Comparative study of measured and modelled number concentrations of nanoparticles in an urban street canyon

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Abstract

This study presents a comparison between measured and modelled particle number concentrations (PNCs) in the 10–300 nm size range at different heights in a canyon. The PNCs were modelled using a simple modelling approach (modified Box model, including vertical variation), an Operational Street Pollution Model (OSPM) and Computational Fluid Dynamics (CFD) code FLUENT. All models disregarded any particle dynamics. CFD simulations have been carried out in a simplified geometry of the selected street canyon. Four different sizes of emission sources have been used in the CFD simulations to assess the effect of source size on mean PNC distributions in the street canyon.

The measured PNCs were between a factor of two and three of those from the three models suggesting that if the model inputs are chosen carefully, even a simplified approach can predict the PNCs as well as more complex models. CFD simulations showed that selection of the source size was critical to determine PNC distributions. A source size scaling the vehicle dimensions was found to better represent the measured PNC profiles in the lowest part of the canyon. The OSPM and Box model produced similar shapes of PNC profile across the entire height of the canyon, showing a well-mixed region up to first ≈ 2 m and then decreasing PNCs with increased height. The CFD profiles do correctly reproduce the increase from road level to a height of ≈ 2 m; however, do not predict the measured PNC decrease higher in the canyon. The PNC differences were largest between idealised (CFD and Box) and operational (OSPM) models at upper sampling heights; these were attributed to weaker exchange of air between street and roof-above in the upper part of the canyon in the CFD calculations. Possible reasons for these discrepancies are given.

Keywords: Dispersion; Modelling; Nanoparticles; Particle number concentration; Street canyon

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1. Introduction

The introduction of stricter emission standards, cleaner fuels and better emission control technology has decreased the particle mass emissions from diesel-engined vehicles but may have increased the particle number emissions because of lower available particle surface area favouring nucleation over adsorption (Kittelson, 1998). This will also lead to a shift of size distributions towards smaller size ranges as discussed by Cheng et al. (2008). The ultrafine particles (those below 100 nm), which are not explicitly the part of current regulatory limits, contribute significantly to particle number concentrations (PNC) but little to particle mass concentrations (PMC) in the ambient environment (Jones and Harrison, 2006; Kumar et al., 2008a–e). Recent toxicological and epidemiological studies suggest strong correlations between adverse health effects and exposure to ambient ultrafine particles at high number concentrations (Oberdorster, 2000; Peters and Wichmann, 2001; Pope III and Dockery, 2006; Brugge et al., 2007). This indicates the need to design effective mitigation strategies to regulate the particles on a number basis in urban areas. The lack of standard methods and instrumentation for particle number measurements, and the detailed understanding of the influence exerted on particle dispersion by ambient meteorology and traffic volume have limited the scope for accurate modelling of particles on number basis in urban areas.

Several simple to complex models are currently available for the dispersion of particles in the urban environment. These include simple Box models, Gaussian models, Computational Fluid Dynamics (CFD) models, Lagrangian / Eulerian models, and models that include particle dynamics. A review of these models can be seen in Holmes and Morawska (2006) and Vardoulakis et al. (2003). Validation studies for particle numbers are not abundantly available. Many models are suitable for the prediction of PMCs and gaseous pollutants in urban

environments, but few are appropriate for the prediction of PNCs. Also, as with PMC models, there are many practical constraints related to the use of PNC models which require a great amount of input information (i.e., emission factors, meteorology, local traffic and the geometry of the site, etc.) rarely available in detail for routine applications. For example, the mass emission factors for various type of vehicles under a range of driving conditions are important input parameters for a street canyon model, but less information is available on a number basis. Moreover, the prediction of particles on a number basis becomes more complicated when particle dynamics modules for various transformation processes are incorporated into the models. This requires more detailed input information. Lohmeyer (2001) reported that predictions of various pollutants from different models can vary by up to a factor of four for identical conditions, depending on the quality of the input information.

This study presents a comparison between measured and modelled PNCs at different heights of a canyon. The PNCs were predicted using a simple modelling approach (a modified Box model), an Operational Street Pollution Model (OSPM) and a CFD code, FLUENT. Every model disregarded particle dynamics. The modified Box model combined a simple Box model with modules for vertical PNCs and the regimes for traffic and wind produced turbulence. The CFD simulations were carried out by assuming a simplified geometry of one of our previously studied streets (Pembroke Street) in Cambridge, UK (Kumar et al., 2008b, c). Four different sizes of emission sources were used in CFD simulations. This allowed the study of the effect of the size of the emission sources due to rapid mixing in the immediate vicinity of a vehicle on the mean PNC distributions. The vertical PNC profiles were produced for both sides (leeward and windward) of the canyon using various models; these are also discussed and compared with the measured vertical PNC profiles.

2. Methodology

2.1 Site description, instrumentation and measurements

Measurements were carried out in Pembroke Street (Cambridge, UK; 52°12' N and 0°10' E), just outside the Chemical Engineering Department building. The studied section is ≈ 167 m long (L), and has height (H) to width (W) ratio of approximately unity ($H = W = 11.6$ m). The orientation of the street canyon is southwest (SW) – northeast (NE), and this has one-way traffic travelling from SW to NE, as seen in Fig. 1.

