Characterisation of nanoparticle emissions and exposure at traffic intersections through fast-response mobile and sequential measurements

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Graphical abstract

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Abstract

Quantification of disproportionate contribution made by signalised traffic intersections (TIs) to overall daily commuting exposure is important but barely known. We carried out mobile measurements in a car for size-resolved particle number concentrations (PNCs) in the 5–560 nm range under five different ventilation settings on a 6 km long busy round route with 10 TIs. These ventilation settings were windows fully open and both outdoor air intake from fan and heating off (Set₁), windows closed, fan 25% on and heating 50% on (Set₂), windows closed, fan 100% on and heating off (Set₃), windows closed, fan off and heating 100% on (Set₄), and windows closed, fan and heating off (Set₅). Measurements were taken sequentially inside and outside the car cabin at 10 Hz sampling rate using a solenoid switching system in conjunction with a fast response differential mobility spectrometer (DMS50). The objectives were to: (i) identify traffic conditions under which TIs becomes hot-spots of PNCs, (ii) assess the effect of ventilation settings in free-flow and delay conditions (waiting time at a TI when traffic signal is red) on in-cabin PNCs with respect to on-road PNCs at TIs, (iii) deriving the relationship between the PNCs and change in driving speed during delay time at the TIs, and (iv) quantify the contribution of exposure at TIs with respect to overall commuting exposure. Congested TIs were found to become hot-spots when vehicle accelerate from idling conditions. In-cabin peak PNCs followed similar temporal trend as for on-road peak PNCs. Reduction in in-cabin PNC with respect to outside PNC was highest (70%) during *free-flow traffic conditions* when both fan drawing outdoor air into the cabin and heating was switched off. Such a reduction in in-cabin PNCs at TIs was highest (88%) with respect to outside PNC during *delay conditions* when fan was drawing outside air at 25% on and heating was 50% on settings. PNCs and change in driving speed showed an

exponential–fit relationship during the delay events at TIs. Short–term exposure for ~2% of total commuting time in car corresponded to ~25% of total respiratory doses. This study highlights a need for more studies covering diverse traffic and geographical conditions in urban environments so that the disparate contribution of exposure at TIs can be quantified.

Keywords: Particle number concentration; Number size distribution; In–vehicle exposure; Respiratory deposition doses; Traffic intersections.

1. Introduction

Epidemiological studies have found an association between airborne nanoparticles (referred here to those below 300 nm to represent majority of particle number concentrations, PNCs) and cardiovascular and respiratory diseases (Gwinn and Vallyathan, 2006; Valavanidis et al., 2008). Recent studies have shown that commuters' exposure to average PNCs in urban areas can be ~1.5–times higher while travelling by car and buses compared with walking along roadsides (Kaur et al., 2005). Even a brief exposure to high PNCs can contribute to significant proportion of daily average exposure. For instance, the work of Fruin et al. (2008) showed that ~6% of total commuting time spent in car in American road environment can contribute ~36% of daily average exposure (i.e. sum of indoor, outdoor and in–vehicle exposure) to PNCs. Similarly, Dons et al. (2012) showed that ~3.6% of daily time spent in transport microenvironment (i.e. car) in Belgium contributed to ~14.5% of average daily exposure to PNCs. Similar contribution of exposure to PNCs during commuting can be expected for individuals living in UK and elsewhere (Knibbs et al., 2011). What is currently unknown from published studies is that how much fraction of total commuting exposure

occurs while travelling through the traffic intersections (TIs), which is one of focus area of this study.

As on 2013, there are 29.1 million cars and 1.2 million motorcycles in UK, comprising ~30 million personal vehicles; about 67% of UK commuters use a personal vehicle (Statistics, 2014). On an average, the whole UK population spends 1.45 h daily in a vehicle (Deborah Lader, 2006). The time use survey by National Statistics Agency in the UK during 2006 suggests an overall increase by ~16% in travelling time against the year 2000 level of 1.25 h per day (Statistics, 2003). Travelling time is increasing with time in the UK and elsewhere (Transportation, 2011), indicating a growing need of accurate characterisation of exposure at pollution hot-spots such as TIs during daily commuting.

Assessing human exposure to PNCs in urban transport environment is challenging (Ragettli et al., 2013) since PNCs varies throughout the commuting route due to constant changes in driving speed, traffic characteristics (i.e. traffic volume and percentage of petrol and diesel fuelled vehicles) and topography (Goel and Kumar, 2014). Signalised TIs are places where higher peaks of PNCs occur quite frequently compared to the rest of the route with relatively non-congested conditions – these locations are generally termed as pollution hot-spots (Goel and Kumar, 2014). Our recent review (Goel and Kumar, 2014) reported ~17- and 5-fold larger values of peak PNCs at TIs ($5.4 \pm 1.7 \times 10^5 \text{ \# cm}^{-3}$; Tsang et al., 2008) compared with average typical roadside PNCs in European ($3.2 \pm 1.6 \times 10^4 \text{ \# cm}^{-3}$; Kumar et al., 2014) and Asian ($1.2 \pm 1.0 \times 10^5 \text{ \# cm}^{-3}$; Kumar et al., 2014) urban environments, respectively. Characterisation of circumstances under which TIs become a hot-spot is therefore important

to understand for reducing the exposure to vehicle emitted PNCs during commuting. However, this aspect is largely unknown and is taken up for investigation in this study.

While a number of commuting exposure assessment studies has emerged in recent past (Hudda et al., 2011; Joodatnia et al., 2013a, b; Knibbs et al., 2010; Zhu et al., 2007), studies focusing on exposure assessment at TIs are still scarce. For example, only a few studies have carried out fixed-site measurements at TIs (Holmes et al., 2005; Morawska et al., 2004; Tsang et al., 2008; Wang et al., 2008). Given that the particle number distributions (PNDs) vary significantly within short distances from roadside, fixed-site measurements away from source may result in underestimation of commuter exposure (Fujitani et al., 2012a; Knibbs et al., 2011; Ragetti et al., 2013). Moreover, the frequency of measurements in fixed-site studies at TIs ranges from 0.0034–1 Hz (see Table 1). These measurements offer limited information about the transformation processes (e.g. nucleation, coagulation, condensation and deposition) that occur on very short time scales i.e. much lesser than 1 s (Carpentieri and Kumar, 2011; Kumar et al., 2009, 2010), clearly highlighting a need for fast response mobile measurements that are capable of capturing the particle dynamics (Kumar et al., 2011) – an aspect that is covered as a part of this study.

A few studies have assessed the effect of vehicle age, ventilation settings, driving speed and transformation of PNCs inside the car cabins during commuting. These studies have reported that in-cabin PNCs increase with the increase in age of vehicles and by switching ventilation setting from recirculation to outdoor air intake (Hudda et al., 2011; Knibbs et al., 2010; Zhu et al., 2007). Dilution is the foremost processes governing PNCs inside the cabin and their concentration at TIs can be as high $\sim 3.8 \times 10^4 \text{ \# cm}^{-3}$ on congested highways (Holder et al., 2014), which are ~ 1.2 -times larger than those generally seen along the roadsides in European

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environments ($\sim 3.2 \times 10^4 \text{ \# cm}^{-3}$) (Kumar et al., 2014). However, as analysed in current study, none of the published work so far have investigated the effect of driving and ventilation conditions on the in-cabin and on-road PNCs at the TIs.

