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A case study of aerosol (4.6 nm < D_p < 10 μm) number and mass size distribution measurements in a busy street canyon in Manchester, UK

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Abstract

The Air Quality Management community is increasingly turning its attention to urban 'hot-spots' where localised high concentrations of pollutants can arise. One such location is the urban street canyon where dispersion is poorly understood or described by regulatory models because of the complexity of the airflow, turbulence and local influences. Similarly, simple metrics such as PM_{10} fail to describe the range of sizes, composition, sources and behaviours encompassed by the term 'particle'. A 2-week experimental case study to measure size-segregated aerosol in the size range 4.6 nm–10 μm at a fine time scale (10 min resolution) was undertaken in a typical street canyon in Manchester. The wind direction incident to the canyon, and hence the vortex flow within the canyon, was found to have a large influence on the number concentrations, with values typically 2–10 times greater in perpendicular flow than the estimated inner-urban background. Concentrations were also inversely related to wind speed and directly related to traffic flow. Coarse mode mass concentrations were generally found to follow urban background PM_{10} concentrations except with a 0–5 $\mu g\,m^{-3}$ enhancement related to traffic-induced re-suspension within the canyon. A small pollution episode consisting of coarse material re-suspended by high winds was extended in time within the canyon.

Keywords: Street canyon; Urban aerosol; Re-suspension; Particle number; Size distribution

1. Introduction

Pollution of the urban air by suspended particulate matter has been identified as a serious health risk, not only in the visibly affected mega-cities of the developing world, but also at the relatively low concentrations found in the air of the more developed world too (Schwartz, 1994), incurring significant loss of productive life and public health burden (Künzli et al., 2000; COMEAP, 2001). Epidemiological studies have linked both acute and chronic exposure to suspended particulate matter to increased morbidity and mortality linked to respiratory and cardiovascular disease in the USA (e.g. Dockery et al., 1993; Samet et al., 2000) and in Europe (Katsouyanni et al., 1997).

Currently, the respirable fraction of suspended particulate matter, PM_{10}, is regulated in the UK (as well as $PM_{2.5}$ in the USA) despite the fact that PM_{10} contains particles with a range of sources, compositions and sizes, with consequent differing spatial and temporal variations in emission and concentration, and differing transport properties. Recently, there have been a number of toxicological studies (reviewed in Donaldson et al., 2001) that have suggested that the toxic effect of

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particles are related to either their total surface area or total number concentration, measures which, in an urban setting, find their maxima in the accumulation (0.1–1 μm) and ultra-fine (<0.1 μm) modes, respectively. However, the effect of coarse particles on mortality and morbidity cannot yet be ruled out, and the composition or solubility, or the interaction of the pollutant mix may also be significant (Harrison and Yin, 2000). Measured and modelled concentrations of PM_{10} are used for both epidemiological studies as well as for the delineation of Air Quality Management Areas in the UK and the assessment of control measures, such as traffic management. However, due to the inhomogeneity of its constituents, PM_{10} does not necessarily represent the true, or a single, adverse health effect.

Although there may be a significant correlation between PM_{10} and particle number in the urban background (Harrison et al., 1999), mass and number metrics are dominated by coarse and ultra-fine particles, respectively, and thus represent different sources and have different responses to meteorology. PM_{10} and $PM_{2.5}$ have increasingly been shown to be poor indicators of ultra-fine aerosol (and hence health effect) in roadside areas due to traffic emissions being less diluted than in the urban background (Shi et al., 1999; Molnár et al., 2002).

The urban street canyon is an important microenvironment for it can contain a high concentration of sources (vehicles), often in congested driving conditions, plus it may be expected that the physical flow isolation of the street canyon will increase concentrations compared to a more open site, and could complicate the differences between mass and number concentrations and between ultra-fine and coarse mode concentrations even further. Although most people spend relatively short periods in street canyons compared to other microenvironments, this close proximity to sources and poor dispersion represents a disproportionately high exposure, especially for ultra-fine particles, and it has been shown that adverse health effects can be triggered by such brief excursions (Michaels and Kleinman, 2000). Some individuals may spend much longer in street canyons (street workers, drivers, workers in buildings along the street) and thus the canyon will have a much greater influence on their total exposure. Consequently, if exposure of the population to the different fractions within PM_{10} are to be understood, and if the efficacy of urban pollution control measures are to be assessed, then it is necessary to determine, by measurement in the field, how particle number and mass concentrations in urban street canyons are related to simple available parameters, such as traffic flow, wind speed and direction, etc., and how this behaviour varies with particle size.

