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Dynamics and dispersion modelling of nanoparticles from road traffic in the urban atmospheric environment – a review

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Abstract

Reducing exposure to atmospheric nanoparticles in urban areas is important for protecting public health. Developing new or improving the capabilities of existing dispersion models will help to design effective mitigation strategies for nanoparticle rich environments. The aims of this review are to summarise current practices of nanoparticle dispersion modelling at five local scales (i.e. vehicle wake, street canyons, neighbourhood, city and road tunnels), together with highlighting associated challenges, research gaps and priorities. The review begins with a synthesis of available information about the flow and mixing characteristics in urban environments which is followed by a brief discussion on dispersion modelling of nanoparticles. Further sections cover the effects of transformation processes in dispersion modelling of nanoparticles, and a critical discussion on associated structural and parametric uncertainties in modelling. The article concludes with a comprehensive summary of current knowledge and future research required on the topic areas covered.

Appropriate treatment of transformation processes (i.e. nucleation, coagulation, deposition and condensation) in existing dispersion models is essential for extending the applicability of gaseous dispersion models to nanoparticles. Some modelling studies that consider the particles down to 1 nm size indicate importance of coagulation and condensation processes on street–scale modelling whereas others neglecting either sub–10 nm particles or Van der Waals forces along with fractal geometry suggest to discard these processes due to negligible effects on particle number and size distributions. Further, it is important to consider those transformation processes e.g. at city scale or in road tunnels because of the much longer residence time or much higher concentration levels compared to the street scale processes. Structural and parametric uncertainties affect the modelled results considerably. In particular, parametric uncertainty in the form of particle number emission factors appears to be the most significant due to considerably large variations in their estimates. A consistent approach to the use of emission factors, appropriate treatment of transformation processes in particle dispersion models and the evaluation of model

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performance against measured data are essential for producing reliable modelled results.

Key words: Aerosol and particle dispersion; Model uncertainty; Nanoparticle modelling; Number and size distribution; Street canyon; Ultrafine particles

1. Introduction

Nanoparticle emissions from road vehicles and their adverse impacts on human health and environment have urged the air quality science and management communities to reinforce research in this area. While emission sources such as power plants (Li et al., 2009), airports (Hu et al., 2009), building demolition sites (Hansen et al., 2008), tyre and road surface wear (Dahl et al., 2006), biofuel derived (Kumar et al., 2010b), natural formation (Holmes, 2007) and other emerging sources such as manufactured nanomaterials (Kumar et al., 2010a) are important contributors to the *number* concentration of atmospheric particles, emissions from gasoline– and diesel–fuelled vehicles remain the dominant source in polluted urban environments. These can alone contribute up to about 90% of the total particle number (ToN) concentrations (Pey et al., 2009).

Atmospheric nanoparticles need to be controlled for several reasons: the toxic nature of fresh emissions (Murr and Garza, 2009), the ability of ultrafine fraction particles (<100 nm) to penetrate the epithelial cells and accumulate in lymph nodes (Nel et al., 2006), the possible association with paediatric asthma (Andersen et al., 2008) and the potential for oxidative damage to DNA which may lead to increased risk of cancer (Møller et al., 2008) are a few examples of adverse health effects to the public due to nanoparticle exposure. Although most epidemiological studies have focused on PM_{10} or $PM_{2.5}$, there is a certain evidence indicating that short–term exposure to high concentrations of nanoparticles may aggravate existing pulmonary and cardiovascular disease or trigger stroke, while long–term exposure may increase the risk of cardiovascular disease and death (Andersen et al., 2010; Brugge et al., 2007; Pope III and Dockery, 2006). For instance, Kumar et al. (2011a) made preliminary estimates of nanoparticle emissions from road vehicles and their exposure related excess deaths in megacity Delhi. They reported that exposure to ambient ToN concentrations may result in a notable number of excess deaths (e.g., ~508 and ~1888 deaths per million people in 2010 and 2030, respectively, under the business as usual scenario). Physico–chemical characteristics of nanoparticles and their dynamic nature play an important role in changing the optical properties of coarse particles in the atmosphere through coagulation or condensation, leading to concerns such as diminishing urban visibility (Horvath, 1994) and global climate change (Strawa et al., 2010). A comprehensive review of nanoparticle characteristics, sources, measurement methodologies, health, environmental and regulatory implications can be found in Kumar et al. (2010c).

An urban area consists of street canyons where pollutant concentrations can be several times higher than in unobstructed locations depending upon traffic characteristics, street canyon geometry, entrainment of emissions from adjacent streets and turbulence induced by prevailing winds, traffic and atmospheric stability (Kumar et al., 2008b; Kumar et al., 2009a). Real–time continuous measurements of nanoparticles at many locations is rarely possible due to practical and technical constraints (Kumar et al., 2011b). Therefore, a better understanding of dispersion modelling and the associated challenges is crucial for designing long– or short–term mitigation strategies.

