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Review

A review of fundamental drivers governing the emissions, dispersion and exposure to vehicle-emitted nanoparticles at signalised traffic intersections

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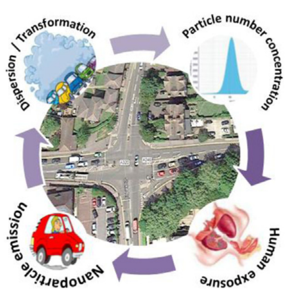
HIGHLIGHTS

- Parameters governing the emissions, dispersion and exposure at TIs are reviewed.
- Extent of PNCs and their exposure at TIs relative to other environments is analysed.
- Critical knowledge gaps in dispersion modelling parameters of PNCs are discussed.
- Peak PNCs at TIs were found 17-fold higher compared with average roadside PNCs.
- Nucleation is a foremost process, followed by dilution, deposition and coagulation.

GRAPHICAL ABSTRACT



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ABSTRACT

Signalised traffic intersections (TIs) are considered as pollution hot-spots in urban areas, but the knowledge of fundamental drivers governing emission, dispersion and exposure to vehicle-emitted nanoparticles (represented by particle number concentration, PNC) at TIs is yet to be established. A number of following key factors, which are important for developing an emission and exposure framework for nanoparticles at TIs, are critically evaluated as a part of this review article. In particular, (i) how do traffic- and wind-flow features affect emission and dispersion of nanoparticles? (ii) What levels of PNCs can be typically expected under diverse signal- and traffic-conditions? (iii) How does the traffic driving condition affect the particle number (PN) emissions and the particle number emission factors (PNEF)? (iv) What is the relative importance of particle transformation processes in affecting the PNCs? (v) What are important considerations for the dispersion modelling of nanoparticles? (vi) What is extent of exposure at TIs with respect to other locations in urban settings? (vii) What are the gaps in current knowledge on this topic where the future research should focus? We found that the accurate consideration of dynamic traffic flow features at TIs is essential for reliable estimates of PN emissions. Wind flow features at TIs are generally complex to generalise. Only a few field studies have monitored PNCs at TIs until now, reporting over an order of magnitude larger peak PNCs ($0.7–5.4 \times 10^5$ cm^{-3}) compared with average PNCs at typical roadsides (~0.3×10^5 cm^{-3}). The PN emission and thus the PNEFs can be up

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to an order of magnitude higher during acceleration compared with steady speed conditions. The time scale analysis suggests nucleation as the fastest transformation process, followed by dilution, deposition, coagulation and condensation. Consideration of appropriate flow features, PNEFs and transformation processes emerged as important parameters for reliable modelling of PNCs at TIs. Computation of respiratory deposition doses (RDD) based on the available PNC data suggest that the peak RDD at TIs can be up to 12-times higher compared with average RDD at urban roadsides. Systematic field and modelling studies are needed to develop a sound understanding of the emissions, dispersion and exposure of nanoparticles at the TIs.

1. Introduction

Airborne nanoparticles (referred here to those below 300 nm to represent majority of particle number concentrations, PNCs) come from a variety of exhaust and non-exhaust sources in the urban environments (Kumar et al., 2013a). Road vehicles are a major source of nanoparticle emissions (Johansson et al., 2007; Keogh et al., 2009; Kumar et al., 2011a; Shi et al., 2001), and these can contribute up to 90% of total PNC in polluted urban environments (Kumar et al., 2010a; Pérez et al., 2010; Pey et al., 2009). Small size of nanoparticles enables them to enter deeper into lungs, causing both acute and chronic adverse health effects such as asthma, cardiovascular and ischemic heart diseases (HEI, 2013). However, the number of excess deaths that occur in cities worldwide due to the exposure to nanoparticles are yet largely unknown (Kumar et al., 2014). A very few preliminary estimates are available on this topic, showing high numbers. For instance, Kumar et al. (2011b) showed that the exposure to particle number (PN) emissions from road vehicles in Delhi caused 11,252 excess deaths in 2010 that were predicted to reach to 58,268 by 2030 under the business as usual scenario.

Majority of cities worldwide are facing challenges associated with the air pollution (Kumar et al., 2013b). For example, a recent report of World Health Organisation on ambient air pollution suggests that annual mean concentration of PM_{10} (particulate matter less than 10 μm) has increased by more than 5% between 2008 and 2013 in 720 cities across the world (WHO, 2014). The issue of air pollution becomes more prominent at certain locations, such as signalised traffic intersections (TIs) with high pollutant concentrations, which are generally termed as “hot-spots”. Whilst some studies (Mohan et al., 2007; Wu et al., 2010; Zhu et al., 2008) define hot-spots as a localised place where maxima of air pollutant concentration can occur, the United Stated Environmental Protection Agency (USEPA) defines these as small geographical locations such as the TIs and the busy roadsides where pollutant concentration is higher than the National Ambient Air Quality Standards (NAAQS). In case of airborne nanoparticles, neither such a definition nor NAAQS are yet available for comparison and distinguishing the hot-spots in a particular area. Nonetheless, the same terminology can be adopted for nanoparticles by using the typical average values of PNCs in urban environments as a reference value to identify the nanoparticle hot-spots. Recently, Kumar et al. (2014) compiled the data on roadside PNCs in 42 different cities worldwide. They found the average values of PNCs as $3.2 \pm 1.6 \times 10^4$ cm^{-3} and $1.2 \pm 1.0 \times 10^5$ cm^{-3} in European and Asian cities, respectively. These or other localised PNCs measured elsewhere can be taken as a preliminary threshold value for determining the nanoparticle hot-spots in urban areas.

Evidences of hot-spots for gaseous pollutants are available in abundance. For instance, Coelho et al. (2005) and Li et al. (2009) found that a frequent stop-and-go situation at TIs often results in excessive delays, speed variations, alleviated fuel consumption and gaseous emissions. Likewise, hot-spots of nanoparticles can frequently occur at TIs due to the creation of pollution pockets by changing traffic conditions (e.g. acceleration-deceleration, stop-go). However, a very few studies have measured PNCs at the TIs to present an exhaustive picture of nanoparticle hot-spots in urban areas (see Table 1). These studies have found up to ~17- and 5-folds larger values of peak PNCs at the TIs (e.g. $5.4 \pm 1.7 \times 10^5$ cm^{-3}; Tsang et al., 2008) compared with the average typical values of roadside PNCs in European and Asian cities, respectively (Kumar et al., 2014). A number of practical and technical constraints such as portable instruments having high sampling response and broad size range, their low-cost and robustness for continuous unattended monitoring, and lack of standardised measurement methods make the study of nanoparticles at TIs even rarer (Kumar et al., 2011a). This is reflected by the fact that there are not many field studies available for TIs (Table 1), clearly indicating a need for more measurement studies to understand PNC levels in diverse traffic and driving conditions. These studies would be instrumental for developing particle number emission factors (PNEF) that are one of the key inputs for dispersion modelling which is, in turn, important for understanding the exposure to vehicle-emitted nanoparticles at TIs.

As seen in Table 2, a number of review articles are currently available in the published literature. Although these articles either deal with the flow and dispersion of gaseous pollutants at TIs (e.g. Ahmad et al., 2005; Tiwary et al., 2011) or particle transformation processes (dilution, nucleation, coagulation, condensation, evaporation and deposition) at various spatial scales (e.g. Kumar et al., 2011c; Carpentieri et al., 2011). For instance, Ahmad et al. (2005) summarised the results of wind tunnel simulations for TIs. They also discussed the effects of building configurations, canyon geometries and variability in approaching wind directions on flow fields and exhaust dispersion at TIs. Tiwary et al. (2011) reviewed the state-of-the-art knowledge on modelling the airflow and concentration fields of inert pollutants at TIs. Kumar et al. (2011c) discussed dispersion modelling techniques of nanoparticles at five local scales (vehicle wake, street, neighbourhood, city and road tunnels). However, the complexities associated with the emissions, dispersion and exposure related to vehicle-emitted PN emissions at TIs have not been discussed in detail until now (see Table 2).

The aim of this review is therefore to assess the fundamental drivers that govern the emissions, dispersion, concentration and exposure to PNCs at TIs. In order to set the background context for our review article, the key traffic and wind flow features at TIs are first briefly presented (Section 2). This is followed by an up to date summary of field studies that have monitored PNCs at TIs over the past one decade (Section 3) and the effect of traffic driving conditions and meteorology on PNEFs (Section 4). Further section presents a discussion on relative importance of particle transformation processes in altering the ambient PNCs at TIs (Section 5). A

Table 1
Summary of relevant field studies covering measurements of PNCs at the TIs.