Most field studies in urban areas have been sited at 'background' locations. Urban street canyon field measurements of particle mass concentrations, other than just PM_{10} and $PM_{2.5}$, have been limited. Coarse mode mass concentrations were investigated in the UK by Namdeo et al. (1999) and by Harrison et al. (2001). Diurnal cycles in ultra-fine particle concentrations measured in a street canyon in Lahti, Finland were presented by Väkevä et al. (1999) and diurnal variations in particle number concentrations, size distributions and mass mode concentrations were investigated at a traffic-influenced urban site in Basel, Switzerland by Junker et al. (2000), but in both studies the effect of wind speed and direction was not tackled. Measured variations in aerosol size distributions in European street canyons were reported by Wählin et al. (2001) and Wehner et al. (2002), but the effect on PM measures was not discussed.

A pilot field study was undertaken in an urban street canyon, called SCAR: Street Canyon Aerosol Research. One of the aims was to investigate how both the health-related ultra-fine particle number concentration, and potentially confounding coarse mode mass concentrations vary at a fine time scale, and such results are presented here.

2. Activities

2.1. Site

An experimental site was chosen alongside the Town Hall building in Princess Street, Manchester, one of the major cities in the northwest of England. The City of Manchester is surrounded by numerous satellite towns, which make up the conurbation of Greater Manchester, which has a population of 2.5 million. The city centre has distinct peaks in weekday traffic flow that occur at 07:00–09:00 and 16:00–18:00 h local time. Manchester is prone to frequent cyclonic conditions and experiences average wind speeds for the UK, with a January mean of $5.0\,m\,s^{-1}$ and a July mean of $3.9\,m\,s^{-1}$ (International Station Meteorological Climate Summary, Version 4.0).

The chosen section of the street (Fig. 1) is 120 m long and asymmetric. The Town Hall along the southwest side has a height of 22–28 m, and a variety of buildings on the northeast side have heights of 10–18 m. Rooftops are complex and varied on both sides. There are open squares at both ends (Albert Square and St Peter's Square) and the canyon is aligned at 130° to north. The street canyon itself is 17 m wide with two lanes of traffic both travelling towards the southeast (the direction referred to as *down-canyon* throughout SCAR), plus parking bays on the south side and a row of bus stops on the north side.

Four experiments were performed. SCAR 1, 2 and 3 in February, April and May 2001 concentrated on turbulence measurements and will be discussed in a separate paper. This paper is concerned with data from

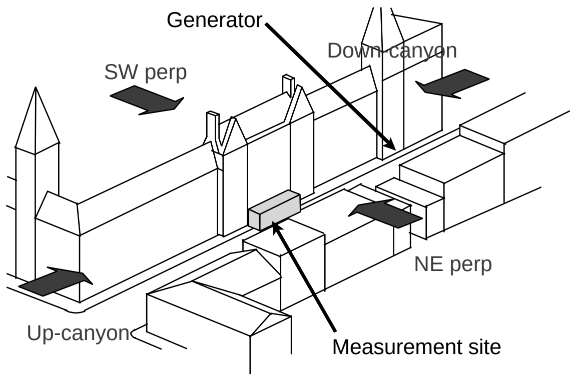

Fig. 1. Princess Street experimental canyon site and wind sectors described in text.

SCAR-4, which lasted for 2 weeks in October 2001 (15th to 26th inclusive) and ran continuously for each Monday to Friday period.

2.2. Instruments and measurements

During SCAR-4 instruments were placed upon a platform 2 m above the road and within a trailer located in two adjacent parking bays on the southwest side of the canyon (the higher wall), at the mid-point of the canyon's length. Two aerosol instruments were deployed on the platform: an SMPS (TSI Model 3936 Scanning Mobility Particle Sizer, covering the diameter range 4–160 nm) and an FSSP-100 (PMS Model Forward Scattering Spectrometer Probe, diameter range 2–47 μm). An ultrasonic anemometer (RM Young Model 81000) was mounted in the centre of the lift platform with its sensor head 3 m above the platform floor. The SMPS sampled air at a rate of 0.3 lpm from an inlet located 1 m below the anemometer at a height of 4 m above the road. It was set to scan across 98 channels in 43 s with a retrace time of 15 s, giving one complete scan across the size range every 60 s. After quality control this data was averaged into 10-min periods. The FSSP's sample head was located 0.5 m above the platform floor on the roadside edge of the platform. The anemometer and FSSP were logged together as an eddy correlation system via a PC with a multi-serial card installed using software developed at UMIST. The anemometer was logged at 20 Hz, and the FSSP at 20 Hz. The third instrument was an ASASP-X, which was located in the trailer. It was logged along with an ultrasonic anemometer (Gill Solent model A1012R). This was fixed on a slender mast so that the sensor head and the copper ASASP-X inlet were 3.5 m above the road, and was left in a fixed position 1.5 m behind the trailer for the duration of the experiment. Data from this system was logged at 20 Hz with aerosol data produced from the ASASP-X at 1 Hz. A chemiluminescence NO_x analyser (Thermo Environmental Instruments Model 42C), which sampled ambient air from a Teflon inlet alongside the Solent anemometer at 3.5 m and was logged at 1 Hz, was also installed in the trailer.

