

H13-5 MODELLING THE DISPERSION OF NANOPARTICLES IN STREET CANYONS

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Abstract: Suitable dispersion models are required for the prediction of nanoparticle number concentrations for adopting mitigation policies. The aim of this work is to model the dispersion of nanoparticle number concentrations in the 10–300 nm range at different heights in an urban street canyon. A modified Box model (Kumar *et al.* 2009b) and an operational street pollution model (OSPM) are used for this purpose and modelled results are compared with the measured nanoparticle concentrations. Further, the article discusses the role of particle dynamics in street-scale modelling and analyses the influence of the uncertainty in particle number emission factors on modelled concentrations. Reasons for discrepancies in modelled results due to particle number emission factors and street-level particle dynamics are given.

Keywords: Dispersion modelling; Nanoparticles; Particle number concentrations; Number emission factors; Street canyons.

INTRODUCTION

Recent Euro 5 and Euro 6 vehicle emission standards include limits for nanoparticles (those below 300 nm) on a *number* basis. Related standards are likely to be enforced for *ambient* nanoparticles in the near future to protect against their possible adverse impacts on public health and the environment. Reliable modelling tools are essential to design effective mitigation policies for nanoparticle rich environments, such as urban street canyons. However, the challenges in modelling of nanoparticle number concentrations grow with the inclusion of the particle dynamics which occur after their release into the atmosphere. Currently, insufficient knowledge is available of the processes that determine the size distribution and its development at all relevant spatial scales. Moreover, the performance of nanoparticle models suffers greatly from the lack of sufficient validation data and routinely required input information, such as particle number emission factors.

This article aims to model the number concentrations of nanoparticles in the 10–300 nm range in an urban street canyon using a modified Box model (Kumar *et al.* 2009b) and the OSPM (Berkowicz 2000). The modelled results are then compared with the measured concentrations and the role of particle dynamics at street-scale modelling and the influence of the uncertainty in particle number emission factors on modelled number concentrations are discussed. Note that this article focuses only on particle *number* concentrations. Therefore, it only includes particles of size less than 300 nm, as this size range comprises more than 99% of total number concentrations of particles in the ambient environment (Kumar *et al.* 2009a). Particles below 10 nm are not discussed because of their significant losses in sampling tubes (Kumar *et al.* 2008e).

METHODOLOGY

Site description, instrumentation and data acquisition

Measurements were carried out in Pembroke Street, Cambridge, UK (52°12'N and 0°10'E). The studied section of the street was about 167 m long and 11.6 m wide. It has almost the same height (i.e. 11.6 m) of buildings on either side of the road. The orientation of the street is southwest (SW)–northeast (NE). This is a one-way street and traffic travels from SW to NE.

A fast response differential mobility spectrometer (DMS500) was used to measure particles in the 5–2500 nm at a sampling frequency of 10 Hz. The measurements were made continuously for 17 days between 7 and 23 March 2007. In this article, only 24 hours of data are included for analysis as this includes continuous pseudo-simultaneous measurements at four different heights (i.e. 1.0, 2.25, 4.62 and 7.37 m; referred to as $z/H = 0.09, 0.19, 0.40$ and 0.64 , respectively) in the street canyon. Meteorological parameters (wind speed, wind direction, temperature and relative humidity) were measured simultaneously at 16.60 m above the road level (i.e. at 5 m above the roof top). Traffic volumes were taken manually and average traffic speed at the site was estimated to be about 30 ± 7 km h⁻¹. Recent articles (Kumar *et al.* 2008a; Kumar *et al.* 2008d) can be referred to for detailed description of the measurement site, instrumentation and data acquisition.

Description of models

The modified Box model

Assuming that concentrations are uniformly distributed in the lower part of the canyon (up to h_0) due to a well mixed region and the concentrations above h_0 decrease exponentially with height, the model shown in equation (1) is formulated. It is termed a 'modified Box model' because a standard box model is modified by including modules for vertical variations in particle number concentrations and regions for traffic- and wind-dependent concentrations. Our recent study (Kumar *et al.* 2008a) demarcated wind and traffic dependent regimes of nanoparticle concentrations depending on the above-roof wind speed (U_r). The results of this study are included in the model with the following assumptions: (i) in traffic-dependent region (when $U_r \ll U_{r,crit}$), number concentrations of nanoparticles were approximately constant and independent of U_r up to a critical wind speed ($U_{r,crit}$), and (ii) in wind dependent regime (when $U_r \gg U_{r,crit}$), these are inversely dependent on U_r .