A fast response differential mobility spectrometer (DMS500) was used to measure the particle number distributions (PNDs) in the 5–2738 nm size range at a sampling frequency of 0.5 Hz, rather than the maximal sampling frequency 10 Hz, to improve the signal-to-noise ratio for achieving the maximal quality of data. In this article, the PNCs in the 10–300 nm range were only considered for analysis. Particles below 10 nm were not used, because of their significant losses in the sampling tubes (Kumar et al., 2008b, e); also particles above 300 nm were disregarded, as their proportion was negligible ($<1\%$) compared to total PNCs in the ambient environment (Kumar et al., 2008a–d).

Meteorological parameters (wind speed, wind direction, temperature and pressure) are measured at 16.6 m above the road level. Traffic volumes were taken manually, and by a movement-sensitive CCTV camera. Measurements were made continuously for 24 h a day between 7 and 23 March 2007 for 17 days. The particle measurements were taken at 1.60 m above the road level on all days except 24 h (between 20 and 21 March 2007) when these were taken pseudo-simultaneously at four heights (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); results of these measurements are presented elsewhere (Kumar et al., 2008b–c). This 24 h data were selected for comparison with modelled results.

These data represented such a period when the wind direction was across the canyon (i.e., NW) and wind speeds were well above 1.5 m s^{-1} i.e., wind-produced turbulence was likely to dominate traffic-produced turbulence (Kumar et al., 2008c; Solazzo et al., 2007). Moreover, these measurements were taken at four heights in the canyon, providing opportunity to compare with the modelled vertical PNC profiles using various models, explained in the next section. Note that the sampling points were on the leeward side of the canyon and no measurements were available for the windward side (Fig. 1). The range of air temperature (T_a) and U_r during the measurements were between -1.2 and $8 \text{ }^{\circ}\text{C}$ and between 2.42 and 4.30 m s^{-1} , respectively. Further information on measurements of PNCs, traffic volume and meteorological data, together with a schematic diagram of the studied canyon and the sampling positions are presented elsewhere (Kumar et al., 2008b–c).

3. Descriptions of models used

3.1 The modified Box model

A simple modelling approach, combining a Box model with modules for vertical PNC variation and the regimes for traffic and wind dependent PNCs, is used to predict the PNCs in selected street canyon. The formulation of a Box model assumes that the selected stretch of road is longitudinally homogeneous and that the source of the particles due to traffic emissions within the canyon and the removal of particles due to exchange with background from the canyon top must be equal apart from any deposition and gravitational settling losses which are considered to be negligible. Furthermore, our recent study (Kumar et al., 2008c) demarcated traffic and wind dependent PNC regions depending on the above-roof wind speed (U_r). These results were included in this model assuming that in the traffic-dependent PNC region (when $U_r \ll U_{r,\text{crit}}$),

the PNCs were approximately constant and independent of U_r up to a critical value of cut-off wind speed ($U_{r,crit}$). In the wind-dependent PNC region (when $U_r \gg U_{r,crit}$), the PNCs are inversely dependent on U_r . The $U_{r,crit}$ is defined as the U_r which separates the regions of traffic and wind dependent PNCs. In addition, the vertical concentration profiles, showing an exponential PNC decay with height above the height of a well-mixed region close to the road level, has been incorporated in to this model. Details of the model formulation are provided in supplementary Section S.1. The final expression for the leeward side of the canyon, as seen in Eq. (S-8), is:

$$C = \frac{\sum_{x=1}^n E_{x,i-j} T_x}{b_1 U_r W} \exp(-k_1 z) + C_b \quad (1)$$

where $z = \max(z, h_0)$, $U_r = \max(U_r, U_{r,crit})$, and $k_1 = 0.11 \text{ m}^{-1}$

In Eq. (1), C and C_b are the predicted and background PNCs ($\# \text{ cm}^{-3}$), U_r and $U_{r,crit}$ are in cm s^{-1} , k_1 is exponential decay coefficient in cm^{-1} , b_1 ($= 0.013$) is an empirical constant, h_0 ($= 2 \text{ m}$) is assumed height of the well-mixed region close to road level, $E_{x,i-j}$ is the particle number emission factor in $\# \text{ veh}^{-1} \text{ cm}^{-1}$ in any particle size range ($i-j$) of any vehicle class x , T_x is the number of vehicles per second of a certain class, W is the width of the canyon in cm, and z is vertical height in cm above the road level in the canyon. The empirical constant b_1 is replaced with b_2 ($= 3.58 b_1$) to predict the PNCs in the windward side of the canyon. The PNCs are assumed constant at all heights in this side of the canyon and k_1 is assumed to be zero (refer Section S.1 for details).

Our fast response measurements in the vehicle wake (Kumar et al., 2007) and street canyon (Kumar et al., 2008b–d) showed that the dilution was very fast in the vehicle wake and the effect of transformation processes was generally complete by the time particles were

measured at road side. Considering this, particle dynamics have been ignored and total particle numbers are assumed to be conserved for Box model and other models described in Sections 3.2 and 3.3.

