MEASUREMENT AND DISPERSION BEHAVIOUR OF PARTICLES IN VARIOUS SIZE (5 NM>D_P<1000 NM) RANGES IN A CAMBRIDGE STREET CANYON

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INTRODUCTION

Current regulations for atmospheric particulate matter include PM_{10} and $PM_{2.5}$ mass concentration. Fine particulates (those below 1000 nm) are not included in the regulatory limits. These particles contribute significantly to particle number concentration (PNC), but little to particle mass concentration. It is widely hypothesised that number concentrations of fine particles are better correlated with acute and chronic health effects and other environmental hazards than are particle mass concentrations *(Donaldson et al.,* 1988*; Pope III,* 2000). Vehicles are a major source of fine particles in urban areas. High PNCs are common in urban street canyons, giving rise to health concerns for those exposed. It is important to understand the dispersion behaviour of fine particles in urban street canyons to enable better prediction of human exposure and design of regulatory mitigation strategies. The introduction of cleaner fuels, better emission control technologies and tighter legislations have decreased the total mass of particles emitted, but not significantly the number.

A recently developed instrument, the 'fast response differential mobility spectrometer DMS500', was used to measure the particle number and size distributions in the 5-1000 nm range. This article addresses the dispersion behaviour of particles due to the changes in wind conditions and traffic activity levels in a Cambridge street canyon. The classification of size ranges: nucleation (5-30nm, $N_{5\text{-}30}$), accumulation (30-300nm, $N_{30\text{-}300}$) and coarse (300-1000nm, $N_{300\text{-}1000}$).

METHODOLOGY

Measurements were carried out on a 200 m long street section on Fen Causeway in Cambridge (UK). The height and width of the selected section were ~20 m giving an aspect ratio of ~1. The DMS500 was deployed to measure the particle number distribution at a sampling frequency of 1Hz continuously for 10 hours on 7 weekdays. These measurements were recorded between 0900 and 1900 on 8, 9, 12, 13, 16, 19 and 21 June 2006. Measurements were taken at three different heights thought to be of relevance to human inhalation i.e., 0.20, 1.00 and 2.60 m. Representative samples were taken for 20 minutes in an hour at each height, on two different occasions (i.e., 2 samples per hour, 10 minutes per sample) by manually repositioning the sampling probe every 10 minutes. *Kumar et al.* (2007) give further details of the site and experimental methodology.

RESULTS AND DISCUSSIONS

Previous analysis (*Kumar et al.,* 2007) showed that automotive traffic was the main source of particles at the measurement site. Total PNCs in the 5-1000 nm range were found to be inversely correlated with the reference wind speed and directly correlated with the traffic volume. The largest PNCs were near to road level, which is near to the pollution sources. The PNCs measured at 0.20 m and 1.0 m were the same to within 0.5-12.5 % indicating a well-mixed region. This could be attributed to the enhanced mixing from traffic produced turbulence. Concentrations at 2.60 m were smaller by 10-40 % than those at 0.20 m and 1.0 m, suggesting a concentration gradient in the upper part of the canyon. In this article, the

results have been analysed by dividing the particle number distributions (PNDs) into various size ranges in order to investigate their modal share, sources, formation mechanisms and dispersion behaviour.

Modal share of particles and their formation mechanism

The average of the PNCs over the entire sampling duration indicated that coarse, accumulation and nucleation modes constituted 1%, 68% and 31% respectively of the total (see Fig. 1). Consistent with this, *Tuch et al.* (1997) reported that the contribution of particles larger than 500 nm to the overall PNC was negligible in the European urban environment. However, our results showed that the particles smaller than 100 nm contributed about 72% to the overall PNC while they found a much smaller contribution. Our results were also in agreement with the measurements made in Manchester, UK (*Longley et al.,* 2003) wherein the dominance of accumulation mode ($N_{30\text{-}300}$) PNCs to the overall (4.6 nm to 10000 nm) PNCs was reported. Accumulation mode particles are formed due to the incomplete combustion of fuel; they consist of soot particles mainly emitted from diesel engined vehicles or direct injection gasoline engined vehicles *(Graskow et al.,* 1998). Gasoline engined vehicles were found to dominate (76%) the traffic fleet (see Fig. 2) and they produce PNCs principally in the nucleation range (*Kittelson et al.,* 2006). Despite this, the nucleation mode ($N_{5\text{-}30}$) particles, which are formed from the condensation of semi-volatile species during the rapid cooling of exhaust emissions by mixing with ambient air, did not dominate the total PNCs. This may be because the overall particle emissions from the diesel vehicles dominated or because for stop-start driving the PNCs for light duty gasoline engined vehicles can be similar to modern heavy duty diesel powered vehicles (*Graskow et al.,* 1998), due to the storage and release of volatile hydrocarbons at the time of increased speed from gasoline engined vehicles (*Kittelson et al.,* 2001). The traffic during our experiments was made unsteady by traffic signals at both ends of the experimental section, causing acceleration and generally congested conditions. A further possible reason for smaller PNCs in the nucleation range could be due to the presence of higher accumulation range particles which act to suppress the formation of nucleation range particles since the carbonaceous agglomerates scavenge the volatile material, reducing the likelihood of gas-to-particle conversion (*Kittelson et al.,* 2004).

