Vertical and horizontal variability in airborne nanoparticles and their exposure around signalised traffic intersections

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Abstract

We measured size-resolved PNCs in the 5–560 nm range at two different types (4– and 3–way) of TIs in Guildford (Surrey, UK) at fixed sites (~1.5 m above the road level), sequentially at 4 different heights (1, 1.5, 2.5 and 4.7 m), and along the road at five different distances (10, 20, 30, 45 and 60 m). The aims were to: (i) assess the differences in PNCs measured at studied TIs, (ii) identify the best fit probability distribution curves for the PNCs, (iii) determine vertical and horizontal decay profiles of PNCs, (iv) estimate particle number emission factors (PNEFs) under congested and free-flow traffic conditions, and (v) quantify the pedestrian exposure in terms of respiratory deposition dose (RDD) rates at the TIs. Daily averaged particle number distributions at TIs reflected the effect of fresh emissions with peaks at 5.6, 10 and 56nm. Despite the relatively high traffic volume at 3–way TI, average PNCs at 4–way TI were about twice as high as at 3–way TI, indicating less favourable

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dispersion conditions. Generalised extreme value distribution fitted well to PNC data at both TIs. Vertical PNC profiles followed an exponential decay, which was much sharper at 4-way TI than at 3-way TI, suggesting ~60% less exposure for people at first floor (4.7 m) to those at ground floor around 4-way TI. Vertical profiles indicated much sharper (~132-times larger) decay than in horizontal direction, due to close vicinity of road vehicles during the along-road measurements. Over an order of magnitude higher PNEFs were found during congested, compared with free-flow, conditions due to frequent changes in traffic speed. Average RDD rate at 4-way TI during congested conditions were up to 14-times higher than those at 3-way TI ($1.20 \times 10^{11} \text{ h}^{-1}$). Findings of this study are a step forward to understand exposure at and around the TIs.

Capsule abstract

Exposure to PNCs at traffic intersections is over an order of magnitude higher during congested “red traffic light” compared with free flow “green traffic light” conditions.

Keywords: Particle number concentration; Number size distribution; Traffic intersections; Vertical variation; Horizontal profile; Pollution hotspots

1. Introduction

Numerous toxicological (Handy and Shaw, 2007; Sharma, 2010; Stone et al., 2009) and epidemiological (Bos et al., 2011; Jacobs et al., 2010) evidences suggest adverse effects of airborne nanoparticles (referred here to those below 300 nm to represent majority of particle number concentrations, PNCs; Kumar et al. 2010) on human health. Signalised traffic intersections (TIs) are known as hotspots of PNCs (Goel and Kumar, 2014, 2015a, b), but there is a limited information on the variability in PNCs along the roads, and in vertical direction, at different types of TIs. This is an important consideration for exposure in urban areas where people live along the roads as well as at the different floors of buildings.

Spatial variations of nanoparticles have been investigated for diverse urban settings such as in street canyons (Kumar et al., 2008a, b), along the highways (He and Dhaniyala, 2012; Zhu and Hinds, 2005) and motorways (Imhof et al., 2005). Yet, there are a handful of studies focussing on horizontal (Kim et al., 2013) and vertical (Nakashima et al., 2014) variations in PNCs at the signalised TIs. Past studies suggest an exponential decay in PNCs with an increasing height within street canyons (Kumar et al., 2008a) while no such vertical decay has been reported at the sides of highway or motorways (Nakashima et al., 2014). Earlier work also suggests an exponential decay in PNCs with increasing horizontal distance from the highways in absences of any obstructions (Zhu et al., 2002). Such profiles are not available for TIs, but are important to understand human exposure at these hotspots. With this in mind, we undertook this study to determine the vertical and horizontal variability in particle number size distributions (PNDs) at two different types (i.e. 3- and 4-way) of TIs.

Particle number emission factors (PNEFs) is a measure that relate the characteristics of an emission source with its strength. There are a number of methods to derive PNEF through laboratory testing based on the engine and chassis dynamometer studies (Jayaratne et al., 2009; Morawska et al., 1998; Ristovski et al., 2004; Ristovski et al., 2005), direct on-road and on-board measurements under real-world driving cycle (Keogh and Sonntag, 2011; Nickel et al., 2013) and inverse modelling techniques (Kumar et al., 2008a; Morawska et al., 2005). These techniques are discussed in detail in Kumar et al. (2011). The main advantage of field studies over chassis dynamometer based studies is that the former takes into account the effects of transformation processes such as nucleation, dilution, condensation, coagulation and dry and wet deposition on the changing PNCs between the source and the receptor (Kumar et al., 2011). A number of past studies have determined PNEFs for road tunnels, highways and street canyons with free flowing traffic conditions based on field measurements, as summarised in Table 1. However, studies determining the PNEFs in

congested driving conditions, which often prevail at TIs, are yet to appear and therefore is taken up for investigation in this study.

Information regarding the distribution of pollutant concentrations in different environments (e.g. TIs, motorways, roadways, households) is important for assessing the variability in exposure estimates (USEPA, 1992), apportioning the contribution of the emission sources and evaluating policies and interventions (Batterman et al., 2007; Jia et al., 2008; Loh et al., 2007). Fitting numerous types of probability distribution functions (hereafter referred as probability function) to the pollution data allows to estimate the frequency of individual concentration ranges to which urban dwellers expose in diverse urban settings. There are a number of such studies available in the literature, which has fitted probability function curves to their air pollution data. For instance, Mage (1984) fitted pseudo-lognormal distribution on carbon monoxide data set that was measured in Washington D.C (USA). Later, Burkhardt et al. (1998) applied lognormal distribution to ammonia measured in downtown of Los Angeles and in the rural areas near Edinburgh. Similarly, Kao and Friedlander (1995) applied lognormal distribution to particulate matter of diameter less than 10 μm (PM_{10}) measured in south coast air basin of California. Likewise, Morel et al. (1999) fitted type V Pearson distribution for PM_{10} , $\text{PM}_{2.5}$ (diameter less than 2.5 μm), nitrogen dioxide and sulphur dioxide data measured in Santiago (Chile). However, there are hardly any studies that have attempted similar distribution fits to the PNC data at the TIs, which is one of the aims of this study.

The distinctive features that aim to fill the existing research gaps of this work are as follows. Firstly, as opposed to previous studies that have analysed the vertical and horizontal variations of nanoparticles in street canyons (Kumar et al., 2008a, b), highways (He and Dhaniyala, 2012; Zhu and Hinds, 2005) or motorways (Imhof et al., 2005), this study has

assessed the horizontal and vertical variability in PNCs at *urban traffic hotspots* (i.e. TIs). Secondly, this is for the first time when the PNEFs are derived for congested driving conditions at the TIs and compared with those obtained during free flow driving conditions. Thirdly, we fitted probability functions to short (1 s) and long-term (1 h) average PNC data measured at 3- and 4-way TIs. These distributions are important in assessing the frequency and variability in PNC exposure at the TIs.

In summary, the specific objectives of this study are to assess the vertical and horizontal profiles of PNDs and PNCs at 4- and 3-way TIs, in order to compare the exposure at these TIs. Furthermore, this study estimated PNEF for congested and free flow driving conditions and fitted statistical distributions to different time averaged PNCs at both types of TIs.

2. Methodology

2.1 Site description

Measurements were conducted at two different types of TIs (i.e. 4- and 3-way) in a typical UK town, Guildford. Four-way TI was located in a suburban area of Guildford and has 4 intersecting roads (Figure 1a). Legs 1 and 2 represent Egerton road that was ~12 m wide. Leg 3 represents Richard Meyjes road that was ~15 m wide. Leg 4 represents Gill Avenue that was ~18 m wide and was 3 m away from the sampling site (Figure 1b). On one side (i.e. leg 4), the TI was surrounded by 3 detached residential buildings (height ~6 m) whilst the other side was bordered by trees (i.e. leg 1); rest of the two sides were having an open ground (Figure 1b). All the 4 legs of the roads were having two-way traffic flow. This TI has a signal cycle (i.e. total time of red, yellow and green lights) of around 116 s; length of the red light varied from 51 to 82 s on different legs of the TI.