3.2 The Operational Street Pollution Model (OSPM)

The OSPM, which contains a simplified empirical description of flow and dispersion conditions for urban street canyons, has been deployed to predict the PNCs at the different receptor heights on the leeward and windward side of selected street canyon. The OSPM estimates the concentrations of pollutants using a combination of a plume model for the direct contribution and a box model for the re-circulating pollution part in the street canyon. In OSPM, the turbulence in the street canyon is modelled by taking into account the effect of atmospheric turbulence produced by wind shear and the traffic-produced turbulence by vehicles. The latter dominates the mixing during low and calm wind conditions. A detailed description of the OSPM can be seen in Berkowicz (2000) and at *ospm.dmu.dk*.

3.3 Computational fluid dynamics (CFD) simulations

A CFD code, FLUENT, is used to predict the dispersion of PNCs in a street canyon. FLUENT is a multipurpose commercial CFD software, and has widely been used to model flow and dispersion in urban applications (Di Sabatino et al., 2007; Garmory et al., 2008; Hamlyn and Britter, 2005a; Lien et al., 2004; Solazzo and Britter, 2007a). The flow field was calculated using steady Reynolds Averaged Navier Stokes (RANS) with the standard $\kappa - \varepsilon$ turbulence model (κ is turbulent kinetic energy and ε is dissipation rate of kinetic energy) with model constants $C_{1\varepsilon} = 1.44$ and $C_{2\varepsilon} = 1.92$, has been deployed for the simulations of flow and turbulence distributions (Hassan and Crowther, 1998; Richards and Hoxey, 1993). The

dispersion of the particles was simulated with the User Defined Scalar (UDS) option in FLUENT. An advection–turbulent diffusion equation was solved using the mean velocity field from the $\kappa - \varepsilon$ model and with a turbulent Schmidt number set to unity (i.e., the turbulent diffusivity was set equal to the effective kinematic viscosity, also calculated by the $\kappa - \varepsilon$ model).

3.3.1 Domain

The canyon has been modelled as an infinitely long canyon for a cross–wind condition. This allows us to use a two–dimensional (2D) domain as shown in Fig. 1. The height of the domain from the street level to domain top was set equal to $6H$; this was sufficiently far above the canyon that its effect is negligible. The domain inflow and outflow length was set equal to $5H$. This configuration was selected as this provides enough length in the upstream region to develop the boundary layer (Sini et al., 1996). A similar domain was used by Solazzo and Britter (2007a). This domain contained a total of 53824 grid cells. The smallest grid size was 0.002 m close to walls. The grid size was increasing with distance from the wall, using an expansion factor equal to 1.10, near street walls, floor and the roof (Kim and Baik, 2004). There were a total of 117 nodes up the wall and similar number of nodes across the width of street. The roughness (z_0) of all the walls was set equal to 0.10 m.

3.3.2 Boundary conditions

A uniform velocity profile was set as a boundary condition at the inlet. The turbulent kinetic energy (k) profile at inlet was set equal to IU_r^2 (Kim and Baik, 2004); where I is the turbulent intensity and set equal to 0.1 and U_r is the wind velocity at inlet. The turbulent dissipation (ε) profile at inlet was set equal to $\varepsilon(z) = C_\mu^{0.75} k^{1.5} \kappa^{-1} z^{-1}$ (Richards and Hoxey, 1993); where $C_\mu = 0.09$, $\kappa = 0.40$, and z is the height above the canyon. A symmetry condition is assumed at the top of the flow domain; no-slip conditions are considered at the side walls, street floor and roof in the upstream and downstream region of the domain. A background concentration was set at the inlet and all points in the grid at the inlet.

3.3.3 Emission Source

There is no standard practice to assign the size of an emission source in CFD simulations. Several CFD studies for street canyon simulations have used various types of sources to simulate the traffic conditions. These may be a point source (Walton and Cheng, 2002), a line source (Baker et al., 2004; Garmory et al., 2008) or an area source (Baker et al., 2004; Park et al., 2004). Of these studies Garmory et al. (2008) and Park et al. (2004) use a 2D representation of an infinitely long canyon whereas Walton and Cheng (2002) and Baker et al. (2004) use a 3D domain. In order to assess the effect of source size on simulated results, in this study we use a 2D domain to simulate an infinitely long canyon and use four different sizes of finite cross section line emission sources with constant discharge on the centre-line of the canyon. All sources are located 0.20 m above the road level to simulate the height of the exhaust pipe. Despite the small direct source area (opening of the exhaust pipe), the emission sources

should be associated with a larger area in the model taking into account the dilution and mixing immediately downstream of the rear of the vehicles that are not present in the CFD model. The descriptions of sources are as follows:

- A smallest emission source with 0.53 m width $\times$ 0.11 m height (hereafter referred to as CFD_S_a), that approximates a finite cross section line source similar to the one used in several other CFD studies (Baker et al., 2004; Garmory et al., 2008).
- A largest emission source with 5.08 m width $\times$ 1.98 m height (hereafter referred to as CFD_S_b), approximating the width of the traffic-lanes and height of vehicles. This was selected to take in to account a maximal initial dispersion due to the rapid mixing in the wake of the vehicle.
- Two intermediate size sources with 1 m width $\times$ 0.75 m height (hereafter referred to as CFD_S_c) and 2 m width $\times$ 1.5 m height (hereafter referred to as CFD_S_d) are also selected. Yasuda et al. (2007) showed in their large eddy simulations for flow and dispersion in the vehicle wake that due to traffic-produced turbulence vertical plume height at the rear end of a vehicle is of the range 0.5–1.0 vehicle height. We used both the extreme cases for selecting a source area by assuming that average vehicle width and height are about 2 m and 1.5 m, respectively.

