
28 air from above so that their number could be assumed to be conserved. In this case
29 the ratios ($N_{5-30}:N_{30-300}$) at any level should indicate the rate of production of new
30 particles. This ratio was about 5 times larger at rooftop level than at street level,
31 indicating a much larger relative production of nucleation mode particles at
32 rooftop level than at street level. This is in conjunction with less efficient
33 scavenging mechanisms at rooftop level, due to the presence of fewer

1 accumulation mode particles (Kerminen et al. 2001). The production of nucleation
2 mode particles (and also a nucleation event) at rooftop level were attributed to
3 both the production of new particles through gas-to-particle conversion and
4 photochemically induced nucleation, since conditions were found to be favourable
5 for both processes. However the relative contributions of both these two
6 nucleation processes could not be identified due to a lack of measurements.

7 The PNCs in each size range at street level were found to be inversely
8 correlated with the wind speed and linearly correlated with the traffic volume.
9 Also, the PNCs in the N_{30-300} size range at street level were found to be largely
10 influenced by both the wind speed and the traffic volume. The correlation was
11 weaker for PNCs in the N_{5-30} size range. These results support our earlier findings
12 (Kumar et al. 2007; Kumar et al. 2008a–c). On the other hand, the PNCs in each
13 size range at rooftop level showed a positive correlation with wind speed and a
14 surprising negative correlation with the traffic volume. Moreover, the PNCs in the
15 N_{5-30} size range at rooftop level showed a much larger influence of wind speed
16 than did the PNCs in the N_{30-300} range.

17 Since these results are based on short-time measurements, a study with
18 simultaneous measurements at both levels, over a much longer duration would be
19 useful.

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FIGURE CAPTIONS

Fig. 1.

Schematic diagram of sampling site, showing the traffic flow, wind direction and a likely vortex, as described in the text (figures not to scale).

Fig. 2.

(a) Hourly averaged PNDs at both street and rooftop levels, and (b) resulting PNDs after subtracting of roof top level PNDs from street level PNDs, at various heights.

Fig. 3.

Effect of wind speed on particle number distributions at (a) street level (averaged over locations A, B and C), and (b) rooftop level. Both figures show averages of half-hourly wind speeds and PND data for various ranges of wind speed.

Fig. 4.

Proportions of hourly averaged PNCs in various size ranges at (a) street, and (b) rooftop level.

Fig. 5.

Comparison of hourly averaged PNCs in various size ranges at both street and rooftop levels. The error bars show the standard deviation of hourly averaged values of the PNCs.

Fig. 6.

Ratios of the PNCs in different size ranges (i.e., N_{5-30} and N_{30-300}) at street and rooftop levels plotted against the hourly averaged wind speed.

Fig. 7.

The peak diameters of hourly averaged particle number distributions plotted against the wind speed at both street and rooftop levels.

Fig. 8.

Plots of particle number concentrations against (a) wind speed and (b) traffic volume at *street* level, and against (c) wind speed and (d) traffic volume at *rooftop* level.

1 TABLES

2 Table 1

3 Comparison of meteorological parameters and traffic volume at both street and
 4 rooftop levels during the entire measurements. The values shown in the table are
 5 the hourly average (Av) over the entire measurements and the standard deviations
 6 (SD) of hourly averaged values.

Date	Wind speed (m s ⁻¹)		Relative Humidity (%)		Temperature (°C)		Traffic Volume (veh h ⁻¹)		Wind Direction
	Av	SD	Av	SD	Av	SD	Av	SD	
16 June	6	1.8	39	2.6	25	0.6	1623	165	Southwest
22 June	6	1.6	44	5.9	20	0.9	1629	125	Southwest

1 **Table 2**
2 Results of correlating PNCs in various size ranges with the wind speed at both
3 street and rooftop levels.

Level	N ₅₋₃₀		N ₃₀₋₃₀₀		N ₅₋₁₀₀₀	
	<i>n</i>	R ²	<i>n</i>	R ²	<i>n</i>	R ²
Street	0.94	0.28	0.95	0.48	0.97	0.20
Rooftop	-2.3	0.80	-0.5	0.58	-1.59	0.81

- 1 **Table 3**
- 2 Correlations of PNCs in various size ranges with traffic volume at both street and
- 3 rooftop levels.

Level	R^2		
	N_{5-30}	N_{30-30}	N_{5-1000}
Street	0.27	0.49	0.57
Rooftop	0.28	0.20	0.28

1 **Table 4**
2 Correlations between the normalised PNCs in various size ranges and the wind
3 speed at both street and rooftop levels.

Level	N_{5-30}/T		N_{30-300}/T		N_{5-1000}/T	
	n	R^2	n	R^2	n	R^2
Street	0.76	0.25	0.76	0.49	0.78	0.20
Rooftop	-2.4	0.78	-0.75	0.55	-1.84	0.77

4