The distinctive features that aim to fill the existing research gaps of this work are as follows. Firstly, as opposed to previous studies (Holder et al., 2014; Hudda et al., 2011; Knibbs et al., 2010; Zhu et al., 2007) that have analysed the effect of velocity variations, traffic conditions and ventilation settings on PNCs at individual *commuting routes*, this study has assessed the effect of above-noted factors on the PNCs inside and outside the car cabin at *urban traffic hot-spots* (i.e. TIs). Secondly, as compared to the previous work (Hudda et al., 2011; Knibbs et al., 2010; Zhu et al., 2007), we have monitored size-resolved distribution of particles, pseudo-simultaneously, inside and outside the car cabin under five different ventilation settings, allowing us to compare the effect of ventilation settings on in-cabin PNCs directly. Thirdly, except a few, e.g. Hudda et al. (2011), the majority of existing commuter exposure studies have focused on total particle number counts but not on PNDs that alter rapidly both spatially and temporally by transformation processes (Kumar et al., 2011). Therefore, instruments with a high scanning time (e.g. in seconds) are not suited to capture the high-resolution data of PNDs. Moreover, more than one instrument is needed to monitor the nanoparticles inside and outside at the same time. Our study overcomes these issues by using a purpose-built solenoid switching system together with a fast response differential spectrometer. This set-up has provided size-resolved data of PNDs at the fastest possible sampling rate by any commercially available instrument (i.e. 10 Hz), pseudo-simultaneously both inside and outside the car-cabin to capture PNC variations at the TIs. This feature of fast

temporal resolution and pseudo-simultaneous measurements has not covered by earlier studies.

In order to fill the above-noted research gaps, this study aims to: (i) identify the traffic driving conditions under which a TI becomes hot-spot of PNCs, (ii) assess the effect of ventilation settings in free-flow and delay conditions on PNCs at TIs, (iii) derive relationship between PNCs and change in driving speed during delay conditions at TIs, (iv) analyse the effect of geometries of TIs on PNCs during delay event, and (v) quantify the contribution of exposure at TIs to overall commuting exposure (i.e. average exposure on a study route).

2. Methodology

2.1 Route characteristics

The mobile monitoring was carried out on a 6 km long fixed route (Fig. 1a). This route was chosen with an intention to pass through maximum number of TIs with different geometrical features, traffic and surrounding built-up conditions. Selected route passed through 1 big roundabout, 10 major TIs (see details in Table 2), and 3 mini roundabouts. All these TIs had either single or double carriageways on each road (Fig. 1a). Study route is built of flexible bituminous pavement. Entire route was broadly divided in three zones according to different traffic and driving conditions,

- Zone 1 covers first 2.25 km of the route that consisted TI₁, TI₇, TI₈, TI₉ and TI₁₀. This zone includes a route passing through the main bus station, railway station and city centre. It is the most congested zone of the route with an average driving speed of 19 ± 16 km h⁻¹ and 24 ± 16 km h⁻¹ during morning and evening runs, respectively. Percentage of heavy duty vehicles (HDVs) in total fleet varied from 2 to 8% on different parts of the

roads in this zone (Transport, 2013). Width of the road in this zone varied from ~11 to 21 m and height of buildings in the 6–12 m range. TI₈ and TI₁₀ were surrounded by a canyon with an aspect ratio of ~0.8 and 0.9, respectively.

- Zone 2 covers next 2.25 km of the route that consisted TI₆, TI₅, TI₄, and TI₃. This zone includes a route passing through the local market area, residential complex and a school. This is the second most congested zone, having an average driving speed of 23 ± 14 km h⁻¹ and 20 ± 14 km h⁻¹ during morning and evening runs, respectively. Percentage of HDVs in total fleet varied from 2–4% on different roads of the zone (Transport, 2013). Width of the road in this zone varied from ~8 to 16 m while the height of buildings in the 6–9 m range. TI₅ was surrounded by a canyon with an aspect ratio of ~0.8.
- Zone 3 covers the remaining 1.5 km of the route that consisted TI₃, TI₂, and TI₁. This zone is near to a Guildford business centre and an industrial estate, the average driving speed in this zone was 25 ± 21 km h⁻¹ and 27 ± 15 km h⁻¹ during morning and evening runs, respectively. Percentage of HDVs in total fleet varied from 2–4% on different roads of this zone (Transport, 2013). Width of the road in this zone varied from ~22 to 34 m while height of buildings in the 6–8 m range.

2.2 Study design

Measurements were conducted during morning and evening peak hours of March to April 2014. Travel was made in the clock-wise direction during morning peak hours and in anti-clock wise direction during the evening hours with the intention of following the most agglomerated lanes. Out of the total 74 runs, 43 runs were made during morning hours and the rest during the evening hours. The corresponding run time during morning and evening hours run was 16 ± 4 and 17 ± 4 min, respectively. In order to assess the effect of ventilation

settings on PNDs, the following five different ventilation settings were used (see Supplementary Information, SI, Table S1).

- Setting 1 (Set₁): Windows were fully open and surface area of all four windows was $\sim 1.1 \text{ m}^2$. Ratio of surface area of windows and outside car (excluding its base area) was ~ 0.08 while the ratio of surface area of windows to those of doors was ~ 0.34 . The sequential data collected inside and outside the cabin suggest only $\sim 2\%$ difference between average inside and outside PNCs when the windows were fully open (see SI Fig. S1). Therefore the inside PNCs are considered to be equivalent to on-road (outside) PNCs when all windows were fully open.
- Setting 2 (Set₂): Windows were closed, ventilation fan was 25% on (air exchange rate, $\text{AER} = 125 \text{ m}^3 \text{ h}^{-1}$), and heating was 50% on (in-cabin temperature = 18°C).
- Setting 3 (Set₃): Windows were closed, fan was 100% on ($\text{AER} = 257 \text{ m}^3 \text{ h}^{-1}$), and heating was off (in-cabin temperature = 14°C).
- Setting 4 (Set₄): Windows were closed, fan was off ($\text{AER} = 16 \text{ m}^3 \text{ h}^{-1}$) and heating was 100% on (in-cabin temperature = 19°C).
- Setting 5 (Set₅): Windows were closed, both fan and heating was off ($\text{AER} = 17 \text{ m}^3 \text{ h}^{-1}$ and in-cabin temperature = 15°C).

AER of experimental vehicle under different ventilation settings were estimated using the methodology described in SI Section S1 that uses decay rates of measured concentrations of carbon dioxide (CO_2) as a tracer gas. In Set₁, PNDs were measured only inside the car cabin while pseudo-simultaneous measurements were conducted inside and outside the car cabin in

Set₂, Set₃, Set₄ and Set₅. Each of five ventilation settings was tested at least for 4 hours, except Set₁, for which 8 hours of measurements were taken.

During the measurement campaign, the wind speed ranged between 0.3 and 7.5 m s⁻¹, with an average of ~1.4 m s⁻¹; wind was blowing from south–west direction for most of the time. Ambient temperature and relative humidity outside the cabin varied in the 1–14 °C and 54–83% range, respectively (SI Table S2). Average temperature inside the cabin during Set₁, Set₂, Set₃, Set₄ and Set₅ was 15, 18, 14, 19 and 15 °C, respectively, while the corresponding average relative humidity was 48, 50, 49, 57 and 59%, respectively (SI Table S2). As expected, irrespective of ventilation settings, air temperature inside the cabin was higher than those outside the cabin while the relative humidity inside the cabin was lower than those outside the cabin, exhibiting an inverse behaviour, i.e. decrease in relative humidity with an increase in ambient temperature (Al-Dabbous and Kumar, 2014).

2.3 Instrumentation

A diesel–fuelled car (Ford Fiesta; 2002 registration; 1400 cc) was used for the study. Including driver, there were three non–smoking occupants in the car during the experiments. The experimental car was equipped with the filter–fitted ventilation and heating systems. A Cambustion DMS50 was deployed to measure PNDs in the 5–560 nm size range at a sampling rate of 10 Hz. A short length (0.50 m) of 5 mm internal diameter, electrically and thermally conductive sampling tube, was used to minimise particle losses during sampling (Kumar et al., 2008a). A particle residence time was ~0.6 s at the instrument’s sampling flow rate of 6.5 l min⁻¹. The sample flow rate was maintained by an electrical pump and a classifier restrictor located within the body of the instrument.

A DC powered automated solenoid switching system was used in conjunction with the DMS50 for making the pseudo-simultaneous measurements at two locations inside and outside the car cabin (Fig. 1b). The switching system was controlled by its software, allowing 15 s of measurements at each location by redirecting the sampling flow between the sampling points. This gave a total of 300 samples during each minute at both sampling points. The first 1 s of data from each measurement was discarded in order to allow for sample clearance and the final 14 s of data was retrieved for analysis.

