

Pseudo-simultaneous measurements for the vertical variation of coarse, fine and ultrafine particles in an urban street canyon

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Abstract

The vertical variation of particle number distributions (PNDs) and concentrations in a street canyon is the result of the competing influences of meteorology, traffic and transformation processes overall and for various particle size ranges. A recently developed instrument, the ‘fast-response differential mobility spectrometer DMS500’, measured PNDs in the 5–2738 nm range, pseudo-simultaneously, at four different heights ($z/H = 0.09, 0.19, 0.40$ and 0.64) on the leeward side of an 11.6-m-deep street canyon which had a height-to-width ratio of near unity. Measurements were made in Cambridge, UK, between 20 and 21 March 2007.

The PNDs were bimodal with the same shape at each height, and with similar values of both the peak and geometric mean particle diameters in each mode. This suggested that transformation processes were not important. Coagulation and condensation time scales were comparable and large, and these processes should have had a negligible effect on the PNDs.

The particle number concentrations (PNCs) changed significantly with height from a maximum at $z/H = 0.19$ and decreasing towards both the lowest ($z/H = 0.09$) and highest ($z/H = 0.64$) sampling points. The decrease in PNCs with height in the upper part of the canyon was attributed to the removal of particles as a result of mass exchange between street canyon and the wind above while the reduction in the PNC towards street level was thought to be due to dilution and dry deposition.

Over 99% of the PNCs were found in 10–300 nm range whereas the particle mass concentrations were almost equally distributed between the 10–1000 nm and 1000–2738 nm size range at each height. The PNCs in the 10–30 nm and the 30–300 nm size range were linearly correlated with the traffic volume but poorly correlated with the rooftop wind speed. © 2008 Elsevier Ltd. All rights reserved.

Keywords: Street canyon; Particle number distributions and concentrations; Vertical concentration profiles; Ultrafine particles

1. Introduction

Regulations to control the emission of atmospheric particulate matter (PM) are based on limits for PM_{10} ($D_p \leq 10 \mu m$) and $PM_{2.5}$ ($D_p \leq 2.5 \mu m$), using particle mass concentrations (PMCs), and not particle number concentrations (PNCs). Recent

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toxicological studies indicate greater toxicity for ultrafine particles ($D_p \leq 100$ nm) than coarser particles, per unit mass (Oberdorster, 2000); 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 ultrafine fraction of $PM_{2.5}$ contributes significantly to PNCs but little to PMCs (AQEG, 2005). The lack of standard methods and instrumentation for particle number measurements and detailed understanding of the influence exerted on fine particle dispersion by ambient meteorology and traffic flows have limited the scope for designing effective strategies for the mitigation of particulate pollution in urban areas.

Vehicles are the major source of ultrafine particles in urban street canyons (Schauer et al., 1996); high PNCs are common since the surrounding built-up environment limits dispersion of exhaust emissions (Van Dingenen et al., 2004). Concentrations can be much greater than in unobstructed locations (Bauman et al., 1982; Kumar et al., 2007). Over the past two decades, several groups have studied 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), but the vertical variation of particulate matter (both PMCs and PNCs) is still a matter of discussion. This variation is affected by factors including traffic volume, meteorology (including flow and turbulence produced by wind, traffic and atmospheric instability), the geometry of the street canyon (including the aspect ratio and street orientation), and any advection from adjacent streets. Removal (dry and wet deposition) and transformation (nucleation and condensation/evaporation) processes may also play an important role in altering PMCs and PNCs at different heights in urban street canyons.

Studies of vertical variation in gaseous and particulate mass concentrations have shown different vertical concentration profiles. Bauman et al. (1982) observed a decreasing concentration of carbon monoxide (CO) with height above road level; Zoumakis (1995) found this decrease to be exponential. Similar findings were reported by Murena and Vorraro (2003) for benzene. Few studies exist of vertical variations of PNCs in urban street canyons (Kumar et al., 2008; Li et al., 2007; Longley et al., 2004b). Kumar et al. (2008) also found a decrease in PNC (in the 5–1000 nm range)

with height in the lower part (first 2.60 m) of an approximately 20-m-high street canyon; similar results were reported for PNCs in the 10–487 nm range by Li et al. (2007) for an asymmetric street canyon having sides 10–18 m and 22–28 m high. Near-surface and rooftop level studies for fine particles had also indicated the decrease of PNCs with height above road level (Longley et al., 2004a; Vakeva et al., 1999). However, other studies (Micallef et al., 1998; Colls and Micallef, 1999; Park et al., 2004; Weber et al., 2006) reported increasing mass concentration of PM_{10} , $PM_{2.5}$ and CO with height in the lower part of the canyon, and then decreasing concentrations in the upper part of the canyon. Longley et al. (2004b) reported similar results for fine PNCs.

European Union directive 1999/30/EC (EC, 1999) suggests sampling heights between 1.5 and 4 m above road level, and up to 8 m above road level under specific local circumstances. The variability of vertical concentration profiles described above raises the question: “What should be the recommended height(s) for pollutant measurements in street canyons in order to represent the exposure of the entire local population?”

Very little information is available on the vertical variation of PNC in the coarse (1000 and 2500 nm), fine (below 1000 nm) and ultrafine (fraction of fine particles below 100 nm) particles in urban street canyons. Here, pseudo-simultaneous measurements were taken at four different heights (i.e., 1.00, 2.25, 4.62 and 7.37 m) of an 11.6-m-deep street canyon in Cambridge (UK). Unlike most other studies, real-time continuous measurements of the particle number distributions (PNDs) in 5–2738 nm range were made, sampling at 0.5 Hz using a four-way solenoid switching system together with a recently developed instrument, the ‘fast-response differential mobility spectrometer (DMS500)’. The aims were to determine the vertical variations of mass and PNCs in a typical European street canyon and to evaluate the influences of meteorology, traffic volume, and transformation processes on dispersion of particles in the coarse, fine and ultrafine size ranges.