It should be noted that the sources CFD_S_b, CFD_S_c and CFD_S_d simulate the rapid dilution (in the region of the source) just after the rear end of the vehicle, but not the effect of traffic-produced turbulence in the rest of the vehicle wake as, for simplicity, there is no extra turbulence source added to the CFD simulation.

3.3.4 Simulations

Twenty four sets of simulations (one simulation for each selected hour) were carried out for each source. This 24 h data was selected from the measurement campaign presented in Kumar et al. (2008b). During this period winds were across the canyon (between 296°N and 337°N). The Reynolds number ($Re = U_r H / \nu$, where ν is the kinematic viscosity of the air) for this period varied between 2.1×10^6 and 3.7×10^6 . The density and viscosity of the ambient air were calculated based on the assumed uniform ambient air temperature, and these were changed for each simulation.

The estimated emission factor $1.33 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$ (as discussed in supplementary Section S.2) is used to estimate the emission source strength (S). This changed for each hour depending on the source area and traffic volume (T) that varied between 140 and 1192 veh h^{-1} during the measurements. Table 1 shows the input parameters used for different sets of simulations. Each set of simulation took $\approx 26,000$ iterations to converge solution to residual values of k , ϵ , x and y velocity and concentrations to 10^{-6} . Initially, the FLUENT model was run until the flow field converged, with no emissions, to establish the turbulent flow fields within the modelled domain and primary vortex within the canyon sub-domain. After this, a constant emission source of inert particles was introduced through the specified source area and the calculations re-started until the solution for concentrations converged.

4. Results and Discussion

4.1 Flow and turbulence distributions

Fig. 2 shows the velocity and turbulence distribution in the selected geometry of the street canyon from the CFD simulations. The mean velocity vectors show an expected primary canyon–vortex and small recirculation zones at the bottom corners of the street canyon (Fig. 2a). Further, Fig. 2b shows the distribution of turbulent kinetic energy (TKE), which also shows the production of TKE in the shear layer at the top of the canyon as well as around the separation region at the top of the windward wall. This TKE is then dissipated as it is swept round the canyon by the primary vortex. Different sizes of emission sources are used and their effect on PNC distributions is discussed in subsequent section.

4.2 Effect of source size on PNCs in CFD simulations

The effect of different source sizes on the PNC distribution has been presented in Fig. 3 for one of the 24 modelled cases (No. 1), and this shows the advection of PNCs from the sources to the leeward side of the canyon. However, the PNCs appear to vary with the change in height and width of the source. For example, in case of smallest source CFD_S_a the bottom corner of the canyon and the region near to the street wall up to ≈ 0.5 m in the leeward side showed the largest concentrations (Fig. 3a). Conversely, in other cases with larger source areas, the particles first accumulate on the upper–leeward side corner of the source where the concentrations are the largest, and then advected upwards on the leeward side by the canyon vortex (Figs. 3b–d), showing relatively smaller concentrations near the road level and the leeward side wall. Interestingly, the effect of source size on the PNCs in the windward side of the canyon seems to

be modest up to a distance ≈ 0.5 m from the wall as the PNCs were the same to within 5% at all heights for all cases (Figs. 3a–d).

Vertical PNC profiles are drawn at distances (w) 0.40, 1.50 and 2.50 m away from both sides of the canyon walls (referred to as $w/H = 0.034$, 0.13 and 0.22, respectively) (Fig. 4). These profiles covered the width of the pedestrian path along both sides of the traffic-lane where the pedestrians are most likely to be exposed to the traffic pollution. The PNCs are normalised (C^*) using Eq. (2) (Ketzel et al., 2001), and these are plotted against the normalised height (z/H) of the canyon for each CFD case in Fig. 4.

$$C^* = \frac{(C_{total} - C_b)U_r L}{E} \quad (2)$$

Where L is the scaling length usually the height or the width of the street canyon and E is the emission flux per unit length in $\# \text{ m}^{-1} \text{ s}^{-1}$. The variability in vertical concentration profiles for various source sizes at different distances suggests that the selection of an appropriate source size is important for CFD simulations (Fig. 4). Interestingly, vertical profiles for the two largest sources CFD_S_b and CFD_S_d are nearly identical in the leeward and windward side of the canyon (Figs. 4a–f), suggesting that after a certain height and width of a source (which could be the cross-sectional area of a vehicle) further increase in source size does not change the vertical PNC profiles appreciably.