Position and speed of vehicle was continuously recorded on a second basis (i.e. 1 Hz) using a Global Positioning System (GPS; Garmin Oregon 350). CO₂ inside the vehicle was measured at a rate of 2 Hz by COZIR CO₂ sensor, which can measure in the 0–10000 ppm range, to estimate AER under different ventilation settings (see SI Section S1). Panasonic HC–V500 camera located on dashboard of the car was used to collect the traffic flow data at the time of measurements. Air temperature, relative humidity, nitric oxide (NO), nitrogen dioxide (NO₂), sulphur dioxide (SO₂), carbon monoxide (CO) and ozone (O₃) were measured on 15 min basis by AQMesh inside the car cabin. Only the data of temperature and relative humidity are used for further analysis. Timestamps of all the instruments were matched in the beginning of the experiment.

2.4 Data acquisition

Air samples were collected at rate of 0.1 s in order to capture frequent variations in on-road and in-cabin PNCs. For Set₁, measurements were conducted for 8 h that gave a total of 2,88,000 data points. One full cycle of measurement (inside and outside) took a total of 30 s. For each of remaining four ventilation settings (Set₂, Set₃, Set₄ and Set₅), 480 full cycles and 144000 data points were collected. The system is designed in such a way that the lag time

between the switching is modest. As the particle residence time in the sampling tube was 0.6 s and we have already discarded 1 s of data, the importance of lag time became trivial.

Macros were developed in Microsoft excel to segregate the PND data inside and outside the cabin for all the five different ventilation settings. These data were then analysed by Microsoft excel with the use of DMS50 data processing tool. GPS data was converted from .gpx to excel file by map source software and spatial variations of PNCs and velocity during different runs were plotted by Arcmap version 10.1.

2.5 Estimation of exposure and delay at TI

Exposure to PNCs was quantified in terms of respiratory deposition doses (RDD) by using the methodology presented in Joodatnia et al. (2013a). RDD are directly related to number of particles inhaled and exhaled, breathing rate and duration of exposure (t). Tidal volume and breathing rate depend on age, gender and the level of activity. In this study, typical breathing rate of 18 min^{-1} and tidal volume of 0.8 l for a sitting Caucasian male adult were applied (ICRP, 1994).

Delay at a TI is the time lost by a commuter due to signal and traffic conditions. There are many types of delays (e.g. approach, stopped-time, travel-time, time-in-queue and control) that can be measured at a TI serving different purposes in transportation engineering (Darma et al., 2005). Control delay is generally measured to analyse the effect of control such as traffic signal and it is a sum of deceleration, stop and acceleration delays (Sekhar et al., 2013). Velocity-time plots were drawn for identifying the locations where deceleration and acceleration happened in the vicinity of a TI. Distance-time plots were drawn for calculating the corresponding delays. The examples of such plots for a delay event at TI₂ are shown in Fig. 2. These types of plots (velocity-time and distance-time) were drawn for each of the

delay events at TIs on the study route. A total of 220 plots, covering each of 10 TIs during 21 runs made under Set₁, which is considered equivalent to on-road measurements, were drawn to estimate the control delay at TIs. Thereafter, deceleration, stop and acceleration delays were estimated by using Eqs. (1), (2) and (3), respectively (Quiroga and Bullock, 1999) and the resulting delay times are presented in Table 2.

$$\text{Deceleration delay } (T_d) = T_{de} - T_{ds} - \left[\frac{(L_{de} - L_{ds})}{V_{ff}} \right] \quad (1)$$

$$\text{Stop delay } (T_s) = T_{se} - T_{ss} - \left[\frac{(L_{se} - L_{ss})}{V_{ff}} \right] \quad (2)$$

$$\text{Acceleration delay } (T_a) = T_{ae} - T_{as} - \left[\frac{(L_{ae} - L_{as})}{V_{ff}} \right] \quad (3)$$

where V_{ff} ($m\ s^{-1}$) is free-flow speed of vehicle. As described in Fig. 2, T (s) and L (m) represent time and location of experimental car, respectively. The first letters in subscripts represent the delay type (i.e. d , s and a are deceleration, stop and acceleration, respectively) and the second letters in the subscripts e and s are end- and start-point of delay types, respectively.

3. Results and discussions

The size of particles is an important parameter to determine their behaviour in the respiratory system and their atmospheric lifetime is a key consideration in assessing the associated health effects. Therefore, measured PNCs have been divided in three size ranges for some of the discussions: 5–30 nm (nucleation mode; and PNC in this range is represented by N_{5-30}); 30–300 nm (accumulation mode, N_{30-300}) and 300–562 nm (coarse mode; $N_{300-562}$). The overall framework of the results presented in this section is presented in SI Fig. S3.

3.1 Particle number size distributions and concentrations during *overall* journeys

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3.1.1 On-road and in-cabin PNDs

Fig. 3a shows average PNDs during the morning and evening runs during Set₁. These show similar trimodal pattern with peaks at ~5, 10 and ~75 nm. This type of PNDs are observed in case of slow moving traffic, leading to somewhat decreased separation between experimental car and the vehicle ahead, compared with free-flow conditions, resulting in higher PNCs in the 5–30 nm range (Section 3.3). During all other settings (Set₂, Set₃, Set₄ and Set₅), the shape of average PND curves inside the cabin are similar to those measured outside the cabin, without an appreciable changes in peak diameters. Irrespective of ventilation settings, the magnitude of in-cabin PNDs was small compared with the corresponding outside PNDs (Fig. 3b). It shows that the in-cabin space serves as a damping chamber for outside particles (Zhu et al., 2007). Most of the differences between inside and outside the average PNDs were observed in ultrafine particle range (diameter <100 nm), as seen in Fig. 3b. Maximum reduction in magnitude of inside PNDs with respect to those found outside was noted in Set₄, followed by Set₅. It is a result of limited intake of outside air and hence limited penetration of particles inside the cabin during these settings (see Table 3).

3.1.2 On-road PNCs

Fig. 3c shows the run-wise total PNCs in selected size ranges for all the five different ventilation settings. In Set₁, average PNCs ($7.6 \pm 3.8 \times 10^4 \text{ cm}^{-3}$) for all morning runs were similar to those obtained during the evening runs ($8.8 \pm 3.5 \times 10^4 \text{ # cm}^{-3}$), except for the run 20 when the experimental car was following a 42 year old diesel car. Due to this event, average PNCs in the 5–30 nm range during run 20 increased by about a factor of 4 relative to average PNCs in the same size range for all other runs ($4.7 \times 10^4 \text{ # cm}^{-3}$). Contribution of average PNCs in the 5–30 nm range during morning and evening runs was 61% and 55%, respectively, and as expected, fraction of PNCs in the 300–562 nm range was negligible

(Joodatnia et al., 2013a, b). Overall average PNCs ($8.1 \pm 3.6 \times 10^4 \text{ \# cm}^{-3}$) during Set₁ were comparable with the previous studies, i.e. $7.0 \pm 2.9 \times 10^4 \text{ \# cm}^{-3}$ in Pirjola et al. (2006), $9.5 \pm 2.9 \times 10^4 \text{ cm}^{-3}$ in Johnson et al. (2009) and $9.3 \pm 3.4 \times 10^4 \text{ cm}^{-3}$ in Kim et al. (2014). In few cases, our PNCs were higher. For example, we found 1.6– and 4.5–times higher PNCs in our case than those reported in Somerville USA (Padró-Martínez et al., 2012) and Germany (Hagemann et al., 2014), respectively (see Table 4). Part of these differences could be explained by the collection of the data at a lower sampling rate, i.e. 1s and 3s in USA and Germany, respectively, compared with 0.1 s in our study. Low sampling rate is most likely to miss the peak PNC events that occur within a fraction of seconds (Kumar et al., 2010; Joodatnia et al., 2013a), resulting in under estimation of PNCs. As also discussed in Kumar et al. (2014), dissimilarities in the characteristics of driving route such as traffic intensity, congestion, differences in the proximity of other vehicles (i.e. sources) to the experimental car and effects of meteorological conditions could have also contributed to the observed differences.