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

where $z = \max(z, h_0)$, $U_r = \max(U_r, U_{r,crit})$, $k_1 (= 0.11 \text{ m}^{-1})$ is an exponential decay coefficient derived from Fig. 1, $h_0 (= 2 \text{ m})$ is the assumed height of the well mixed region close to road level, $E_{x,i-j}$ is the particle number emission factor (# veh⁻¹ cm⁻¹) in a size range $i-j$ for a vehicle type x , C and C_b are predicted and background nanoparticle concentrations. Note that

equation (1) is for the leeward side of the canyon and the empirical constant b_1 is taken as 0.013 in this case. Assuming that the predicted concentrations on the leeward side of the canyon are equal to the concentrations at all heights on the windward side of the canyon, the empirical constant b_1 should be replaced with b_2 [$= b_1/\exp(-0.11 \times 11.6) = 3.58 b_1$] to predict concentrations at the windward side. A detailed description of the model formulation can be seen in Kumar *et al.* (2009b).

The Operational Street Pollution Model (OSPM)

The OSPM is a widely used model that is used here to predict number concentrations of nanoparticles. It contains a simplified empirical description of the flow and dispersion conditions for urban street canyons. 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 street canyons. A detailed description of the OSPM can be seen in Berkowicz (2000) and at www.ospm.dmu.dk.

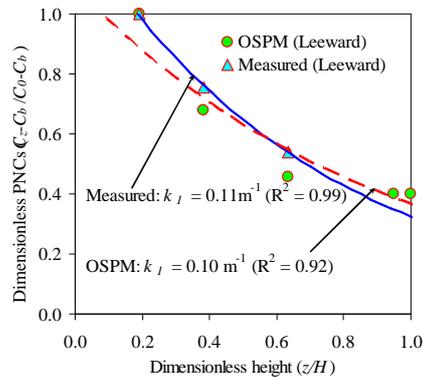


Figure 5. Estimation of exponential decay coefficient (k_1) using measured and OSPM modelled data.

RESULTS AND DISCUSSIONS

Role of particle dynamics in street-scale modelling

Figure 2 shows hourly averaged measured particle number distributions at different heights in the canyon. These were bimodal and lognormal in form at each height. Two distinct modes peaking at 13.3 nm (nucleation mode) and 86.6 nm (accumulation mode) were observed. Time scales for removal and transformation processes (e.g. dilution, dry deposition, coagulation and condensation) were estimated to analyse their effect on particle number distributions. The deduced time scales were of the order of 40s for dilution, 30 to 130s for dry deposition on the road surface, and 600 to 2600s for the dry deposition on the street walls, about 10^5 s for coagulation, and about 10^4 – 10^5 for condensation (Kumar *et al.* 2008d). Comparison of these estimated time scales shows that dilution is quick and it does not allow others (except dry deposition on the road surface) to alter the size distributions. This is also evident from the similarity in shape and the negligible shift in peak and geometric mean diameters of particle number distributions in both modes at each height (Fig. 2). Our recent study also supported this hypothesis; this study compared time scales for evolution of particle size distributions due to transformation processes in the wake of a moving vehicle with the time for these particles to reach the road side in an urban street canyon (Kumar *et al.* 2009c). Results of this study found that the competing influence of transformation processes were nearly complete by the time particles reach the roadside, suggesting that it is possible to neglect the effect of particle dynamics and assuming total particle numbers as conserved; a similar assumption is adopted here. This assumption is in accordance to the results reported by Ketzel and Berkowicz (2004) on street level particle dynamics, as is also the case in existing dispersion models such as the OSPM used in this study.

It should be noted that particle dynamics should be included in *city scale* models as they may affect the total number concentrations considerably (Gidhagen *et al.* 2005). Ketzel and Berkowicz (2005) found that changes in total nanoparticle number concentrations due to the combined effect of transformation and loss processes can lie between a loss of 13 and 23% compared to an inert treatment. Similarly, a study by Kumar *et al.* (2009a) compared particle dynamics between street and rooftop levels. They found about a five times greater formation rate of new particles at rooftop level than at street level, attributing this to weaker scavenging mechanisms and favourable conditions for gas-to-particle conversion at rooftop.

Comparison of measured and modelled nanoparticle number concentrations

Figure 3 shows the comparison of hourly averaged measured and modelled nanoparticle number concentrations at various heights on the leeward side of the canyon for the 24 hours of data. The overall performance of both the modified Box and OSPM models is compared using commonly applied statistical methods in Table 1. Predictions from the Box model were generally within a factor of two of the data and within a factor of three for OSPM.