The equipment used included two sources of aerosol that could lead to misleading results. The first source was the diesel-powered platform scissor lift. The lift's engine was switched off as soon as the lift had been raised or lowered. To avoid sampling the cloud left behind from this operation all aerosol data in the 5-min period that began with the engine being started was discarded. Inspection of the aerosol time series indicated that this 5-min period was sufficient to allow for the dispersal of the extra aerosol. The second source was the diesel-powered generator used to power the equipment, located in a parking bay approximately 30 m NW from the instruments. The generator was down-wind of the instruments for most of the experimental period. However, there were occasions when the generator was up-wind of the instruments and had a significant influence on number concentrations of ultra-fine aerosol measured by the SMPS. Thus, data recorded when the generator was up-wind (9% of the data set) has also been discarded.

Wind speed and direction at rooftop height were acquired from the permanent ultrasonic anemometer (Gill Solent, Model Windmaster) on the roof of the UMIST Main Building. This building is 750 m from the subject canyon. It is taller than average for Manchester city centre and is not overlooked, thus providing an indication of wind immediately above the experimental canyon. The wind speeds recorded here are referred to as U_r or rooftop wind speed in this paper. During the experimental period the mean temperature in the canyon was 13°C, the mean rooftop wind speed was 5 m s^{-1} and the wind direction was mostly southerly, especially in week 2. This wind speed and direction are both very typical of Manchester.

PM_{10} and NO_x data from three sites from the UK Automatic Urban Network has also been used: Manchester Town Hall, Manchester Piccadilly and Salford Eccles. Manchester Town Hall site (NO_x only) is on the opposite side of the Town Hall building to the Princess Street canyon, <100 m away, and is 25 m above a quiet street. It is classed as 'Urban Background'. Manchester Piccadilly is 350 m from the Princess Street canyon and is at ground level in an open space with gardens, albeit with busy traffic on one side and a busy bus station on another. It is classed as an 'Urban Centre' site. Salford Eccles is 6 km due west of the SCAR site and is at ground level in a suburban street 100 m from Eccles town centre. Although classed as 'Urban Industrial' it is characteristic of the suburban background in Greater Manchester.

Traffic is regulated by signals at both ends of the canyon leading to three distinct traffic flows adjacent to the instruments: free flow, zero flow (traffic being stopped from entering the canyon) and queues forming from the signals controlling the exit from the canyon. Traffic flow rates were not measured by instruments but have been derived from manual observations. Observation was not continuous, but occurred over blocks of about an hour at various periods throughout the experiment. This data has been averaged and smoothed to produce an idealised diurnal pattern, which shows that traffic flow varied between 30 h^{-1} in the early morning periods to 1100 h^{-1} during the evening peak. In the analysis that follows this idealised traffic data has been applied to each day, and day-to-day variations disregarded. Systematic observations of the split between light- and heavy-duty vehicles were not made. However, two brief periods of observation suggested that heavy-duty vehicles constituted 30–40% of all vehicles between 10:00 and 11:00 while this proportion fell from 7% to 3% between 17:30 and 21:30. This is consistent for such a one-way road that carries more home-bound cars in the evening than city-bound cars in the morning.

A mobile anemometer system was also deployed in three of the SCAR experiments to investigate horizontal variation of turbulence within the canyon. The system employed either a RM Young 81000 or a Gill Solent R2 mounted on a small mast so that the sonic head was 2–4 m above the pavement. After 10 min of recording it was moved 10 m along the pavement, completing a circuit of both sides of the canyon in 3–4 h. More data from this part of SCAR will be presented elsewhere.

3. Results

3.1. Fine aerosol: source and composition

Aerosol size distributions were derived by combining data from the SMPS, ASASP-X and FSSP. The mean aerosol number size distribution observed over the 2-week period at the lowest level (4 m) exhibited a clear mode at 25–30 nm in diameter (see Fig. 2). The 25–30 nm roadside modal diameter, or one very similar, has also been observed in the urban canopy and has been related to motor vehicle (especially petrol) emissions (Harrison et al., 1999; Shi et al., 1999) and is thus consistent with expectations for a receptor only a few metres from busy traffic.