3.1.3 In-cabin PNCs and their effect on inside to outside ratios

Average PNCs inside the cabin were found to be the lowest in Set₅, followed by Set₄, Set₂ and Set₃ (see Fig. 3c and Table 3). It is the result of limited air intake, which is evident from ~88% and 95% smaller AER values in both settings (Set₅ and Set₄) compared with Set₂ and Set₃, respectively. Irrespective of ventilation settings, average PNCs in the 30–300 nm range were almost similar both inside and outside the cabin (Fig. 3c). This is expected due to a much larger sink of nucleation mode (5–30 nm) particles due to their relatively higher diffusivity compared with particles in the 30–300 nm range (Kumar et al., 2008b; Lee et al., 2014). Whilst average PNCs in the 5–30 nm range inside the cabin were found to be dependent on individual settings, these increased with the outdoor air intake due to change in

fan settings. Therefore, the ratio of inside to outside average PNCs (*I/O*) was mainly influenced by the changes in PNCs in the 5–30 nm size range. This is mainly because this size range contributes to a major fraction of total outside (~60%) and inside (~50%) PNCs during different ventilation settings (Table 3). Consequently, the *I/O* was highest for Set₂ (AER = 277 m³ h⁻¹), followed by Set₃ (AER = 128 m³ h⁻¹), Set₄ (AER = 16 m³ h⁻¹) and Set₅ (AER = 14 m³ h⁻¹); see Table 3. We have not investigated the effect of relative humidity on nanoparticles behaviour inside the cabin. This is mainly due to small variability in relative humidity ($\pm 5\%$) observed during our measurement campaign, which do not allow a thorough investigation of the effect of relative humidity on in-cabin PNCs.

The values of *I/O* presented in Table 3 are not directly comparable with the published studies because of measurements being made under varying ventilation settings on different type of vehicles (in terms of age, mileage, and filter efficiency). However, we have made an attempt to put our results in the perspective of available published studies so that any factors contributing to these differences can be highlighted. For instance, the *I/O* reported in our study for Set₅ is substantially smaller than those reported in Zhu et al. (2007), Hudda et al. (2011) and Knibbs et al. (2010) for the identical ventilation setting (see Table 5). These studies were conducted in *free-flow* traffic conditions such as on a freeway (vehicle speed 82 km h⁻¹; Zhu et al., 2007), on the roads with little vehicular traffic (52 km h⁻¹; Hudda et al., 2011), or in tunnel environments (80 km h⁻¹; Knibbs et al., 2010) compared with our study on an urban route in a congested traffic environment (27 km h⁻¹). Outside PNCs during free-flow traffic conditions were generally found to be ~29-times lower as compare to congested driving conditions in our case (Section 3.3). Therefore, even if inside PNCs are

identical in free-flow and congested traffic conditions, the *I/O* will still be higher for the free-flow conditions as compare to congested driving conditions.

Fig. 4 shows the effect of ventilation settings on size-dependent *I/O*. Interestingly, these *I/O* in Set₂, Set₃, Set₄ and Set₅ fit well with a power-law function, showing R^2 as 0.94, 0.94, 0.66 and 0.69, respectively. Moreover, slope of curve is almost similar in Set₄ (slope = 0.48) and Set₅ (slope = 0.40) that is about twice the slope obtained in Set₂ (slope = 0.29) and Set₃ (slope = 0.21), as seen in Fig. 4. It clearly shows the impact of the fan settings on the *I/O*. For instance, when the fan setting was at 25% and 100% on during Set₂ and Set₃, respectively, *I/O* increased the in-cabin PNCs in the 5–30 nm range by ~13% and 26% in Set₂ and Set₃ as compare to both Set₄ and Set₅, respectively (Table 3).

3.2 Spatial variations in *on-road* PNCs

In order to assess the spatial variations, average PNCs inside and outside the cabin were normalised and the resulting variations are presented in SI Fig. S4. These normalised values were obtained by dividing the PNCs at each point of time by the peak PNC value for every individual run. Since the total run time was nearly identical for each individual setting, normalised values obtained for each individual run were averaged to present an overall picture of inside and outside the PNCs for different settings (SI Fig. S4). In each setting, PNC series could clearly be divided into three zones, as described in Section 2.1. First 400 s of the run was the travel time taken by the vehicle to traverse the portion of the route in zone 1, which is the most congested and highly built-up part of the route. This resulted in 1.3- and 1.1-times higher on-road PNCs compared with those in zones 2 and 3, respectively. Relatively lower concentrations in zones 2 ($8.0 \times 10^4 \text{ \# cm}^{-3}$) and 3 ($9.4 \times 10^4 \text{ \# cm}^{-3}$) where our experimental vehicle travelled during the next 400–900 s and remaining ~300 s, respectively, were mainly

due to a less congested route in zone 2 and relatively larger green cover around the road and least vehicle congestion compared with the rest of the route in zone 3.

PNDs showed trimodal shapes with peaks at ~5, 10 and 65 nm in each of the three zones, but their magnitude varied depending on traffic intensity in each zone (see SI Fig. S5). Furthermore, spatial variation of on-road 1 s average PNCs during a typical morning run clearly indicates that peak PNCs mostly occur in the vicinity of TIs (Fig. 5a). Based on visual interpretation of video footage, it was found that acceleration after idling at TIs was a dominant factor that correlated well with the peak PNCs on the study route. Further discussions on the effect of driving conditions on PNDs are discussed in Sections 3.3 and 3.4.

3.3 Effect of driving conditions on *on-road* and *in-cabin* PNCs at the TIs

A vehicle can travel through the TI in uninterrupted (i.e. free-flow) or interrupted (i.e. delay) traffic flow conditions. PNDs showed similar shapes of trimodal distributions with peaks at about 5, 10 and 80 nm during both the free-flow and delay conditions (Fig. 5b). However, the magnitude of average PNDs at 5, 10 and 80 nm was 14, 12 and 3-times higher during delay conditions compared with free-flow conditions. This was a result of sudden acceleration in speed of vehicles, quickly emitting a large amount of particles (Jayaratne et al., 2010). Larger differences between the peaks in average PNDs during free-flow and delay conditions at ~5 and 10 nm as compare to 80 nm show that nucleation is a dominant process under peak PNC conditions at the TIs.

Furthermore, peak PNCs at TIs during delay events were up to 29-times higher than average PNCs under free-flow conditions (Fig. 5c). Some of this difference is brought by the PNCs in the 5–30 nm range. Their contribution in peak PNCs was ~11% higher during delay conditions compared with free-flow conditions. Further differences could be due to a number

of varying driving conditions, as evident from the distinct PNDs observed at TIs when experimental vehicle was (i) accelerating with no other diesel vehicles around, (ii) idling during congestion, and (iii) accelerating in proximity of diesel vehicle (Fig. 5d). For instance, PNDs during (i) and (iii) were bimodal in shape, with a much higher peak in the 5–10 nm range during (iii) than during (i). On the other hand, PNDs during (ii) were dominated by particles in the 20–40 nm range (Fig. 5d). The dominance of these particles could be due to the following reasons. Firstly, engine emissions from the idling vehicles are generally dominated by 20–100 nm size particles (Wang et al., 2006) and therefore these appears to be the fresh exhaust emissions from the vehicles. Secondly, small contribution may come from growth of smaller size particle (5–10 nm) due to coagulation as time scale of coagulation during idling is ~ 100 s (based on Fig. 3 in Kumar et al. (2011) for our PNC values of $4.25 \times 10^5 \text{ \# cm}^{-3}$) and maximum delay time at TI was $\sim 69 \pm 26$ s (see Table 2).

Average on-road PNCs during the acceleration ($0.8 \times 10^5 \text{ \# cm}^{-3}$) were nearly twice to those measured during the idling ($0.4 \times 10^5 \text{ \# cm}^{-3}$), with a maximum difference being up to ~ 6 -fold (Fig. 5f). Furthermore, the proximity of the experimental car to diesel-fuelled vehicle was found to further increase the average PNCs over 20-times compared with average PNCs during acceleration when away from a diesel vehicle (Figs. 5e–f). Thus, on-road peak PNCs depends predominantly on the number and type of surrounding vehicle, frequency of traffic light, acceleration and congestion. On the other hand, ventilation settings showed a major influence on the in-cabin PNCs, as discussed in the subsequent text.