Three-way TI was located in the city centre of Guildford and has three intersecting roads (Figure 1c). Legs 1 and 2 represent Onslow street and were 18 and 21 m wide, respectively.

Leg 3 represents Woodbridge road and was 12 m wide. Sampling location was around 4 m away from the leg 2 in front of St. Savior's church. On all three sides of the TI either commercial or residential buildings were present; these buildings varied from 6 to 14 m in height. All the three roads were having two-way traffic flow; the direction of traffic flow is represented by arrows in Figure 1c. This TI has a signal cycle of around 83 s with length of red light varying from 31 to 68 s on different legs of the TI.

Average daily traffic flow on different roads intersecting at both of these TIs was obtained from 5-minute manual traffic count every hour. Total traffic volume at a TI was estimated by summing the traffic flow on each of the roads intersecting at a TI. Total traffic volume (i.e. sum of traffic on all three legs) and the percentage of heavy duty vehicles at 3-way TI were 64400 veh day⁻¹ and 6%, respectively. The corresponding values at the 4-way TI were 60171 veh day⁻¹ and 4%, respectively (Figure 2). Further details of traffic volume are presented in Supplementary Information (SI) Table S1.

2.2 Study design

Four different sets of measurements were conducted at the studied TIs. These included monitoring at: (i) the corners of the TIs (i.e. ~1.5 m above the road level close to the breathing zone; this is referred hereafter as "fixed-site" measurements), (ii) four different heights in vertical direction, (iii) five different points away from the centre of TIs, and (iv) a background location. Detailed setup of location of these measurements is shown in Figure 1.

Measurements at the corner of the 4- and 3-way TIs were conducted between 4 February and 28 April 2015 (from 0800 to 2000 h; local time) and between 22 January and 2 February 2015 (from 08:00 to 18:00 h), respectively. A total of 4536000 data points of size-resolved PNDs were collected during 126 h of fixed-site measurements at both the TIs. Vertical profiles were obtained through pseudo-simultaneous measurements of PNDs at 4 different

heights (i.e. 1, 1.5, 2.5 and 4.7 m) on 2 February 2015 at the 3-way TI while on 21 and 22 April 2015 at 4-way TI (Figure 1b–d). Since health and safety concerns constrained us to use a tall mast near the intersections, measurements were carried out at heights of less than 6 m. These measurements were conducted by using a DC powered automated solenoid switching system in conjunction with the DMS50. Detailed description of switching system is provided in Section 2.3. Four different sampling tubes (i.e. tube 1, 2, 3 and 4) were used to measure PNDs at the respective four heights at both the TIs. All the four tubes were mounted on a single pole, which was securely fastened to the nearby building at the 3-way TI and to nearby pole of street light at 4-way TI. A total of 480 and 1320 full cycles of measurements were obtained at the 3- and 4-way TIs, respectively, giving a total of 1152000 size resolved PNDs on both the TIs.

Horizontal profiles of PNCs were measured at the legs 3 and 4 of the 4-way TI on two days (23 and 27 April 2015) between 0800 and 2000 h. A total of 648000 data points of size-resolved PNDs were obtained during 18 h of measurements at both the legs of 4-way TI. The high built-up area around the 3-way TI restricted us to perform horizontal monitoring at this TI. Reference point of measurement is referred as P_R (Figure 1e), followed by sampling points, P_1 , P_2 , P_3 and P_4 which were 10, 20, 30, 45 and 60 m away from the centre of this 4-way TI (Figure 1e). PNDs were measured sequentially at a frequency of 10 Hz at the sampling points. To acquire a representative data set at each of the sampling points, the samples were taken for 11 min in an hour at each sampling point, by manually repositioning the location of the instrument.

Both the studied TIs were far apart and therefore two different approaches were used to determine the background PNDs for both the TIs. For the 4-way TI, we firstly monitored PNDs continuously between 08:00 and 20:00 h on 17 May 2015 in an open Performing Arts

Studio field within the University of Surrey at a height of 1.5 m, giving a total of 43200 size-resolved PND points. This sampling location was about 1.3 km away from the sampling site of the 4-way TI in North-East (NE) direction (Figure 1a). There were no major roads near the background sampling location and this was upwind of sampling locations to present a fairly good representation of local background levels of the PNCs. Furthermore, the wind direction, speed, temperature and relative humidity on this day was similar (SI Table S2) to those observed on the day of measurements at the 4-way TI. Therefore these background measurements were assumed to be representative of the background PNCs for the 4-way TI. Comparison of this background and the site data indicated that the total background PNCs were equal to 5th percentile of 1s average PNCs of same day. Following this finding, the background PNCs were estimated for the rest of the day at 4-way TI and for whole monitoring duration at 3-way TI. A similar approach has been used by previous studies to use 5th percentile of the observed PNCs to deduce local background levels (Goel and Kumar, 2015b; Hudda et al., 2014).

Figure 3 shows the wind rose diagram for the measurements at both the TIs. This diagram classified wind direction in eight categories, which were northwest (292.5°–337.5°), north (337.5°–22.5°), northeast (22.5°–67.5°), east (E; 67.5°–112.5°), southeast (112.5°–157.5°), south (157.5°–202.5°), southwest (202.5°–247.5°) and west (247.5°–292.5°). The hourly meteorological data (i.e. wind speed, wind direction, temperature and relative humidity) during all the measurements was obtained from the nearest meteorological station (i.e. Royal Horticulture Society's garden in Wisley). This station is located about 10 km away from the site to the NE of Guildford at a height of 10 m, which offsets the effect of the ground-level turbulence, and maintained by the UK Met Office. The meteorological data of this station have also been used by other studies carried out in Guildford (e.g. Al-Dabbous and Kumar, 2014). Average wind speed, ambient temperature and relative humidity during the

measurements were $3.0 \pm 1.3 \text{ m s}^{-1}$, $9.0 \pm 4.8 \text{ }^{\circ}\text{C}$ and $52.6 \pm 14.6 \%$ at 4-way TI, respectively. Corresponding values at 3-way TI were $3.5 \pm 1.9 \text{ m s}^{-1}$, $5.3 \pm 2.7 \text{ }^{\circ}\text{C}$ and $63.5 \pm 7.9 \%$, respectively. By using the average wind speed at the studied TIs and considering moderate incoming solar radiation during the measurements, atmospheric condition is classified as Pasquill stability class B at both the TIs during the measurements (Mohan and Siddiqui, 1998).

2.3 Instrumentation

A differential mobility spectrometer (Cambustion DMS50) was deployed to measure the PNDs in the 5–560 nm size range at a sampling rate of 10 Hz at both the TIs. The instrument works on electrical mobility detection technique to classify the particles in 34 size channels. Details of the working principle, detection efficiency and noise level can be seen in Kumar et al. (2010). The DMS50 has been successfully deployed in our previous studies in diverse settings such as fixed-site roadside measurements (Al-Dabbous and Kumar, 2014), vehicle wake (Carpentieri and Kumar, 2011), within the car cabins (Goel and Kumar, 2015a; Joodatnia et al., 2013a, b) and indoor environments (Azarmi et al., 2014; Azarmi et al., 2015; Kumar and Morawska, 2014; Kumar et al., 2012). A thermally conductive silicone tube of 0.5 m length with an internal diameter of 5.5 mm was used for the fixed-height, horizontal profiles and background measurements in order to minimise particle losses in the sampling tube (Kumar et al., 2008c). A DC powered automated solenoid switching system was used in conjunction with the DMS50 for measuring the vertical profiles at four different heights, pseudo-simultaneously at both the TIs (Figure 1d).