Time and size-resolved aerosol chemical composition measurements were also taken with an aerosol mass spectrometer (AMS) 3 months later at a nearby street canyon site, located 25 m above the street and close to the top of the canyon. Full details are given in Allan et al. (2003). The data indicated that the aerosol sampled in Manchester, in common with other urban areas, had two mass modes. The finer mode covered the diameter

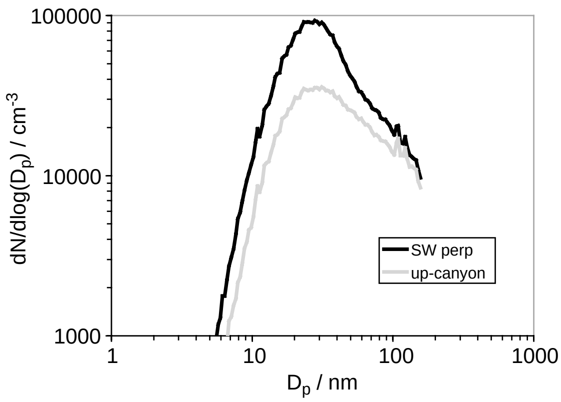

Fig. 2. Number size distributions from the SMPS for perpendicular and parallel wind regimes.

range 30–200 nm (therefore corresponding to the majority of the number of particles) and the volatile fraction was dominated by aliphatic organics representative of unburnt petrol and diesel fuel. Mass concentrations of those particles of diameter <200 nm, which were identified as organic, were strongly correlated with NO_x concentrations measured at the Manchester Town Hall AUN site. This agrees with the findings of Williams et al. (2000), who found that Manchester ambient particle number concentrations in the size range 100–500 nm were linked to traffic activity, and results obtained by Kleeman et al. (2000), in which different engine types produced a mode at 100–200 nm, which consisted mainly of organic carbon (OC). The second (coarser) mass mode was dominated by sulphate and nitrate, which is generally advected into the canyon with occasional contributions from oxygenated organic compounds that have undergone significant atmospheric processing.

3.2. Urban background ultra-fine aerosol number concentrations

Ultrafine aerosol is defined here as aerosol within the range (4.6 nm $< D_p <$ 100 nm) as measured by the SMPS, and shall be referred to as $N_{0.1}$. To establish the enhancement in ultra-fine number concentrations over inner-urban background due to the presence of the canyon, background $N_{0.1}$ concentrations were estimated. The $N_{0.1}$ concentrations measured by the SMPS at 4 m were related to NO_x (in ppb) measured at 3.5 m height by $N_{0.1} = 150\ NO_x$ ($R^2 = 0.62$) within the range of background NO_x concentrations (12–87 ppb) during SCAR-4. This relationship was then used to estimate inner-urban background $N_{0.1}$ from NO_x measurements made at the UK Automatic Urban Network site at Manchester Town Hall. This gave values in the range 4000–9000 cm^{-3} for typical Manchester NO_x which is

Table 1
Typical values of $N_{0.1}$ concentration (Units: cm^{-3})

Flow regime	Range[a]	Max	Typical[b] night	Typical[b] day	Low wind high traffic
Up-canyon	4000–46 000	136 000	7000[c]	15 000[c]	41 000[c]
NE perpendicular	8000–21 000	107 000	Not applicable	Not applicable	Not applicable
SW perpendicular	5500–120 000	188 000	10 000[d]	30 000[d]	80 000[d]

Notes: For comparison, estimated city centre background $N_{0.1}$ is 4–9000 cm^{-3}.
[a] 5th to 95th percentile.
[b] Typical conditions: $U_r = 4\,ms^{-1}$, traffic = 700 h^{-1} (day), 200 h^{-1} (night). Poor conditions: $U_r = 2\,ms^{-1}$, traffic = 1000 h^{-1}.
[c] Based upon parameterisation described in text.
[d] Typical conditions in SW perpendicular: $U = 2\,ms^{-1}$, traffic = 700 h^{-1} (day), 200 h^{-1} (night). Poor conditions: $U = 1\,ms^{-1}$, cross-canyon flow.

comparable to measurements made in Manchester (Williams et al., 2000) and elsewhere (e.g. Väkevä et al., 1999).

3.3. Ultra-fine aerosol number concentrations

$N_{0.1}$ concentrations ranging from 2000 to 190 000 cm^{-3} were measured. The 5th and 95th percentiles were 4500 and 120 000 cm^{-3} respectively, and the mean throughout the experiment was 27 000 cm^{-3}. These results are comparable to other urban canyon measurements (Väkevä et al., 1999; Wehner et al., 2002) (Table 1).