Unlike the leeward side, concentration profiles taken at various positions in the windward side of the canyon (Figs. 4b, d and f) show a similar trend with a consistent increase in concentrations with increasing distance from the windward wall. The difference in vertical PNC profiles was the smallest at 0.40 m (Fig. 4b). This suggests that the effect of source size is minimal on the PNCs in first ≈ 0.50 m near the windward wall. This could be due to the inflow

1 of cleaner air from the top of the canyon close to the windward wall that is slightly decoupled
2 from the higher concentrations in the middle of the canyon.

3 The shapes of the vertical PNC profiles at various distances in the leeward side of the
4 canyon are more complex (Figs. 4a, c and e). For example, between $z/H = 0.3$ and 0.7 , as the
5 distance from the leeward side wall increases from 0.40 m to 2.5 m, the PNCs increase for
6 CFD_S_b and CFD_S_d (the largest sources by area) but decrease for the smallest source CFD_S_a
7 and is constant for the source CFD_S_c. Interestingly, the vertical PNC profiles were identical for
8 CFD_S_b, CFD_S_c and CFD_S_d at 0.40 m (Fig. 4a), suggesting that the effect of source size is
9 negligible on vertical PNC profiles in the first ≈ 0.50 m near to the leeward side wall. However,
10 the profile for CFD_S_a is different, with average PNCs being $\approx 18\%$ larger than others; this is
11 due to the emission of particles through a smaller area near to road level and their advection
12 very close to the wall, as is also shown in Fig. 3a.

13 Furthermore, the shapes of vertical PNC profiles are different than generally be
14 expected, that is decreasing with height. The size of the source, especially the height, seems to
15 play a critical role in determining the shapes of these profiles. The PNCs increase from the road
16 level to a certain height and then decreases with height and eventually for some cases increases
17 again towards the roof-height (Figs. 4a, c and e). The height, where the maximum of PNC
18 occurs, could be related to the height of various sources used. As marked in Figs. 3a–d, these
19 heights are about 0.3 , 2.2 , 0.9 and 1.7 m above road level for CFD_S_a, CFD_S_b, CFD_S_c and
20 CFD_S_d, respectively. It should be noted that this includes 0.20 m that is the height between the
21 lowest edges of the sources and the road level.

22 As seen in Fig. 3, the PNCs are uniformly emitted throughout the source area and then
23 advected by the canyon-vortex towards the upper leeward side corner of the source where the

maximum PNCs are seen, and then these decrease towards the road and roof-top level of the canyon. The two smallest sources by area (i.e., CFD_S_a and CFD_S_c, height 0.11 and 0.75 m, respectively) emit particles close to the ground where they are then swept around the edge of the canyon leaving a relatively low concentration in the centre, which leads to concave vertical profiles as seen in Fig. 4. The other two sources emit the particles at larger heights (1.5 and 2 m source heights) for them to be swept in to the centre of the canyon leading the convex vertical profiles observed. However, measurement studies and different models show different vertical profiles and these details are discussed in the next section.

4.3 Comparison of vertical PNC profiles

The turbulence from the moving traffic will scale on the traffic speed and the turbulence from the wind will be linked to the wind speed. In either the traffic produced or wind-shear produced turbulence cases, the mixing close to the source will be determined mainly by the flow around the vehicle. This will lead to rapid mixing in this wake region close to the vehicle. Consequently, the source size should scale with the vehicle dimensions, not that of the exhaust pipe. These arguments suggest that one of the three larger sources (not CFD_S_a) might be most appropriate for the comparisons with measured and other modelled (OSPM and Box) results. Since our measurements were at 0.40 m away from the wall of the leeward side and all three CFD sources (except CFD_S_a) showed identical profiles at this height, one of these sources (CFD_S_c) has been selected for further comparisons.

Apart from the CFD simulations, as discussed in Section 4.2, the vertical PNC profiles were produced by using the OSPM and modified Box model for both sides of the canyon (Fig. 5). These profiles were plotted with the measured vertical PNC profiles though the measured data was only available for the leeward side of the canyon.

1 It is generally expected that PNCs would be larger near to the road level due to the
2 presence of the emission sources. The PNCs are then expected to decrease with height due to
3 removal of particles as a result of mass exchange between the street and the less polluted wind
4 above. Interestingly, various modelled and measured concentrations show different shapes of
5 vertical profiles. We concluded from our previous discussions on measured PNC profile close to
6 the road level (Kumar et al., 2008a) and across the entire height (Kumar et al., 2008b) of the
7 canyon that the flow close to road level in a real street canyon is considerably more complex
8 than the simple descriptions that we and other typically use, in reality involving along and cross
9 street flows, recirculating vortex and flow intermittency (Britter and Hanna, 2003). These
10 complexities will probably be specific to each individual street canyon. Therefore, it is not
11 straightforward to describe the vertical PNC profiles close to road level. The empirical models
12 OSPM and the Box model assume a well-mixed region in the first few meters of the canyon
13 leading to constant concentrations in this region. Similar to our earlier studies, present study also
14 indicated decreasing PNCs (except CFD simulations) with increased height above ≈ 2 m (Fig.
15 5a). This observation is in agreement with several other studies for particle number
16 concentrations (Kumar et al., 2008a,b; Li et al., 2007; Longley et al., 2004), particle mass
17 concentrations (Chan and Kwok, 2000; Colls and Micallef, 1999; Kumar et al., 2008b; Li et al.,
18 2007; Micallef and Colls, 1998; Weber et al., 2006) and gaseous pollutants (Li et al., 2007;
19 Berkowicz et al., 2002; Murena and Vorraro, 2003; Park et al., 2004; Vogt et al., 2006;
20 Zoumakis, 1995). A review of these studies is presented in supplementary Table S.1.