At both the TIs, length of the tube to measure at the sampling locations z_1 , z_2 , z_3 and z_4 were 0.6, 1.0, 1.5 and 4.17 m, respectively. The switching system was designed in our previous work (Kumar et al., 2008b) and then updated recently (Al-Dabbous and Kumar, 2014; Goel

and Kumar, 2015a; Joodatnia et al., 2013a) to automate its switching time through a software so that the lag time between the switching is modest. The switching system automatically shifted the sampling flow between each of the four heights once every 60 s. This allowed us to make 15 s of measurements at the rate of 10 scans per second. This gave a total 150 sample scans in a minute at each location by redirecting the sampling flow between the sampling points. The first 2 s of data from each measurement was discarded, as the particle residence time in the tube at sampling locations z_1 , z_2 , z_3 and z_4 was 0.2, 0.4, 0.5 and 1.5 s, respectively, and therefore the importance of lag time of particle within the sampling tube became trivial. The final 13 s of data at each sampling point was retrieved for analysis at studied TIs. Particle losses (especially for below 20 nm in diameter) in long sampling tube due to their diffusion is an important issue and should be taken in consideration for tubes longer than 1 m (Kumar et al., 2008c). Therefore, we corrected our measured data for particles losses in the sampling tube, following the approach described in Kumar et al. (2008c). Corrected values of particle number and size distributions were then used in our subsequent analysis at both the TIs. Macros were developed in Microsoft excel to segregate the PND data at different heights at the studied TIs.

Traffic flow videos at the TIs were continuously recorded for the entire monitoring period using Panasonic HC-V500 camera. In addition, AQ Mesh was used to measure 15-min average concentration of oxides of nitrogen (NO_x), carbon monoxide, sulphur dioxide and ozone during the entire sampling period. Timestamps of all the instruments were matched in the beginning of each experiment. All the collected data were then analysed by Microsoft excel with the use of DMS50 data processing tool.

2.4 Estimation of exposure rates and the PNEFs

Exposure to PNCs is estimated in terms of RDD rate (i.e. number of particles deposited in lungs per unit time, $\# \text{ h}^{-1}$) for a male for light exercise condition. RDD rates are a product of number of particles inhaled and exhaled, breathing rate, deposition fraction and duration of exposure. Tidal volume and breathing rate depend on age, gender and the level of activity. In this study, typical breathing rate of 20 min^{-1} and tidal volume of 1.25 l for a male adult under light exercise were applied (ICRP, 1994). The deposition fraction depends on the size distribution of particles. Size-dependent deposition fractions were estimated as 0.72 ± 0.08 and 0.75 ± 0.07 for the 3- and 4-way TIs, respectively (Hinds, 1982; Kumar et al., 2014). Further details on the method to estimate exposure rates can be seen in Joodatnia et al. (2013a).

Based on 15 min average PNC data, PNEFs were calculated for the mixed vehicle fleet during congested driving conditions at the TIs. This averaging time for estimating the PNEFs was selected to match the sampling rate of NO_x concentrations, which was 15 min. Congested driving condition is referred here to simultaneous occurrence of free-flow as well as stop and go conditions at the TIs due to changing colour of the traffic signal on different legs of the TI. For example, stop and go conditions will persist at the road leg with a red signal but at the same time, the leg with a green signal will contain free flow traffic conditions. In addition, separate emission factors for free flow conditions at TIs were calculated from our previous field campaigns (Goel and Kumar, 2015a). The NO_x -tracer method, which has been used by earlier studies (Gehrig et al., 2004; Jones and Harrison, 2006; Nickel et al., 2013), was applied to determine the size dependent PNEFs. This approach assumes dispersion of particles similar to gaseous pollutants during their transport from the point of emission (i.e. tailpipe) to the point of measurement (i.e. near roadside). The PNEFs were then measured for a mixed vehicle fleet by using an Eq. (1).

$$PNEF = \frac{R \times \sum (EF_{i,NOx} \times n_i)}{\Delta NOx} \times \frac{\Delta PNC}{n_{fleet}} \quad (1)$$

where

$$\begin{aligned} \sum EF_{i,NOx} \times n_i = & EF_{diesel\ car,NOx} \times n_{diesel\ car} + EF_{petrol\ car,NOx} \times n_{petrol\ car} + EF_{LDV,NOx} \\ & \times n_{LDV} + EF_{Bus,NOx} \times n_{bus} + EF_{truck,NOx} \times n_{truck} + EF_{motorcycle,NOx} \\ & \times n_{motorcycle} \end{aligned}$$

ΔPNC s and ΔNOx are the net PNC and NOx concentrations at the TIs; these were obtained after taking the background values from the total measured concentrations. n_{fleet} is the traffic volume counts; i is the category of vehicles (i.e. diesel and petrol fuelled cars, light duty diesel vehicles, bus, truck and motorcycle). $EF_{i, NOx}$ is emission factors of NOx for different categories of vehicles based on urban driving cycle from the National Atmospheric Emission Inventory (DEFRA, 2013). In the NAEI (2011), emission factors are estimated based on urban driving cycle which has less frequent stop and go traffic conditions as compare to those observed at the TIs (Goel and Kumar, 2015a). Therefore, to modify the emission factors taken from NAEI for more congested traffic driving conditions such as those at the TIs, the ratio ($R = NOx_C/NOx_F$) of NOx concentrations during congestion (NOx_C) and free flow (NOx_F) were estimated at the studied TIs using the methodology explained in SI Section S1. Thereafter, these ratios were used to correct the emission factors of NOx based on the NAEI (2011). In order to calculate the PNEFs for different legs of TIs, the whole data set at both the TIs is divided based on the wind directions.

At the 4-way TI, direction of the wind from the north, east, south and west represented pollution from road legs 1, 2, 3 and 4, respectively. There was only a sparse data recorded when wind was blowing from the south (Figure 3a), therefore no PNEFs were estimated for leg 3 of this TI. At the 3-way TI, winds from southwest and southeast were assumed to

represent pollution from legs 1 and 3, respectively, while north, northwest and northeast represented pollution from leg 2 (Figure 3b). Only a limited data points were available when wind was blowing from SE direction; therefore PNEFs for leg 3 of 3-way TI are not estimated for this reason.

2.5 Fitting of probability function curves

Statistical distributions depend on a number of factors such as: (i) space (i.e. monitoring location), (ii) time (i.e. averaging time of the data), and (iii) size range of PNCs. To assess the space dependence at TIs, statistical distributions were fitted at two different types of TIs (i.e. 3- and 4-way) by using Easyfit software (version 5.6). Time dependence was assessed by fitting distributions to 1 s (short-term), 15 min (medium-term average) and 1 h (long-term) averaged PNCs at the studied TIs. Dependence on size range was assessed by fitting distribution to three different size ranges (i.e. 5–30, 30–300 and 5–560 nm). Total PNCs and N_{5-560} are used interchangeably and so is the case with the N_{5-30} (nucleation mode) and N_{30-300} (accumulation mode) size ranges throughout the text.

Statistical distributions were fitted to a total of 18 combinations (i.e. two monitoring locations $\times$ three different time averages $\times$ three different size ranges). After finalising the total number of combinations of PNCs, a total of 61 different statistical distributions were tried on each of the 18 combinations by using a two-step process: (i) selection of the candidate distributions and evaluating these distributions based on goodness-of-fit criteria (Sharma et al., 2013), and (ii) estimation of their parameters such as shape, scale and location (Jia et al., 2008; Sharma et al., 2013). After a thorough visual examination of probability function plots and histograms and considering the goodness-of-fit test using Anderson-Darling (A-D) method (Sharma et al., 2013), all the 61 distributions were ranked based on the A-D statistics and the “best fit” distribution was selected for each of the 18 combinations. The ranking of distributions for all

the 18 combinations is provided in SI Tables S4–S9. The A–D test served as the primary criterion since it is suitable for fitting distributions in both the middle and upper percentile ranges (Sharma et al., 2013). Smaller value of A–D statistics indicate a better probability function fits (Jia et al., 2008). In order to identify the type of a distribution that could fit to majority of the time averaged series of PNCs, a common distribution among the top ten ranked distributions was chosen (SI Tables S4–S9). Thereafter the parameters (i.e. shape, location and scale) of the identified “best” and “most common” probability functions were estimated based on the method of moments (Sharma et al., 2013). The above–noted approach was also applied to RDD data to identify these fit distributions to 6 different combinations (i.e. two monitoring locations at corners of TIs $\times$ three different time averages) of RDD rate at the studied TIs and the corresponding results are provided in SI Tables S10 (4–way) and S11 (3–way).