To assess the effect of ventilation settings on in-cabin PNCs at TIs, events corresponding to free-flow, acceleration and idling in each of four ventilation settings (Set₂, Set₃, Set₄ and Set₅) are extracted. Detailed discussion on PNDs inside and outside the cabin for a

combination of ventilation settings and driving conditions is provided in SI Section S2 and size-resolved I/O are shown in the Fig. 6. Set₂ was observed to be the worst setting since it had the highest I/O (0.90) in the 5–300 nm range during *free-flow* traffic conditions as opposed to Set₃ (I/O = 0.63) that turns out to be the worst setting during acceleration during *delay conditions*. Set₅ emerged as the best setting during *free-flow* traffic conditions, with the lowest I/O (0.30) in the 5–300 nm range (Fig. 6a–b). Set₂ was found to be the best setting (I/O = 0.12) during acceleration in delay conditions. As far as the idling conditions are concerned, Set₂ (I/O = 0.75) and Set₄ (I/O = 0.06) were found to be the worst and the best settings, respectively (Fig. 6c). Based on the above discussion, it was found that effect of ventilation setting is not similar in all the traffic driving conditions. Therefore, consideration of driving conditions is important for an accurate estimation of in-cabin PNC exposure that can be approximated using the above-derived I/O and the on-road PNC data.

3.4 Effect of geometrical features of TIs on PNCs

Geometrical features such as shape and built-up area around a TI is another important factor that affects the dispersion of particle number emissions and hence their distributions and concentrations. As summarised in Goel and Kumar (2014), some numerical modelling and wind tunnel studies have analysed effect of geometrical features on the dispersion of pollutants at TIs. For example, the effect of built environment on wind flows at TI was studied by Brixey et al. (2009). They found that placing a small tower at the corner of street canyon around a TI enhanced the wind speed in (and ventilation from) the street canyon. Both the shape of TIs and built-up area around them in individual zones were nearly identical (Section 3.2). Therefore one TI from each of the three zones (TI₂, TI₄ and TI₈ in zones 1, 2 and 3 respectively), having the highest median of short-term (~0.1 s) average PNCs during delay conditions at that TI, is selected as an example case for further discussion.

The relationship between PNCs and acceleration of vehicle is greatly influenced by geometrical features of TIs. For instance, at four-way perpendicular TIs (e.g. TI₂ and TI₄), PNCs increased with an increase in acceleration (Figs. 7b–c). While at a densely packed three-way TI (e.g. TI₈), PNCs decreased with an increase in acceleration (Fig. 7d). This can be explained by counteracting effect of built-up area, which limits the dispersion and the traffic produced turbulence (TPT) that enhances the mixing of exhaust emissions. Irrespective of any shape of a TI, the TPT will always have an effect on mixing of emissions within the first few meters of the height above the road (Al-Dabbous and Kumar, 2014b; Kumar et al., 2008b), depending on the driving speed of a vehicle (Di Sabatino et al., 2003). At a three-way TI, average driving speed of the experimental car during acceleration was measured to be about twice the average driving speed at the four-way TI, explaining a decrease in PNCs with increasing acceleration at the three-way TI. Of course exchange between the polluted air at the road level and the clean air above in urban canopy layer is expected to be limited due to the surrounding built-up area at the three-way TI, leading to a possible build-up of PNCs. This effect of exchange seems to have been overshadowed by the much higher mixing effect of TPT at the three-way TI compared with four-way TI, giving a declining trend in PNCs during acceleration. For an in-depth understanding, there is clearly a need for detailed modelling (physical or numerical) studies to apportion the impact of built-up area and TPT on pollutant dispersion around such TIs.

As for acceleration, measured data did not show any clear trend to derive a relationship between the delay time and the PNCs but some interesting observations were noted. Fig. 7a shows the comparison of short term (~0.1 s) average PNCs during delay conditions at 10 different TIs in all the three zones (see Table 2 for statistics). 95th percentile of average PNCs

was highest at TI₂ in zone 3, followed by TI₄ in zone 2 and TI₈ in zone 1, respectively. Although the video footage suggested a highest traffic volume at TI₄, highest average PNCs were yet seen at TI₂. This was predominantly the result of following a 42-years old diesel car by our experimental vehicle at TI₂. Median of average PNCs was noted to be the highest at TI₈, TI₄ and TI₂ among the TIs in zone 1, 2 and 3, respectively. These highest values were presumably due to frequent start and stop driving conditions that were also highlighted by higher standard deviation of driving velocity at these TIs, as seen in Table 2, compared with the other TIs in the respective zones.

3.5 Relationship between PNCs and driving speed at TIs

Regression analysis between PNCs and acceleration of the vehicle (i.e. driving speed at time i subtracted by speed at time $i-1$) was performed for all the 10 TIs. However, a presentable relationship was only found for 6 TIs (i.e. TI₁, TI₂, TI₄, TI₇, TI₈ and TI₁₀), as seen in Figs. 7b–d (see also SI Fig. S10). Further analysis of these (exponential) relationships (SI Section S3), allowed us to develop a generalised form that is seen through Eq. (4).

$$(PNC)_i = (PNC)_{avg} \times \exp \left[a \left(\frac{dv}{dt} \right)^3 + b \left(\frac{dv}{dt} \right)^2 + c \left(\frac{dv}{dt} \right) \right] \quad (4)$$

where PNC_i is the PNC at any time i during a delay event at the TIs (see Section 2.5); PNC_{avg} is the average PNC during the delay event; dv/dt is change in driving speed (i.e. difference in driving speed at time $i-1$ and time i), and a , b and c are coefficients based on geometry and built-up area around TIs. The values of ' a ', ' b ' and ' c ' differ based on geometry and built-up area around an individual TI. Table 6 presents a range of their values, which were derived from our experimental data presented in Figs. 7b–d and SI Fig. S10 for different types of TIs. Relationship between coefficients ' a ', ' b ', ' c ' and physical parameters (i.e. delay time, wind

speed, road width and building height) is shown in SI Fig. S11. In case of four-way TIs, a strong linear correlation ($R^2 = 0.83$; p-value <0.05) is found between ' a ' and delay time (SI Fig. S11a) while ' a ' depends linearly on wind speed ($R^2 = 0.81$; p-value <0.05 ; SI Fig. S11b) in case of three-way TIs. Coefficient ' b ' showed a linear dependency with delay time ($R^2 = 0.72$; p-value <0.05) at four-way TIs (SI Fig. S11c) while no such dependency was observed at three-way TIs. A strong linear correlation ($R^2 = 0.80$ and p-value <0.05) is found between ' c ' and ratio of delay time and building height at four-way TIs while even a stronger correlation ($R^2 = 0.91$ and p-value <0.05) was found between ' c ' and wind speed at three-way TIs (SI Fig. S11d-e). These values are TI specific and should be generalised cautiously. However, relationships obtained here could be used to estimate the preliminary values of these coefficients at the TIs elsewhere.

Since the PNCs are directly proportional to particle number emission factors (PNEFs), the above presented relationship can be used to modify the average roadside PNEFs to take into account of frequent driving changes at the TIs. Apart from driving conditions and geometrical features, transformation processes also play an important role in changing PNCs between the source and receptors at TIs, as discussed in Section 3.6.

3.6 Fine scale analysis of peak PNC events at TIs

Temporal variability in in-cabin peak PNCs corresponding to outside peak PNCs is discussed in SI Section S4. It is found that in-cabin peak PNCs follows similar temporal variations as of on-road peak PNCs in Set₂ and Set₃ with a lag period of 30–60 s for outside and inside peak PNCs. This lag period in Set₄ and Set₅ increased up to ~90 s due to lesser ingress of outdoor air during these settings (Set₃ and Set₄).

To assess the effect of transformation processes, we have selected inside and outside PNDs averaged over 1 s duration during the peak PNC events for detailed discussion. These PNDs are normalised by dividing with their corresponding total PNC (Fig. 8). This normalisation was carried out to highlight the effect of relative transformation processes for removing the effect of dilution (Carpentieri and Kumar, 2011).

