NUMBER CONCENTRATION, DISTRIBUTION AND TRANSFORMATION OF NANOPARTICLES IN AND OUTSIDE A CAR CABIN

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ABSTRACT

City dwellers are regularly exposed to nanoparticle (i.e. particles <300 nm in diameter), emitted by fossil fuelled vehicles, whilst commuting by transport modes such as taxis and buses. Exposure to these nanoparticles can lead to significant adverse effects on human health. This study aims to investigate spatial distribution of particle number concentrations (PNCs) and distributions (PNDs) in and outside a car cabin during driving. Possible influences of particle transformation processes on PNC and PNDs in the car cabin are also investigated. Another objective is to predict the PNCs in the car cabin using those measured outside.

Measurements of particles in the 5-560 nm size range were conducted using a fast response differential mobility spectrometer (DMS50) in conjunction with an automated switching system. The DMS50 was used to measure size–resolved sequential distributions at: (i) four seats in the car cabin during about 700 minutes of driving, and (ii) two points at the driver's seat, inside and the front bonnet outside the cabin, during about 500 minutes of driving. The emission penetration and spatial distribution in the car cabin through (i) the ventilation system (Vent), and (ii) door/window sealing (CG) was simulated by means of three-dimensional computational fluid dynamics (CFD) using the Fluent code. Standard k- ω turbulence model was employed to simulate turbulence flow in the cabin. Vent/CG emission ratio was altered for the two different scenarios (0.9/0.1 and 0.7/0.3), indicating; (i) no filter fitted Vent and high vehicle sealing efficiency, (ii) filter fitted Vent and reduced sealing efficiency.

Four-point measurements indicated that the average PNCs at the front seats (3.96 and 3.85×10^4 cm⁻³) were almost identical to those found at the rear seats (3.82 and 4.00×10^4 cm⁻³). The very small differences (~0.1%) suggest that the car cabin is very close to a well-mixed microenvironment. Two-point measurements revealed that the ratio of average PNCs in ($2.72 \pm 1.03 \times 10^4$ cm⁻³) and outside the car cabin ($3.75 \pm 1.62 \times 10^4$ cm⁻³) was about 0.72. A semi-empirical box mode model was introduced to predict PNCs in the car cabin as a function of those measured outside and cabin air exchange rate. Performance evaluation of the box model against statistical measures was within the recommended guidelines for urban air quality modelling. Overall, PNCs calculated by the model demonstrate a satisfactory correlation with the measured values. CFD simulations indicate that away from the Vent, emission is dispersed almost uniformly in the car cabin. Vent / CG ratios indicated that despite changes of emission filtering into the cabin, the dispersion characteristics remained almost identical at passengers' breathing height (i.e. 1.2 m from the floor).

Key words: Car cabin; Particle number concentration; Transformation processes; Nanoparticles; CFD Simulation

INTRODUCTION

Emissions from fossil fuel driven vehicles are the major source of atmospheric nanoparticle pollution in urban areas. A number of studies have indicated that the vehicular emissions substantially contribute to the adverse effects on human health (Bos et al., 2013; Hofmann, 2011). Joodatnia et al. (2013a) demonstrated that freshly emitted nanoparticles (i.e. those below 300 nm in diameter) contributed to more than 99% of PNCs inside a car cabin during journeys on the typical UK urban roads. A number of other recent studies also addressed passenger exposure to PNCs during commuting. These studies indicate that exposure rates in the car cabins are affected by their key determinants such as ventilation system, routes, traffic parameters and meteorological conditions (Knibbs et al., 2011). Our recent work has shown that the close proximity to the tail pipe of the preceding vehicle, in slow moving and congested traffic conditions, increases one second averaged PNC measurements up to two order of magnitude greater than hourly average values in a car cabin (Joodatnia et al., 2013a).