Author (year)	City (Country)	Instruments	Size range (nm)	Maximum PNC ($\times 10^5$ cm^{-3})	Traffic density (h^{-1})	HDV (%)	Months	Remarks
Morawska et al. (2004)	Salzburg (Austria)	SMPS	13–830	0.2[a]	3600	20–30%	September	Sampling point was around 5 m away from the road and 1.2 m above the ground.
Holmes et al. (2005)	Brisbane (Australia)	SMPS	9–407	2.6	200–1920	20%[c]	January	Monitoring is carried out at one fixed site that is surrounded by river on south side and buildings on other three corners.
Tsang et al. (2008)	Mong Kok of Kowloon (Hong Kong)	WCPC	5–2000	5.4	840	29%	July	Monitoring was carried out at three fixed sites. Intersection was street canyon intersection bounded on all sides by high rise buildings. These sites were located at 1m, 5 m and 6 m distance from intersecting roads, respectively.
Wang et al. (2008)	Texas (USA)	CPC & SMPS with DMA	7–290	2.4	10452–11897	3.7%	December–June	Mobile sampling was carried out at four corners of an intersection along with sampling at one fixed site on the south-east corner of the intersection. Measurements were conducted both in upwind and downwind direction of both roadways.
Oliveira et al. (2009)	Optro (Portugal)	CPC	6–700	1.07	2500	25%[c]	July	Sampling site was located in the city centre at 3 m distance from intersecting roads.
Fujitani et al. (2012)	Kawasaki City (Japan)	SMPS	8–300	~1.4	2167	25%	January	Monitoring was carried out at intersection of industrial road and main highway.
Holder et al. (2014)	North Carolina (USA)	EEPS	6–560	0.7[b]	–	–	April	Mobile measurements were carried out on a specified route to assess the spatial variability.

Note:
[a]Average PNC;
[b]90th percentile;
[c]Proportion of diesel-fuelled vehicles out of total vehicle fleet; EEPS = Engine Exhaust Particle Sizer; SMPS = Scanning Mobility Particle Sizer; CPC = Condensation Particle Counter; WCPC = Water-based CPC.

simplified approach to carry out dispersion modelling of nanoparticles at TIs is then presented (Section 6). Further, critical synthesis of published information on intermittent exposure experienced by urban dwellers at these hot-spots compared with exposures in other urban environments is discussed (Section 7). The review finally concludes with summary, conclusions and grey areas requiring further research (Section 8).

2. Traffic and wind flow features at TIs

Detailed study of both the traffic and wind flows at TIs is important to understand the PN emissions, dispersion and transformation of nanoparticles. Motorised road-traffic is the main source of both the PN emissions and traffic produced turbulence (TPT) at TIs. On the other hand, wind flow plays an important role in dispersion of nanoparticles released by the traffic at TIs. Since there are already specialised reviews and research articles available on this topic, as summarised in Table 2, we have briefly discussed the key traffic and wind flow features in subsequent sections for the sake of completeness and setting up context for the dispersion modelling of nanoparticles (Section 6).

2.1. Key feature of traffic flow

The estimation of PN emissions from road traffic at TIs requires in-depth understanding of traffic characteristics such as category-wise volume of traffic, technology distribution, and driving conditions (André and Hammarström, 2000). Traffic emission estimates, which are based on traffic-counts and use of different statistical methods, are often not accurate and do not reflect the dynamic behaviour of the traffic flow (Pandian et al., 2009). In-depth analysis of 'speed' and 'acceleration' in specific situations (e.g. stop-and-go at traffic lights and overcrowded roads) using the traffic-flow models can provide reliable emission estimates (Pandian et al., 2009; Schmidt and Schäfer, 1998). Underestimation of vehicle speed or flow rate may lead to drastic increase in emissions (Negrenti, 1999). For instance, Eisele et al. (1996) reported that 10–30% of underestimation of traffic volume can result in up to 50% of underestimation of carbon monoxide emissions on local arterial roads. Such an underestimation can also be expected for nanoparticle emissions. Estimates of traffic flow features based on both the traffic count and traffic-flow models are therefore necessary for accurate assessment of PN emissions at TIs (Pandian et al., 2009).

Traffic flow models can be broadly classified into three categories – *microscopic*, *macroscopic* and *mesoscopic* – based on their functionality. For instance, *microscopic* models describe both the space-time behaviour (i.e. car following, lane changing, merging, and diverging) of vehicles in short time steps (down to 0.1 s). These models are used for small geographical areas such as the TIs in urban areas. *Macroscopic* flow models describe traffic flow at a high level of aggregation without distinguishing its constituent parts. The traffic stream is represented in an aggregate manner using characteristics such as their flow-rate, density, and velocity (Tolujew and Savrasov, 2008). These models are generally used for regional or city scale transport planning and management. *Mesoscopic* models falls between *microscopic* and *macroscopic* models. These models simulate individual vehicles, but describe their activities and interactions based on aggregate (macroscopic) relationships. These models are used for simulating traffic characteristics on large highway networks.

Mesoscopic and *macroscopic* models are used to assess the larger geographical area such as large highway network and city. They cannot capture the detailed effect of traffic control at the TIs (Zhang and Ma, 2012). Therefore, *microscopic* simulation models are often preferred for TIs since these can capture dynamic movement of vehicles in detail. In-depth review of the capabilities and usefulness of these models for traffic flow modelling can be seen elsewhere (Chowdhury et al., 2000; Pandian et al., 2009).

Table 2
Summary of review article discussing flow field and dispersion modelling of inert pollutants at TIs, besides studies focussing on nanoparticle dispersion modelling and regulation implications during the past one decade.

Author (year)	Study focus
Ahmad et al. (2005)	Reviewed the effect of building configurations, canyon geometries, traffic induced turbulence and variable approaching wind directions on flow fields and exhaust dispersion in urban street canyons and intersections, based on wind tunnel simulations studies.
Biswas and Wu (2005)	Reviewed the state of knowledge on formation and potential use of manufactured and anthropogenic airborne nanoparticles.
Holmes and Morawska (2006)	Reviewed the dispersion modelling techniques that can be applied within different environments, in regards to scale, complexity of the environment and concentration parameters.
Nowack and Bucheli (2007)	Classified different types of nanoparticles and summarised their formation, emission, occurrence and fate in the environment.
Buseck and Adachi (2008)	Discussed physical and chemical properties of airborne nanoparticles and their significance from health and climate change perspective.
Ju-Nam and Lead (2008)	Discussed physicochemical aspects of manufactured and natural aquatic nanoparticles to assess their toxicity and fate in the natural aquatic environment.
Morawska et al. (2008)	Reviewed information on vehicle generated ultrafine particles related to their characteristics and dynamics in the air in the context of the human exposure and epidemiological studies as well as in relation to their management and control in vehicle affected environments.
Morawska et al. (2009)	Reviewed the existing instrumental methods to monitor airborne nanoparticles in different types of indoor and outdoor environments.
Simonet and Valcárcel (2009)	Described some methodological aspects relating to the fields of nanoparticle analysis, nanometrology and analytical chemistry.
Kumar et al. (2010a)	Reviewed potential prospects of regulatory control for atmospheric nanoparticles, recent advances on this topic and future research priorities.
Kumar et al. (2010b)	Compared the behaviour of manufactured and vehicle derived airborne nanoparticles and discussed the consequences for prioritising research and regulation activities.
Kumar et al. (2010c)	Discussed the potential impact of the particle number concentrations derived from biofuel vehicles on existing regulatory concerns over atmospheric nanoparticles.
Morawska (2010)	Summarised the state of knowledge on possible health impacts of airborne engineered nanoparticles generated in commercial and research facilities.
Carpentieri et al. (2011)	Reviewed the research work relevant to modelling the dispersion of nanoparticles in vehicle wake.
Knibbs et al. (2011)	Reviewed the state of knowledge on determinant, variability and transport mode-dependence of exposure to ultrafine particles during commuting.
Kumar et al. (2011b)	Synthesised information related to current practices of nanoparticle dispersion modelling at five different local scales (i.e. vehicle wake, street canyons, neighbourhood, city and road tunnels).
Kumar et al. (2011a)	Discussed the technical challenges that are needed to be tackled before developing a regulatory framework for atmospheric nanoparticles.
Morawska et al. (2011)	Reviewed the existing regulations, policy measures and health guidelines related to reduction of airborne particulate matter (both on mass and number based) concentration.
Tiwary et al. (2011)	Reviewed the current practice in monitoring, modelling flow fields and inert pollutant concentrations at urban road intersections and the implications for commuter exposure.
Kumar et al. (2012)	Discussed the importance of nanoparticles generated by building and construction activities and their associated exposure.
Kumar et al. (2013b)	Synthesised the existing information on 11 non-vehicle exhaust sources of urban nanoparticles.
Kumar et al. (2014)	Reviewed the studies related to road traffic-emitted particle number emissions and concentrations in European and Asian cities and presented an integrated evaluation of emissions and population exposure.