The magnitude of $N_{0.1}$ concentration was strongly influenced by the mean airflow in the canyon, which was principally controlled by the general wind direction. Data from the ultrasonic anemometers indicated that the mean flow pattern within the canyon could be divided into four regimes (see Fig. 1) and the data was split accordingly:

- 'Up-canyon' flow is defined as flow when the wind direction above the canyon is within $\pm 40°$ of the canyon axis, blowing against the traffic flow. In such cases, the wind was channelled along the canyon with very little vertical component.
- 'NE perpendicular' occurred when the wind approached the canyon over the shorter NE wall. There was only limited data in this regime.
- 'SW perpendicular' occurred when the wind approached over the taller wall. Although there was some evidence of vortex flow it appeared that this vortex might not always penetrate the full depth of the canyon.
- 'Down-canyon' winds did not occur during SCAR-4.

In 'up-canyon' flow, $N_{0.1}$ was related to traffic flow rate and rooftop wind speed, U_r. The data could be reproduced with the following parameterisation:

$$N_{0.1} = 92TU_r^{-1.45} + (4.2T + 3300) \quad R^2 = 0.745,$$

where T = traffic flow rate (vehicles $hour^{-1}$) and the term in brackets gives an estimate of background concentration. The parameterisation under-predicts concentrations for those periods immediately preceded by a perpendicular flow regime.

All of the high concentrations occurred during periods when the wind was approaching the canyon from a southwesterly direction ('SW perpendicular', see Fig. 1) over the taller canyon wall. In this regime the concentrations were acutely dependent upon the wind direction at street level. When the wind direction at street level was channelled up-canyon to within 20° of the canyon axis the maximum $N_{0.1}$ concentration recorded was 54 000 cm^{-3}. This occurred more often when the wind direction above the canyon was between 40° and 60° to the canyon axis.

However, within the canyon, down- and cross-canyon flow were also occasionally observed. As noted above, down-canyon data was discarded due to contamination from the generator. Cross-canyon flow (between 30° and 90° to the canyon axis, 26% of the retained data in SW perpendicular) meant that air was being blown from the roadway directly to the instruments over a distance of a few metres. Also, in cross-canyon flow wind speed was restricted to below 1.5 $m\,s^{-1}$. The combination of poor dispersion in low winds, and also the shorter distance from source to receptor over which dilution may occur, led to very high $N_{0.1}$ concentrations (in the range 30 000–190 000 cm^{-3}). The relative importance of weak dispersion and limited dilution could not be distinguished with this small data set. The SMPS size distributions illustrated that the extra particles were predominantly of a diameter below 60 nm. (see Fig. 2).

What decided whether the in-canyon flow was channelled or cross-canyon could not be clearly determined. However, some insight into the mean flow within the canyon can be gained from data recorded by the mobile turbulence system. This system gave 10-min averages of wind speed and direction at pavement locations during several daytime periods. Seven of those periods were during SW perpendicular flow. This data illustrated how, as the wind direction above the canyon exceeds 50° to the canyon axis, inflow is seen at the NW (down-wind) end of the canyon and meets the up-canyon flow at some point along the canyon length. At this point cross-canyon flow is more likely to occur. This point appears to move towards the upwind end of the canyon as the wind direction becomes more perpendicular. Care must be taken with this interpretation, as the data for the various points is not simultaneous; however, it may indicate how the variation of in-canyon wind direction occurs. It also indicates how inflow from the down-wind end of the canyon appears to be important. This aspect of canyon flow may be influenced by the large open square beyond this end of the canyon. In general, cross-canyon flow occurred more often than up-channel flow when the approach wind direction exceeded 240° (60° to the canyon axis).

The frequency distribution of $N_{0.1}$ concentrations showed how unrepresentative the mean concentration can be, especially if considering the health effects of brief excursions in particle numbers. In week 1, which had an even balance of parallel and perpendicular flow, the 99th percentile concentration was 3.7 times the mean. However, if one considers the raw 1-min data then the 99th percentile was 6.2 times the mean.

In SW perpendicular flow $N_{0.1}$ concentrations could not be related to roof-level wind speed. However, it was also found that wind speed within the canyon was not simply related to wind speed at roof level, suggesting that airflow at street level became isolated from the flow above in this regime. There was a weak inverse relationship between $N_{0.1}$ and in-canyon wind speed, U (see Fig. 3).