3. Results and discussion

3.1 PNDs and PNCs within breathing zone at the 3– and 4–way TIs

Figure 4a shows the average PNDs at both the 3– and 4–way TIs. PNDs showed 3 distinct peaks at 5, 10 and 56 nm at both the TIs. Peaks at 5 and 10 nm are attributed to nucleation mode particles that are formed due to the condensation of volatile particles and gasses. Peak at 56 nm is attributed to primary emissions from the combustion of fuel in the engine. Inter–comparison of the magnitude of PND peaks at 5 and 10 nm at the 4–way TI shows them to be about 3– and 4–times to those at the 3–way TI as opposed to the peak at 56 nm that was almost identical at both the TIs (Figure 4a). The measured PND spectrum corroborate well to previous studies (Holmes et al., 2005; Wang et al., 2008), reporting similar bimodal PNDs in 10–35 nm and 50–80 nm range at 4–way TIs.

328 The hourly averaged PNCs at 3- and 4-way TIs were found to be $3.44 \pm 0.41 \times 10^4$ and
 329 $7.29 \pm 2.11 \times 10^4 \text{ cm}^{-3}$, respectively. Studies limited for TIs are as seen from Table 2, which
 330 presents a summary of past studies that have conducted the fixed-site measurements at the
 331 TIs. It can be clearly seen that our results of 4- and 3-way TIs are within a factor of 2 to
 332 those compared with the published literature of 4- and 3-way TIs, respectively (Table 2).
 333 The cross-comparison of PNCs measured at our TIs show nearly two-fold (i.e. 1.6-times)
 334 PNCs at the 4-way TI than those observed at the 3-way TI ($3.44 \pm 0.41 \times 10^4 \text{ # cm}^{-3}$) (Figure
 335 4b-c). PNCs in nucleation mode were 20% higher at 4-way TI as compared to 3-way TI. On
 336 the other hand, concentration of accumulation mode particles was almost similar at both the
 337 TIs (Figure 4b). There are a number of factors such as traffic volume, traffic composition,
 338 built-up area around a TI, signal cycle length, wind speed and wind direction that would have
 339 contributed to the differences in the PNCs. One of the major contributors among these factors
 340 is traffic volume. Therefore, we normalised the PNCs with respect to traffic volume for
 341 assessing the effect of traffic volume on the normalised PNCs (Figure 4d). Even after this
 342 normalisation, the normalised PNCs at 4-way TI were nearly the same (i.e. 1.6-times higher)
 343 to those at 3-way TI, suggesting that there are other factors such as built-up area around a TI,
 344 traffic signal cycle length, wind speed and direction play a role in producing these PNC
 345 differences. Built-up area around the 3-way TI was higher compared with those at 4-way TI
 346 but the PNCs were still higher at the 4-way TI. These observations indicate that the effect of
 347 relatively denser built-up area around the 3-way TI is negated by the other factors such as
 348 signal cycle length, wind speed and wind direction. Length of green phase of the signal cycle
 349 at 3-way TI was about 1.5-times than that at 4-way TI (30 s), leading to shorter delay time at
 350 3-way TI compared with 4-way TI. Since road vehicles are expected to contribute relatively
 351 more to nucleation mode particles during delay periods compared with their larger-size
 352 counterparts, a major part of this PNC difference could be attributed to the delay conditions at

the TIs (Goel and Kumar, 2015a). The rest of the differences could be attributed to the predominant wind direction, which was NE at the 4-way TI advecting emissions from the legs 1, 2 and 3 of the TI (Figure 3a) to the sampling location compared with contribution of emissions from one leg (i.e. leg 2) only for 3-way TI, which experienced winds from northwest (Figure 3b). The SI Section S2 shows the details of measured PNDs at two different types of TIs during different wind directions. The wind speed is a remaining factor, which was somewhat similar ($3.0 \pm 0.7 \text{ m s}^{-1}$) at 4-way TI to those found at 3-way TI ($3.5 \pm 0.3 \text{ m s}^{-1}$) and the differences in relative humidity and ambient temperature were also modest during the measurements at the two TIs.

3.2 Probability functions of particle number concentrations at intersections

Probabilistic methods using frequency histogram over point estimates (i.e. average or median) are potentially more representative of actual concentrations and their corresponding frequency of occurrence (Jia et al., 2008; Sielken Jr and Valdez-Flores, 1999). Based on the frequency histograms, hourly averaged PNCs at 3- and 4-way TI for 99% of the total sampling duration were found to be less than or equal to 7.59×10^4 and $1.56 \times 10^5 \text{ cm}^{-3}$, respectively. Similarly, half of the time, hourly averaged PNCs are less than or equal to 3.14×10^4 and $4.30 \times 10^4 \text{ cm}^{-3}$ at 3- and 4-way TIs, respectively.

Frequency histograms are usually used to assess the rate of violation of pollution concentrations against any ambient air quality standards (Sharma et al., 2013). No such standards are available in case of PNCs but a value of *urban roadside* PNCs was indicated by Kumar et al. (2014) as $3.2 \pm 1.6 \times 10^4 \text{ cm}^{-3}$, based on the review of numerous studies. Furthermore, the typical on-road PNC concentrations during free-flow driving conditions were reported as $6.4 \pm 1.9 \times 10^4 \text{ cm}^{-3}$ by Goel and Kumar (2015a). We used these PNCs as a reference value to compare with our measured PNCs at the TIs. The hourly averaged PNCs at the 3- and 4-way TIs were found to exceed the urban roadside PNC values for 43% and 62%

of the total time, respectively. The hourly averaged PNCs at our 3- and 4-way TIs were found to exceed the average on-road PNCs during free flow traffic conditions for 12% and 38% of the total time, respectively.

Many types of probability functions have been used in the past on air pollution data (Burkhardt et al., 1998; Kao and Friedlander, 1995; Mage, 1984; Morel et al., 1999). The types of probability functions vary for different time averages, therefore to assess the effect of time averages, distribution were fitted to 1 s, 15 min and 1 h averaged *total* PNCs at the 3- and 4-way TIs (see Section 2.6 and SI Tables S4–S5). Figure 5 shows the “best” and the “common” fit distributions on the *total* PNC data for three different averaging periods. Generalised logistic distribution fitted “best” the 1 s average PNC at both the TIs and 15 min averaged PNCs at 4-way TI. While Weibull distributions fitted “best” the 1 h averaged PNCs at both the TIs and 15 min averaged PNCs at 3-way TI (Figure 5). Irrespective of a TI type, generalised extreme value (GEV) distribution was found to be the “common” fit distribution among the top ten fits at both the TIs (Table 3).

Furthermore, different probability functions fit may suite better to nucleation and accumulation mode particles. We assessed the variability in type of “best” and the “common” fit probability functions to PNCs in 5–30 and 30–300 nm size ranges, using the methodology described in Section 2.6. A total of 12 combinations (three time periods $\times$ two size ranges $\times$ 2 types of TIs) were assessed. The SI Tables S6–S9 present the ranking of probability functions fitted to PNCs in the 5–30 nm and 30–300 nm size ranges at both the TIs. Out of these 12 combinations, GEV and exponential distribution fitted well to 4 and 3 combinations, respectively, whilst five different distributions fitted well to rest of the 5 combinations (see Table 4). GEV is found to be a “common” fit for both size ranges at 4-way TI as well as for the PNCs in 5–30 nm range at 3-way TI. On the other hand, generalised Pareto distribution is

found to be a “common” fit probability functions for PNCs in 30–300 range at 3–way TI. Interestingly, 5 out of top 10 best fit probability functions were same for different time averaged of PNCs in 5–30 nm range at 4–way TI, showing that the negligible impact of averaging period on underlying distribution at this TI.

3.3 Vertical variations of PNCs at the TIs

Following the methodology presented in Section 2.2, we plotted the size–resolved PNDs measured at 4 different heights at each of the 3– and 4–way TIs to understand their vertical variability (Figure 6). The PNDs at each height were found to be similar to each other with the peaks at 6, 10, 20 and 65 nm at 3–way TI as opposed to 3 peaks at 6, 9, and 27 nm at 4–way TI (Figure 6a–b). Negligible changes in peak values of PNDs indicate that the competing influences of transformation processes are usually complete by the time particles reached to the monitoring location at both the TIs. Further confirmation comes from normalised PND profiles against the corresponding total PNCs, which show perfectly superimposed curves at all four heights (see Figure 6c–d), indicating dilution as a key process (Carpentieri and Kumar, 2011; Goel and Kumar, 2015a).