2.2. Key feature of wind flow

Wind flow within and above the TIs is challenging to describe. This is because of the complex geometry, TPT, roadway design and atmospheric stability (Carpentieri et al., 2012; Tiwary et al., 2011). Wind flow at TIs is typically studied through wind tunnel experiments, numerical simulation or combination of both. A systematic review of key wind tunnel and numerical simulation studies at TIs is presented in Table 3. The findings of these studies suggest that two different types of turbulences (atmospheric and mechanical) affect the wind flows at TIs. Atmospheric turbulence is produced by: (i) the interaction of wind with the complex geometry of TIs, and (ii) the turbulence generated by the atmospheric stability conditions. Mechanical turbulence is produced due the interaction of ambient air with the moving traffic that is generally referred to as TPT. Detailed description of the effect of these turbulent mechanisms on wind flow at TIs and their relevance to nanoparticle modelling is presented in subsequent sections.

2.2.1. Effect of wind produced turbulence (WPT) on wind flows

When aerodynamically rough and inhomogeneous surface interacts with wind flows, turbulence is created due to the formation of an intense shear layer near the top of the canopy and by the wakes behind individual roughness elements such as towers and buildings. This turbulence is generally termed as WPT. This efficiently mixes and diffuses momentum, heat, moisture or any other scalar quantity (Roth, 2000). Detailed assessment of interaction between wind flow and surrounding geometry is therefore important in order to quantify the WPT.

The wind flow features at TIs are more complex than the flow features in a single street or road, due to the interaction of flow around several buildings and streets (Carpentieri et al., 2012). Hoydysh and Dabberdt (1994) carried out wind tunnel experiments for a grid of orthogonal streets, measuring concentrations of a tracer gas at a symmetrical TI. This study demonstrated that concentrations vary significantly at various locations around the TIs, with maximum values of tracer gas concentration being consistently seen at street corners. Their work also showed that the street aspect ratio had an important influence on dispersion conditions at these TIs.

Small asymmetries in geometry or wind directions can lead to a very different flow and dispersion pattern at the TIs (Balogun et al., 2010; Kastner-Klein et al., 1997; Robins et al., 2002). Scaperdas and Colvile (1999) performed Computational Fluid Dynamics (CFD) simulations to study the detailed wind flow features at a TI of symmetrical and asymmetrical canyons. Later, Soulhac et al. (2009) carried out wind tunnel experiments to study the wind flow features at a TI of symmetrical canyons. Their findings are summarised in Table 4, which suggest that pollutant transfer from one street to another is driven by the mixing at TIs in case of symmetrical street canyon geometry. However, pollutant transfer becomes significant as soon as there are minor departures from symmetrical to asymmetrical geometry of street canyons (Aristodemou et al., 2009; Balogun et al., 2010; Robins et al., 2002). Area of influence of a TI (AII) changes radically along with the changes in wind directions.

Table 3
Review of some of the key wind tunnel and numerical simulation references (studies) explaining features of wind flow at the TIs.

Studies	Study focus
Wind tunnel simulations	
Hoydysh and Dabberdt (1994); Dabberdt et al. (1995)	Studied the dispersion of tracer gas at TI of orthogonal streets by means of wind tunnel experiments. They found that maximum tracer gas concentration values were consistently located at street corners, and the street aspect ratio had an important influence on dispersion conditions at the TIs.
Kastner-Klein et al. (1997)	Studied the tracer gas concentration field at simple perpendicular TIs of symmetrical street canyons for reference wind direction of 90° by means of wind tunnel experiments.
Robins et al. (2002)	Studied the dispersion of tracer gas at a simple TI of two perpendicular streets through wind tunnel experiments. They found that the exchange of wind flow between the main street and side street were negligible in symmetrical situation. Small departures in symmetry were sufficient to establish significant exchanges.
Klein et al. (2007)	Studied the effect of building height on wind flow and pollutant dispersion pattern at urban TI through wind tunnel experiments.
Carpentieri et al. (2009)	Studied the mean flow, turbulence and flow path lines at a street canyon TI by using flow visualisation and Laser Doppler Anemometry methods in wind tunnel experiments.
Brixey et al. (2009); Heist et al. (2009)	Carried out wind tunnel measurements and CFD simulations of wind flow features in an idealized urban array of three story row houses. They also studied the effect of tall tower located at downwind edge of one of these houses on wind flow features.
Soulhac et al. (2009)	Studied wind flow and dispersion mechanism at urban TIs of orthogonal streets. By using the results of wind tunnel and numerical modelling, they developed a new operational model for pollutant exchanges at the TIs.
Carpentieri et al. (2012)	Studied mean and turbulent tracer flux balance in geometries of real street canyon TI (DAPPLE site) through wind tunnel experiments.
Kukačka et al. (2012)	Studied the vertical advection and turbulent scalar fluxes at X-shaped TI for five different reference wind directions by means of wind tunnel experiments.
Ahmad (2013)	Studied the combined effect of traffic induced turbulence and natural wind flow on tracer gas dispersion at urban TI through wind tunnel experiments.
CFD simulations	
Gadilhe et al. (1993)	Carried out numerical simulation of wind flow for a complex and realistic TI using a standard k-ε model of turbulence. They found fairly good agreement between wind tunnel measurements and model results.
Scaperdas and Colvile (1999)	Carried out CFD simulations to understand the effect of wind direction on small scale dispersion patterns at TIs.

For example, in case of a symmetrical TI, at reference wind direction $(ø) = 0°$, AII penetrates to no more than two street widths into either side of the street (Garbero et al., 2010). While in case of asymmetrical TI, at $ø = 0°$, the AII varies from H to $5H$ into either side of the street; where H is the height of tallest building around the studied TI (Scaperdas and Colvile, 1999). AII becomes more extensive in case of an oblique reference wind direction. For instance, at symmetrical TI, at $ø = 10°$, AII increases beyond five street widths in one side street and falls to zero in other side street (Garbero et al., 2010). A few wind tunnel (Brixey et al., 2009; Heist et al., 2009) and CFD simulation (Brixey et al., 2009; Heist et al., 2009; Scaperdas., 2000) studies have also assessed the influence of small and tall towers placed at the corners of street canyons on the flow and turbulence field at corners of the TIs. They found that the presence of a tower enhanced wind speed in (and ventilation from) surrounding street canyons and forced a strong lateral flow into the side streets (see Table 3).

Interaction of the wind flow characteristics among the intersecting streets at the TIs is challenging to model and is still poorly understood (Balogun et al., 2010). This is mainly because the flow field data at TIs are scarce and have just started to become available. Wind flow features at the TIs affect the dilution of traffic emissions and dilution affects the transformation processes of nanoparticles. Therefore, there is a need to carry out more wind flow modelling studies by means of physical and numerical modelling to understand the key flow features and develop nanoparticle dispersion models for TIs.

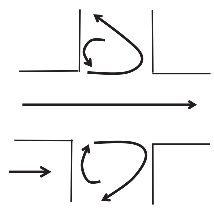
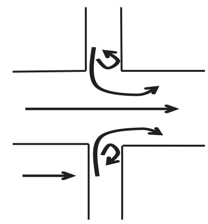
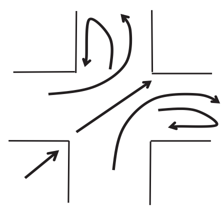
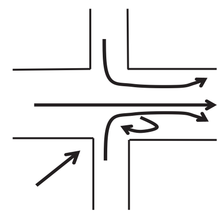
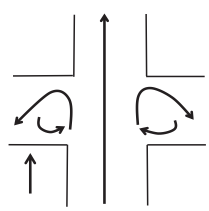
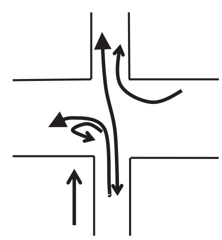
2.2.2. Effect of TPT on wind flows

TPT is turbulent kinetic energy generated due to the movement of road traffic (Wang and Zhang, 2009). This plays an important role in the dispersion of nanoparticles near roadways, especially during low prevailing winds. For instance, Jicha et al. (2000) carried out CFD simulation and Kastner-Klein et al. (2001) performed wind tunnel experiment to study the effect of moving traffic on wind flow features in a street canyon. They both found that one-way traffic increases ventilation of a canyon during perpendicular winds by enhancing the circulation in the canyon. Berkowicz et al. (2002) found from their CFD simulations that the TPT can affect the vertical dispersion of pollutants up to a height of ~4 m (of ~21 m high canyon) in an urban canopy layer. Di Sabatino et al. (2003) proposed a theoretical framework to estimate the TPT in street canyons at low wind speed conditions. They derived the parameterisation for TPT, which was suitable for low-, intermediate- and high-traffic density in street canyons. The follow-up study by Kastner-Klein et al. (2003) modified the parameterisation derived for TPT by incorporating the combined effect of WPT and TPT. Recently, Ahmad (2013) carried out a wind tunnel experiment for investigating the effects of heterogeneous traffic on line source dispersion in close proximity of a TI surrounded by symmetric street canyons. They observed that TPT greatly influences the tracer gas concentration at various points around the studied TI due to generation of large size eddies. It is clear from available studies that the effect of TPT on line source dispersion in street canyons has been carried out in some detail, but corresponding information for TIs is yet to become available in abundance.