These PNDs showed a number of distinct shapes during different time steps, depending on traffic driving conditions and ventilation settings which were important for inside PNDs. For instance, bimodal PNDs, with peaks at about 10 and 100 nm, were observed during free-flow traffic driving conditions (Fig. 9). These changed to unimodal PNDs, with a peak at about 10 nm, during the acceleration of experimental car and the vehicles ahead (Fig. 9). Three main conclusions can be drawn these normalised PNDs. Firstly, some of events showing perfectly superimposed PNDs at different time steps indicate dilution as a key process. For instance, inside and outside PNDs during Set₂, and inside PNDs during Set₅, demonstrated this PND behaviour at different time steps (Figs. 8a and 8d). Secondly, shapes of PNDs *inside* the cabin were nearly unchanged compared with those measured outside (Figs. 8 and 9). These unchanged shapes can be explained by restricted penetration of outside air inside the cabin due to the effect of ventilation settings. Thirdly, a number of diverse shapes of *outside* PNDs and their corresponding GMDs were found to be a function of distance from the source and this distance varied frequently during stop- and go-situation at TIs (Fig. 9). For instance, in Set₄, the GMD increased from 16 nm to 26 nm when distance between experimental and preceding vehicle at TI increased from about 2 to 5 m.

3.7 Contribution of exposure at TIs to overall commuting exposure

On-road average RDD rate varied from run to run, showing the largest RDD during those runs when the PNCs were dominated by the freshly emitted particles in 5–30 nm range (Fig. 10a). For instance, the highest RDD values were ~3-times the overall average ($2.8 \pm 2.4 \times 10^9 \text{ \# min}^{-1}$ or $7.6 \pm 6.5 \times 10^6 \text{ \# m}^{-1}$) for run 20 where the PNC contribution of 5–30 nm size particles towards the total PNC was 82% compared with the overall average of 58%.

Average RDD rate during delay period at TIs were found to be up to 7-times higher than those during the remaining run period. Short-term exposure under delay conditions at TIs contributed to a significant proportion of commuting exposure. For instance, percentage of time spent under delay conditions at TIs during run 19 was only 27 s (i.e. 2% of total commuting time) but the RDD was 1.1×10^{10} particles (i.e. 25% of total RDD during commuting) (Fig. 10b). One of the prominent reasons for this exposure are the changes in driving conditions during delay periods at the TIs, which lead to increased fuel consumption and result in greater amount of particle number release (see Section 3.3). Availability of RDD rate for a direct comparison is limited, but we have made an attempt to compare our results with other best possible matching studies. For instance, Int Panis et al. (2010) estimated the average RDD rate for a male cyclist as $4.6 \times 10^6 \text{ \# m}^{-1}$ in Brussels. Our RDD estimates for our entire route and TIs are 1.6- and 14-times higher, respectively, than those reported by Int Panis et al. (2010) in their study in Brussels. Some of the differences can be explained by the instrument employed (ultrafine particle counter, UPC) that measured particles in the 20–1000 nm size range, whereas the cut-off size range for the DMS50 employed in this study was 5 nm. Based on measured data in our study, PNCs in the 5–20 nm size range can contribute ~48% of total PNCs (see Section 3.1). Therefore, a factor of

two differences is not surprising for the overall route, given also the influence of other factors such as traffic composition and driving conditions along the route that could have contributed to the dose estimates. For example, maximum traffic volume in their study was about 50,000 veh day⁻¹ while this more than two-times (1,13,680 veh day⁻¹) on our study route. Although average driving speed on their route (~19 km h⁻¹) was almost similar to those (~22 km h⁻¹) observed in our study.

We used the RDD and AER data to derive a relationship between them. Average in-cabin RDD rate were found to increase linearly ($R^2 = 0.98$) along with an increase in AER (see SI Fig. S12d). As expected, this linear trend between in-cabin RDD and AER mimics the trend between the in-cabin PNCs and AER since RDD rate is directly proportional to in-cabin PNCs (see SI Fig. S12). As expected, median RDD rate during Set₃ is highest ($1.30 \times 10^7 \# \text{ s}^{-1}$; AER: 257 m³ h⁻¹), followed by Set₂ ($0.86 \times 10^7 \# \text{ s}^{-1}$; AER: 125 m³ h⁻¹), Set₄ ($0.38 \times 10^7 \# \text{ s}^{-1}$; AER: 16 m³ h⁻¹) and Set₅ ($0.28 \times 10^7 \# \text{ s}^{-1}$; AER: 17 m³ h⁻¹). Alternatively, reduction in in-cabin RDD rate with respect to on-road RDD rate was highest during Set₅ (75%), followed by Set₄ (71%), Set₂ (67%) and Set₃ (19%), respectively (Fig. 10c). These data suggest that keeping the fan setting at minimum favours the reduction in in-cabin RDD rate throughout the route. As discussed in Section 3.1.3, AER increases with increasing fan settings and therefore an increase in *in-cabin* PNCs is expected that results in higher RDD. It is evident from the above discussion that TIs contribute notably to overall daily overall commuting exposure and that in-cabin exposure at TIs can be reduced significantly by keeping the fan setting at minimum level.

4. Synthesis and future research

A DMS50 in conjunction with a solenoid switching system was used to measure number and size distribution of particles in the 5–562 nm range. These measurements were carried out pseudo–simultaneously inside and outside the car cabin under five different ventilation settings during commuting on a typical urban route. The study aims to identify the conditions under which TIs becomes hot–spots and to analyse the effect of different ventilation settings, driving conditions and layout of TIs on particle number and size distributions. The contribution of exposure doses towards the overall commuting exposure at TIs is estimated.

The key conclusions from the study are as follows:

- Measurements were taken inside and outside the cabin in five different ventilation settings. Irrespective of ventilation settings, trimodal PNDs were observed both inside and outside the cabin showing peaks at about 5, 10 and 75 nm. Magnitude of inside PNDs was always lower than that of outside the cabin. During delay and free–flow conditions at TIs, PNDs were similar to those measured for overall route. Peak in nucleation mode during delay condition was found to be ~14–times higher than that observed during the free–flow conditions.
- In–cabin average PNCs were found to be largely dependent on the AER. For example, maximum reduction (~75%) in in–cabin PNCs with respect to outside PNCs was observed when AER was nearly the lowest (e.g. $\sim 17 \text{ m}^3 \text{ h}^{-1}$ during Set₅). Variations in heating inside the car did not show any significant impact on in–cabin PNCs. A strong correlation ($R^2 = 0.96$) in a power form is found between the *I/O* and AER for the studied runs. Driving conditions were found to have an effect on ingress of particles inside the

cabin. Therefore, it is important to derive driving condition specific *I/O* and the ones reported in our study can be used to approximate the in-cabin PNCs elsewhere.

- As vehicles produce higher particle number emission during congestion, dispersion of exhaust emission is also limited by the surrounding built-up area, resulting in highest PNCs at the most congested and highly built-up part of the route. *Average* PNCs at TIs during *delay* conditions can be up to 29-times higher as compare to *free-flow* conditions due to changes in driving (deceleration, idling and acceleration) conditions at the TIs. The largest PNCs were found during delay conditions at the TIs when test vehicle was accelerating either in proximity of tailpipe of a preceding vehicle or running in a parallel lane. During a typical delay condition at the TIs, average PNCs during acceleration were found up to ~6-times higher than those during idling conditions.
- Change in driving speed and average PNCs in delay conditions at TIs were found to follow an exponential relationship. Geometrical shape of TIs and the built-up area around them significantly changed the values of coefficients (e.g. a, b and c) in this relationship, which can applied to *average* PNEFs for making them suitable for use in dynamic driving conditions at the TIs.
- It is found that RDD rate during delay period at TIs can be up to 7-times higher than those during the rest of the journey period. In fact, about 2% of journey time exposure under delay conditions at TIs found to contribute up to ~25% of total commuting exposure doses.