It has been suggested by previous studies (Capannelli et al., 1977; Dabberdt and Hoydysh, 1991; Kumar et al., 2008a; Vardoulakis et al., 2002) that the vertical profile of gaseous pollutants and PNCs in a street canyon follow an exponential decay profile. To test whether a similar variation occurs for the PNCs at different types of TIs, we first applied the best fit line to our dimensionless PNC data and against the dimensionless height (z/H) (Figure 6e–f). The best fit line on dimensionless PNCs showed an exponential decrease with height at both the TIs, with R^2 as 0.99 (4–way) and 0.86 (3–way) TIs (Figure 6e–f). This suggests that the exponential decay profile also fit well to the TIs (Figure 6e–f), irrespective of their types, such as the case was with the street canyons for PNCs (Table 5). Fitted exponential profiles

are expressed in the form of Eq. (2) that was given by Kumar et al. (2008a) for street canyons.

$$\frac{(C_z - C_b)}{(C_0 - C_b)} = \exp\left(-k\left(\frac{z}{H}\right)\right) = \exp(-k_1 z) \quad (2)$$

Where C_z and C_b are the PNCs at any height z and background, respectively; C_0 is the PNC at the lowest (i.e. at 1.0 m) sampling point. H is the height of the highest building around a TI and it is 6 and 14 m for 4-way and 3-way TI, respectively; k/H ($=k_1$) is the exponential decay coefficient in m^{-1} and k_1 is inverse of the characteristic dispersion height which corresponds the height above the road level at which dimensionless concentration is $e^{-1} = 0.37$ (Kumar et al., 2008b).

By applying Eq. (2) at two different types of TIs, k_1 is found to be 0.66 and 2.12 m^{-1} at 3- and 4-way TIs, respectively. This indicates that the characteristic dispersion height above the lowest sampling point at 3-way TI is 3.2-times of those at 4-way TI. These observations also suggest that the flow is restricted at the 3-way TI due to surrounding built-up area, resulting in build-up of PNCs to a greater height compared with 4-way TI.

Figures 6g–h show vertical profiles of size-resolved PNCs at both the TIs. These figures also show a similar decaying trend at both the TIs between 1.5 and 4.7 m. At the lower heights, there was a contrasting trend (i.e. an increase in PNCs at the 3-way TI as opposed to a decrease in PNCs at the 4-way TI from 1.5 to 1 m height). Given that the height ($\sim 0.70 \text{ m}$) of the tailpipes of heavy duty vehicles was close to the sampling height at 1 m, an increase in PNCs towards the lowest height at the 3-way TI could possibly be explained by 2% higher volume of heavy duty vehicles at this TI than those at 4-way TI.

Table 5 shows the summary of the studies that have measured vertical profiles of PNCs in the

roadside environments. Taking together the measured profiles and those summarised in Table 5, vertical profile at 3-way TI is similar to those observed in asymmetric street canyons (Kumar et al., 2008a) whilst the profile at 4-way TI is similar to those observed near freeway (Zhu and Hinds, 2005), motorway (He and Dhaniyala, 2012; Imhof et al., 2005) and in symmetric street canyons (Kumar et al., 2008b).

In terms of vertical decay, PNCs decreased by 10, 5 and 15% at 1.5, 2.5 and 4.7 m, respectively, compared to those at 1 m (i.e. where largest PNCs were observed) at 3-way TI. PNCs increased by 15 and 13% at the heights 1.5 and 2.5 m, respectively, and decreased by 40% at 4.7 m, compared with those at 1 m at 4-way TI. This indicates that the people residing at upper heights (i.e. 4.7 m) close to the 4-way TI will have almost half (i.e. 40%) of the exposure to PNCs as compare to those at ground floor (i.e. 1 m). While only 15% reduction in exposure to PNCs with height (i.e. from 1 m to 4.7 m) were observed in case of 3-way TI.

3.4 Horizontal variations of PNCs along the TIs

To analyse the horizontal variation in the PNCs, measurements were carried out at five different distances from the centre of the TI on two different legs (3 and 4) of the 4-way TI were monitored (Figure 1f). Figures 7a–b show the PNDs at five different points (i.e. P_R , P_1 , P_2 , P_3 and P_4 at 10, 20, 30, 45 and 60 m away from the centre of the TI, respectively) on legs 3 and 4 of the 4-way TI, respectively. The PND remains consistent in their shape for all the points, except P_2 , on both the legs, and show a general decay pattern with increasing distance from the centre of the TI. At the point P_2 , PND with 3 peaks in nucleation mode (5.6, 15 and 24 nm) and one peak in accumulation mode (65–100 nm) is similar to PNDs reported by Goel and Kumar (2015) during acceleration of the experimental car. Since P_2 is near to stop line (i.e. where vehicle stops when signal light turns to red) on both the legs, this PND

pattern could be attributed to an acceleration of vehicles after the signal turns from red to green.

Horizontal profiles of PNC at legs 3 and 4 of the 4-way TI are shown in Figure 7c–d. An exponential decay of PNCs is found with increasing distance from the centre of a TI at both the legs of the studied TI (Figure 7c–d). However, the rate of decay indicated by the slope of curve is about 2-times, higher on leg 3 (i.e. slope = 0.03) as compare to leg 4 (i.e. slope = 0.01). This can be explained on the basis of predominant wind direction during the measurements at these two different legs of the TI (Figure 7e–f). There are two components of wind acting at each of the five sampling points on both the legs: (i) parallel to the traffic flow, and (ii) perpendicular to traffic flow (Figure 7e–f). Parallel component of wind will always transport exhaust particles from one point to another. On the other hand, the perpendicular component of wind can bring fresh exhaust emissions either away or towards the sampling point, depending on the wind direction. At leg 4, $u\cos\phi$ represents perpendicular component of wind that brought the traffic exhaust emissions towards the sampling point (Figure 7f). While at leg 3, $u\sin\phi$ is the perpendicular component of the wind, which transports traffic exhaust emissions away from the sampling points (Figure 7e). Therefore the difference in PNCs at any two points, and hence the slope of decay profile, is expected to be larger at leg 3 compared with leg 4, mainly because only exhaust emissions transported by parallel component of the wind are contributing to PNCs at each of the five sampling points on leg 3. This observation is further supported by higher percentage (13, 14, 25, 27 and 46% at P_R , P_1 , P_2 , P_3 and P_4 , respectively) of accumulation mode (i.e. particles in the 30–300 nm size range) particles at various points on leg 3 compared with leg 4 (14, 13, 12, 20 and 18% at P_R , P_1 , P_2 , P_3 and P_4 , respectively). As for the comparison with the literature, we could only locate one study by Kim et al. (2013) that measured horizontal profiles of particles below 100 nm at 5, 30 and 80 m away from the centre of a 3-way TI in Tokyo (Japan), which was

surrounded by buildings. They found that the organic carbon in particles decays exponentially with increasing distance from the TI. The slope of their exponential decay was 0.02, which is reasonably well within a factor of 2 to those observed in our study at 4-way TI.

3.5 Estimation of size dependent PNEF for different driving conditions at the TIs

PNEFs present a functional relationship between particle number emissions and the activity that generate emissions (Goel and Kumar, 2014). Therefore, PNEFs are one of the most important input parameters for computing nanoparticle emissions and carrying out reliable dispersion modelling. There are several studies that have measured PNEFs either in laboratory or estimated using inverse modelling technique for highway, tunnel environment or on rural motorway. Review of these studies can be seen in Kumar et al. (2011). Based on the review, Kumar et al. (2011) reported that PNEFs of mixed vehicle fleet is in the range of 2.15-5.20, 0.31-2.7 and 0.23-2.8 $\times 10^{14}$ veh⁻¹ km⁻¹ on highways, tunnels and motorways, respectively. However, such information for TIs rarely exists. Keeping this in mind, we estimated size-dependent PNEFs for congested and free flow driving conditions at two different types of TIs using the methodology explained in the Section 2.4 and results are summarised in Table 6.