2.2.3. Effect of atmospheric stability on wind flows

Atmospheric stability is defined in terms of the tendency of a parcel of air to move upward or downward after it has been displaced vertically by a small amount. This is thus an important parameter affecting dispersion or build-up of pollutants in the atmospheric environment. Uehara et al. (2000) performed the wind tunnel experiments to study the effect of atmospheric stability on wind flows in regular urban street canyons. They found that the turbulence within the canyon became weaker under stable atmospheric conditions. The mixing in the street canyon was enhanced during unstable atmospheric conditions. Later, Kikumoto et al. (2009) performed large eddy simulation to study the effect of atmospheric stability on dispersion conditions in an urban street canyon. They found that turbulence is accelerated by the buoyancy effects in street canyons during unstable atmospheric conditions. Conversely, the flow is depressed by thermal stratification in stable atmospheric conditions, and the pollutant stagnates near the bottom of the canyon. Although the effect of atmospheric stability on flow field has been studied in detail to some extent for street

Table 4
Comparison of flow field features at a TI of symmetrical street canyons of aspect ratio of 1 (Case 1) and TI of symmetrical and asymmetrical street canyons (Case 2) (Scaperdas and Colvile, 1999; Soulhac et al., 2009).

Wind direction with respect to street reference	Case 1 (street canyons in both the directions are symmetrical and they all have aspect ratio = 1)	Case 2 (street canyons in *y*-direction are asymmetrical and street canyons in *x*-direction are symmetrical)	Key characteristics
$\varnothing = 0^{\circ}$			In both cases, flow separation at the upstream corner of the side streets leads to the formation of recirculating vortices at the entrance of two side streets.
$\varnothing = 45^{\circ}$			In Case 1, flow field is symmetrical about diagonal with identical recirculation regions in two downstream streets. Whereas in Case 2, flow field is symmetrical about *X*-direction with a recirculation vortex forming around one of the corners.
$\varnothing = 90^{\circ}$			In Case 1, flow field is simply a rotated and version of those obtained for $\varnothing = 0^{\circ}$. However in Case 2, flow field shows symmetry about *Y*-axis with pronounced recirculation vortex forming around only one of the corners.

canyons (Tiwary et al., 2011; Uehara et al., 2000), but no such studies are available for TIs.

3. Field measurements of particle number and size distributions at TIs

Only a handful of studies have monitored the PNC at TIs and findings of these studies are summarised in Table 1. The peak PNC measured by these studies have been found to vary in the 0.7–5.4 × 10^5 cm^{-3} range, showing up to ~7-times differences among the PNCs measured by them. The lowest peak PNC (0.7 × 10^5 cm^{-3}) was observed at a TI site in the USA where mobile measurements were taken and sulphur content in diesel and gasoline at the time of measurements was less than 10 and 30 ppm, respectively. The highest peak PNC (5.4 × 10^5 cm^{-3}) was observed at a TI in Hong Kong where measurements were taken 1 m away from the roadside and sulphur content in diesel and gasoline at time of measurement was less than 50 ppm (EPD, 2014). Kumar et al. (2014) highlighted a number of factors that are likely to be responsible for the variability observed in peak PNCs. For instance, lower cut-off for PNC measurements varied between 5 and 9 nm in studies listed in Table 1 and this can account for up to ~12% of total PNC (Kumar et al., 2009a). Distance of measurement location from TI is another consideration. In case of unobstructed topographic setting, PNC can decrease up to ~40% of their kerbside level within a distance of ~10 m (Kumar et al., 2014). Some of variability in PNC can be explained by seasonal effects (e.g. temperature inversion) that have been found to significantly increase the PNC during cold months (Buonanno et al., 2013). Average PNCs have been found up to ~300% higher during winters than those during rainy season for identical traffic emission conditions (Byčenkienė et al., 2014). Sulphur content of diesel and gasoline used in road transport is another important factor. Reduction in sulphur content of diesel from 50 to 10 ppm can result up to ~30% reduction in PNCs (Jones et al., 2012). In summary, nearly ~400% of variability can be expected among PNC values reported in Table 1 due to experimental set-up, fuel types and seasonal conditions. The rest of the variability can be attributed to the other local factors such as traffic volume, background concentration, and interrupted traffic flow and driving conditions specific to individual sampling locations.

For a detailed understanding of particle dynamic and dispersion at TIs, the combined effect of various factors such as wind and traffic flow, driving conditions, metrology and road grade on PNCs must be assessed. Out of the reviewed studies (Table 1), Fujitani et al. (2012) measured the PNCs at a TI in an open area. Holmes et al. (2005) and Oliveira et al. (2009) examined spatial distribution of PNCs around urban TI sites. Tsang et al. (2008) analysed the effect of driving conditions on PNCs at a TI. Holder et al. (2014) carried out mobile measurements to study the effect of driving conditions on concentrations of ultrafine and black carbon at a TI. Except Wang et al. (2008), none of these studies assessed the effect of flow (wind and traffic) dynamics on PNCs at TIs since this was not the original focus of these studies. Greater numbers of field studies are clearly needed to improve our understanding of the dispersion of nanoparticles at and around the TIs.

Most of the studies listed in Table 1 have used CPC (Condensation Particle Counters) or mobility particle size spectrometers that are often referred to as SMPS (Scanning Mobility Particle Sizer) or

Table 5
Description and limitations of various PNEFs derivation techniques.

Method	Description	Influencing factors	Limitations	Source
Laboratory testing (engine and chassis dynamometer studies)	Emission measurements are carried out under controlled conditions in laboratories. PNEFs derived by this method represent the strength of source, but do not take into account the effect of transformation processes on PNEFs.	Vehicle type, driving condition, engine load	PNEFs derived by this method may not represent PN emissions under real-world conditions with sufficient accuracy.	Jayaratne et al. (2009); Morawska et al. (1998); Ristovski et al. (2005); Ristovski et al. (2004)
Measurement under real-world conditions (road tunnel, remote sensing, on-road chasing technique and on-board emissions measurements)	Emission measurements are carried out under real-world conditions to yield the data regarding the actual emission behaviour of road vehicles in real world conditions. Since measurements are carried out near the receptor, PNEFs derived by this method takes in to account the effect of transformation processes but does not represent the actual strength of the source.	Vehicle type, driving condition, engine load, road grade and metrology	In case of road tunnel studies, it is challenging to apportion emissions to specific vehicle classes unless different tunnel bores are dedicated to them. In case of remote sensing technique, instantaneous emission rates associated with driving conditions at a particular point on the road are measured. Therefore these may not be a representative of average emissions over a full drive cycle. On-road chase techniques are best conducted on a test track due to traffic safety considerations. On-board measurements such as portable emission measurements systems add additional mass (~30–70 kg) to the vehicle that may bias the measurements, especially for light-weight cars.	Samaras et al. (2005); Holmen et al. (2005); Janhäll et al. (2004); Rosenbohm et al. (2005); Morawska et al. (2005); Nickel et al. (2013); Keogh and Sonntag (2011); Wehner et al. (2009); Hak et al. (2009); Franco et al. (2013)
Inverse modelling technique	PNEF are derived on basis of roadside measurements after accounting for dispersion of particles using an inverse modelling approach.		Since the method uses a nanoparticle dispersion model to estimate the PNEFs, the accuracy of the estimated PNEFs depends on the ability of the model to reproduce the dispersion of PN emissions.	Kumar et al. (2008a); Kumar et al. (2008b); Morawska et al. (2005); Zhang et al. (2004)

DMPS (Differential Mobility Particle Sizer) to monitor particle number size distributions at TIs (Wiedensohler et al., 2012). Depending on the manufacturer and the model number, the scanning time of SMPS varies and can be typically in the 30–300 s range, with a detection limit of up to 10^8 cm^{-3} (Kumar et al., 2010a; TSI, 2014a). Same is the case with the CPC, which have a typical response time of about 5 s and detection limit of 10^7 cm^{-3} (TSI, 2014b). These instruments are suitable for fixed-site measurements at TIs, however their portability may be an issue for mobile measurements within the vehicles. The challenges for mobile monitoring arise due to instruments' size and a need of clean and continuous source of power (e.g. from batteries), which itself does not produce exhaust emissions (e.g. diesel electricity generators). Some of these instruments (e.g. CPC) contain a reservoir of volatile liquid butyl alcohol, which may spill during mobile measurements and there may be loss of data until it returns to normal position (PMS, 2013). Most of currently available instruments are able to measure the maximum level of concentrations expected during fixed or mobile monitoring at TIs (Kumar et al., 2010a). The instruments with fast sampling response can even capture the rapid evolution of particle number size distribution due to competing influences of transformation processes (Kumar et al., 2011c).