21 It is interesting to compare the shape and magnitude of vertical PNC profiles produced
22 by the CFD with other modelled and measured vertical PNC profiles (Fig. 5a). The OSPM and
23 Box models assume constant PNCs up to ≈ 2 m, while the measurements show an increase in

PNCs from road level up to ≈ 2 m. This increase is reproduced by the CFD model. However, the CFD profile does not show the decrease to roof level seen in the measured data. These results suggest that size of the source which is closest to the vehicle dimensions may be a better representation for setting up a source in CFD simulations. As possible reasons for the positive concentration gradient close to the road level were identified (Kumar et al., 2008b): dry deposition, a recirculating vortex structure in the canyon transporting the pollutants from the windward side along with the sweeping of near road concentrations to the more elevated sampling points on the leeward side, and the trailing vortices in the vehicle wake transporting the pollutants from the lowest sampling points to the upper sampling points. The CFD simulations presented in this study support previously found positive PNC gradient close to road level as the selected street canyon had one-way traffic and the counter-effect of trailing vortices may not present to produce a well-mixed region close to road level. Moreover, a canyon vortex and its effect on PNC distributions are clearly evident from Figs. 2a and 3. It should be noted that the effect of traffic-produced turbulence is not considered in CFD simulations which can produce a well-mixed region close to road level. However, this effect can be ignored considering that above-roof wind speeds were always in excess of 1.5 m s^{-1} during selected duration where wind-produced turbulence is likely to dominate traffic-produced turbulence (Di Sabatino et al., 2003; Kumar et al., 2008c; Solazzo et al., 2007). As deposition was not modelled by the CFD, the elevated source (0.20 m above ground) might be a reason for the concentration gradient near the ground.

In the upper part (above ≈ 2 m) of the canyon, OSPM and Box model predict similar shape of measured PNC profiles. However, CFD results do not show the large decrease in PNC with increased height as seen in other models and measurements. This suggests that the CFD

1 model does not predict enough mixing in the region of the leeward wall. However, Walton and
2 Cheng (2002) compare RANS and large-eddy simulations (LES) to the wind tunnel data of
3 Hoydysh and Debberdt (1998) and both show trends on the leeward side of the canyon in
4 agreement with our CFD predictions i.e., only a small decrease up to rooftop level. In common
5 with our CFD simulation the wind tunnel data was obtained for an idealised case of a canyon in
6 a perpendicular wind, therefore it may not be the case that the difference from the field studies
7 of Kumar et al. (2008b) is due to the inability of our 2D CFD solution to capture real-world 3D
8 effects. Moreover, the small decrease in some of the vertical near-ground CFD profiles reveal
9 that the CFD model produces reasonable dilution in the lower part of the canyon, but does not
10 seem to produce enough dilution in the upper part of the canyon. This might be because the real
11 structure of the roof, and actual flow conditions in the field, are more complex than assumed
12 simplified structure, resulting in a weaker exchange of air between street and canyon top in the
13 upper part of the canyon. Conversely, the operational models (OSPM and modified Box model)
14 assume a larger decrease in concentration across the entire height of the canyon as these are
15 calibrated using experimental results from various field studies.

16 The vertical PNC profiles for the windward side of the canyon are nearly similar in
17 shape for all models (Fig. 5b). This is expected for the OSPM and Box models as they both
18 assume identical PNCs at all heights in the windward side. Also, the CFD results show almost
19 identical PNCs at each height of the canyon. This nearly constant vertical profile was also
20 observed by Hoydysh and Dabberdt (1998) and Walton and Cheng (2002). However, the
21 average PNCs for CFD were about 1.8 and 4.8 times larger than for the Box and OSPM models,
22 respectively. The higher PNCs predicted by the CFD on this wall are due to the higher values
23 predicted at the top of the leeward wall being advected to the other side of the canyon, as
24 discussed previously.

4.4 Comparison of measured and modelled PNCs

Fig. 6 shows the comparison of measured and modelled PNCs at various heights in the leeward side of the canyon for the 24 h simulations. The overall performance of the models applied in this study has been compared using commonly used statistical parameters, as shown in Table 2 (Kumar et al., 2008c). Predictions of modelled results from CFD and Box models were generally within a factor of two (FAC2), and within a factor of three (FAC3) for OSPM. Differences between modelled results and measurements can be largely attributed to a large difference (up to a factor of three) in particle number emission factors (PNEF), as discussed in Section S.2. Although a change in PNEF will not bridge the difference in the predictions by different models. In general, the predictions are still in fairly good agreement as might be expected between experiments and modelling. Each model showed a good correlation coefficient (R) at all heights, but relatively larger values were noticed for the OSPM at all heights (except $z/H = 0.19$) than for the Box and CFD models. As illustrated in Fig. 6, the OSPM consistently under-predicts the PNCs at all heights; this is indicated by the positive values of fractional bias (FB) in Table 2. Conversely, the Box and CFD models slightly over-predict the PNCs. However, these observations indicate that predictions using a simple modelling approach (modified Box model), idealised CFD simulations or widely used operational model (OSPM) were within an acceptable range, despite ignoring the particle dynamics and using different mixing mechanisms.