$N_{0.1}$ was also positively related to traffic flow rate when cross-canyon flow periods were discarded, although this simple pattern was broken by high concentrations during the period between the morning and evening traffic peaks. This seems to indicate that the ultra-fine aerosol released into the canyon atmosphere

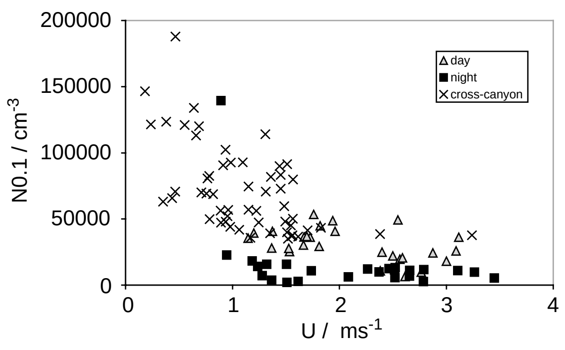

Fig. 3. Dependence of $N_{0.1}$ upon in-canyon wind speed in SW perpendicular flow.

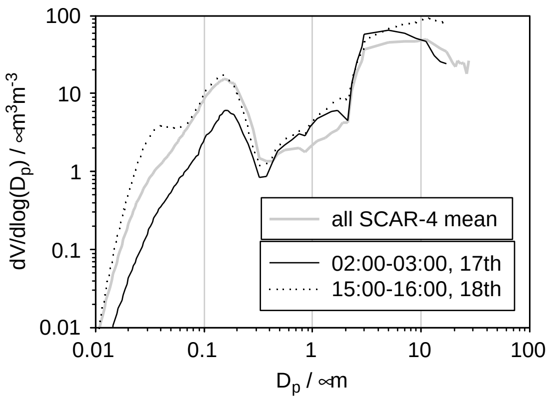

Fig. 4. Mean roadside aerosol mass size distribution. Curve for 17th represents low emission, high dispersion conditions and 18th represents high emissions and low dispersion conditions.

during the morning rush hour remains suspended once the traffic flow has reduced to a daytime plateau, and is slow to be dispersed. However, as the traffic data is a smoothed 'idealised' cycle (see above), higher than-predicted traffic flows during this mid-day period cannot be ruled out. However, this effect is only clearly seen in SW perpendicular flow indicating the importance of vortex re-circulation within the sheltering lee of the tall canyon wall. A similar effect is seen in the coarse aerosol data, as discussed below.

3.4. *Mass concentrations*

The number concentrations measured with the SMPS, ASASP-X and FSSP were used to estimate mass size distributions, by assuming all measured particles were perfect spheres of density $1000\,\mathrm{kg\,m^{-3}}$. The full, mean aerosol mass size distribution for the whole of SCAR-4 is presented in Fig. 4. Three persistent mass modes are identifiable: a fine mode ($D_p < 0.5\,\mu m$, incorporating the ultra-fine number mode) and a coarse mode ($D_p > 2\,\mu m$). Fig. 4 also shows two periods of opposing conditions, which bracket all other conditions in week 1. On the 17th, 02:00–03:00 was a period of up-canyon flow with moderately high winds in the canyon, and low traffic, while 15:00–16:00 on the 18th had SW perpendicular flow with low wind speeds and higher traffic. The effect of the extra ultra-fine particles can clearly be seen.

3.5. *Fine mode*

The fine mass mode concentration, $PM_{0.5}$, was related to $(N_{0.1})^{0.5}$ and was consequently largely controlled by diurnal variation in traffic-source emission, wind-driven dispersion and the incident wind direction, although the large excursions in $N_{0.1}$ had a smaller effect on $PM_{0.5}$.

Daytime values tended to 5–12 $\mu g\,m^{-3}$ with moderate to high winds. Elevated values were seen principally in SW perpendicular winds and when wind speed was low ($<3\,m\,s^{-1}$).

3.6. Coarse mode

The coarse mode particle mass concentrations were derived from the FSSP ($2\,\mu m < D_p < 10\,\mu m$), and will be referred to as PM_{coarse}. A good correlation was found between PM_{coarse} and the mass concentration measured by the ASASP-X data in the range $1.0 < D_p < 2.5\,\mu m$, or $PM_{2.5-1}$:

$$PM_{coarse} = 30(PM_{2.5-1})^2 \ (R^2 = 0.8).$$

Below 0.8 μm the correlation breaks down. This relationship was used to estimate PM_{coarse} for those periods when FSSP data was not available. Combining the estimated and measured PM_{coarse} it is possible to see that during week 1 PM_{coarse} generally followed the PM_{10} concentrations measured at the Manchester Piccadilly AUN monitoring site. SCAR PM_{coarse} exceeded Piccadilly PM_{10} for at least 12 h following a high-wind event on the afternoon of 16 October, a period during which up-canyon flow and medium-to-high wind speeds led to low fine aerosol concentrations (see Fig. 5).