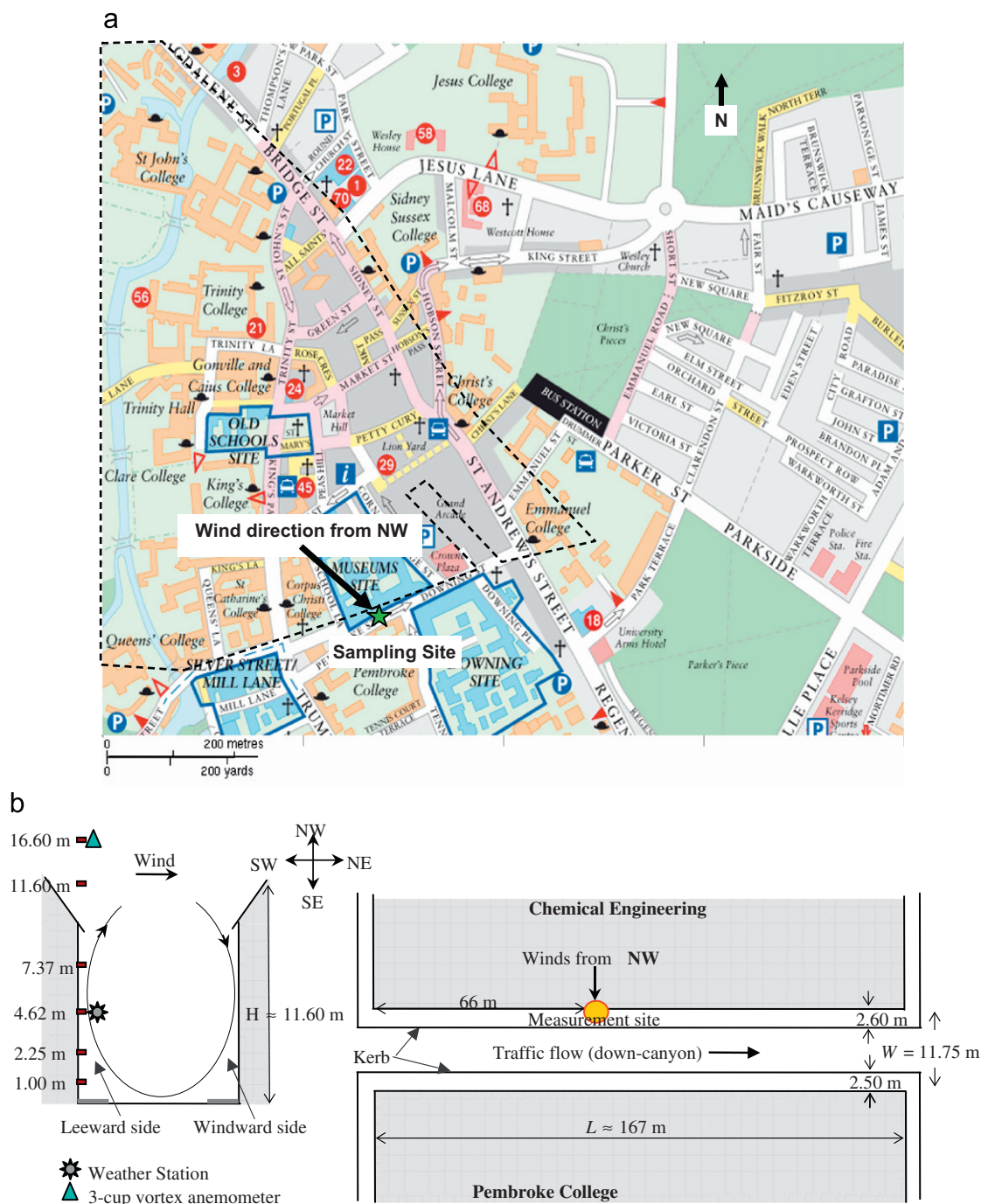
2. Methodology

2.1. Site description

Field experiments were performed between 20 and 21 March 2007 in Pembroke Street (Cambridge, UK; 52°12'N and 0°10'E) just outside the Chemical

Engineering Department building (Fig. 1a). Pembroke Street is close to the city centre; distinct peaks in weekday traffic occur at 08:00–09:00 h, 11:00–12:00 h and 19:00–20:00 h local time. The

studied section of street canyon (Fig. 1b) is 167 m long, running approximately northeast (NE) to southwest (SW). The Chemical Engineering Department is on the northwest (NW) side of the street and



(Figures not to scale)

Fig. 1. (a) Map of the location of the street canyon in the Cambridge area (map reproduced with permission of Cambridge University Press). The measurement site is marked by the green star and the dotted line covers the traffic-restricted area. (b) Schematic diagram of Pembroke Street, showing the traffic flow and wind directions, as described in text.

Pembroke College on the southeast (SE) with mean building heights (H) of about 11.6 and 11.5 m, respectively. 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 11.75 m wide (W) with one lane (6.65 m wide) of traffic travelling towards the northeast. Traffic flow at the NE end of the street is regulated by traffic signals while the traffic flow is free at the SW end. The studied section has an aspect ratio (H/W) of about unity (0.98) and has a length-to-height ratio (L/H) of about 14, making it a 'long-length' street canyon (Vardoulakis et al., 2003). The sampling points were 66 m from the SW end of the street canyon, 0.40 m from the wall of the Chemical Engineering Department building and set back 2.20 m from the kerb. During the studied period, winds were from the NW direction (see Section 3.1), and the next adjacent parallel street (away from the traffic restricted area) in the upwind direction was ~ 800 m away (Fig. 1a). Therefore, it is most unlikely that emissions from adjacent streets affected our measurements.

2.2. Instrumentation

A novel four-way solenoid switching system, constructed for this study, was used with the particle spectrometer (DMS500) to measure PNDs pseudo-simultaneously (see Section 2.3) in the 5–2738 nm range at four heights. The DMS500 can measure PNDs at a frequency of 10 Hz. However, our experiments recorded the average of 20 measurements (i.e., 0.5 Hz sampling frequency) to improve the signal/noise ratio. A detailed description of the working principle of the DMS500 and its application in different scenarios can be found in Collings et al. (2003), Biskos et al. (2005) and Symonds et al. (2007). The instrument was calibrated by the manufacturer, immediately before the study, using polystyrene spheres of known diameter and by comparing the results from sampling an aerosol with those from a scanning mobility particle sizer. Calibration errors in particle diameter measurements and sample flow rates were 3.4% and 2.3%, respectively. A cyclone, with a steel restrictor with a 0.52 mm-diameter hole, at the head of the sampling tube maintained a sample flow rate of 2.51 min^{-1} , and reduced the pressure within the sampling tube to 0.16 bars, improving the instrument's time response and reducing particle agglomeration (Biskos et al., 2005).

The switching system was designed to take pseudo-simultaneous measurements by automatically switching the sampling flow between each height once every 60 s, though it was capable of switching times between 20 and 150 s.

An automatic vortex, pole-mounted, three-cup anemometer (Windware, UK; maximum measurable wind speed: 56 m s^{-1}) recorded the wind speed 5 m above the rooftop (i.e., 16.60 m above road level). A wireless weather station (Thermor, UK) 4.62 m above road level recorded ambient temperature, humidity, atmospheric pressure, wind speed, and direction. Readings from the Cambridge University operated AT&T weather station were correlated with these local observations, which were found to be in reasonable agreement, these readings were used to determine the wind direction.

2.3. Measurements

Measurements were recorded continuously for 24 h (16:00–16:00 h), between the 20 and 21 March 2007, at four different heights (z) (1.00, 2.25, 4.62 and 7.37 m; referred to as $z/H = 0.09, 0.19, 0.40$ and 0.64 , respectively). To reduce particle losses in the sampling tubes and to reduce the lag-times, four independent sampling heads were used, one at each height. All four heads were mounted on a single pole, which was securely fastened to the building using guy ropes. It was thought to be unwise to leave the pole and sampling heads unattended so one or two of the researchers constantly observed the system. It is for this reason that the study only took place over one 24-h period. A DMS500 was used with the switching system that changed the sample height once every 60 s taking a total of 15 measurements in 1 h at each height. The results for the first 15 s of each 60 s sample were discarded to enable the previous sample to clear from the sampling tube and to equilibrate the pressure within the sampling tube. Exactly simultaneous measurements at each height could not be performed because only one instrument was available, but since the sampling was done in 360 separate time periods in total during the measurements whilst the PNC changed in an essentially random manner with respect to time, sufficient measurements were made to draw conclusions on the vertical variations of the PNC with height.

The wind speed was recorded every minute during the entire sampling period both within the street (at 4.62 m or $z/H = 0.40$; hereafter called U_s) and above

the rooftop (i.e., at 16.6 m or $z/H = 1.43$; hereafter called U_r). Outdoor temperature, relative humidity and pressure were recorded at approximately every 15 min. Traffic volumes were sampled through the measurement period by a movement-sensitive CCTV camera. Manual traffic counts were also made throughout the day to ensure that the sampling was reliable.