The PNEFs at 3- and 4-way TIs during congested conditions are found to be $6.41 \pm 3.32 \times 10^{14}$ and $7.69 \pm 10.90 \times 10^{14}$ veh⁻¹ km⁻¹, respectively (Table 6). A few studies (Gramotnev et al., 2003; Kumar et al., 2008a; Morawska et al., 2005) representing congested traffic driving conditions (i.e. traffic driving speed of <30 km h⁻¹) are selected from the literature for comparison with our results (Table 7). For example, Gramotnev et al. (2003) estimated PNEFs of 4.05×10^{14} veh⁻¹ km⁻¹ for a motorway in Brisbane for start and stop traffic conditions of mixed traffic fleet. Later, Morawska et al. (2005) estimated PNEF of 5.67 ± 2.80

521 $\times 10^{13} \text{ veh}^{-1} \text{ km}^{-1}$ for a busy street canyon in Brisbane (Australia) for start and stop traffic
522 conditions for a mixed traffic fleet (20% heavy duty vehicles). Further, Kumar et al. (2008a)
523 estimated PNEF in the range of $1.43\text{--}2.63 \times 10^{14} \text{ veh}^{-1} \text{ km}^{-1}$ for a street canyon in Cambridge
524 (UK) for a mixed traffic fleet (2% heavy duty vehicles) and traffic speed about 30 km h^{-1} .
525 Our average PNEFs at 3- and 4-way TIs were found to be within a factor of 3 to those
526 reported in above-mentioned studies (Table 7), except Morawska et al. (2005) where our
527 PNEFs for both TI are over a magnitude higher than those reported in Morawska et al.
528 (2005). There could be two possible reasons for this difference. Firstly, about 28% of the fleet
529 consisted of diesel cars in our study as opposed to negligible number of diesel fuelled
530 passengers cars in their study. Secondly, Morawska et al. (2005) used 15–700 nm size range
531 to estimate their PNEFs. Based on our measurements, PNCs in 5–15 nm size ranges
532 accounted for 48% and 69% of total PNCs at 3- and 4-way TIs (Figure 4a). Fleet
533 characteristics, their maintenance and inspection policies vary in different European
534 countries, leading to differences in the PNEFs.

535 Frequent changes in traffic driving conditions are one of the main reasons of high PNEF
536 during *congested* driving conditions at TIs that those during the free flow conditions (Goel
537 and Kumar, 2014, 2015a). A comparison has been made between PNEFs during congested
538 and free flow (i.e. during green traffic lights) conditions at TIs (Table 6). It is found that
539 PNEF during congested driving conditions at TIs can be up to 10-times of those during free
540 flow conditions. This is in line with the findings of Jayaratne et al. (2010) that found up to an
541 order of magnitude higher particle number emissions during acceleration compared with free
542 flow driving conditions from diesel and compressed natural gas buses.

543 The above discussions clearly show that the PNEFs during congested conditions are over a
544 magnitude higher than those during free flow conditions at the TIs. Inadequate treatment of

the effect of driving conditions on particle number emissions can result in inaccurate estimation of PNCs at the TIs.

3.6 Pedestrian exposure to PNCs at the studied TIs

To compare pedestrian exposure, hourly averaged RDD rates were estimated at both the TIs using the methodology described in Section 2.5. Average RDD rate at both our TIs was found to be $5.33 \pm 4.77 \times 10^{10} \text{ h}^{-1}$ (with $3.74 \pm 1.73 \times 10^{10}$ and $6.91 \pm 5.44 \times 10^{10} \text{ h}^{-1}$ at 3- and 4-way TIs, respectively; Figure 8). Average RDD rate at 4-way TI is about 85% higher than those at 3-way TI. These higher RDD rates at the 4-way TI could be due to both 60% higher PNC and up to 61% higher deposition fraction than those at the 3-way TI (Section 2.5).

The estimates of RDD rates on TIs are not available in the literature for direct comparison with our results. Therefore, we have converted the PNCs to RDD rates for some of the studies (Holmes et al., 2005; Wang et al., 2008) that made PNDs available in their research reports. Average RDD rate were estimated for Wang et al. (2008) and Holmes et al. (2005) as $0.34 \times 10^{11} \text{ h}^{-1}$ and $0.15 \times 10^{11} \text{ h}^{-1}$ for a 4-way TI in the Corpus Christi (USA) and Brisbane (Australia), respectively. Average RDD rate at the studied 4-way TI ($0.69 \times 10^{11} \text{ h}^{-1}$) is about 2.0 and 4.6-times of those estimated for Wang et al. (2008) and Holmes et al. (2005), respectively. In case of Wang et al. (2008) the average PNCs was similar at their and our TIs (Table 2). However, shape and magnitude of PNDs were different in their and our studies. For example, about 81% of PNCs were in the 5–30 nm size range at our 4-way TI compared with 68% of PNCs in the study of Wang et al. (2008). In case of Holmes et al. (2005), average PNC was 50% of that observed at our 4-way TI. Since RDD rate is directly proportional to the product of PNCs and deposition fraction, 50% lesser RDD rate can therefore be expected. Also, percentage of PNCs in 5–30 nm range was 68% in Holmes et al. (2005) compared with 81% at our 4-way TI.

Above discussions show that the exposure at TIs can be many folds higher than the other urban environments. The exposure varies depending on the PNC and deposition fraction at different types of TIs. Therefore there is a need to carry out more studies at different types of TIs in diverse geographical settings. Such studies could also assist in developing a database, showing the contribution of exposure at TIs towards the overall daily exposure during commuting in diverse city environments.

3.6.1 Frequency of exposure doses at the TIs

Similar to PNCs, frequency histograms were plotted for RDD rate at both the TIs. These histograms showed that 99% of the time hourly averaged RDD rate was less than or equal to $0.7 \times 10^{11} \text{ h}^{-1}$ and $2.37 \times 10^{11} \text{ h}^{-1}$ at 3- and 4-way TIs, respectively. While 50% of the time, hourly averaged RDD rate was less than or equal to $3.51 \times 10^{10} \text{ h}^{-1}$ and $5.62 \times 10^{10} \text{ h}^{-1}$ at 3- and 4-way TIs, respectively. Most of the time (i.e. 62% and 70% of the time at 3- and 4-way TI, respectively), the hourly averaged RDD rate at studied TIs were found to exceed the average RDD rate in the *urban roadside* environments in European cities (i.e. $3.12 \times 10^{10} \text{ h}^{-1}$; Kumar et al., 2014). As expected, the hourly averaged RDD rate were found to exceed the average RDD rate during free flow driving conditions (i.e. $2.44 \times 10^{10} \text{ h}^{-1}$; Goel and Kumar, 2015a) for 75% and 79% of the time at 3- and 4-way TIs, respectively.

Probability functions were fitted to identify the “best” and “common” fit distributions for three averaging periods – short (1 s), medium (15 min) and long (1 h) term average RDD rates – at both the TIs. GEV and exponential probability functions were found to be a “best” fit distribution for 1s averaged RDD rate at 3- and 4-way TIs, respectively. While Log-Logistic was found to be the “best” fit distribution for 15 min and 1 h average RDD rate at both the TIs. GEV is found to be the “common” fit distribution for different time averages of RDD rate at both the TIs (SI Table S10 and S11). The best fit distributions identified for

PNCs in Section 3.2 are different than those identified for RDD rate at each of the studied TIs. This can possibly be explained by the dependence of RDD rate on the product of PNCs and deposition fractions.

4. Summary and conclusions

We measured the size-resolved PNCs in the 5–560 nm size range at the 3– and 4–way TIs. The aims were to present vertical and horizontal variations of PNCs, and estimate PNEFs and RDD rate at two different types of TIs. Probability functions were fitted to PNC and RDD data using a statistical distribution fitting tool (Easyfit) in order to understand the ranges of exposure doses at different types of TIs. Based on the ranking of fitted distributions, the “best” and “common” fit probability functions were identified at both the TIs. Pseudo-simultaneous measurements using a custom-built solenoid system with the DMS50 were made to measure vertical profiles at 4 different heights at both the TIs. Horizontal profiles were drawn by measuring PNCs at 4 different points in a sequential manner on two perpendicular legs of 4–way TI.