A very few studies have recorded the particle number size distributions at TIs. Data extracted from these studies are summarised in Supplementary Information (SI) Fig. S1, which shows particle number size distributions at: (i) a TI of a highway (Fujitani et al., 2012), (ii) a TI of an arterial road (Holder et al., 2014), (iii) a TI surrounded by two street canyons that have building height between 15 and 30 m (Holmes et al., 2005), and (iv) at a roadside in an urban street canyon (Kumar et al., 2008a) to show their comparison with those recorded at TIs. As expected, all the TIs show much higher magnitude of particle number size distributions compared with roadside measurements in street canyons. In general, a much higher nucleation mode particles at TI can be expected due to diverse driving behaviour (e.g. acceleration, deceleration, idling) compared with the free flow traffic conditions on non-congested roads (see Section 4.1.2).

4. PNEFs and PN emission modelling

4.1. PNEFs at TIs

PNEF presents a functional relationship between PN emissions and the activity data that generate emissions. This is one of the most important input parameters for computing nanoparticle emissions and carrying out dispersion modelling. Broadly there are three methods to derive emission factors: (i) laboratory testing based on engine and chassis dynamometer studies, and (ii) direct on-road and on-board measurements under real-world driving cycle, and (iii) using inverse modelling techniques. Brief comparison of these approaches and examples are presented in Table 5.

As summarised in Table 5, a number of factors influence the estimation of PNEFs, including meteorology, road grade, vehicle types, speed, load and driving condition, lower and upper cut-off values of particle size range measured, and sulphur content of the fuel. For instance, meteorology affects the time scale and importance of various transformation processes and hence the estimates of PNEFs that are based on the ambient PNCs using the inverse modelling approach (Kumar et al., 2011c).

4.1.1. Effect of road grade

Road grade, which is a percentage rise or drop in vertical distance with respect to horizontal distance, also affects the PNEF estimates due to the change in the engine power demand of a vehicle. For instance, there is a less demand of engine power during downhill movement of a vehicle as compared to uphill movement, as shown in SI Fig. S2. A conventional diesel bus engine was found to emit ~8-times less PNCs for downhill movement as compare to uphill movement (Holmen et al., 2005). The PNEFs are therefore expected to change in the similar fashion as do the PNCs. This effect was demonstrated by Zheng et al. (2013a) where they found up to an order of magnitude larger PNEF from a heavy duty diesel truck during uphill driving compared with downhill driving. Their study measured only *solid* particles using a CPC that had cut-off size range of 23 nm and temperature of primary dilutor and evaporation tube was 150 and 350 °C, respectively. Furthermore, different levels of PN emissions are expected to be released by vehicles due to differences in engine technology and fuel use. For instance, PNEF of heavy duty vehicle was found in the range of ~10^{14}–10^{15} veh^{-1} km^{-1}, which are up to an order of magnitude larger than those for gasoline–fuelled (~10^{12}–10^{14}) and diesel-fuelled (~10^{14}) cars (Kumar et al., 2011c).

4.1.2. Effect of interrupted traffic flow

Traffic situation at TIs remains generally complex since the traffic flow is interrupted due to the restrictions laid by traffic signals. These restrictions lead to frequent changes in driving conditions such as deceleration, idle, acceleration and cruise (Papson et al., 2012). The PN emissions released during all these conditions and hence the corresponding PNEFs can also vary accordingly, due to constantly changing fuel consumption and engine load (Chen and Yu, 2007; Lei et al., 2010).

Numerous studies have measured an increase in PNCs as a result of vehicle acceleration, confirming the increased PN emissions due to accelerating conditions. For instance, Tsang et al. (2008) carried out a study to assess the pedestrian exposure to PNCs at a busy TI in Mong Kok, Hong Kong. They observed a sharp increase in PNCs as a result of vehicle acceleration after ~3 s when the traffic signal changed from red to green. Wang et al. (2008) found that average PNCs at TIs during red-light periods are nearly 5-times higher compared to those during green-light periods (Fig. 1). A most recent study by Johnston et al. (2013) monitored nanoparticles from motor vehicles at a TI in Wilmington, Delaware, USA. They observed abrupt peaks in PNCs that varied from a few second to tens of seconds after the traffic signal changed from red to green. Jayaratne et al. (2010) found up to an order of magnitude higher PN emissions during acceleration compared with steady driving conditions for diesel and CNG buses. Sulphur content of the fuel also plays a major role in the formation of nanoparticles and consequently influences the PNEFs, as discussed in Section 3

4.1.3. PNEF databases

A number of individual studies have measured PNEFs under laboratory and real-world conditions, as summarised in Table 5. In addition to these studies, two comprehensive databases (Computer Programme to calculate Emission from Road Transport, COPERT4; and PARTICULATE; Luhana et al., 2004) based on chassis dynamometer testing are also available for the PNEFs under different driving conditions. COPERT4 provides PNEFs for solid particles in

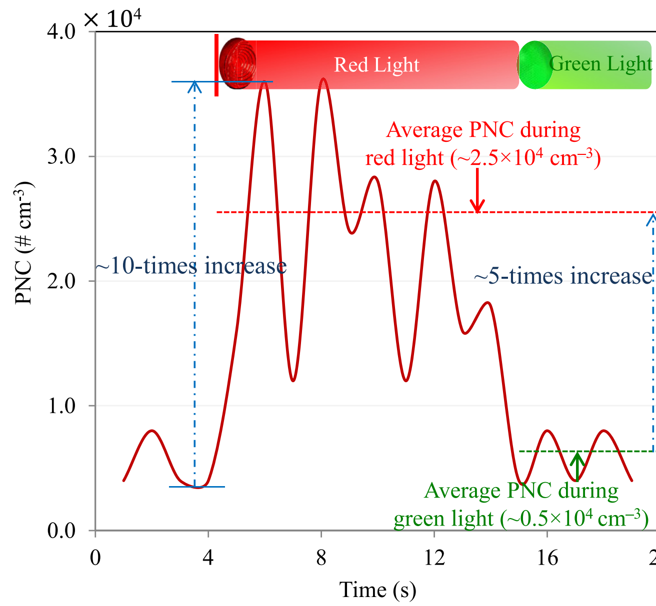


Fig. 1. Temporal variation in PNCs as a function of traffic light at a TI (Wang et al., 2008).

the size range of 50–1000 nm for different types of vehicles (e.g. passenger car, buses, coaches and heavy duty vehicles) under urban, rural and highway driving conditions (Ntziachristos et al., 2000). Similarly, PARTICULATE program was launched by European Union in the year 2000. The aim of this project was to study both the nucleation as well as solid particles in the 7–1000 nm size range by measuring them for a variety of vehicles under a range of engine capacities, fuels and technologies (Kulmala et al., 2011). However, studies covering the dynamic and complex situation of traffic emissions to estimate the PNEFs at TIs are yet rarely available. One way of accounting the effect of driving changes on nanoparticle emissions at the TIs is the estimation of PNEFs with respect to delay events (see SI Section S1). However, frequent driving changes at the TIs make the PNEFs derived by roadway or highway studies unsuitable to TIs. Despite the availability of numerous PNEF databases (Keogh et al., 2010, 2009; Kumar et al., 2011c) there is clearly a lack of PNEF databank that could explicitly be applicable to emission modelling of nanoparticles at the TIs.