2.4. Particle losses in tubes

Four different lengths (5.17, 5.55, 8.90 and 13.40 m, referred to as L_1 , L_2 , L_3 and L_4 , respectively, 7.85 mm internal diameter) of thermally and electrically conductive sampling tube, made of silicon rubber to which carbon has been added, were used to obtain samples from each of the sampling heights. To quantify the particle losses in these tubes together with the switching system, particle measurements were made, using the same time frequency, etc., from a stationary diesel-engined car's exhaust (at a distance of approximately 500 mm), and compared with separately taken measurements using a reference tube of much

shorter length ($L_{\text{ref}} = 1.0$ m). We assumed that the losses in L_{ref} would be equivalent to the losses in first metre of each of the other four tubes, and determined the losses in each tube relative to their “corrected” length (i.e., their actual length minus 1 m). The size-dependent penetrations for the effective length of different tubes were then defined as the number concentration through the effective L_1 , L_2 , L_3 and L_4 of tube divided by the number concentration through the L_{ref} of tube. A correlation for penetration as a function of effective length and particle diameter was determined and was used to estimate the penetration for the actual lengths used. The penetrations for each tube length are shown in Fig. 2. Particle losses below 10 nm diameter are highly significant; as high as ~80% for L_4 , so have been discarded for subsequent analysis. Particle losses between 10 and 20 nm ranges are significant; size-dependent corrections have been made for this range. No corrections were performed for particle sizes > 20 nm. Comparison of experimental results with laminar and turbulent flow regime models (Hinds, 1999) were also made. The results were better described by the turbulent

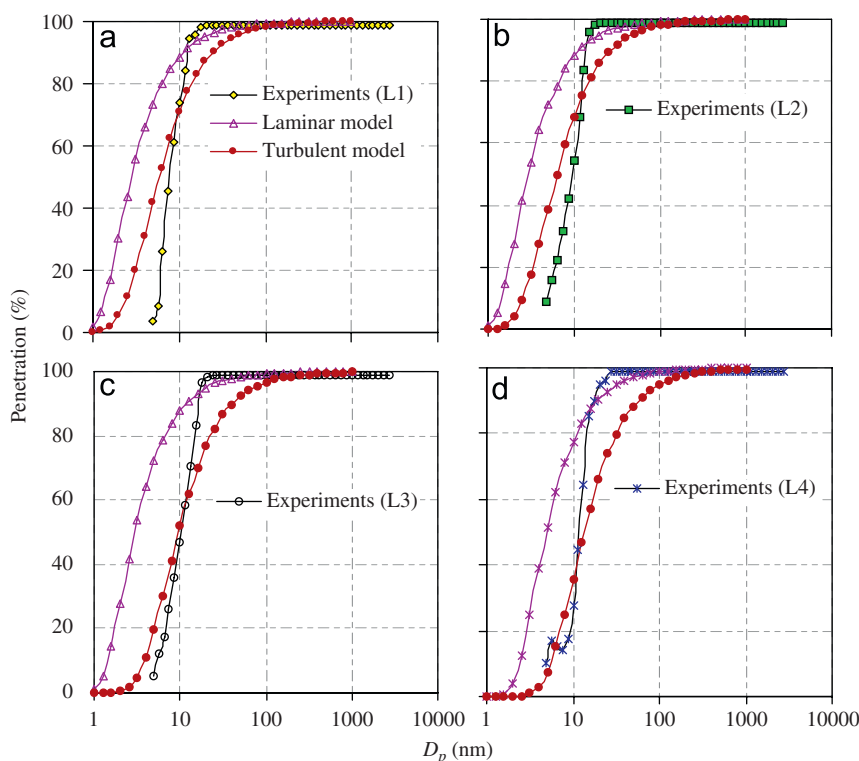


Fig. 2. Penetration of particles in different lengths of sampling tube, and their comparison with laminar and turbulent regime models. Turbulent and laminar model estimates are made for the following field conditions: ambient temperature, 8.2 °C; sample flow, 2.51 min⁻¹; sample line pressure, 0.16 bars; and Reynolds number, 461.

flow model, even though the Reynolds number in all the sample line lengths was within the laminar regime.

3. Results and discussions

3.1. Meteorology and traffic volume during experiments

The variation of the rooftop (U_r) and in-street (U_s) wind speeds during the sampling period is shown in Fig. 3. The hourly averaged values of U_r and U_s over the entire sampling period were $3.1 \pm 0.8 \text{ m s}^{-1}$ and $0.3 \pm 0.2 \text{ m s}^{-1}$, respectively. The values of U_s were found to be well correlated to U_r (i.e., U_s ranged between $0.10U_r$ and $0.15U_r$). This value of U_s seems to be considerably smaller than expected. The typical recirculation velocity is $1/3$ – $1/2$ of U_r for a street canyon of near-unity aspect ratio with U_r perpendicular to the street axis and exceeds 1.5 – 2 m s^{-1} (Britter and Hanna, 2003).

Fig. 4 shows the wind rose diagram for the 1-min averaged U_r for the entire sampling period. The predominant wind direction was from the NW (i.e., a cross-canyon flow perpendicular to the street

axis). Measurement points were on the leeward side of the street canyon (Fig. 1). Table 1 shows the frequency of the 1-min averaged U_r at different heights. For most of the time (88%), U_r was between 1.5 and 4.5 m s^{-1} . It exceeded this range for 8% of the time, and was $<1.5 \text{ m s}^{-1}$ for only 4%.

The hourly average traffic volume through the test site was determined over the sampling period (mean 536 veh h^{-1} , S.D. 266 veh h^{-1}). Traffic volume was smallest between 00:00 and 07:00 h

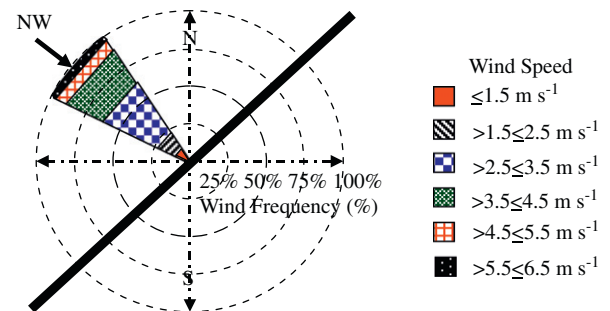


Fig. 4. Wind rose diagram for the 1-min averaged rooftop wind speed over the entire sampling duration. The wind was blowing from NW during the entire sampling duration. The thick black line represents the street canyon.

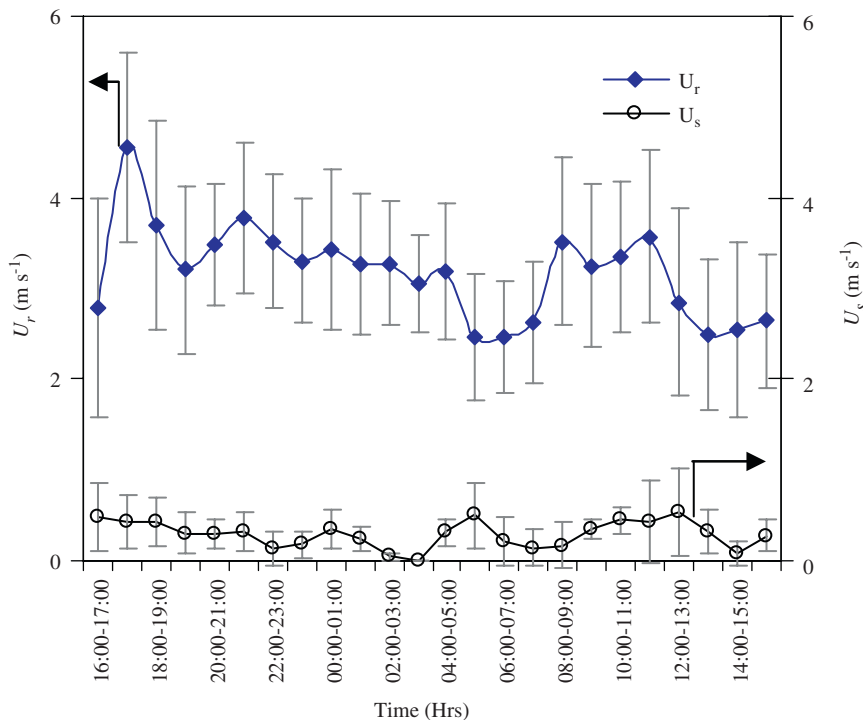


Fig. 3. Diurnal variation of hourly averaged U_r and U_s . Bars show the standard deviation of hourly averaged values.