The correlation between PNCs in a car cabin and those measured outside are characterised by a number of recent studies. The ratio of PNCs in the car cabin to those measured outside the cabin (I/O) is highly influenced by the air exchange rate (A_E) (Fruin et al., 2011; Hudda et al., 2012). General consensus is that A_E increases at higher travelling speeds (Fruin et al., 2011). However, the A_E increase was greater for

older vehicles compared to newer ones due to reduction of sealing efficiency of windows and doors in older cars, making them less air tight (Fruin et al., 2011). Zhu et al. (2007) indicate that commuters in the cabin of new cars experience lower exposure to emitted nanoparticles (i.e. $I/O \sim 0.4$) compared to the older ones (i.e. ~ 0.8). Knibbs and de Dear (2010) found that I/O reduced to ~ 0.84 for filter fitted vehicles compared to vehicles with no filter (~ 1.04), with the ventilation set to intake outside air into the car cabin. Moreover, the I/O reduced further to 0.66 at lower fan settings (Knibbs and de Dear, 2010). Furthermore, significant reduction of I/O (i.e. to 0.08) was recorded when the ventilation was set to recirculation instead of intake outside air into the car cabin (Knibbs and de Dear, 2010). Particles emitted from fossil fuel driven vehicles undergo a series of complex transformation processes, which are constantly competing against each other on different time scales (i.e. nucleation $\sim 10^{-7}$ - 10^{-8} s near tail pipe, dilution $\sim 10^{-1}$ - 10^{-2} s near kerbside) (Ketzel and Berkowicz, 2004; Kumar et al., 2011). However, such information is not available for particles within the car cabin microenvironments.

A fast response differential mobility spectrometer (Cambustion DMS50) was used in conjunction with an automated switch system to measure PNCs and PNDs at multiple locations in and outside the car cabin. Such measurements provided an opportunity to study PNC distributions at four locations in the car cabin and to obtain a realistic insight into particle transformation processes in the car cabin. Moreover, in order to correlate PNCs in the car cabin to those measured outside, a semi-empirical model were introduced using a quantitative method of estimating I/O for different particle size. Using CFD simulations, the effects of filtering ratios on pollutant spatial distribution in the car cabin were also examined.

METHODOLOGY

Measurements were made on car journeys in a typical UK town, Guildford, which has about 137,000 inhabitants. Measurements were conducted on a 2.7 km long route that connects Guildford town centre to the University of Surrey. The maximum speed limit on the route was 48 km h⁻¹. The average speed of test vehicle on the route was 20 ± 5 km h⁻¹, with corresponding journey time of 9 ± 3 minutes. Measurements were made in the cabin of an unleaded petrol-fuelled car (Volkswagen Golf, 1998 registration; 1600cc engine size). All windows remained closed throughout the study periods. The car is not equipped with a heating, ventilation and air condition system (HVAC) filter. The fan-driven ventilation system (on medium speed; 2 of a scale of 1-4) was the main source of ventilation, which maintained a flow rate of 4.2×10^{-2} m³ s⁻¹ of outdoor air into the cabin (Vent). The difference between the total volumetric air flow rate in the cabin and those provided by fan assisted ventilation is the air flow through cracks and gaps (CG) in the vehicle body which is mainly due to air leak through the cabin sealing.

Instrumentation and data collection

The DMS50 was used to measure number and size distributions of particles in the 5–560 nm range at a sampling frequency of 10 Hz in conjunction with an automated four-way solenoid switching system. Two separate sets of sequential measurements were made. Firstly, at four seats in the car cabin during about 700 minutes of driving, near the breathing height of all four occupants (i.e. 1.2 m above the car floor) and, secondly, two points at the driver's seat, inside and the front bonnet outside the cabin, during about 500 minutes of driving. To minimise particle losses in the sampling tube, a short length (0.50 m) of 5 mm internal diameter tube was used. The switching system allowed 10 s sequential measurements at each location; one complete cycle for measurements at all four locations took 40 s in total. The first 2 s of data from each measurement location was discarded in order to allow for sample clearance and the remaining 8 s data at each sampling point was used for data analysis.