4.2. Microscopic emission model

An approach to model the nanoparticle emissions at the TI is the use of microscopic emission models, which can provide a precise description of vehicle emission behaviour by relating emission rates to vehicle operation during a series of short time steps. These models can broadly be classified into the following categories:

- *Statistical models*: These models adopt mathematical functions of instantaneous speed and acceleration to predict the emission rates. These estimates are of generally high quality, but lack a physical interpretation and can also over-fit the calibration data (Lei et al., 2010; Rakha et al., 2004).
- *Load-based models*: These models estimate the fuel consumption rate of a vehicle to derive tailpipe emissions on the basis of engine out emission and efficiency of after treatment technology (Huang, 2009; Li et al., 2009). The major disadvantage with this type of models is their complex numerical structure and need of high computational efforts.

- *Emission map models*: These models are typically matrices that contain the average emission rates for combination of speed and acceleration in the driving cycles used. These are often based on steady-state data and are highly sensitive to the driving cycle, offering modest flexibility to account for important factors such as road grade, driver characteristics, or the interaction between the driver and different roadway elements (Barth et al., 1996; Huang, 2009).

A summary of the capabilities and characteristics of microscopic emission models is presented in Table 6. At present such models are available for gaseous pollutants and coarse particulate matter (on a mass basis), but not for the nanoparticles. Nonetheless, these can be adopted for nanoparticles by incorporating suitable PNEFs that are able to reflect dynamic traffic conditions seen at the TIs.

5. Importance of particle dynamics at the TIs

Vehicle emissions consist of hot gases and primary particles, which are highly dynamic and reactive in nature (Kumar et al., 2011c). Just after the release of PN emissions from vehicular exhaust, physical and chemical composition of particles changes rapidly due to the effect of transformation processes (Carpentieri and Kumar, 2011; Kumar et al., 2009b). In order to assess the relative importance of various transformation processes on particle number and volume concentrations, we derived their timescales for typical TIs that are presented in Table 7, using the methodology described in SI Section S2. These preliminary values of time scales can be taken as a relative measure of the time taken to reduce the PNCs at TI, if the source was turned off. Thus a short time scale indicates a strong effect of that particular process on the PNCs. The time scale analysis suggests that nucleation as the most important process at TIs, followed by dilution, deposition, coagulation and condensation. Brief description of these processes, along with a comparison of their timescales for the TIs to those for the street canyons is presented. Such information is essential since an inadequate treatment of these processes may result in inaccuracies in prediction of PNCs at the TIs (Section 7).

Table 7
Time scale analysis of various transformation processes at TIs along with their effect on PN and particle volume concentration. Where symbol +, −, 0 represent gain, loss and no effect on number and volume concentrations, respectively.

Transformation processes	Effect on concentration		Time scale (s)[a]
	Number	Volume	
Nucleation	+	+	80
Dilution	±	±	10^2
Dry deposition	−	−	10^3
Wet deposition	−	−	−
Coagulation	−	0	5×10^3
Condensation	0	+	$0.4–8 \times 10^4$
Evaporation	0/−	−	−

[a] Detailed calculations of time scale analysis are explained in SI Section S2.

Nucleation leads to formation of new particles (initial size around 1.5–2 nm) through gas-to-particle conversion (Kulmala et al., 2004). This happens when cooling and condensation of hot gases generated from tailpipe of vehicles are mixed with the ambient air (Kumar et al., 2011c). Time scale of nucleation process at TIs is ~80 s for the nucleation mode particles production rate of 10^3 cm^3 s^{-1} (Table 7). This time scale is ~8 times higher than those for the nucleation in street canyons (~10 s; SI Section S2). Due to formation of new particles, this process increases the particle number and volume concentration at the TIs.

Dilution occurs directly after the release of emissions from the tailpipe of vehicles. It is a key process that induces the other transformation processes to act and alter the number and size distributions. Time scale of dilution process at TI is estimated ~10^2 s (Table 7), which is up to ~3 times higher than those for regular street canyons (~40 s; Kumar et al., 2008b). It may increase or decrease the number and volume concentrations at the TIs, depending on the dilution ratio, meteorological parameters and gas phase chemistry.

Dry and wet deposition can be explained as the removal of the particles either at air–surface interfaces or by precipitation, respectively (Laakso et al., 2003; Seinfeld and Pandis, 2012). Dry deposition is mainly driven by Brownian diffusion and inertial impaction (Kumar et al., 2011c). Wet deposition is mainly driven

Table 6
Comparison of different types of microscopic emission models.

Source	Model	Type	Road grade	Vehicle categories	Pollutant types	Cold start emission	Remark
Rakha et al. (2003)	Comprehensive modal emission model (CMEM)	L	N	LDV and HDV	CO_2, CO, HC and NOx	Y	Model exhibit abnormal behaviour at low speed and high acceleration.
Boulter et al. (2007)	Digitized Graz Model (DGV)	EM	N	Cars	CO_2, CO, HC, PM and NOx	Y	–
Boulter et al. (2007)	MODEM	EM	N	Cars	CO_2, CO, HC, PM and NOx	N	Model estimation for particulate matter emissions is not good. Therefore there is a need to look at PM emission estimates cautiously.
Boulter et al. (2007)	Passenger car and Heavy-duty Emission Model (PHEM)	EM	N	Cars and HDVs	CO_2, CO, HC, PM and NOx	N	HDV part of the model does not include the distortion in emissions due to traffic signals. Emission maps are solely based on the regulatory testing and hence significantly underestimate the emission levels.
Boulter et al. (2007)	Vehicle Transient Emissions Simulation Software (VeTESS)	EM	Y	Cars and HDVs	CO_2, CO, HC, PM and NOx	N	Model also contains emission maps for transient conditions.
Rakha et al. (2003)	Virgina Tech Microscopic energy and emission model (VT-Micro)	S	Y	LDVs and trucks	CO, HC and NOx	N	Emission estimates by model are found consistent with the laboratory measurements.

Note: Y = Yes; N = No; L = Load based model; S = Statistical model; EM = Emission map based model; LDV = Light duty vehicle; HDV = Heavy duty vehicle.

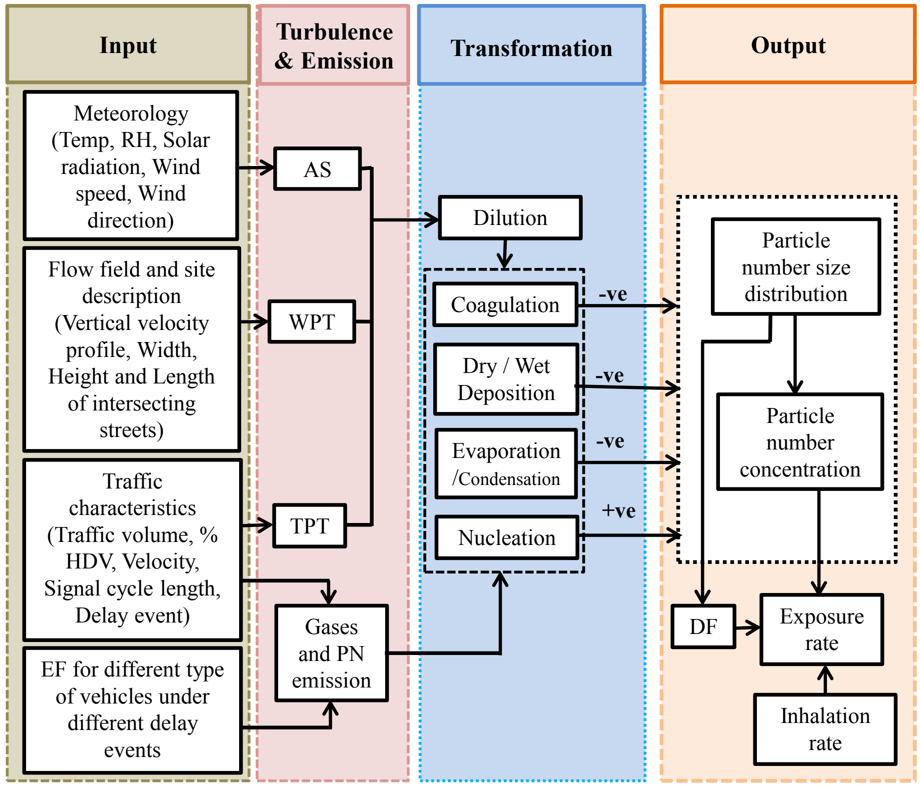


Fig. 2. A simplified modelling framework for estimating PNC and associated exposure at the TIs. Acronyms AS, DF, PN, EF, HDV, TPT and WPT refer to atmospheric stability, deposited fraction, particle number, emission factors, heavy-duty vehicles, traffic-produced turbulence and wind-produced turbulence, respectively.

by nucleation scavenging (i.e. rainout) and aerosol-hydrometeor coagulation (i.e. washout). Dry deposition is one of the dominant removal mechanisms for the nucleation mode particles (Hinds, 1982). On the other hand, wet deposition (rainout) plays an important role in removing the larger-sized particles (Jacobson and Seinfeld, 2004). Time scale of dry deposition at TIs is estimated ~10^3 s (Table 7), which is over an order of magnitude larger than those for regular street canyons (~30 s and 130 s for 10–30 nm and 30–300 nm size of particles, respectively; Kumar et al., 2008b). Deposition process can reduce both the number and volume concentrations of particles to significant levels at the TIs.