Table 1

Frequency of 1-min average rooftop wind speed (U_r) in different ranges during sampling at different heights

U_r (m s^{-1})	Frequency (%) of U_r during the time of particle measurements				
	At $z/H = 0.09$	At $z/H = 0.19$	At $z/H = 0.40$	At $z/H = 0.64$	Average of all heights
≤ 1.5	3.6	3.8	4.1	3.9	3.9
$> 1.5 \leq 2.5$	21.7	21.2	22.5	26.4	23.0
$> 2.5 \leq 3.5$	42.1	40.2	40.4	35.2	39.5
$> 3.5 \leq 4.5$	25.5	26.3	25.5	24.9	25.6
$> 4.5 \leq 5.5$	6.2	5.1	6.3	6.9	6.1
$> 5.5 \leq 6.5$	0.9	3.5	1.2	2.7	2.1

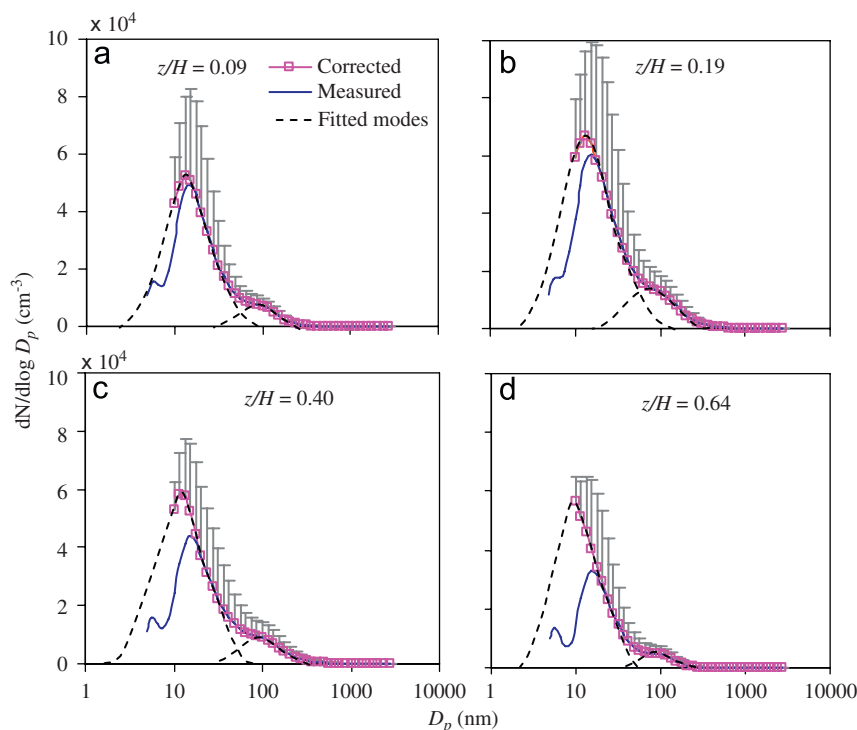


Fig. 5. Hourly averaged corrected and measured particle number distributions at (a) $z/H = 0.09$, (b) $z/H = 0.19$, (c) $z/H = 0.40$, and (d) $z/H = 0.64$. Dotted lines represent mode fitting curves to corrected PNDs. Bars show the standard deviation of the hourly averaged PNDs on each height; only positive standard deviation values are plotted for the clarity of the figures.

(mean 204 veh h^{-1} , S.D. 70 veh h^{-1}) and was largest between 07:00 and 16:00 h (mean 758 veh h^{-1} , S.D. 190 veh h^{-1}). Between 17:00 and 24:00 h, the hourly average and standard deviation were 501 and 111 veh h^{-1} , respectively. The maximum traffic volume occurred between 08:00 and 09:00 h at 1092 veh h^{-1} ; the minimum (140 veh h^{-1}) was between 00:00 and 01:00 h. These traffic volumes were generally consistent with traffic volumes on other weekdays that were collected as a part of a wider experimental campaign. Traffic speed was manually measured to be $30 \pm 7 \text{ km h}^{-1}$. Since the SW end of

the street canyon had a free flow of traffic, periods of congestion were rarely observed.

3.2. Vertical variations of particle number distributions (PNDs) and particle number concentrations (PNCs)

Fig. 5a–d shows the mean PNDs at each height throughout the measurement period. The PNDs could be described as consisting of distinct populations in different modes and quantified with their total particle number, geometric mean diameter and

shape. The modes were categorized as nucleation (10–30 nm, N_{10-30}), accumulation (30–300 nm, N_{30-300}) or coarse (300–2738 nm, $N_{300-2738}$) particles. Nucleation mode particles are believed to be formed by gas-to-particle conversion after rapid cooling and dilution of exhaust emissions when the saturation ratio of gaseous compounds of low volatility (e.g., sulphuric acid) reaches a maximum (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 coming mainly from diesel-engined or direct injection gasoline-engined vehicles (Graskow et al., 1998). A fraction of these particles (those between 30 and 100 nm, called Aitken mode particles) arise from the growth or coagulation of nucleation mode particles, and are also produced in high numbers by primary combustion sources such as vehicle exhausts (Kulmala et al., 2004). Coarse particles are mainly produced from brake wear, tyre wear and the resuspension of particles by traffic and wind-produced turbulence.

The PNDs at each height were bimodal with each mode having a lognormal form. Two distinct modes peaking at 13.3 nm (nucleation mode) and 86.6 nm (accumulation mode) were seen at each height (Fig. 5a–d). The shape of the PNDs at each height were similar but the maximum height of PNDs was seen at $z/H = 0.19$ decreasing towards the lowest ($z/H = 0.09$) and the highest ($z/H = 0.64$) sampling heights. The similarity in shape and negligible shift in peak and geometric mean diameter of PNDs in both modes at each height suggests that transformation processes were generally complete by the time the particles were measured and total particle numbers were conserved. Variations in PNDs with height were believed to be due to dilution and mixing caused by wind-produced and traffic-produced turbulence, and the removal processes at the road and rooftop levels and the walls rather than particle transformation processes. The relevance of various particle transformation and removal processes is estimated from time scale analyses in Section 3.5.

It is expected that PNCs will be larger in the lower part of the canyon due to the presence of the emission sources. The PNCs are expected to decrease with height due to removal of particles as a result of the mass exchange between the street canyon and the (less polluted) wind above. Interestingly, the vertical profiles show smaller PNC at

$z/H = 0.09$ and a maximum at $z/H = 0.19$ in each size range, as seen in Fig. 6. It should be noted that the background PNCs, which could be assumed to be the y -intercept of the fit to the data (PNCs vs traffic volume) with zero traffic volume and emissions (see Table 3), are not subtracted before plotting the vertical profiles since these values were much smaller ($< 10\%$ of PNCs in each size range as shown in Table 3) than the values measured and do not change the general shape of the profiles.