This study presents hitherto missing information related to effect of dynamic driving conditions on nanoparticle concentrations and exposure at the TIs. Further studies covering a diverse range of TIs in different geographical settings are needed to develop a database

that could assist in estimating contribution of exposure at TIs towards the daily commuting exposure in diverse city environments. There is also a need of physical modelling (i.e. wind tunnel) studies that can help apportioning the effect of mixing due to surrounding built-up environment and the TPT on the PNCs at the TIs. Together, the findings of field studies and the physical modelling studies could help in developing better suited dispersion models that can reliably predict the PNCs and associated exposure at the TIs.

5. Supplementary Information

This section includes Sections S1–S4, Figures S1–S20 and Tables S1–S2.

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List of Figure Captions

Fig. 1. (a) Map of the study route showing velocity variations during a typical morning run.

Traffic intersections covered are marked with the blue circles; (b) experimental setup showing instruments and monitoring locations inside (i.e. 1) and outside (i.e. 2) the car cabin.

Fig. 2. An example of (a) distance versus time, and (b) velocity versus time, plots of an experimental car during a delay event at TI₂. Acronyms used in this figure are the same, which are used in Eqs. (1) to (3) and explained in Section 2.5.

Fig. 3. (a) In-cabin PNDs for Set₁, Set₂, Set₃, Set₄ and Set₅, respectively. (b) Outside PNDs for Set₁, Set₂, Set₃, Set₄ and Set₅, respectively. (c) Outside (*O*) and in-cabin (*I*) PNCs in three different size ranges (5–30, 30–300 and 300–560 nm) during Set₁, Set₂, Set₃, Set₄ and Set₅. Please note that run numbers 1–12, 1–5, 1–6, 1–9 and 1–14 were taking during morning hours for the Set₁, Set₂, Set₃, Set₄ and Set₅, respectively, and the rest of the runs represent evening hours. For the sake of clarity only positive standard deviations are shown.

Fig. 4. Size-dependent *I/O* for all the studied settings (i.e. Set₁, Set₂, Set₃, Set₄ and Set₅), showing a power-law increase in *I/O* with the increasing diameter.

Fig. 5. (a) Spatial variations of PNCs during a typical morning run of Set₁. (b) Average PNDs during delay and free-flow events at TIs; these PND values are computed by averaging 96 and 124 number of delay and free-flow events (each having a duration of about 52 and 4 seconds, respectively) at all the TIs. For the sake of clarity, only positive standard deviations of delay events are shown in this figure. (c) Box plot of short-term (~0.1 s) average PNCs during the delay and 4-s average PNCs during free-flow conditions at all TIs on the studied route. Pie chart represents the overall proportion of PNCs in two size ranges (5–30 and 30–300 nm). The upper and lower whiskers represents 5th and 95th percentile. The upper boundary of the box is the 75th percentile; line inside the box is the median and the lower

boundary of the box is 25th percentile; square signs represent the mean value. (d) comparison of PNCs and velocity during idling and acceleration at TI when traffic volume was low, (e) comparison of PNCs and velocity during idling and acceleration at TI when experimental car was in proximity to diesel vehicle, and (f) PNDs for selected events for three different traffic conditions at TIs: (i) Idling, (ii) acceleration with no other diesel vehicle around, (iii) acceleration in vicinity of diesel vehicle.

Fig. 6. Size-dependent *I/O* in four different size ranges i.e. 5–30, 30–100, 100–300 and 5–300 nm during (a) free-flow, (b) acceleration and (c) idling conditions at TIs.

Fig. 7. (a) Box plot of short term (~0.1 s) average PNCs during delay events at 10 TIs. Relationship between acceleration of vehicle and natural logarithm of PNCs during delay period at (b) TI₂, (c) TI₄, and (d) TI₈. Rectangular box on each of the plots represents the geometry of TI along with width of its different legs.

Fig. 8. Temporal changes in normalised PNDs averaged over 1s during peak PNC events of selected runs (a) 10, (b) 12, (c) 6, and (d) 13 of Set₂, Set₃, Set₄ and Set₅, respectively. Each line represents 1s averaged PNDs during peak PNC events. Here inside PNDs during Set₂ and Set₅ showed perfectly superimposed PNDs at different time steps representing dilution as a key process.

Fig. 9. Separated distinct 1s average PNDs and GMD during Set₃, Set₄ and Set₅. PNDs in each of ventilation settings are marked by same colour rectangle and number in bracket in the title of each figure represents the total number of distinct PNDs in respective ventilation setting. Traffic driving condition corresponding to each of distinct PND is also noted in each figure.

Fig. 10. (a) Respiratory deposition dose (RDD) rate of peak PNCs under delay at TIs and remaining journey (i.e. overall journey–delay period) in Set₁ (windows open, fan and heating

off). (b) Box plot of average RDD rate of overall journey under four different ventilation settings. Percentage reduction in average RDD rate with respect to average outside RDD rate under these different ventilation settings is also presented on secondary axis. The upper and lower whiskers of box plot represents 5th and 95th percentile. The upper boundary of the box is the 75th percentile; line inside the box is the median and the lower boundary of the box is 25th percentile. Square represents the mean value. (c) Percentage contribution of delay period (red bar) and remaining journey in total commuting RDD. (d) Percentage contribution of time spent during delay period (red bar) and remaining journey.

List of Tables

Table 1. Summary of fixed site monitoring studies at TIs.

Author (year)	City (Country)	Instrument	Sampling Frequency (Hz)	Size range (nm)	Maximum PNC ($\times 10^5 \text{ cm}^{-3}$)	Traffic Density (h^{-1})	HDV (%)	Months
Morawska et al. (2004)	Salzburg (Austria)	SMPS	300 ^a	13–830	0.2 ^b	3600	20–30%	September
Holmes et al. (2005)	Brisbane (Australia)	SMPS	120 ^a	9–407	2.6	200 – 1920	20% ^c	January
Tsang et al. (2008)	MongKok of Kowloon (Hong Kong)	WCPC	1	5–2000	5.4	840	29%	July
Wang et al. (2008)	Texas (USA)	CPC & SMPS with DMA	120 ^a	7–290	2.4	10452 – 11897	3.7%	December–June
Fujitani et al. (2012b)	Kawasaki City (Japan)	SMPS	–	8–300	~1.4	2167	25%	January

Note: ^aScan time in seconds; ^b90th percentile; ^cPercentage of diesel vehicles including cars.

Table 2. Description of studied TIs and the summary of PNC, GMD and delay periods obtained during measurements; ‘ σ ’ stands for standard deviation and ‘ $\bar{N}$ ’ represents average PNCs.

Traffic intersections	Description	$\bar{N}_{5-560} \pm \sigma_{5-560}$ during delay event (cm^{-3}) $\times 10^5$		GMD (nm)	Average velocity during delay $\pm \sigma$ (km h^{-1})	Control delay $\pm \sigma$ (s)
		5–30 nm	30–300 nm			
TI ₁	Intersection of two A-class roads near commercial zone of the city. It caters traffic from town centre towards commercial hub in north direction, business park in west direction and slyfield industrial estate in south direction	1.01 $\pm$ 0.69	0.64 $\pm$ 0.34	22 $\pm$ 3	5 $\pm$ 7	57 $\pm$ 31
TI ₂	Intersection of two A-class roads. Slyfield industrial estate is located in north direction and Guildford business park in west direction	3.15 $\pm$ 5.42	0.42 $\pm$ 0.22	13 $\pm$ 2	6 $\pm$ 9	69 $\pm$ 26
TI ₃	T-type intersection of A-class and b-class road. Slyfield industrial estate in north and stroke park in the south side of the intersection.	0.84 $\pm$ 0.84	0.29 $\pm$ 0.22	16 $\pm$ 3	8 $\pm$ 7	42 $\pm$ 8
TI ₄	Intersection of two A-class roads. Intersection is surrounded mainly by residential area	2.64 $\pm$ 3.35	0.40 $\pm$ 3.10	11 $\pm$ 2	7 $\pm$ 13	53 $\pm$ 24
TI ₅	T-type street canyon intersection. Intersection is surrounded by local market area.	0.36 $\pm$ 0.32	0.31 $\pm$ 0.24	26 $\pm$ 3	10 $\pm$ 7	28 $\pm$ 10
TI ₆	Intersection of two A-class roads. City centre in west direction and Slyfield industrial estate in north direction	0.88 $\pm$ 0.56	0.59 $\pm$ 0.30	24 $\pm$ 3	6 $\pm$ 8	55 $\pm$ 24
TI ₇	T-type intersection of A-class and b-class road. Main bus park situated in South east corner of the intersection.	1.28 $\pm$ 0.82	0.97 $\pm$ 0.54	26 $\pm$ 2	7 $\pm$ 8	49 $\pm$ 6
TI ₈	Y-type street canyon intersection. Railway station is situated in west side of the intersection and city centre is situated in south side. Surrounded by densely packed buildings.	1.61 $\pm$ 1.57	1.04 $\pm$ 0.82	24 $\pm$ 2	7 $\pm$ 8	44 $\pm$ 21
TI ₉	T-type intersection of two A-class roads. Situated near city centre and railway station.	1.30 $\pm$ 0.76	0.77 $\pm$ 0.27	22 $\pm$ 3	7 $\pm$ 7	26 $\pm$ 9
TI ₁₀	Four way intersection of two a-class roads. Railway station on north– west corner, city centre in south east corner. Surrounded by densely packed buildings.	3.86 $\pm$ 3.74	1.13 $\pm$ 1.19	13 $\pm$ 3	9 $\pm$ 7	29 $\pm$ 14