The inter-comparison of modelled PNCs is of particular interest to see why these models predict different values of PNCs for the same input parameters. The modelled PNCs from Box and CFD models were close to each other at $z/H = 0.19$, but those from OSPM were about a factor of two smaller than these models. The difference between the modelled PNCs using CFD

and Box models at other heights increased. The modelled PNCs using OSPM at each height were consistently smaller than those from Box and CFD models; these were about a factor of 4 and 5 smaller at $z/H = 0.40$ and 0.64 , respectively, than those from the CFD model (Fig. 6). The large differences in PNCs at upper sampling heights could be because the CFD model considers weaker exchange of air in the upper part of the canyon as discussed in Section 4.3. Some differences in PNCs across the entire height of the canyon could be because the OSPM explicitly takes in to account the turbulence created by the wind and traffic, but the Box and CFD models do not.

5. Summary and Conclusions

A modified Box model, OSPM and CFD simulations were used to predict the PNCs at different heights in a regular (aspect ratio of unity) street canyon for cross-wind conditions. The modelled PNCs were compared with measured PNCs in the 10–300 nm range. Four different sizes of finite cross section line emission sources were selected in the CFD simulations to assess their effect on mean PNC distributions in the street canyon. Modelled vertical PNC profiles were compared with the measured vertical PNC profiles.

In the CFD simulations, vertical PNC profiles were drawn at various distances away from the leeward walls of the canyon. These showed large variations for various sizes of sources, indicating that selection of an appropriate source size is important to determine the PNC distributions. However, the effect of source size on the windward side of the canyon was modest. The source with the smallest area (CFD_ S_a) produced the largest PNCs near (up to ≈ 0.50 m) to the leeward side wall. This is because the smallest source is close to the ground and hence the particles are emitted into the edge of the vortex sweeping round the canyon, leading to high concentrations there. The larger sources are centred further away from the ground and emit

1 the particles nearer to the centre of the vortex, leading to higher concentrations away from the
2 wall. A source size scaling the vehicle dimension, not the size of the exhaust pipe, appears to
3 better represent the measured PNCs profiles in the lowest part of the canyon since this
4 accounted for the effect of traffic-produced turbulence through rapid mixing in the source
5 region.

6 The models used in this study produced different shapes of vertical PNC profiles in both
7 sides of the canyon. These shapes were particularly different in the leeward side. Both the non-
8 CFD (OSPM and Box) models showed constant PNCs up to h_0 (i.e., ≈ 2 m) and decreasing PNCs
9 above this height. The CFD model showed an increase from road level to a height of ≈ 2 m; this
10 observation is in agreement with the measurements. However, they do not predict the measured
11 decrease in PNC towards the top of the canyon above ≈ 2 m, suggesting that the CFD model
12 does not predict enough dilution in the region of the leeward side wall. Considering the wind
13 speeds used in this study the wind-produced turbulence is likely to dominate, however it may be
14 the case that traffic-produced turbulence may have some effects.

15 In the windward side of the canyon, both OSPM and Box models predicted constant
16 PNCs at each height. The CFD model also produced similar shape of vertical PNC profiles, but
17 with far higher PNCs than found by both non-CFD models. The higher PNCs predicted by the
18 CFD on this side of the wall are due to the higher values predicted at the top of the leeward wall
19 which is advected to the windward side of the canyon with relatively little further mixing.

20 The measured PNCs compared well (between a factor of 2 and 3) with those modelled
21 using Box, OSPM and CFD models, suggesting that if the model inputs are chosen carefully,
22 even a simplified approach can predict the PNCs as well as more complex models. The inter-
23 comparison between the models for idealised (CFD and Box) and operational (OSPM)

conditions showed larger PNC differences at the upper sampling heights when compared to the PNC differences near to the road level. The largest PNC difference between idealised and operational models at upper sampling heights were attributed to the weaker exchange of street and above-roof air in the upper part of the canyon by idealised models. This is because the real structure of the roof, and actual flow conditions in the street canyon, are expected to be more complex than assumed idealised conditions. Moreover, some differences in PNCs over the entire height of the canyon could be because the OSPM explicitly takes in to account the turbulence created by the wind and traffic, but the other models do not.

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Figure Captions

Fig. 1. Schematic diagram of computational domain representing Pemborke Street, description of boundary conditions and measurements points (figure not to scale). H and W are the height and width of the canyon, respectively (both 11.60 m), whereas H_s and W_s are the height and width of the source, respectively.

Fig. 2. Flow and turbulence distributions showing (a) mean velocity vectors (m s^{-1}), and (b) distribution of mean turbulent kinetic energy ($\text{m}^2 \text{s}^{-2}$). These figs. are for a constant inlet velocity 3.8 m s^{-1} , and for κ and ε inlet profile as described in Section 3.2.2. High density of vectors at the top of Fig. (a) is due to the close grid spacing in this region.