Coagulation is a process in which particles collide due to their random (Brownian) motion and coalesce to form larger-sized particles (Kumar et al., 2011c). Time scale of polydisperse coagulation at TIs is estimated ~5×10^3 s (Table 7), which is up to two orders of magnitude lower than those for regular street canyon (~5×10^5 s for 30–300 nm size particles; Kumar et al., 2008b). Coagulation process reduces the number concentration of smaller particles but shows no effect on volume concentration.

Condensation and evaporation are diffusion-limited mass transfer process between the gas-phase and the particle-phase, governed by the higher vapour pressure of condensable species in the air surrounding the particles (Kumar et al., 2011c). Time scale of condensation process at TIs is estimated as ~$0.4–8 \times 10^4$ s for growth rate of 1 and 20 nm h^{-1}, respectively (Table 7), which is similar for 1 nm h^{-1} (~10^4 s) but about an order of magnitude lower for 20 nm h^{-1} (~10^5 s) than those for regular street canyons (Kumar et al., 2008b). Condensation helps to grow the volume of particles but does not change their number concentrations. Evaporation works as an opposite process to condensation where the volume of the particles reduces and in some cases it may cause the volatile particles to completely disappear (Kumar et al., 2011c).

These transformation processes are responsible for some of spatial and temporal variability in particle number and size distribution (Birmili et al., 2013). Relative contribution of various transformation processes in altering the PNCs at the TIs is not yet been experimentally quantified, but these are important to consider for more accurate dispersion modelling of PNCs at the TIs (see Section 7).

6. Dispersion modelling techniques

6.1. Important considerations for dispersion modelling of nanoparticles at TIs

Traffic generated PN emissions often increase in the vicinity of TIs. Numerous factors such as complex wind flow patterns and transformation processes determine the concentrations of nanoparticles in the intersecting streets at TIs. A simplified approach to perform dispersion modelling of nanoparticles and associated exposure at the TIs is presented in Fig. 2. Summary of a number of governing factors that can be used to assess the suitability of currently available dispersion models at TIs is presented below.

(i) Disrupted stop-and-go traffic flows at TIs compel the vehicles to accelerate and decelerate, and thereby increasing the PN emissions. Therefore the PNEFs capable of capturing the effect of these dynamic conditions at TIs are required in order to make reliable PN emissions estimates in dispersion models (Section 4.1).

(ii) TIs are regions with a significant exchange of pollutants between the intersecting streets. Therefore the dispersion models for nanoparticles should be able to take account of the complex flow field induced by these exchanges at TIs (Section 2.2).

(iii) Dispersion models should be able to adequately treat dilution and complex transformation processes that occur after the release of exhaust gases into the ambient environment (Section 5).

Consideration of (i) is related to uncertainties associated with input parameters whilst the latter two considerations (ii + iii) relate to structural uncertainties in the dispersion models (see Section 6.2).

6.2. Suitability of currently available aerosol and inert pollutant models for TIs

There are currently a very few models that are especially designed to predict PNCs by taking into account the particle dynamics. The summary of these models, which can be used at various spatial scales, is provided in Kumar et al. (2011c). Table 8 includes the detailed characteristics of some of these models that take into account the detailed particle dynamics at local scales and can be used at the TIs after appropriate modifications.

There are infrequent studies which have performed PNC modelling at the TIs. One such study is by Wang et al. (2013) that used CTAG (Comprehensive Turbulent Aerosol Dynamics and Gas Chemistry) model and divided the study area in two domains: “exhaust-to-road” and “road-to-ambient”. This model taken in to account the effect of WPT, TPT and atmospheric stability on wind flow features. To incorporate the effect of TPT at a TI, the study considered vehicles as stationary objects and resultant wind velocity as vector sum of external wind speed and vehicle velocity. However, interaction of the wakes of individual vehicles and external air depends upon the traffic density and surrounding geometry (Di Sabatino et al., 2003). This simplification may not truly represent the actual turbulence features at TIs. Also, this study has used the PNEF derived on the basis of average vehicle velocity and percentage of HDVs at intersecting highways. As evident from discussions presented in Section 4.1, PNEFs at TIs are highly dependent on driving conditions and consideration of average velocity is likely to affect accuracies of PN emission estimations. However, CFD models can provide detailed flow and dispersion characteristics, these are generally complex to use, require extensive computation and expertise, and are not easily accessible for free use.

A few models addressing dispersion of gaseous pollutants and particulate matter (on a mass basis) at TIs are currently available. These include Gaussian puff and hybrid models (Tiwary et al., 2011). The USEPA has recommended several Gaussian type operational air quality models such as California Line Source Model (CALINE-4), California Line Source Model with Queuing and Hotspot Calculation (CAL3QHC/CAL3QHCR), Hybrid Roadway Intersection Model (HYROAD) and Canyon Plume Box Model (CPB-3) suitable for modelling air quality near TIs (EPA, 2008). However, majority of these models do not take into account the full effect of TI's geometry and TPT on pollutant dispersion (Tiwary et al., 2011; Vardoulakis et al., 2007). This inadequate treatment of flow features may result in large uncertainties in predicted pollutant concentration at TIs. Theoretically, models for gaseous pollutants based on CFD or hybrid modelling could be modified by incorporating particle dynamics module in them and providing appropriate PNEFs for the dispersion modelling of nanoparticles at TIs. Likewise, models developed for inert pollutant dispersion, especially for TIs (e.g. SIRANE; Soulhac et al., 2009), can be modified by incorporating dynamic PNEFs and particle dynamics modules in order to incorporate the (i + iii) consideration (Section 6.1).

One of the major limitations is that the currently available dispersion models are developed for inert pollutants, based on the simplified geometries of TIs, and therefore may not be applicable elsewhere. At the same time, it is not feasible to develop a single “universal” model that can be used for all different types of geometric configurations of TIs. Development of geometry-specific dispersion models, which can also account for particle transformation, are therefore needed for reliable estimation of PNCs and exposure at the TIs.

7. Exposure assessment at TIs

Understanding of spatio-temporal distribution of nanoparticles in urban environments is of significant concern for the accurate exposure assessment (Birmili et al., 2013; Kumar et al., 2014). To compare the PNC exposure observed in different urban environments, an extensive review of existing studies, falling into 4 different categories (urban background, street canyon, roadside and traffic intersections), is carried out. Details of these studies and summary of their results are presented in SI Table S1 and Fig. 3, respectively. Average PNC and geometrical mean diameter of particle number size distribution for each environment was calculated by averaging studies in individual categories. Fig. 3a clearly shows that the highest average PNCs are observed at TIs, followed by the roadside, street canyons, and urban background. Average PNCs at TIs were found to be ~1.5 and 1.9-times higher than those in roadside and street canyons, respectively. It is worth noting that these comparisons are based on the averaged PNCs and if short term averaging (e.g. 1 s) during peak conditions is considered the corresponding differences were found to increase to ~17 and 21-times, respectively. The higher PNCs at TIs are expected due to complex wind flow conditions (Section 2.2) accompanied by frequent changes in driving conditions of vehicles (Section 4.1.2). An interesting trend emerged from this analysis, showing an exponential increase in PNCs from urban background, to street canyons, to roadside, to PNCs at TIs with a significant correlation

Table 8
Characteristics of a few currently available aerosol dynamic models.

Model	Type	Nucleation	Coagulation	Condensation/ Evaporation	Deposition	Source
MAT	Combination of a plume model with a 1-D Langrangian trajectory framework	N	Y	Y	Y	Ketzel and Berkowicz (2005)
GATOR -GCMM	Unified fully coupled online model that account for major feedbacks among metrology, chemistry, aerosol, cloud and radiation	Y	Y	Y	Y	Zhang (2008)
ADCHEM	Langragian	Y	Y	Y	Y	Roldin et al. (2011)
CTAG	CFD	Y	Y	Y	Y	Wang et al. (2013)

Note: Y = Yes; N = No; MAT = Multi-plume Aerosol dynamic and Transport; GATOR-GCMM = Gas, Aerosol, Transport, Radiation, General Circulation and Mesoscale Meteorological; ADCHEM = Aerosol Dynamic, gas and particle phase CHEMistry; CTAG = Comprehensive Turbulent Aerosol Dynamics and Gas Chemistry.