Table 3. Summary of results from mobile measurements in five different ventilation settings. N and I/O refer to PNC, and in-cabin to on-road ratios in a particular size range reflected by the size range noted in the sub-script; over bar represents average of a particular parameter. σ , GMD and AER stand for standard deviation, geometrical mean diameter, and air exchange rate, respectively.

Parameters	Set ₁	Set ₂	Set ₃	Set ₄	Set ₅
In-cabin $\overline{N}_{5-560} \pm \sigma_{5-560}$ ($\times 10^4 \text{ cm}^{-3}$)	—	2.7 $\pm$ 1.3	4.8 $\pm$ 2.3	1.3 $\pm$ 0.63	1.1 $\pm$ 0.63
In-cabin $\overline{N}_{5-30}$ (%)	—	52	58	46	45
In-cabin GMD $\pm \sigma$ (nm)	—	31.4 $\pm$ 10.1	27.0 $\pm$ 4.6	30.8 $\pm$ 4.5	30.2 $\pm$ 5.5
On-road $\overline{N}_{5-560} \pm \sigma_{5-560}$ ($\times 10^4 \text{ cm}^{-3}$)	8.1 $\pm$ 3.6	5.7 $\pm$ 2.4	8.0 $\pm$ 3.6	4.6 $\pm$ 2.4	4.4 $\pm$ 1.9
On-road $\overline{N}_{5-30}$ (%)	58	63	68	58	61
On-road GMD $\pm \sigma$ (nm)	39.9 $\pm$ 8.6	23.4 $\pm$ 4.1	22.4 $\pm$ 3.4	25.2 $\pm$ 4.9	24.4 $\pm$ 4.8
$\overline{(I/O)}_{5-560}$	—	0.47	0.60	0.28	0.25
$\overline{(I/O)}_{5-30}$	—	0.41	0.51	0.24	0.20
$\overline{(I/O)}_{30-100}$	—	0.60	0.75	0.31	0.32
$\overline{(I/O)}_{100-300}$	—	0.78	0.87	0.56	0.48
AER ($\text{m}^3 \text{ h}^{-1}$)	—	128	277	16	17

Table 4. Comparison of PNCs reported by various studies under Set₁.

City (Country)	Month (year)	Instrum ent	Size range (nm)	Average PNC (# cm ⁻³) ×10 ⁴	HDV (%)	Sulphur content in diesel (ppm)	Source
Helsinki (Finland)	August (2003)	ELPI and SMPS	5–1000	7.0±2.9	10%	350	Pirjola et al. (2006)
Karlsruhe (Germany)	2009	CPC	4–3000	1.8	–	10	Hagemann et al. (2014)
Los Angeles (USA)	April (2003)	CPC	10–1000	5.5– 20.0 ^a	1.4, 3.6 and 14% ^b	500	Westerdahl et al. (2005)
Minnesota (USA)	Summer (2006 and 2007)	EEPS	6–500	9.5±2.9	10%	15	Johnson et al. (2009)
Seoul (Korea)	December (2011)	FMPS	5.6–560	9.3±3.4	31.7±2.1	10	Kim et al. (2014)
Somerville (USA)	Septembe r (2009)– June (2010)	CPC	4–3000	5.0	–	15	Padró- Martínez et al. (2012)
Guildford (UK)	March– April (2014)	DMS 50	5–560	8.1±3.6	2–4%	10	This study

Note: ^arange of PNCs on freeway; ^baverage truck count; EEPS: Engine Exhaust Particle Sizer; ELPI: Electric Low Pressure Impactor; SMPS: Scanning Mobility Particle Sizer; CPC: Condensation Particle counter; FMPS: TSI Fast Mobility Particle Sizer; DMS: Differential Mobility Spectrometer

Table 5. Comparison of *I/O* ratio reported in other published studies in different ventilation settings.

Study	Automobile	Model year	Road environment	Ventilation condition ^a	<i>I/O</i>
Zhu et al. (2007)	Volkswagen Jetta	2000	Freeway	A	0.8
				B	0.7
				C	0.6
	Audi	2004		A	0.35
				B	0.5
				C	0.18
	PT Cruiser	2005		A	0.18
				B	0.3
				C	0.05
Knibbs et al. (2010)	Mazda 121	1989	Tunnel	A	0.95
				B	1.04
				C	0.47
				D	0.39
	Mitsubishi Magna	1998		A	0.89
				B	1.01
				C	0.29
	Toyota Hilux	2005		A	0.91
				B	1.04
				C	0.25
	Volkswagen Golf	2005		A	0.66
				B	0.84
				C	0.08
				D	0.17
	Subaru Outback	2007		A	0.88
				B	0.91
				C	0.45
				D	0.68
Hudda et al. (2011)	Ford Contour	1999	Arterial road with less traffic	A	0.53
				D	0.06
	Honda civic	2009		A	0.67
				D	0.03
	Toyota Prius	2010		A	0.52
				D	0.02
Joodatnia et al. (2013)	Volkswagen Golf	1998	Congested arterial road	B	0.72
This study	Ford Fiesta	2002	Congested arterial road	A	0.25
				E	0.6
				F	0.47
				G	0.28

Note: ^aWindows closed; A = Outside air intake (low fan/off), no recirculation; B = Outside air intake (medium fan), no recirculation; C = Outside air intake (low fan), with recirculation on; D = Indoor air recirculation only, no outdoor air intake; E = Outside air intake (fan full), no recirculation; F = Outside air intake (fan low), no recirculation and heating medium; G = Outside air intake (fan off), no recirculation and heating full.

Table 6. Range of ‘a’, ‘b’ and ‘c’ for different types of TIs during morning and evening peak hours, respectively.

Type of TI	Morning/ evening	$a (\times 10^{-3})$	$b (\times 10^{-3})$	$c (\times 10^{-3})$
Four-way perpendicular TI with 2–3 buildings around the TI (TI ₁ , TI ₂ and TI ₄).	Morning	–0.9 to –4.9	–27.5 to 3.3	155.4 to 195.1
	Evening	–2.7 to –16.1	–2.3 to 45.5	187.4 to 310.3
Four-way perpendicular TI with dense built-up area (TI ₁₀).	Morning	1.5	0.3	–0.1614
Three-way TI in a densely built-up area (TI ₈).	Morning	1.2	0.9	–65.2
	Evening	0.7	4.5	130.6
Three-way TI with 2–3 building in surroundings (TI ₇).	Morning	–4.3	12.1	225.3
	Evening	–2.1	–12.4	157.8

Figure 1 (Colour)

(b)

Figure 2 (Colour)

Figure 3 (Colour)

Figure 4 (Colour)

Figure 5 (Colour)

Figure 6 (Colour)

Figure 7 (Colour)

Figure 8 (Colour)

Figure 9 (Colour)

Figure 10 (Colour)