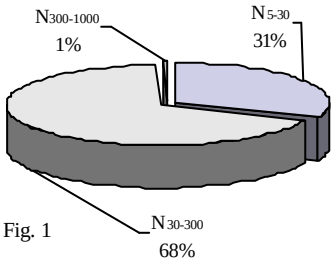

Fig. 1

Fig. 1; Modal share of particles in each size range.

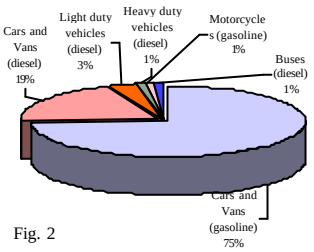

Fig. 2

Fig. 2; Modal share of each vehicle type.

Dispersion behavior of particles in different modes

The reference wind speed (RWS), measured on an adjacent rooftop, wind direction and traffic volume were the main factors influencing the measured concentrations of particles in the 5-1000nm range (*Kumar et al.,* 2007). Similarly, a clear 5-1000nm range (*Kumar et al.,* 2007). Similarly, a clear dependence of the concentration of nucleation, accumulation and coarse

particles on the RWS was seen (Fig. 3). To investigate this percentage changes in daily averaged nucleation and accumulation mode PNCs obtained from their overall averaged PNCs (averaged over the entire sampling duration) were plotted against the inverse of percentage changes in daily mean RWS from the overall mean RWS (averaged over the entire sampling duration). Daily changes in nucleation and accumulation mode particles were found to follow the daily changes in inverse RWS (Figs. 4a and 4b). The dilution of nucleation and accumulation mode particles with the background air appears to be the main factor responsible their concentration changes.

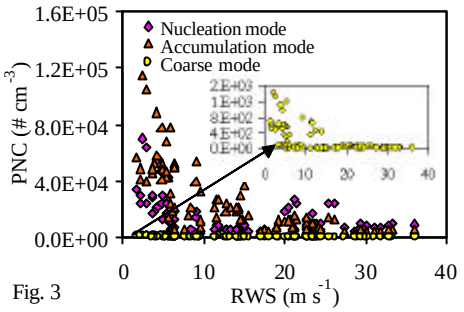

Fig. 3

Fig. 3; Effect of half-hourly averaged RWS on nucleation, accumulation and coarse mode particles during the entire sampling duration.

It was found that changes in accumulation mode particles were better correlated with changes in inverse RWS (R^2=0.91) than were changes in nucleation mode particles (R^2=0.74). *Zhang and Wexler* (2004) reported that transformation processes such as coagulation play a minor role for accumulation mode particles in the urban environment. The total number of particles can therefore be assumed to be conserved. Good correlation of the concentration of accumulation mode particles with RWS^{-1} during our measurements supports the view that their PNCs are inversely proportional to the dilution.

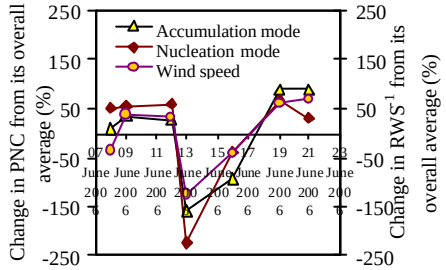

Fig. 4a

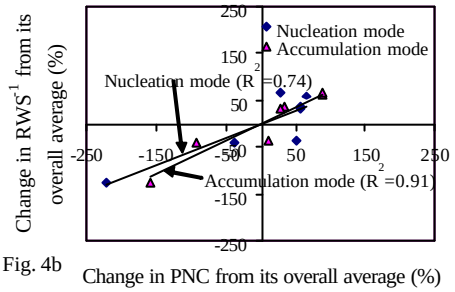

Fig. 4b

Fig. 4 (a) & (b); Relation between change in nucleation and accumulation mode PNCs and the changes in the inverse of RWS. The PNCs in both modes are the hourly average of the three measurement positions on each day.