To compare our vertical profiles with other studies, we reviewed the literature (Supplementary Table S1) showing the vertical profiles for PNCs, PMCs (PM_{10} , $PM_{2.5}$) and gaseous pollutants (CO , NO_x , benzene). Most studies provide data only above ~ 2.5 m, and above this height all show concentration decreasing with height, as does our current study. However, a few studies (Micallef et al., 1998; Colls and Micallef, 1999) measured PM_{10} , $PM_{2.5}$ in the lowest 3 m of the canyon; their concentration profiles were not monotonically decreasing with height; both the largest and the smallest concentrations were observed at 0.81 m on some occasions. Conversely, our previous study (Kumar et al., 2008) covered only the first 2.60 m of the canyon, and the concentrations were found to be decreasing with height. Similarly, Li et al. (2007) reported concentrations at 1.5 m, and found the exponential decrease in concentrations with height though the difference in the their first and second

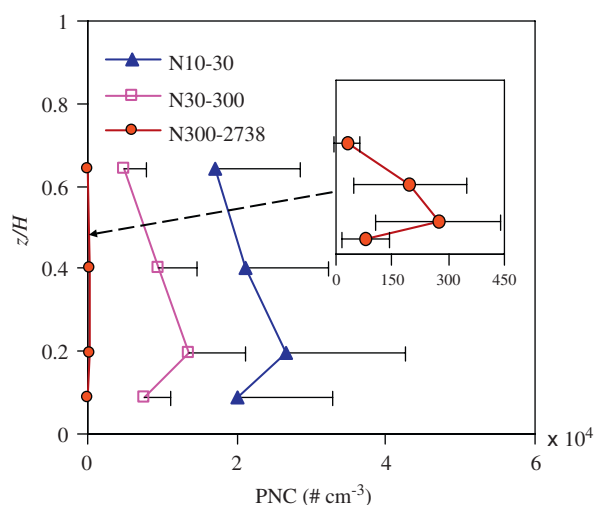


Fig. 6. Vertical profiles of particle number concentrations in N_{10-30} , N_{30-300} and $N_{300-2738}$ size ranges. Bars show the standard deviation of the hourly averaged PNCs over the entire sampling period. Only negative or positive standard deviations are plotted for the clarity of the figures.

measurement points are quite high (i.e., 1.5 and 8 m). Both studies represent asymmetric canyons where the mixing and flow conditions would be different from the symmetric canyon studied here. Similar to our current work, profiles were shown by Weber et al. (2006) with the largest PM_{10} and $PM_{2.5}$ at 3.9 m, decreasing towards road level (2.5 m) and rooftop (8.84 m). Furthermore, Longley et al. (2004b) reported the largest PNCs (10–487 nm) at 10 m on some occasions, decreasing towards road (3.5 m) and rooftop (18 m) levels. Apart from differences in canyon geometry, the main difference for our study is that the traffic flow was one-way as compared with two-way traffic in most other studies. Also, the present study only represents the cross-canyon (NW, leeward situation) winds as compared with several other wind conditions in other studies.

Some possibilities are proposed for the smaller concentrations at $z/H = 0.09$ than at $z/H = 0.19$. Firstly, ignoring the effect of traffic-produced turbulence (likely to be similar at the first two sampling heights), dilution and dry deposition are the important processes, as evident from their time scales (see Section 3.5). Dilution can also be assumed to be comparable for both heights, leaving dry deposition as the important additional process (due to the small time scale) for the concentration differences near the road surfaces. Other evidence of this can be seen by the good linear correlation between N_{10-30} and N_{30-300} (shown in Table 2), with negative N_{10-30} intercept only at $z/H = 0.09$, indicating a sink of these particles at this height. We expect that dry deposition should remove the smaller particles (i.e., N_{10-30}) more effectively due to their high diffusion coefficient compared with that for larger particles (i.e., N_{30-300}). This difference can be seen by the concentration profiles for N_{10-30} and N_{30-300} particles between $z/H = 0.09$ and 0.19; the PNCs in N_{10-30} range at $z/H = 0.19$ were only 1.33 times higher than at $z/H = 0.09$, indicating much higher removal of smaller particles, as compared

with 1.79 times larger at $z/H = 0.19$ for the PNCs in N_{30-300} range than at $z/H = 0.09$.

Secondly, the flow in a real street canyon is likely to be considerably more complex than the simple descriptions that we and others typically use, in reality involving along and cross street flows, recirculating vortex and flow intermittency; these complexities will probably be specific to each individual street canyon. A recirculating vortex structure in the canyon can transport the pollutants from the windward side, along with the sweeping of near road concentrations ($z/H = 0.09$) to the more elevated sampling points on the leeward side. Some variation in observations due to canyon asymmetry, other nearby streets, one-way or two-way traffic and other individualities are also to be expected. It is possible that the one-way traffic in our study may have also influenced our results. This would accentuate the formation of trailing vortices in the vehicle wake (Baker, 2001). These trailing vortices may transport the pollutants from the lowest sampling point to the upper sampling points. Since the sampling points were quite close to the traffic lane (see Section 2.1), the influence of this on the first two sampling points may be significant. Our results show that the dispersion of pollutants in the lower part of the canyon may not be straightforward and there is clearly a need for furthermore detailed studies.

3.3. Vertical variations of particle mass distributions (PMDs) and concentrations (PMCs) with height

PMDs ($dM/d\log D_p$) were obtained by multiplying the “corrected” number distribution ($dN/d\log D_p$) by mass per particle $M(D_p)$ (Park et al., 2003), i.e.

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

Detailed description of the estimation of $M(D_p)$ can be seen in Symonds et al. (2007) and Park et al. (2003). The density of particles is assumed to be 1 g cm^{-3} . The hourly averaged PMDs at each height for the entire sampling period are shown in Fig. 7a–d. The PMDs at each height were similar in shape, with a large PMD peak at $183 \pm 46 \text{ nm}$ and another at 649 nm . As with the PNDs, the highest PMDs were observed at $z/H = 0.19$, with peaks at 237 and 649 nm . Using the corrected PMDs, PMCs were obtained in two broad categories i.e., PM_1 (mass concentrations of particles having

Table 2

Cross-correlation between N_{10-30} and N_{30-300} size particles at each height

z/H	Correlation	R^2
0.09	$N_{10-30} = 2.99N_{30-300} - 2835$	0.64
0.19	$N_{10-30} = 1.60N_{30-300} + 4662$	0.56
0.40	$N_{10-30} = 1.21N_{30-300} + 9780$	0.51
0.64	$N_{10-30} = 2.99N_{30-300} + 2346$	0.59

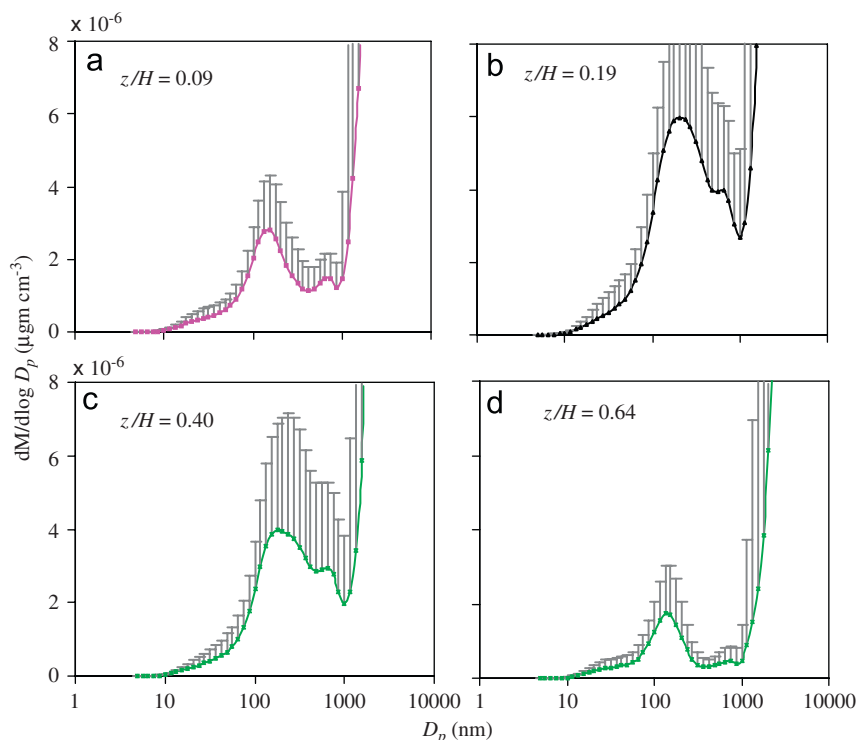


Fig. 7. Hourly averaged corrected particle mass distributions at (a) $z/H = 0.09$, (b) $z/H = 0.19$, (c) $z/H = 0.40$, and (d) $z/H = 0.64$. The PMDs are estimated from “corrected” PNDs. Bars show the standard deviation of the hourly averaged PMDs; only positive standard deviation values are plotted for the clarity of the figures.