The following conclusions are drawn:

- Irrespective of any type of a TI, daily averaged PNDs were found to be trimodal in shape with peaks at 5.6, 10 and 56 nm. The magnitude of peaks at 5.6 and 10 nm were found to be 2.7– and 3.6–times larger at the 4–way TI compared with 3–way TI. Despite the relatively higher traffic volume at the 3–way TI, average PNCs at 4–way TI were found to be about twice to those observed at the 3–way TI, showing the dominating impact of signal cycle length (i.e. delay time) on the PNCs at a TI.
- Irrespective of any type of a TI, similar shape of PNDs were observed at all the 4 different heights corroborating with our previous findings in the context of street canyons (Kumar et al., 2008a) that the competing influences of transformation

processes is generally over by the time plume reach to vertically placed sampling points. At both the TIs, vertical profiles of PNCs followed an exponential decay, with a much sharper decay at the 4-way TI compared with 3-way TI.

- Horizontal profiles also exhibited an exponential decay profile of the PNCs with the increasing distance from the centre of a TI at both the legs of a 4-way TI. The slope of horizontal decay was flatter than that seen in case of vertical profiles, indicating that PNC decay is up to 132-times faster in vertical direction compared with horizontal direction. This decay is mainly due to measurements taken in the close vicinity of sources in the horizontal direction.
- The average PNEF at the 4-way TI was found to be 20% higher than those at the 3-way TI. Frequent changes in traffic driving condition are one of the main reasons of higher PNEFs during congestion compared with free flow conditions at TIs. We found up to an order of magnitude higher PNEFs during congested conditions at TIs compared with those estimated for free flow conditions. Consideration of the effect of driving condition on the PNEFs has implications for dispersion modelling of PNCs at the TIs since neglecting the effect of congestion can result in underestimation of PNCs and associated exposure at the TIs.
- Hourly average RDD rates at the 4-way TI during entire monitoring period were found to be 1.8-times of those at 3-way TI ($3.74 \times 10^{10} \text{ h}^{-1}$). It shows that even if the strength of emission sources (i.e. PNEF) are almost similar, exposure rates could vary notably at different types of TIs. Based on frequency histograms plotted for RDD rate, it was found that the hourly averaged RDD rate at 3- and 4-way TI, exceeded the average RDD rate at roadside urban locations in European cities (i.e. $3.12 \times 10^{10} \text{ h}^{-1}$; Kumar et al., 2014) for 62 and 70% of time, respectively.

The vertical and horizontal profiles as well as the PNEFs derived during the congested traffic conditions at the TIs could be useful to dispersion models for the TIs, which can help in accurately estimating the PNCs and hence people's exposure at the TIs. This work also opened up a number of questions for further research. For example, our study measured vertical profile of PNCs at most commonly occurring types of TIs i.e. 3- and 4-way in a typical town of UK. It showed that the exposure to PNCs decreases with an increase in height at both the TIs. However, the rate of decrease in exposure to PNCs with height vary at different types of TIs, depending on the built-up area around the TI, traffic volume, characteristics of traffic volume (i.e. number of heavy and light duty vehicles) and meteorological parameters (i.e. temperature and wind speed). In this study, we assessed the effect of built up area, traffic volume and characteristics of traffic volume on vertical profiles of PNCs. Further measurements focusing on the effects of meteorological parameters (i.e. temperature, wind speed and wind direction) on PNCs will be useful. We fitted probability functions to average PNC data at two types of TIs. Our work could be expanded to the TIs having diverse traffic and geographical conditions to build a database and develop fast parameterised dispersion models based on the probability functions to predict PNCs at the TI.

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Figure 1. (a) Google map showing the location of background sampling point and the (b) 4– and (c) 3–way TIs in Guildford. Sampling points for the: (d) vertical profile measurements at both the TIs, and (e) horizontal profile measurements at legs 3 and 4 of the 4-way TI. Reference point is marked as P_R , which was 10 m away from the centre of the TI. Sampling points P_1 , P_2 , P_3 and P_4 on each of the leg were at a distance of 20, 30, 45 and 60 m from the centre of the TI, respectively.

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891 **Figure 8.** The box plot of hourly averaged RDD rate at 3 and 4-way TI. The upper and lower
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894 percentile. Diamond represents the mean value. Yellow coloured circles represent the
895 percentage of PNCs in nucleation mode at two TIs.

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Table 1. Review of studies that have estimated PNEFs based on the field observations. The symbol “ σ ” and acronyms HDV and LDV refer to standard deviation, heavy duty diesel vehicle, and light duty diesel vehicle, respectively.

Study	Vehicle type	Range (nm)	PNEF $\pm \sigma$ (# veh ⁻¹ km ⁻¹) $\times 10^{14}$	Environment
Corsmeier et al. (2005)	Mixed fleet	10–400	4.70	Rural motorway
Morawska et al. (2005)	Petrol vehicle	18–880	0.22	Stop and start condition in street canyon
Morawska et al. (2005)	Diesel vehicle	18–880	2.04	Stop and start condition in street canyon
Hueglin et al. (2006)	Mixed fleet	7–3000	7.90	Motorway
Jones and Harrison (2006)	HDV	11–450	6.36	Street canyon
Jones and Harrison (2006)	LDV	11–450	0.12	Street canyon
Rose et al. (2006)	Cars	3–800	0.58 $\pm$ 0.2	Street canyon
Rose et al. (2006)	Trucks	3–800	25.0 $\pm$ 9.0	Street canyon
Kumar et al. (2008a)	Mixed fleet	5–1000	1.21–2.23	Street canyon
Wang et al. (2010)	Mixed fleet	10–700	2.15 $\pm$ 0.05	Highway
Wang et al. (2010)	Mixed fleet	10–700	1.87 $\pm$ 0.03	Urban site
Nickel et al. (2013)	HDV	14–750	11.10	Motorway
Nickel et al. (2013)	LDV	14–750	2.10	Motorway
Ripamonti et al. (2013)	Mixed fleet	3–950	6.03 $\pm$ 0.19	Road in a semi-urban area
This study ^a	Mixed fleet	5–560	6.41 $\pm$ 3.32	3–way TI
This study ^a	Mixed fleet	5–560	7.69 $\pm$ 10.9	4–way TI

^aThe details of methodology used to estimate them are available in Section 2.5 and the results are discussed in Section 3.5.

903 **Table 2.** Summary of fixed-site monitoring studies at different TIs.

Author (year)	City (Country)	Type of a TI	Instrument	Sampling Frequency (Hz)	Size range (nm)	Average PNC ($\times 10^4 \text{cm}^{-3}$)	Traffic Density (h^{-1})	HDV (%)	Months
Morawska et al. (2004)	Salzburg (Austria)	4-way located in a street canyon	SMPS	300 ^a	13–830	2.2	3600	20– 30%	September
Holmes et al. (2005)	Brisbane (Australia)	4-way	SMPS	120 ^a	9–407	2.6 ^b	200 – 1920	20% ^c	January
Tsang et al. (2008)	MongKok of Kowloon (Hong Kong)	3-way located in a street canyon	WCPC	1	5–2000	4.5	840	29%	July
Wang et al. (2008)	Texas (USA)	4-way	CPC & SMPS with DMA	120 ^a	7–290	5.1	10452 – 11897	4%	December –June
Fujitani et al. (2012)	Kawasaki City (Japan)	4-way	SMPS	120 ^a	8–300	9.1	2167	25%	January
This study	Guildford (UK)	4-way	DMS50	10	5–560	5.3	5014	4%	February– April
This study	Guildford (UK)	3-way	DMS50	10	5–560	3.4	5498	6%	January

904 Note: ^aScan time in seconds; ^bMaximum PNCs; ^cPercentage of diesel vehicles including cars.

905 **Table 3.** Shape, location and scale value of common fit for three different time averages of PNCs (N_{5-560}), N_{5-30} , N_{30-300} and RDD rate at two
906 different types of TIs.