A difference in the behavior of nucleation mode particles may be due to the dissimilar effect of dilution on their formation in the ambient atmosphere. Nucleation mode particles are in the vapor phase in vehicle tail pipes and their formation is most strongly affected by dilution rate and dilution ratio. This can change PNCs at the tail pipe by two orders of magnitude (*Kittleson*, 1998). Their growth due to condensation of gaseous vapors, suppression due to adsorption onto existing accumulation mode particles, removal from self-coagulation and surface deposition can significantly change the total number of particles (*Kerminen et al.*, 2004). In short, the complex dynamics of the dilution processes resulted in poorer correlation with RWS^{-1} than was observed for accumulation mode particles because the total number of nucleation mode particles was not conserved.

Correlation between traffic volume and particles in various modes

The hourly traffic volume (averaged over the whole sampling duration) in both lanes was found to be 1566 vehicles h^{-1} with a standard deviation of 232 vehicles h^{-1}. To remove the prime dependence of the inverse RWS on PNCs, the half-hourly average of PNCs in various size ranges were plotted with the product of traffic volume and the inverse of RWS. Figure 5 reveals that nucleation and accumulation mode particles were directly correlated with traffic volume. However, the correlation for accumulation mode particles was better than that for nucleation and coarse mode particles. The better correlation for accumulation mode particles was expected because under unsteady driving conditions even gasoline engined vehicles may emit more particles in the accumulation mode range. Furthermore, this could suggest that although nucleation and accumulation mode particles mainly come from exhaust emissions, their concentrations are highly dependent on traffic type and driving conditions. Hence, PNCs and subsequently the correlation of various mode particles with traffic volume will change significantly with changes in traffic types and driving conditions. Additionally, the notion that majority of accumulation mode particles are directly emitted by diesel engined vehicles and majority of nucleation mode particles are emitted by gasoline engined vehicles (*Kittelson et al.*, 2004) may change for our study because of the higher number of particles emitted in accumulation mode by gasoline engined vehicles during unsteady traffic speed and congested conditions; these typical driving conditions are more likely to occur in urban street canyons.

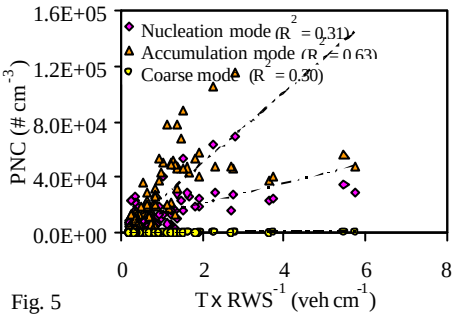

Fig. 5

Fig. 5; Correlation of half-hourly averaged traffic volume (T), RWS with PNCs in various modes.

SUMMARY AND CONCLUSIONS

This work investigated the dispersion behaviour of particles in various size ranges due to the changes in traffic volume and reference wind speed, under typical urban streets and driving conditions. The traffic fleet was dominated by gasoline engined vehicles. Particle number concentrations were dominated by the particles smaller than 300 nm. Nucleation ($N_{5\text{-}30}$) and accumulation ($N_{30\text{-}300}$) mode particles constituted about 31 % and 68 % of the total ($N_{5\text{-}1000}$) particles respectively. Coarse ($N_{300\text{-}1000}$) mode particles contributed the remaining percentage; this small concentration was in agreement with studies by *Tuch et al.* (1997) and *Longley et al.* (2003). Despite the dominance of gasoline engined vehicles in the traffic fleet, nucleation mode ($N_{5\text{-}30}$) particles did not dominate the total PNCs even though gasoline engined vehicles emit a higher proportion of smaller particles than the diesel engined vehicles. Of course, this should simply be because the diesel engined vehicles though fewer, produced far more particles in total. Unsteady traffic speed and the congested traffic conditions on the measurement site were possibly the main cause of the larger number of accumulation mode particles since the gasoline engined vehicles may have been emitting particles similar to diesel engined under these driving conditions.

The PNCs in various size ranges were found to be inversely correlated with the reference wind speed. However, the accumulation mode particles were better correlated with RWS than were nucleation mode particles. This difference in the dispersion behaviours may have been due to the dissimilar effect of dilution on their formation in the ambient atmosphere. Nucleation mode particles are mainly formed from gas-to-particle conversion, and their total number changes rapidly after the exhaust emissions are released into the atmosphere, whereas

changes in accumulation mode PNCs are due to dilution in the ambient atmosphere. Particles in each mode were directly correlated with traffic volume. Accumulation and nucleation mode particles were highly dependent on traffic type and driving conditions. The better correlation of accumulation mode PNCs with traffic volume when compared with nucleation mode PNCs substantiated the view that under unsteady traffic speeds and congested conditions, gasoline engined vehicles emitted larger numbers of accumulation mode particles, similar to diesel engined vehicles, and scavenging by accumulation mode particles reduced the total number of nucleation mode particles.

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