Results from both models follow the expected change in concentration at different heights. As anticipated, modelled concentrations are largest near the road level (i.e. at $z/H = 0.09$) due to the presence of emission sources and then decrease with height due to removal of particles as a result of mass exchange between the street and the less polluted wind above. However, both these models were unable to reproduce the increase in measured concentrations at $z/H = 0.19$ as they both assume well-mixed concentrations up to about 2 m and decreasing thereafter. Both models showed good correlation coefficients (R) at all heights; OSPM producing relatively better values at all heights except $z/H = 0.19$ (see Table 1). As

indicated by positive values of fractional bias (FB) in Table 1, OSPM consistently under predicted the concentrations at all heights, as opposed to the Box model that slightly over-predict the concentrations.

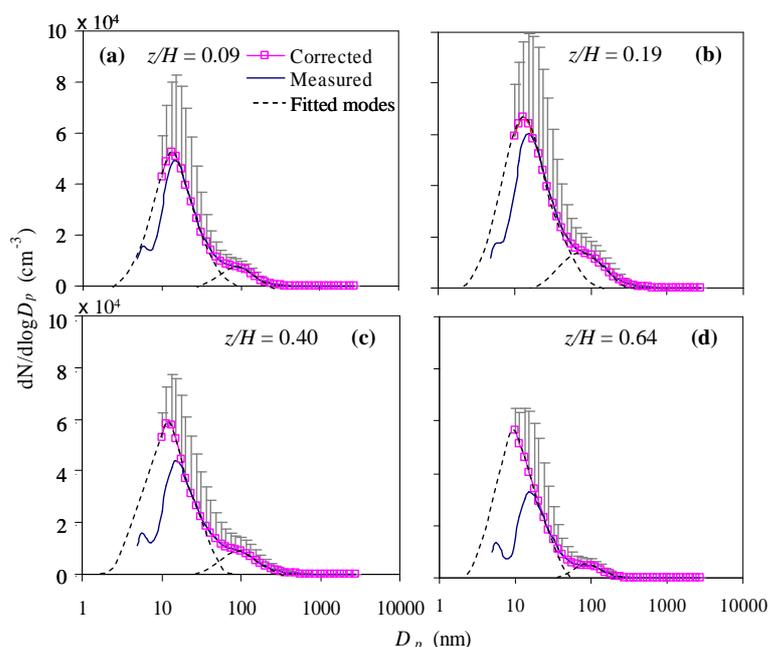


Figure 2. Hourly averaged measured particle number distributions at (a) 1.0, (b) 2.25, (c) 4.62, and (d) 7.37 m of an 11.6 m deep canyon. Distributions have been corrected for particle losses in the sampling tubes of different lengths (Kumar *et al.* 2008d).

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

z/H	Parameters	Box	OSPM	z/H	Parameters	Box	OSPM
0.09	R	0.80	0.84	0.40	R	0.70	0.75
	FAC2	63%	67%		FAC2	88%	17%
	FB	-0.56	0.56		FB	-0.03	0.96
0.19	R	0.90	0.85	0.64	R	0.71	0.74
	FAC2	96%	13%		FAC2	79%	21%
	FB	0.02	0.88		FB	-0.09	1.01

It is useful to assess why these models predict different concentrations for identical input parameters. One reason for this could be the better mixing mechanisms used within the OSPM. It implicitly takes into account both the effect of atmospheric turbulence produced by the wind and the traffic-produced turbulence by the vehicles. On the other hand, the Box model does not take such considerations explicitly, except by defining the vertical profile of concentrations through an exponential decay. The other reason for bias relative to the data could be the large variability in particle number emission factors (discussed in the next section), although a change in particle number emission factors will not of course affect the difference in the predictions by these models. In general, our modelled results indicate that predictions using a simple modelling approach are within an acceptable range, despite ignoring the particle dynamics and using simple mixing mechanisms. It also indicates that if model inputs are chosen carefully, even a simplified modelling approach can predict concentrations as well as more complex models.