Parameter	Type of the TI	Name of fit	Probability function	Averaging time	Shape (ξ)	Scale (σ)	Location (μ)
PNC (# cm ⁻³)	3-way	Generalised extreme value Generalised extreme value	$\frac{1}{\sigma} t(x)^{\xi+1} e^{-t(x)}$ where $t(x) = \left(1 + \left(\frac{x-\mu}{\sigma}\right) \xi\right)^{-\frac{1}{\xi}} \quad \text{if } \xi \neq 0$ $t(x) = e^{-\frac{(x-\mu)}{\sigma}} \quad \text{if } \xi = 0$	1s	0.1345	18453	25994
				15 min	0.0249	16751	26642
				1h	0.0439	17528	25044
	4-way			1s	0.5649	12064	12370
				15 min	0.1080	26172	34053
				1h	0.0440	29774	34926
N ₅₋₃₀ (# cm ⁻³)	3-way			1s	0.3017	10143	15094
				15 min	0.0448	9609	18979
				1h	0.1936	9199	22037
	4-way			1s	0.7599	11613	11088
				15 min	0.3563	11468	13516
				1h	0.3465	12139	14685
N ₃₀₋₃₀₀ (# cm ⁻³)	3-way	Generalised Pareto	$\frac{1}{\sigma} (1 + \xi z)^{-\frac{1}{\xi}}$ Where $z = \frac{x - \mu}{\sigma}$	1s	0.5501	33698	-2535
				15 min	0.5965	31087	-1739
				1h	0.7092	32676	-716
	4-way			1s	0.5592	4065	6938
				15 min	0.0880	5404	8865
				1h	0.1061	5412	9345
RDD rate (# h ⁻¹)	3-way	Generalised extreme value	$\frac{1}{\sigma} t(x)^{\xi+1} e^{-t(x)}$ where $t(x) = \left(1 + \left(\frac{x-\mu}{\sigma}\right) \xi\right)^{-\frac{1}{\xi}} \quad \text{if } \xi \neq 0$ $t(x) = e^{-\frac{(x-\mu)}{\sigma}} \quad \text{if } \xi = 0$	1s	0.201	1.57 × 10 ¹⁰	2.47 × 10 ¹⁰
				15 min	0.077	1.52 × 10 ¹⁰	2.76 × 10 ¹⁰
				1h	0.208	1.44 × 10 ¹⁰	3.27 × 10 ¹⁰
	4-way			1s	0.731	1.52 × 10 ¹⁰	1.63 × 10 ¹⁰
				15 min	0.337	1.49 × 10 ¹⁰	2.02 × 10 ¹⁰
				1h	0.332	1.55 × 10 ¹⁰	2.17 × 10 ¹⁰

907

Table 4. Best fit distributions for PNCs in 5–30 and 30–300 nm size ranges at 3– and 4–way TIs.

Type of TI	Size range	Averaging time	Best fit
3-way TI	PNC in 5–30 nm range	1 s	GEV
		15 min	Exponential
		1 h	Normal
4-way TI		1 s	Exponential
		15 min	Burr
		1 h	Lognormal
3-way TI	PNC in 30–300 nm range	1 s	Exponential
		15 min	Log-Logistic
		1 h	GEV
4-way TI		1 s	Weibull
		15 min	GEV
		1 s	GEV

Table 5. Review of studies for vertical concentration profiles of nanoparticles and gaseous pollutants.

Study/ authors	Size range (nm)	Location	Measurement heights	Observations on vertical profiles
Imhof et al. (2005)	30–10000	60 m from the motorway surrounded by land of agricultural use without trees or buildings	3.0, 18.0 and 33.0 m	Largest PNCs at 3 m and then decreasing with increasing height
Zhu and Hinds (2005)	7–3000	50 m downwind from the centre of freeway near a residential area	0.6, 3.0, 5.5, 8.0, 10.4, 12.8, 15.3 and 17.7 m	Largest PNCs at 3 m and then decreasing with increasing height
Kumar et al. (2008a)	5–1000	Asymmetric street canyon	0.2, 1.0 and 2.60 m	Largest near the bottom decreases with increasing height
Kumar et al. (2008b)	5–2738	Symmetric street canyon	1.0, 2.3, 4.6 and 7.4 m	Smallest at 1.0 m, largest at 2.3 m and then decreases with increasing height
He and Dhaniyala (2012)	> 2.5	Highway with no buildings and tree	0.6, 2.0, 2.7, 3.4, 4.1, 4.9, 6.3, 7.7 and 9.1 m	Smallest at 0.6 m and largest PNCs at 3.4 m and then decreasing with increasing height
Nakashima et al. (2014)	250–3000	3–way TI with tall buildings (6–12 stories) located alongside all roads near to the site	0.3, 0.5, 1, 1.5 and 2 m	Largest PNCs at 0.3 m and then decreasing exponentially with increasing height
Present study	5–560	3–way TI with buildings (2–4 stories) located alongside all roads near to the site	1, 1.5, 2.5 and 4.7 m	Largest PNCs at 1 m and then decreasing with increasing height
Present study	5–560	4–way TI with 3 2 stories houses located on one side	1, 1.5, 2.5 and 4.7 m	Smallest PNCs at 1 and largest at 2.5 m and then decreasing with increasing height

Table 6. Summary of the estimated PNEF at the studied TIs. Please note that PNEF during free-flow conditions are average emission factors for ten different TIs, which are estimated based on the findings of our previous campaign (Goel and Kumar, 2015a). The PNEFs at 3- and 4-way TIs refer to those estimated using all the data while on different legs to those using segregated data for each leg based on approaching wind direction.

TI types	Congested = Free flow + stop-and-go				Free flow	Ratio of total PNEF at TI/PNEF free-flow
	$\text{PNEF}_{\text{total}}$ (# veh ⁻¹ km ⁻¹) × 10 ¹⁴	PNEF_{5-30} (# veh ⁻¹ km ⁻¹) × 10 ¹⁴	PNEF_{30-300} (# veh ⁻¹ km ⁻¹) × 10 ¹⁴	$\text{PNEF}_{300-562}$ (# veh ⁻¹ km ⁻¹) × 10 ¹¹	$\text{PNEF}_{\text{total}}$ (# veh ⁻¹ km ⁻¹) × 10 ¹⁴	
3-way TI	6.41±3.32	4.45±2.61	1.96±0.94	1.48±2.40	0.74±0.60	9
Leg 1 of 3-way TI	6.63±5.01	4.15±4.75	2.48±0.61	5.22±8.57	0.74±0.60	9
Leg 2 of 3-way TI	6.60±2.75	4.74±1.93	1.85±0.84	0.75±1.21	0.74±0.60	9
4-way TI	7.69±10.9	6.05±9.60	1.64±1.74	2.00±3.68	0.74±0.60	10
Leg 1 of 4-way TI	2.49±2.22	1.88±1.99	0.61±0.36	0.48±0.58	0.74±0.60	3
Leg 2 of 4-way TI	6.61±2.96	4.75±2.45	1.86±0.65	0.83±0.27	0.74±0.60	9
Leg 4 of 4-way TI	6.56±6.46	5.40±5.45	1.16±1.06	0.23±0.19	0.74±0.60	9

923 **Table 7.** Published PNEFs from on-road studies during last decade.

Study	Vehicle type	Range (nm)	PNEF $\pm\sigma$ (# km ⁻¹ veh ⁻¹) $\times 10^{14}$	Environment
Gramotnev et al. (2003)	Total traffic = 2928 vehicles h ⁻¹	15–700	4.05	Motorway for start and stop traffic conditions
Morawska et al. (2005)	The share of HDV = 15%	15–700	0.57 $\pm$ 0.28	Busy street canyon for start and stop traffic conditions
Kumar et al. (2008a)	Total traffic = 1566 vehicles h ⁻¹ the share of HDV = 2%	5–1000	1.43–2.63	Street canyon having traffic with traffic speed of 30 km h ⁻¹
Present study	Total traffic = 5014 vehicles h ⁻¹ the share of HDV = 4%	5–562	7.69 $\pm$ 10.9	4-way TI
Present study	Total traffic = 5498 vehicles h ⁻¹ the share of HDV = 6.3%	5–562	6.41 $\pm$ 3.32	3-way TI

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