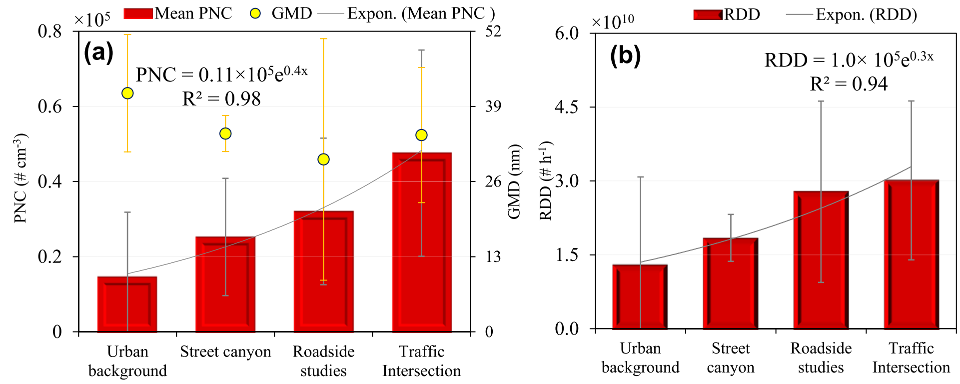


Fig. 3. (a) Summary of average PNCs and the corresponding geometrical mean diameters (GMD) observed in different environments; these values are taken as average of various studies summarised in SI Table S1. (b) Summary of average RDDs estimated for different environments, based on the average PNC values presented in SI Table S1. In studies where GMD was not given, average GMD of that environment is considered to quantify appropriate RDD. Also are shown the correlation among the PNC values (and RDD) in different environments. For the correlation equations shown in both the figures, *x* is equal to 1, 2, 3 and 4 for urban background, street canyon, roadside and traffic intersection, respectively.

factor ($R^2 = 0.98$; Fig. 3a). An interpretation of this relationship could be that if PNC of any of the above-mentioned environments is known, the PNC in other environments can be approximated by using the exponential relationship seen in Fig. 3a. For example, Birmili et al. (2013) measured PNCs at the urban background and roadside locations in Dresden, Germany. By using the relationship shown in Fig 3a, roadside PNCs are predicted as 2.1×10^4 cm^{-3} based on measured urban background PNCs (9.8×10^3 cm^{-3}). These predicted values show a fractional bias of ~0.29 compared with those actually measured at the roadside (2.8×10^4 cm^{-3}), indicating these within the generally expected fractional bias range of ±0.5 (Rim et al., 2013). However, this is a statistical relationship based on a limited dataset and therefore should be generalised cautiously.

Higher PNC does not mean higher respiratory deposited doses (RDD), as the fraction of nanoparticle deposited in respiratory system depends upon the size of particles (ICRP, 1994). RDD is higher for small size particles and decreases in power form for larger particles, as demonstrated by a variety of urban PNC studies (Al-Dabbous and Kumar, 2014; Kumar and Morawska, 2014; Kumar et al., 2014). Therefore, understanding of particle number size distribution is crucial for accurate estimation of RDD. In this study, RDD rate in each environment is calculated using methodology presented in SI Section S3. As expected, the highest RDD rate based on the average PNCs is found at the TIs (~$3.0 \pm 1.6 \times 10^{10}$ h^{-1}), followed by roadside (~$2.8 \pm 1.8 \times 10^{10}$ h^{-1}), street canyon (~$1.8 \pm 0.5 \times 10^{10}$ h^{-1}) and urban background (~$1.3 \pm 1.8 \times 10^{10}$ h^{-1}) locations (Fig. 3b). It is worth noting that these estimates are based on the average PNCs observed in each environment and consideration of peak PNCs might further increase the RDD rate. For instance, total RDD rate becomes 34.2×10^{10} h^{-1} based on peak PNC observed at TIs (see Table 1), which is ~12-times higher than those estimated on the basis of average roadside PNCs.

Short-term exposure under peak PNC conditions at TIs is not very well characterised, but this may contribute to significant portion of daily exposure of urban dwellers. For instance, a commuter will get exposed to ~4.3×10^9 particles over the period of delay time, which is typically ~46 s at many TIs (Zheng et al., 2013b). Assuming that an individual crosses one TI during a day, exposure to this individual at that TI may contribute as much as 13% of total exposure during a typical daily commuting time of ~1.5 h (Fruin et al., 2008; Ragettli et al., 2013) that give a *total* RDD of ~34.4×10^9 particles.

It is evident from the above discussions that some studies are conducted for TIs of regular street canyons (Table 1), but there is clearly a need for more experimental investigations in order to understand the extent of exposure at TIs under diverse geometrical configurations as well as flow and driving conditions of traffic. 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.

8. Summary, conclusion and future directions

The article presents a critical assessment of the important aspects of traffic and wind flow features, emissions, particle dynamics and dispersion modelling of nanoparticles at TIs. Implications of PNCs at TIs towards the exposure to traffic-emitted nanoparticles are also discussed. Numerous types of models available for traffic flow modelling at TIs are reviewed and the effects of atmospheric- and mechanical-produced turbulence on wind flow features at TIs are discussed.

Although some information is currently available on the effects of wind direction and surrounding geometry on wind flow feature at the TIs, information on the effect of TPT and atmospheric stability on wind flow feature at TIs is still limited. So far, only a few field studies have measured the PNCs at the TIs and up to date summary of these field studies is collated. PNEFs at TIs are dynamic and information on them is hardly available. Relative importance of transformation processes is assessed based on the time scale analysis. The features and limitations of currently available aerosol and inert pollutant models are presented that can be considered for dispersion modelling of nanoparticles at the TIs. A need of more field and modelling studies is recognised since these are crucial for improved understanding of particle transformation, dispersion and associated exposure at the TIs. Comparative assessment of exposure to PNCs at TIs with different urban environment is also performed, along with highlighting key areas for further research. The key conclusions drawn from this review are summarised below:

- Microscopic models are found to be suitable for traffic flow modelling at TIs since they can capture the dynamic behaviour of road vehicles in short time steps.
- Wind flow features at TIs are highly sensitive to local geometry, atmospheric stability, TPT and wind direction. Intensity of WPT varies significantly at various points at and around the TIs, and therefore the TIs cannot be considered as uniformly-mixed zones.
- Majority of the current studies have monitored PNCs only at one fixed location at a particular TI. Whilst such measurements provide indicative levels of PNCs, these measurements do not provide detailed insight on the effects of complex wind flow

features and variable emission on particle dynamics and dispersion around the TIs.

- The PNEFs are highly variable and depend upon meteorology, driving conditions, engine speed, engine load, fuel sulphur content, and road grade. Data available on PNEFs under real-world driving conditions, capturing the effect of frequent start-go and acceleration-deceleration experienced at TIs, is nearly non-existent.
- The time scale analysis suggested that the nucleation is the most important transformation process among others at the TIs, followed by dilution, deposition, coagulation, condensation and evaporation for the consideration in dispersion modelling.
- A very few aerosol dynamic models are suitable for dispersion modelling of nanoparticles at the TIs, but these models are complex to use and require excessive computation resources. Models available for gaseous and particulate matter can possibly be modified by incorporating appropriate PNEFs and particle dynamic modules to predict nanoparticles at TIs.
- RDD rate based on peak PNCs at TIs is found to be ~12-times higher than those based on the average PNCs at urban roadsides. Short-term exposure to nanoparticles at TIs may contribute a significant portion of total exposure during daily commuting. A very few studies have assessed exposure to PNCs at TIs and therefore the extent of exposure at a broad variety of TIs is yet poorly understood.

There are a number of key questions that need to be addressed through further research. For example, limited information is available on wind flow features at TIs and presently available studies have focused on physical transfer processes (mass and momentum), but how this knowledge can be extended for exposure evaluation is needed to be explored. Only a handful of studies have tried to assess the effect of driving conditions on PNEFs in real world situation, but the effect of delay event on PN emission at TIs is poorly understood. Information on relevance of various transformation processes at TIs is scarcely available. Moreover, the contribution of these transformation processes in changing the PNCs between the traffic exhaust and receptor locations at TIs is still poorly studied. Currently available models for dispersion modelling at TIs are developed for simplified geometries that cannot be generalised. Adequate characterisation of complex geometry requires consideration of numerous factors such as size and shape of TIs, details of roofs, and building walls. Combination of all these complexities suggests a need to understand the science behind the nanoparticle dispersion at the TIs. There is also a need of more field studies in order to map the PNC around TIs and understand the particle dynamics and their dispersion. Such studies will be of great relevance in evaluation of PNC dispersion models and accurate assessment of exposure at the TIs.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.08.037.

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