Semi-empirical model to predict PNCs in a car cabin

A semi-empirical model was developed to predict PNCs in the car cabin as a function of those measured just outside, as seen in Eq. (1). The box model is based on the mathematical models introduced previously by Jamriska et al. (2000) and Knibbs et al. (2010). The introduced model assumes that particle losses due to transformation processes are modest and PNCs are "well-mixed" in the cabin. The four-point measurements, time scale analysis and CFD simulations proved that these assumptions were appropriate. The losses within the ventilation system were treated by using the empirical constant I/O (He et al., 2007). Eq. (1) calculates PNCs in the cabin (N_{ci}) for particles in the size class *i* at any time (t_{n+1}) based on those measured or estimated outside (N_{Oi}) and inside the cabin in previous time step (t_n):

$$N_{ci}(t_{n+1}) = N_{oi}(t_n) X (I/O)_i + (N_{ci}(t_n) - N_{oi}(t_n) X (I/O)_i) X e^{-AE(\Delta t)}$$
(1)

Where A_E is the air exchange rate into the car cabin. The subscript *i* indicate values (e.g. N_c , *I/O* and N_O) in the D_p to D_p+d_p size range, with D_p and d_p being particle diameter and the increment between two sizes, respectively. Due to fluctuating nature of N_O , a time averaged value is employed at each time step.

Three-dimensional CFD investigation of in-cabin emission dispersion

The spatial distribution of emission dispersion in a car cabin was investigated by means of threedimensional computational fluid dynamics (CFD) using Fluent 6.3 code. Standard k-ω turbulence model was employed to simulate turbulence flow in the car cabin environment. In order to optimise the simulation time and cost, the car cabin was longitudinally split into two parts and symmetrical boundary conditions were assumed along this split plane. The grid independency of the numerical solution was carried out before employing the optimum grid number of 3 million cells. The Standard Wall Function was used for near-wall treatment. CFD simulations were performed for two scenarios: (i) emission penetration into the cabin is mainly through the ventilation system (Vent/CG = 0.9/0.1) and (ii) emission is filtered in the ventilation system (Vent/CG = 0.7/0.3). The first case indicates that the Vent is not filter fitted and vehicle sealing is most efficient allowing penetration of 90 and 10 % of the outside particulate emissions into the cabin through Vent and CG, respectively. On the other hand, the second case indicate that filter fitted ventilation is in use and the vehicle sealing are allowing penetration of only 70 and 30 %outside particles into the cabin through Vent and CG, respectively. In both cases the total pollutant concentration entered into the cabin was assumed to have a unit value and particle transformation processes (i.e. dry deposition) were ignored given the measurement results of our recent field campaigns (Joodatnia et al., 2013b). In order to simulate emission dispersion through the CG into the cabin, arbitrary inlet and outlet were defined far away from the Vent inlet.

RESULTS

Despite great temporal variability of the data at each location, the average PNCs over the 70 return trips at all four locations indicated a relatively well mixed distribution of nanoparticles in the cabin; 3.96, 3.85, 3.82 and 4.00×10^4 cm⁻³ at driver, front passenger and rear seats, respectively. Particles in the 5-30, 30-300 and 300-560 nm size ranges contribute to 35.3, 64.5 and 0.2% of average PNCs measured in the cabin, respectively. The proportion of these size ranges was nearly identical for the front and back seats. Two-point measurements indicated that the PNCs outside the car cabin ($3.75\pm1.62 \times 10^4$ cm⁻³) are ~70% greater than those measured in the cabin ($2.72\pm1.03 \times 10^4$ cm⁻³). Using the measured PNCs in and outside the car cabin, an empirical relationship was derived to estimate *I/O* for particles in 5-560 nm size range, as shown in Eq. (2).

$$(I/O)_i = 0.15 \ln(D_p)_i + 0.175$$
⁽²⁾

Time scale analysis was performed for dilution, coagulation, dry deposition and condensation processes for particles in 5-560 nm size range. Details of this analysis can be found in Joodatnia et al., (2013b). General consensus is that for all processes the shortest time scale occurs at the smallest size particle. Dilution was found to be the shortest process (36s) in the cabin, following the coagulation (620s), dry deposition processes (830s) and condensation process (3.5×10^{17} s). The time scale analysis indicates that the dilution is the dominant process in this car cabin and the other particle transformation processes have modest effect on particle size distribution. This observation indicates that the emissions in the car cabin can be assumed as passive.