$D_p \leq 1000$ nm) and $PM_{2.7}$ (mass concentrations for the fraction of particles having $1000 \text{ nm} < D_p \leq 2738$ nm) for further analysis. Since at all heights, the PMDs showed maximum mass in the 30–300 nm range, the PMCs in this range ($PM_{0.03-0.3}$) were also derived (Fig. 9). Hourly averaged values for $PM_{2.7}$ and PM_1 over the entire sampling period at all heights were 51% and 49% of total mass, respectively. The proportion changed to 58% and 42% at $z/H = 0.09$; this was expected because of the larger amount of coarse particles near to the street level. Otherwise, this proportion was consistent at $z/H = 0.19, 0.40$ and 0.64 having about $48 \pm 0.58\%$ and $52 \pm 0.58\%$ of total mass for $PM_{2.7}$ and PM_1 , respectively. As also expected, $PM_{0.03-0.3}$ contributed significantly ($67 \pm 9\%$) to PM_1 mass at each height. The PMCs for the nucleation mode (those between 10 and 30 nm range) contributed much less ($4.1 \pm 1.9\%$) to the PM_1 at each height. It is important to note that the $PM_{2.7}$ mass, which is similar to the PM_1 mass, represents only some 10's of particles per cm^3 in the $N_{1000-2738}$ range in contrast to the 10^4 's of particles per cm^3 in $N_{10-1000}$ range.

The vertical profiles for $PM_{2.7}$, PM_1 and $PM_{0.03-0.3}$ are shown in Fig. 8. The shape of the PMC profiles are similar to the PNC profiles, showing the smallest concentrations at $z/H = 0.09$. As shown in Table S1 and discussed in Section 3.2, this is not a common feature of vertical profiles for PMCs since most studies (Li et al., 2007; Vogt et al., 2006; Zoumakis, 1995) have reported maximum concentrations near the canyon bottom decreasing with increasing height above road level. However, similar vertical profiles for $PM_{2.5}$ and PM_1 were reported by Weber et al. (2006), and for PM_{10} and $PM_{2.5}$, in the first 3 m of an urban street canyon by Micallef et al. (1998) and Colls and Micallef (1999), attributed to larger variations in pollution mixing due to traffic-produced turbulence close to ground.

3.4. Modal share of particle number concentrations (PNCs), total particle surface area concentration (ToS) and total particle volume concentration (ToV) at different heights

Particles in the N_{10-30} range dominate (~ 65 – 77% of $N_{10-2738}$) the PNCs at each height and particles in

the N_{10-300} range accounted for nearly all ($\sim 99.5\%$ of $N_{10-2738}$) of the PNCs (Fig. 9a). Similar observations were reported by Wehner and Weidensohler

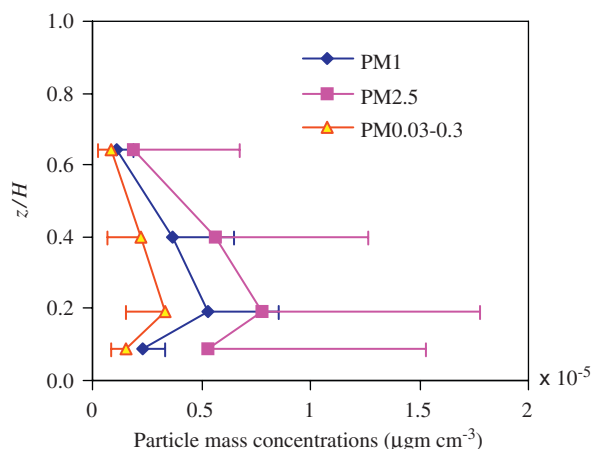


Fig. 8. Vertical profiles of particle mass concentrations in various size ranges. Bars show the standard deviation of the hourly averaged PMCs. Only positive or negative standard deviations are plotted for the clarity of the figures.

(2003) in their long-term study (over 4 years) for sub-micron particles in Leipzig, and by Tuch et al. (1997) for European cities. The ratio of N_{10-30} to N_{30-300} was smallest (1.94) at $z/H = 0.19$ and increased to 2.61 at $z/H = 0.09$ and to 3.49 at $z/H = 0.64$. Low-number concentrations of pre-existing particles have been reported to favour both production of new particles and their growth to detectable sizes in the atmosphere (Kulmala et al., 2004), while high-number concentrations of pre-existing particles promote both the condensation of semi-volatile vapours and disfavour the growth of fresh nuclei and their survival from high-coagulation scavenging (Kerminen et al., 2001). Despite conditions being favourable for nucleation, changes in N_{10-30} and N_{30-300} ratios seems to be largely due to relatively higher decrease (44%, 32% and 64% at $z/H = 0.09$, 0.40 and 0.64, respectively, from $z/H = 0.19$) in PNCs in the N_{30-300} range due to dilution as compared with PNCs decrease (25%, 20% and 36% at $z/H = 0.09$, 0.40 and 0.64, respectively, from $z/H = 0.19$) in N_{10-30} range.

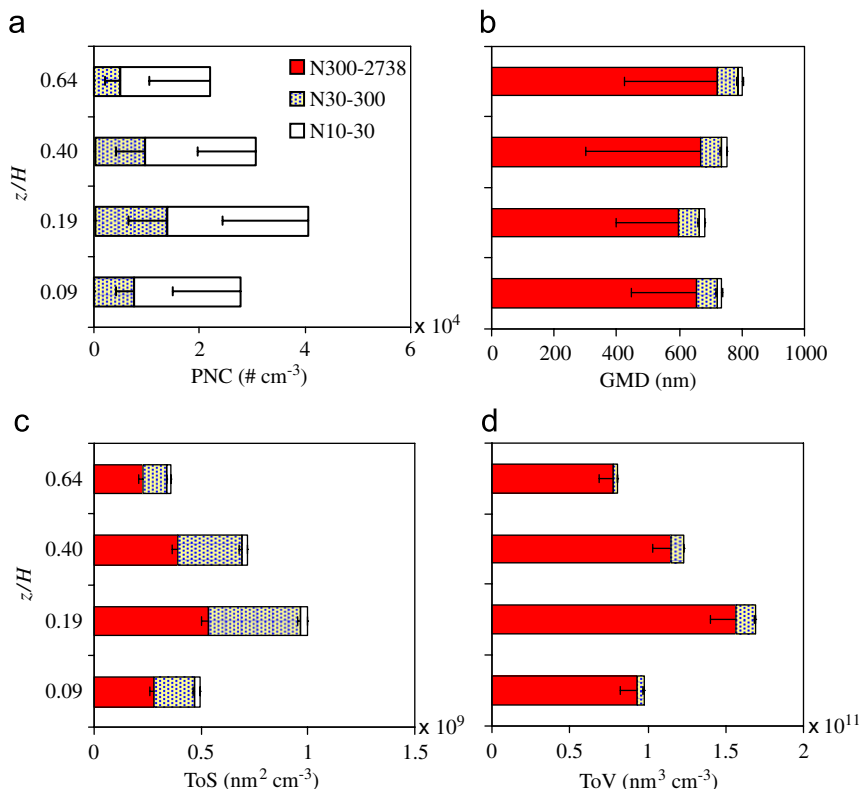


Fig. 9. Hourly averaged (a) particle number concentrations, (b) geometrical mean diameters of PNCs, (c) total particle surface area concentrations, and (d) total particle volume concentrations in different size ranges at each height. Bars show the standard deviation of the hourly averaged values. Only negative standard deviations are plotted for the clarity of the figures.