The sudden rise in PM_{coarse} that evening was slightly delayed in comparison to the rise in wind speed and rise in local PM_{10}, but the rise did coincide with the veering of the wind from a SW perpendicular approach to an up-canyon approach. It seems possible that the SW perpendicular flow partially and temporarily isolated the canyon from the newly suspended coarse particles. However, it is also possible that the PM_{coarse} concentrations did not rise until the wind speed had fallen and dispersion had reduced. Shortly after the evening traffic peak, wind speed was reduced and PM_{10} fell quickly at the relatively open gardens site at Piccadilly (from 91 to 21 $\mu g\,m^{-3}$ in the first hour). At the SCAR site PM_{coarse}, however, fell much more slowly following an exponential decay with a half-life of 7.6 h. As the SCAR instruments were placed much closer to moving vehicles than those at the Network site at Piccadilly, the reduced decay could be related to traffic-induced turbulence keeping the particles suspended, or re-suspending them, as well as re-circulation in the canyon. Two other periods of exponential decay in PM_{coarse} were identified, on the afternoons of 16 and 23 October, with half-lives of 3.4 and 4.3 h, respectively. As the traffic flow rate was similar or higher at these times, these faster decays may be related to the fact that in-canyon wind speeds were lower (an average $1.5\,m\,s^{-1}$ on both of these other occasions compared to $2.2\,m\,s^{-1}$ following the high wind event on the 16th). This observation suggests that whereas Harrison et al. (2001) declare that periods of raised coarse-only concentrations brought about by wind-driven re-suspension are relatively rare compared to coarse-and-fine high pollution episodes, when such events do occur the duration of the high concentrations could be prolonged within urban canyons. A similar peak in wind speed on the early morning of the 18th seems to have had less of an effect due to precipitation scavenging that night.

In week 2, FSSP data was not available, so all PM_{coarse} data is estimated as described above. PM_{10} concentrations at Piccadilly were generally higher than they had been in week 1, but PM_{coarse} was much lower (a mean of 4 $\mu g\,m^{-3}$, compared to 19 $\mu g\,m^{-3}$ for week 1). This may be explained by noting that during week 2, flow was almost exclusively SW perpendicular, partially isolating the canyon from the extra-canyon atmosphere. There was no significant difference in wind speeds between weeks 1 and 2 (means and 95th percentiles were within $0.1\,m\,s^{-1}$). PM_{coarse} has a clear diurnal pattern in

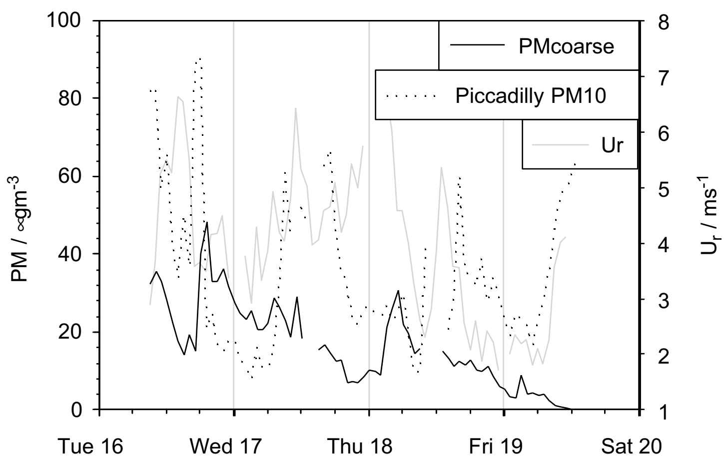

Fig. 5. Coarse mode mass concentrations and urban centre PM_{10}, week 1.

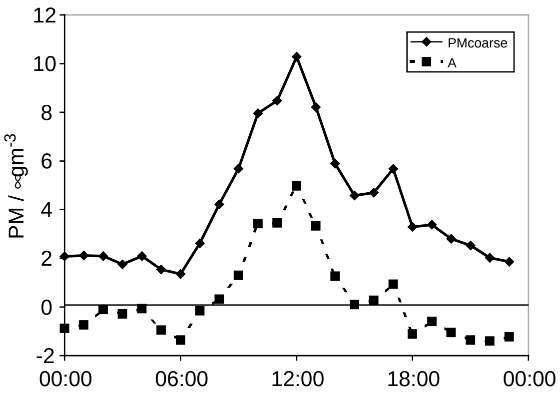

Fig. 6. Diurnal means of estimated PM_{coarse} and intercept A (related to traffic-induced re-suspension at canyon site, see text) from week 2.