CFD simulations indicated that dispersion of passive pollutant is significantly different near the Vent and CG inlets for the studied cases. However, the pollutants were found to be approximately well-mixed away from the Vent and CG inlets. This is in agreement the previous finding from four-point measurements which indicated that the PNC differences were modest at the breathing height of driver and passengers. The study of flow streamlines showed that emissions follow a complex mixing process in the cabin.



Fig. 1: Isotropic view of half of a simplified car cabin for two scenarios: (a) emission penetration into the cabin is mainly through ventilation system (Vent/CG = 90/10 %), and (b) emission is filtered in the ventilation system (Vent/CG = 70/30%). The outer boundary of the vehicle is removed and the cabin boundary is only visible.

The semi-empirical mathematical model (Eq. 1) was used to predict PNCs in the cabin using those measured outside. The total 49 trips were split into two segments by a random selection method to evaluate the performance of the proposed model against measured values. The first 25 trips was used to derive the penetration factors $(I/O)_{i}$, and the remaining 24 trips were used for comparison purposes. At each time step, the $N_{ci}(t_{n+1})$ were estimated in 10 s increments (Δt), where $N_{ci}(t_n)$ and $N_{Oi}(t_n)$ were obtained from actual measured values in the previous time step. Fig. 1.a indicates that, for each trip, the averaged predicted PNCs are in reasonably good agreement with measured PNCs, with coefficient of determination close to unity ($R^2 = 0.97$). However, Fig. 2a shows that in some cases there are over and under predictions within \pm 10% of the 1:1 ratio line. Furthermore, the performance of the box model was investigated for 10000 seconds at 10 seconds time steps. Fig. 2b shows that the coefficient of determination (R^2) between measured and predicted values are about 0.6 which is significantly reduced compared to the R^2 for the trip average predictions. The efficiency of the model is reduced due to the fact that a constant A_E was used at each time step. However, it was shown that the A_E changes due to variations in travel speed in real operational conditions. Hence, despite good estimation of cabin averaged PNCs by the model, its performance might be further improved by using appropriate A_E values at each time step.



Fig. 2: (a) Predicted and measured average PNCs in the cabin for 24 trips and (b) Predicted and measured average size-resolved PNCs in the cabin for 10000 seconds at 10 seconds time steps for particles in 5-560.

CONCUSIONS

Sequential measurements at multiple points in the car cabin indicated that PNCs at the front seats and the rear seats were almost identical, with modest difference of ~0.1%. This shows that the car cabin is a wellmixed microenvironment at the breathing height of driver and passengers. Using the two-point measurements, an expression was developed to estimate size-resolved penetration factor (I/O) as a function of particle size diameter. The average I/O was about 0.72 which is in agreement with those reported for similar ventilation system, vehicle mileage and age. The proposed expression is not universal and is highly dependent on vehicle characteristics; therefore, it will be beneficial to examine the given relation under different ventilation setting/system for other vehicles. Time scale analysis indicated that dilution was by far the shortest process in the car cabin and that transformation processes (e.g. nucleation, coagulation, condensation) have modest effect on nanoparticles size distribution. This also shows that the variation of PNCs in the car cabin was almost entirely due to dilution process. This analytical finding is in agreement with measurements at four locations in the car cabin (Joodatnia et al., 2013b). This finding should be examined under different ventilation system and settings and also for other transport microenvironments (e.g. busses, trains). CFD simulations showed that pollutants at the breathing height of passengers (i.e. 1.2 m from the floor) are dispersed almost uniformly in the car cabin away from ventilation duct. Vent/CG ratios indicated that despite changes of emission filtering into the cabin, the dispersion characteristics remained almost identical at passengers' breathing height.

The proposed semi-empirical box model predicted trip averaged PNCs close to the measured values (i.e. $R^2 = 0.97$). The size-resolved PNC predicted at 10 seconds time intervals performed within the accepted criteria for urban air quality modelling. However, it was found that the overall performance of the box model could be possibly improved by using a time dependent A_E .

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