Fig. 3. Typical distribution of mean PNC ($\# \text{ cm}^{-3}$) contours for (a) CFD_ S_a , (b) CFD_ S_b , (c) CFD_ S_c , and (d) CFD_ S_d . Rectangular boxes represent the source area. These figs. are for Case No. 1, as described in Table 1.

Fig. 4. Vertical profiles of normalised PNCs in the leeward and windward side of the street canyon, respectively, at (a–b) 0.40 m ($w/H = 0.034$), (c–d) 1.5 m ($w/H = 0.13$), and (e–f) 2.50 m ($w/H = 0.22$) away from both sides of walls. Same simulations as in Fig. 3.

Fig. 5. Measured and modelled vertical normalised concentration profiles at (a) leeward and, (b) windward side of the canyon. Note that no measured data is available on the windward side.

Fig. 6. Comparison of hourly averaged measured and modelled PNCs on 0.40 m away from the leeward side of the canyon at heights (a) 1.0 m, (b) 2.25 m, (c) 4.62 m, and (d) 7.37 m. Dotted lines cover the range of PNC predictions with in a factor of two (FAC2).

1 Tables

2 **Table 1.** Input parameters used for each set of simulation. Each case represents hourly averaged
3 values of U_r , S , Re and C_b .

Case No.	Time (h)	U_r (m s ⁻¹)	S^* ($\times 10^9$ # m ⁻¹ s ⁻¹)	Re ($\times 10^6$)	C_b ($\times 10^9$ # m ⁻³)
1	16:00–17:00	3.80	1.78	3.22	1.88
2	17:00–18:00	4.30	1.55	3.66	1.73
3	18:00–19:00	3.54	4.12	3.02	4.99
4	19:00–20:00	3.24	4.36	2.78	5.37
5	20:00–21:00	3.31	2.38	2.85	2.88
6	21:00–22:00	3.75	1.40	3.23	1.52
7	22:00–23:00	3.56	1.47	3.07	2.84
8	23:00–00:00	3.26	1.71	2.82	2.54
9	00:00–01:00	3.72	0.52	3.22	1.25
10	01:00–02:00	3.49	0.63	3.02	1.90
11	02:00–03:00	2.95	1.03	2.56	2.30
12	03:00–04:00	3.19	1.13	2.78	2.57
13	04:00–05:00	3.33	0.59	2.90	1.03
14	05:00–06:00	2.69	0.63	2.34	0.82
15	06:00–07:00	2.42	1.40	2.10	2.37
16	07:00–08:00	2.47	4.40	2.13	5.84
17	08:00–09:00	3.79	4.03	3.19	4.81
18	09:00–10:00	3.14	2.58	2.58	3.85
19	10:00–11:00	3.23	3.07	2.65	3.41
20	11:00–12:00	3.44	3.41	2.82	3.64
21	12:00–13:00	2.88	3.36	2.37	4.18
22	13:00–14:00	2.52	2.90	2.09	3.95
23	14:00–15:00	2.83	2.51	2.33	4.42
24	15:00–16:00	2.66	2.30	2.20	2.78

4 *Source strength (S) has been estimated separately for each source depending on its area and traffic
5 volume during each hour; the values shown in table are for source CFD_ S_a .

Table 2. Overall performance of models used for the prediction of PNCs in the leeward side of the canyon. The correlation coefficient (R) reflects the linear relationship between two variables and the ability of a model to predict the measured PNCs. The fractional bias (FB) reflects the differences between average measured and modelled results. FAC2 is fraction of predictions with in a factor of 2. Ideally expected values for R, FAC2 and FB are 1, 100% and 0, respectively.

z/H	Parameters	Box	OSPM	CFD Simulations			
				CFD_S _a	CFD_S _b	CFD_S _c	CFD_S _d
0.09	R	0.80	0.84	0.80	0.80	0.80	0.80
	FAC2	63%	67%	71%	83%	83%	83%
	FB	-0.56	0.56	-0.47	-0.20	-0.22	-0.20
0.19	R	0.90	0.85	0.90	0.90	0.90	0.90
	FAC2	96%	13%	100%	83%	83%	83%
	FB	0.02	0.88	-0.06	0.10	0.09	0.11
0.40	R	0.70	0.75	0.69	0.69	0.69	0.69
	FAC2	88%	17%	79%	83%	83%	83%
	FB	-0.03	0.96	-0.28	-0.22	-0.21	-0.21
0.64	R	0.71	0.74	0.69	0.69	0.69	0.69
	FAC2	79%	21%	58%	58%	63%	58%
	FB	-0.09	1.01	-0.58	-0.55	-0.53	-0.54

Note: FAC3 for OSPM is 92, 67, 58 and 67% at $z/H = 0.09, 0.19, 0.40$ and 0.64 , respectively.

Fig.1.ppt

Fig.2.ppt (COLOUR)

Fig.2.ppt (Black and White)

Fig.3.ppt (Colour)

Fig.3.ppt (Black and White)

Fig.4.ppt

Fig.5.ppt

Fig.6.ppt