week 2, rising above 5 μg m^{-3} between 09:00 and 15:00 and again between 17:00 and 17:30, peaking around 12:00 at approximately 10 μg m^{-3} (see Fig. 6). During this period wind speed also repeated its diurnal pattern, with remarkably little day-to-day variation, but did not peak until around 16:00. The pattern in PM_{coarse} could be interpreted by assuming that coarse particles are first suspended by the traffic in the morning peak and kept suspended by vehicle-induced turbulence within the canyon, as well as some input of extra wind-driven re-suspension from outside the canyon as wind speeds rise. When week 2 SCAR PM_{coarse} is compared to the urban background AUN site of Salford Eccles a closer correlation is found than with Piccadilly that varies during the course of the day:

$$\text{SCAR } PM_{coarse} = 0.24 \times \text{Eccles } PM_{10} + A,$$

where A varies between -1.4 and 5 during the day (see Fig. 6). The Eccles site is within a town centre but located away from busy traffic, and therefore less likely to affected by local (street-scale) variations than Piccadilly. The extra 0–5 μg m^{-3} seen at SCAR during the day can be at least partly related to the extra traffic-related coarse concentration within the Princess Street canyon. The very low PM_{coarse} concentrations seen at various times throughout SCAR-4 suggest either the absence of any other significant sources of coarse particles (such as direct emission from natural sources) or isolation of the canyon from them.

3.7. Mass concentrations: general

These two major mass modes (fine and coarse) varied independently throughout the 2 weeks of SCAR-4 so that their relative contributions to PM_{10} varied also, mainly as a result of large temporary increases in the coarse mode fraction in week 1. Week 2, on the other hand, was characterised by vortex flow and the fine mode became the dominant mode. The linear correlation between $PM_{2.5}$ and ultra-fine number concentrations ($N_{0.1}$) was not strong ($R^2 = 0.505$), but was greatly improved when a power law was used:

$$N_{0.1} = 304(PM_{2.5})^2 \quad (R^2 = 0.709).$$

This relationship could be improved again by separating data by flow regime:

$$N_{0.1} = \begin{cases} 378(PM_{2.5})^{1.7} \quad (R^2 = 0.855) \text{ in up-canyon flow,} \\ 513(PM_{2.5})^{1.9} \quad (R^2 = 0.680) \text{ in SW perpendicular flow.} \end{cases}$$

However, these relationships are based on less than two full weeks of data and may not apply generally.

4. Conclusions

Compared to the urban background, both fine and coarse mode mass concentrations, and particle mass and particle number concentrations, were found to vary independently in an urban street canyon. Ultra-fine particle concentrations were increased by the presence of the canyon, but were also particularly dependent upon the incident wind direction with strong evidence of raised concentrations on the taller lee-side of the canyon up to at least 0.6 × wall height. Concentrations were also inversely related to local wind speed and directly or exponentially related to traffic flow rate. In 'SW perpendicular' flow, when the wind approached the canyon over the taller wall, ultra-fine aerosol concentrations were inversely related to wind speed within the canyon which was constrained when cross-canyon flow occurred rather than channelling. In-canyon wind speed could not be simply related to roof-level wind speed. $N_{0.1}$ concentrations of 30 000–150 000 cm^{-3} were typical when the vortex blew directly from the road to the instruments.

Coarse mode mass concentrations generally followed the nearby local background PM_{10} concentrations, with traffic within the canyon contributing an extra 0–5 μg m^{-3} during the daytime period. However, when wind speeds were higher than 6 m s^{-1} a high concentration episode was brought about by enhanced re-suspension. The canyon appeared to trap coarse particles so that they remained suspended and concentrations remained high relative to local PM_{10}, decaying exponentially until concentrations in the urban background again rose above in-canyon levels. The rate of decay was related to in-canyon wind speed. Perpendicular flow regimes may have partially isolated the canyon restricting transport of coarse particles into or out of the canyon. This needs further verification. Mass concentration of particles in the range

$0.8\,\mu m < D_p < 2.5\,\mu m$ measured by the ASASP-X were found to be correlated with mass concentration of particles in the range $2\,\mu m < D_p < 10\,\mu m$ indicating that wind-driven re-suspension was at least as significant as wind-driven dispersion in the transport of particles above $0.8\,\mu m$ diameter.

SCAR generated a database of turbulence measurements, including determination of fluxes in the size range $0.1 < D_p < 32\,\mu m$ by eddy correlation, which will be used to support and expand the analysis of this work which will be presented in future papers.

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