The geometric mean diameters (GMDs) in Fig. 9b, averaged over the entire sampling period and all heights in the N_{10-30} and N_{30-300} size ranges, were consistently 16.4 ± 0.9 nm and 64.7 ± 5.1 nm, respectively. However, the GMD in the $N_{300-2738}$ size range was 661 ± 268 nm, showing large variation, probably due to the limited number of particles in that dataset. The total particle surface area and volume concentrations (ToS and ToV) in Fig. 9c and d reflect the PNC but with increased weighting towards larger particles.

3.5. Time scale analysis for various transformation processes

As discussed in Section 3.4, removal and transformation processes such as dry deposition, coagulation and condensation may be important for changing the PNCs and the associated total particle number (ToN), ToS and ToV in any size range. Though these processes are not treated in detail here, we have estimated the time scales for these processes to give a first indication of their relevance on the PNCs at different heights, and to test the above-assumed (Section 3.2) hypothesis that the transformation processes (except possibly condensation) were generally complete by the time particles were measured and the particle numbers were also conserved. These time scales might be thought of a relative measure of the time taken to reduce the concentration of particles in the street, if the source was turned off. Thus, a short time scale indicates a strong process. The main body of these detailed calculations is available in the Supplementary Section S1; the conclusions are summarized here.

The deduced time scales were of the order of 40 s for dilution, 30 and 130 s for dry deposition to the road surface, and 600 and 2600 s for the dry deposition to the road walls for the N_{10-30} and N_{30-300} ranges, respectively. For coagulation, the time scale for N_{10-30} particles was of the order of 10^5 s and for N_{30-300} , it was of the order of 5×10^5 s. For condensation, the time scales were of the order of 10^5 – 10^4 s for extreme growth rates at 1 and 20 nm h⁻¹, respectively.

Comparison of the estimated dilution time scale with those for other processes shows that dilution is comparatively quick and does not allow other processes (with the exception of dry deposition on road surface) sufficient time to act and alter the PNDs. This supports our proposed hypothesis that transformation processes are not important for

changing the PNDs at different heights. As noted, dry deposition on the road surface does seem to be a major removal process, reducing the ToN near road level as is clearly seen in vertical profiles nearest to the road level.

3.6. Effect of meteorology and traffic volume on particle number concentrations

Some of the factors influencing the PNCs may be more important than others in producing the hourly variations. The hourly variations of PNCs in each size range were quite marked, as shown in Fig. 10a–d. The hourly average PNCs in the $N_{10-2738}$ size range over the entire sampling duration were maximum (4.95×10^4 #cm⁻³) and minimum (2.19×10^4 #cm⁻³) at $z/H = 0.19$ and 0.64, respectively. As more than 99% of the total particles by number were in the 10–300 nm size range (shown in Section 3.4), there was a negligible presence of coarse particles suggesting that traffic was the main source of particles. A large peak at $z/H = 0.19$ was observed between 07:50:20 and 07:50:48 h; this was caused by a heavy-duty diesel truck standing near the sampling points. The average PNCs during this period increased from overall average value of $\sim 10^4$ to $\sim 10^7$ #cm⁻³, resulting in an increase of overall average PNCs for the entire hour (07:00–08:00 h) at this height. Similar to the diurnal variation of traffic volume (Section 3.1), as expected, the lowest and the highest average concentrations at all heights were during the periods of the lowest traffic (between 00:00 and 07:00 h) and the highest (between 07:00 and 16:00 h) traffic volume, respectively.

The PNCs at each height do not show strong correlation with the variation in rooftop wind speed (Fig. 10a–d). For example, the highest (17:00–18:00 h, 20 March) and the lowest (16:00–17:00 h, 21 March) wind speeds correspond to very similar values (relatively lower than over all average) of PNCs at each height. However, there were also the periods of higher wind speeds (e.g., between 04:00 and 05:00 h) corresponding to the lowest PNCs, and lower wind speeds (e.g., between 19:00 and 20:00 h) corresponding to the relatively higher PNCs.

The traffic volume and rooftop wind speed are generally the principle variables influencing the PNCs in the street canyon. In order to show their effects on PNCs, the PNCs in N_{10-30} , N_{30-300} and $N_{300-2738}$ size ranges are correlated with traffic volume, as shown in Table 3. PNCs linearly depended on traffic volume, showing stronger

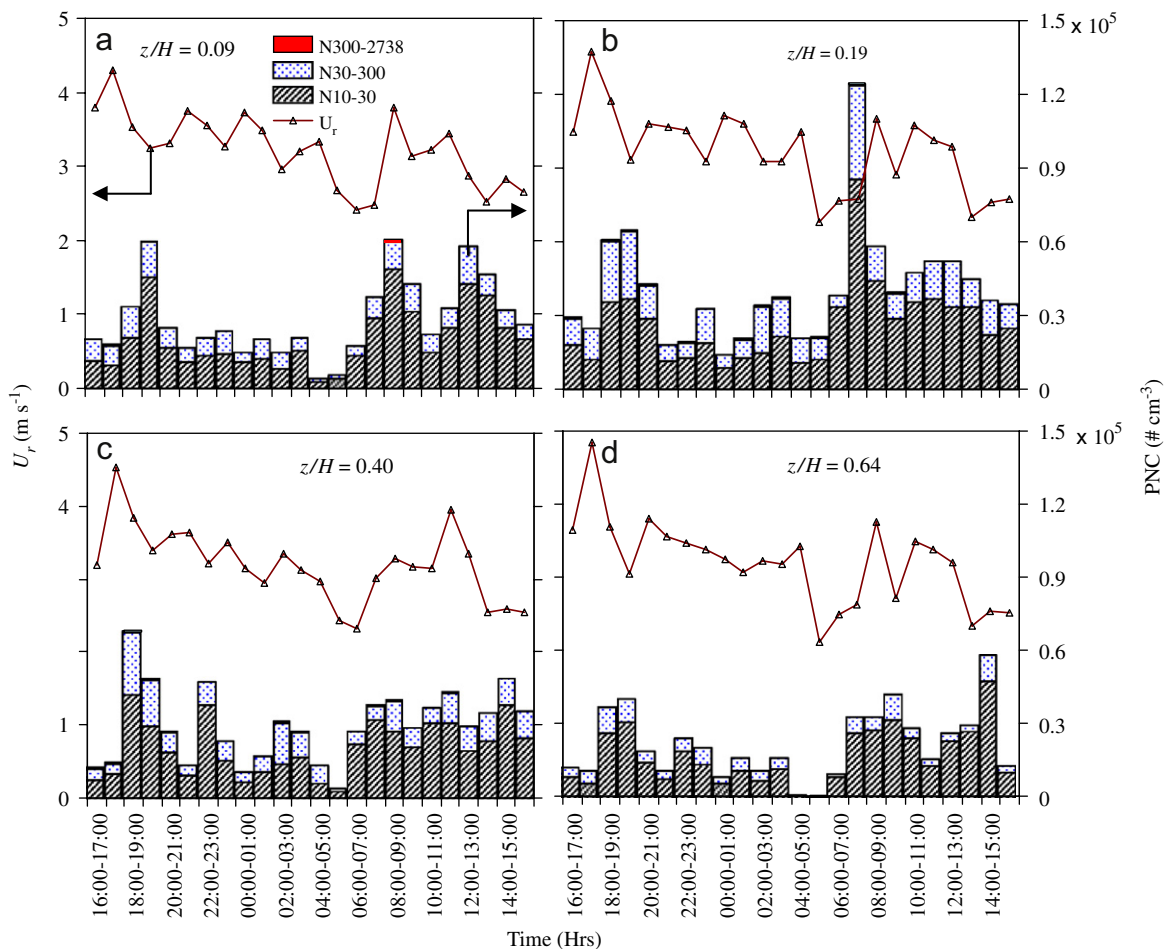


Fig. 10. Diurnal variation of the hourly averaged rooftop wind speed and particle number concentrations in various size ranges at (a) $z/H = 0.09$, (b) $z/H = 0.19$, (c) $z/H = 0.40$, and (d) $z/H = 0.64$, starting from 16:00 h on 20 March till 16:00 h on 21 March 2007. Particle number concentrations are the 1-min average of measurements taken at each height during each hour, and the wind speeds on each height are the 1-min average during the periods of particle measurements at each height.