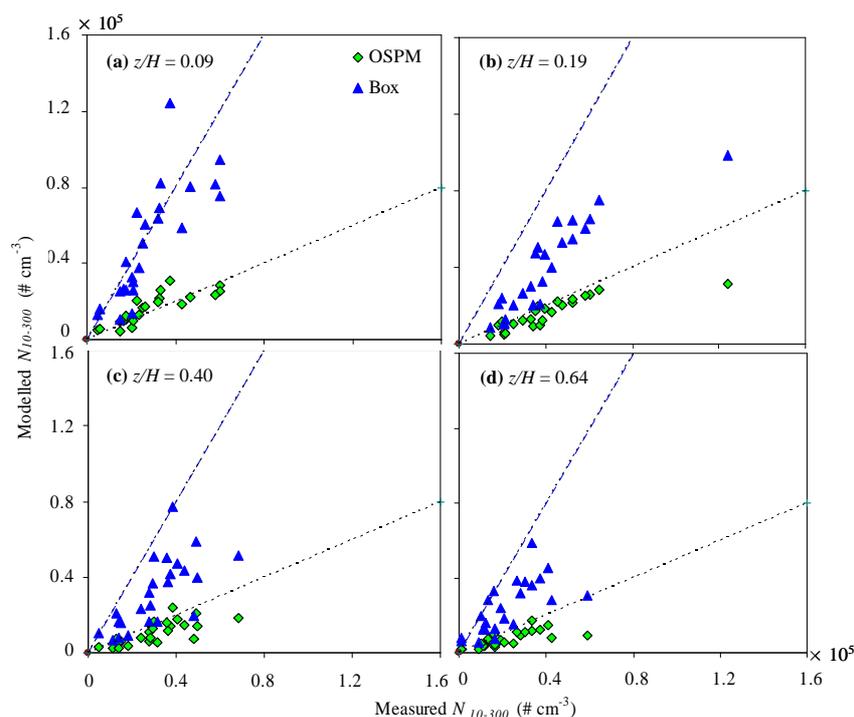


Figure 3. Comparison of hourly averaged measured and modelled nanoparticle number concentrations at the leeward side of the canyon at (a) 1.0, (b) 2.25, (c) 4.62, and (d) 7.37 m heights. Dotted lines cover the range of predictions within a factor of 2 (FAC2).

Influence of the uncertainty in particle number emission factors on modelled concentrations

Particle number emission factors are an essential model input parameter but are not reliably available for routine applications. The value of particle number emission factors used in this study is $1.33 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$. These values were derived using an inverse modelling approach (Palmgren *et al.* 1999) on the data collected during a field measurement campaign in Cambridge (Kumar *et al.* 2008a). This represents a mixed traffic fleet having 6 to 8% heavy duty vehicles (buses and trucks) and an average fleet speed between 20 and 30 km h^{-1} . However, studies of particle number emission factors show up to an order of magnitude difference for a given vehicle type under near-identical conditions (Jones and Harrison 2006; Keogh *et al.* 2009; Kumar *et al.* 2008b). For example, a comprehensive literature review (Kumar *et al.* 2010) of this topic suggested values as low as $5.67 \pm 2.80 \times 10^{13} \text{ \# veh}^{-1} \text{ km}^{-1}$ for particles in the 15 to 700 nm size range for stop-start mixed traffic fleet (i.e. typical urban driving conditions) moving at speeds less than 60 km h^{-1} (Morawska *et al.* 2005). A study by Imhof *et al.* (2005) found values as high as $3.9 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$ for about the same vehicle speed (50 km h^{-1}) in nearly identical driving conditions; these values increased to 11.7×10^{14} and $13.5 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$ for vehicle speeds 100 and 120 km h^{-1} , respectively. These observations clearly reflect a large uncertainty in particle number emission factors, meaning that modelled results are likely to be affected to a similar degree irrespective of the accuracy of a model. For example, if we change the current emission factors values from 1.33×10^{14} to about $2.2 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$ in OSPM, which is still within the above described low and high end ranges and within the range ($2.8 \pm 0.5 \times 10^{14} \text{ \# veh}^{-1} \text{ km}^{-1}$) found by Ketzel *et al.* (2003) for typical urban driving conditions, over 90% of the modelled results at different heights came closer (within a factor of 1.5) to measured concentrations; changes in particle number emission factors obviously affect the modelled results from the modified Box model in similar manner. Clearly, accurate information on emission factors is essential for reliable modelling of nanoparticle number concentrations.

SUMMARY AND CONCLUSIONS

The article compares measured nanoparticle number concentrations at different heights in an urban street canyon with modelled concentrations from the modified Box model and the OSPM. Results suggest that particle dynamics may be disregarded for street scale modelling as the competing influence of transformation processes on particle number distributions seems to be nearly over by the time particles are measured at roadside. Both the models predicted the concentrations to within a factor of two to three of those actually measured, suggesting that even a simplified approach can predict the concentrations as well as more complex models if model inputs are chosen carefully. The particle number emission factor is one of the most important model input parameters, but inadequate information on this may result in large inconsistencies in modelled nanoparticle concentrations. Long-term field measurements (including number and size distributions) are needed for the development and validation of reliable nanoparticle dispersion models and hence for developing mitigation policies.

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