Table 3

Correlation between the PNCs in various size ranges and traffic volume (T) at different sampling heights

z/H	N_{10-30}	N_{30-300}	$N_{300-2738}$
0.09	$30.8T + 1986$ ($R^2 = 0.66$)	$7.7T + 3321$ ($R^2 = 0.58$)	$-0.02T + 90$ ($R^2 = 0.01$)
0.19	$39.1T + 3435$ ($R^2 = 0.66$)	$16.1T + 4053$ ($R^2 = 0.50$)	$0.10T + 210$ ($R^2 = 0.04$)
0.40	$24.1T + 6963$ ($R^2 = 0.54$)	$8.7T + 3970$ ($R^2 = 0.31$)	$0.13T + 111$ ($R^2 = 0.09$)
0.64	$24.5T + 2428$ ($R^2 = 0.52$)	$4.6T + 2062$ ($R^2 = 0.28$)	$-0.01T + 34$ ($R^2 = 0.01$)

association between PNCs in the N_{10-30} size range and traffic volume than the PNCs in N_{30-300} size range; however, the correlations were very poor for the largest particle size range ($N_{300-2738}$) which could be due to the limited data available for these particles, though it is also probable that many of

these particles were wind-blown rather than emitted directly from vehicles. Therefore, the PNCs in $N_{300-2738}$ size range are not considered for further analysis.

Considering the effects of both traffic volume and U_r on the PNCs, number concentrations in any size

range at various heights could be represented as

$$N_{i-j} = aT^m U_r^n + b, \quad (2)$$

where N_{i-j} are the PNCs in any size range; a , b , m and n are constants (here, m is assumed to be unity); T is traffic volume; b represents background PNCs. The correlations using Eq. (2) are shown in Table 4. The values of R^2 shown in Tables 3 and 4 are broadly similar for both (excluding and including U_r) conditions, showing comparatively stronger correlations for N_{10-30} than N_{30-300} at all heights.

To remove the prime dependence of the PNCs on traffic volume, the PNCs were divided by the traffic volume; this was used as a primary variable and plotted against U_r during the periods of particle measurement at each height (see Table 5). It can be clearly deduced from Table 5 that the correlations between the primary variable (PNCs divided by traffic volume) and U_r are very poor as compared to the more robust correlations between PNCs and traffic volume. These observations are in contrast to the results reported by Charron and Harrison (2003) for road-side measurements in London where they found a inverse correlation between the PNCs in both size ranges and U_r . The reason for poor correlations in our case could be an artifact of the small dataset collected since the variations in U_r were significantly smaller (with in a factor of 3, see Table 1) when compared with the variations in

traffic volume (a factor of 10) throughout the campaign. Also, the flow within the measurement period was consistently cross-canyon. The other reason for the poorer correlations could be the dissimilarity in street canyon geometry and sampling position since our measurements were taken nearer to the road side (see Fig. 1) where the influence of vehicle emissions on measurements can be greater because of the advection of traffic emissions towards the leeward side of the canyon by wind-produced street canyon vortex (Britter and Hanna, 2003), though this has not been explicitly shown here. This is especially true since the average time to reflect emissions of individual vehicles with the DMS500 was between 40 and 60 s. This is comparable to the predicted time scale for most significant transformation process (e.g., dilution) under the average wind conditions.

4. Summary and conclusions

The particle number distributions (PNDs) were measured at four heights on one side of a regular street canyon in Cambridge, UK. Particle number distributions and concentrations in various size ranges varied significantly and increased to a peak at $z/H = 0.19$, decreasing towards both the lowest ($z/H = 0.09$) and the highest ($z/H = 0.64$) sampling points. Our results seem to be in accordance with the EU directive, suggesting sampling heights between 1.5 and 4 m since it is within this range that the maximum concentrations were observed. Vertical concentration profiles did not show an exponential decrease with height. The reduction in measured particles at the bottom of the street canyon was not straightforward to describe but was thought to be due to the dry deposition of the particles on to the road surface. The steadily decreasing concentration of particles in the upper part (i.e., between $z/H = 0.19$ and 0.64) of the canyon was attributed to the removal of particles due to the mass exchange between the street canyon and the wind above. The relative changes in the total masses of particles in various size ranges with height ($PM_{0.03-0.3}$, PM_1 and $PM_{2.7}$) were similar to those obtained for number concentrations. The majority (>99% of $N_{10-2738}$) of the PNCs were found in the N_{10-300} nm size range, whereas the particle mass concentrations were found to be almost equally distributed between PM_1 and $PM_{2.7}$ at each height.

Table 4
Correlations of PNCs in various size ranges with traffic volume and U_r

z/H	For N_{10-30}					For N_{30-300}				
	a	m	n	b	R^2	a	m	n	b	R^2
0.09	39	1	−0.14	2044	0.67	6	1	−0.23	3186	0.54
0.19	133	1	−0.98	3095	0.76	35	1	−0.43	3980	0.52
0.40	70	1	−0.64	6481	0.55	15	1	−0.04	4329	0.30
0.64	74	1	−0.87	2228	0.57	7	1	−0.02	2061	0.28

Table 5
Correlations between the product of PNCs in various size ranges and inverse of traffic volume, and the U_r at different heights

z/H	N_{10-30}/T	N_{30-300}/T
0.09	$47.3U_r^{-0.23}$ ($R^2 = 0.01$)	$19.5U_r^{-0.21}$ ($R^2 = 0.19$)
0.19	$107.1U_r^{-0.67}$ ($R^2 = 0.12$)	$27.8U_r^{-0.01}$ ($R^2 = 0.01$)
0.40	$82.4U_r^{-0.64}$ ($R^2 = 0.06$)	$19.1U_r^{-0.01}$ ($R^2 = 0.01$)
0.64	$42.7U_r^{-0.33}$ ($R^2 = 0.01$)	$9.6U_r^{-0.01}$ ($R^2 = 0.04$)

The PNDs at each height were bimodal, similar in shape and showed similar values of peak and geometric mean diameters in each size range; this suggested that transformation processes were essentially complete by the time measurement were made. Differences in PNDs at different heights largely reflect dilution by the wind from above the canyon, and dry deposition (at the lowest height), rather than other particle transformation processes. This possibility was supported by an order of magnitude determination of the time scales for removal and transformation.

Traffic was the main source of particles in the street canyon. The PNCs in each size range (i.e., N_{10-30} and N_{30-300} except $N_{300-2738}$) correlated directly with traffic volume, showing stronger correlation with PNCs in the N_{10-30} size range than those in the N_{30-300} size range at all heights. Little correlation was found between rooftop wind speeds and the PNC, most probably due to the small range of the wind speed in our study.

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Appendix A. Supplementary materials

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.atmosenv.2008.01.010](https://doi.org/10.1016/j.atmosenv.2008.01.010).

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