As seen in Fig. 1, traffic emissions in urban areas generally occur within the urban canopy layer where the atmospheric flow is heavily disturbed by buildings and obstacles (COST732, 2010). This leads to varying flow and dispersion characteristics of pollutants in different urban settings (Britter and Hanna, 2003), and in turn influencing the dilution of those emissions. When nanoparticles and their dynamics are considered for dispersion modelling, dilution remains a very crucial process and it is additionally accompanied by transformation processes such as nucleation, coagulation, condensation, evaporation and also deposition (Ketzel et al., 2007). Occurrence of these processes just after the release of emissions from vehicle tailpipes in the atmosphere continuously change the number and size distributions of nanoparticles and makes their dispersion modelling challenging and distinct from that for gaseous air pollutants. There is currently limited and partly contradicting information available on the effects of transformation processes in nanoparticle dispersion models. One of the main objectives of this article is to discuss them in some detail.

Section 3 summarises a number of reviews available on the dispersion modelling of air pollutants at different urban scales. The main focus of these reviews has been on gaseous pollutants or on various fractions of particulate matter on a mass basis, but dispersion modelling of nanoparticles is generally not covered. This review focuses on dispersion modelling of the number and size distributions of nanoparticles at five urban scales. Considering that there is plenty of information already available on flow characteristics and pollutant transport in different urban settings, this article discusses these topics only very briefly (see Sections 2 and 3) but covers the effect of particle dynamics in the dispersion modelling of *number concentrations* in detail (see Section 4). Furthermore, we discus the uncertainties in the dispersion modelling of nanoparticles (Section 5), which is followed by conclusions and discussions on future research required on the topic areas covered.

The discussions presented in this article cover the following five local scales: (i) vehicle wake scale, (ii) street scale, (iii) neighbourhood scale, (iv) city scale, and (v) road tunnels. The focus remains on the modelling of total number concentrations (ToN*)* in the atmospheric urban environment. It is worth noting that about 99% of ToN concentrations in the urban atmosphere are of sizes below 300 nm (Kumar et al., 2009a) down to around 1.5–2 nm which is the size of stable nucleated particles (Kulmala et al., 2007) . Therefore, the term 'nanoparticle' used in this work generally refers to this size range (Kumar et al., 2010c). In what follows, the words transformation and dynamics are used interchangeably as are the terms particle and aerosol (according to the context).

2. Key flow and mixing features in urban areas

Wind flow and/or the mixing of the pollutants in that flow, through or above the urban areas are not straightforward to describe. This is because of the complex networks of streets and buildings, synoptic scale winds, surface heating and various pollution sources such as moving traffic (Belcher, 2005), as seen in Fig. 1. Incorporation of detailed turbulent mixing mechanisms (vehicle–induced turbulence, road–induced turbulence and atmospheric boundary layer turbulence) improved predictions of the spatial gradients of air pollutants near roadways (Wang and Zhang, 2009; Heist et al., 2009). Britter and Hanna (2003) proposed a simple approach to describe urban scales; length scales such as street (L_s, less than ~100 to 200 m), neighbourhood (L_N, up to 1 or 2 km), city (L_C, up to 10 or 20 km) and regional (L_R, up to 100 or 200 km) scales. The smallest length scale is that of the vehicle wake (L_V) where the mixing and dilution of pollutants occurs faster than at any other scale (Baker, 2001; Carpentieri et al., 2010). Knowledge of both the flow and mixing at various urban scales is essential for dispersion modelling of nanoparticles. The following sections briefly explain these characteristics using an *inside–out* advection approach. A summary of the key flow and mixing features at these urban scales is given in Table 1.

2.1 Vehicle wake

For instance, a parcel of exhaust emission, containing pre–existing particles and various precursor gases for condensation and new particle formation exits the tail pipe. The vehicle wake is the first spatial scale where the emitted nanoparticles will disperse into the ambient environment. The extent of any transformation of particles depends on the flow characteristics and the turbulent mixing that govern the dilution, and the background concentrations (Carpentieri et al., 2010). The vehicle wake consists of two regions: (i) the near wake which is normally considered to be up to a distance of about 10–15 times the vehicle height, and (ii) the far or main wake which is a region beyond the near wake (Hucho, 1987). Number and size distributions of nanoparticles change rapidly in the near wake due to the influences of various transformation processes that are encouraged by the rapid turbulent mixing and dilution (Kumar et al., 2009c; Solazzo et al., 2007). In the diluting and cooling exhaust new particles form by homogeneous nucleation and immediately grow by condensation of condensable vapours. Also, the high number concentration of newly formed particles results in immediate coagulation of many of these particles, transforming the particle size distribution. According to the on–road measurements by Rönkkö et al. (2007), the nucleation mode was already formed after 0.7 s residence time in the atmosphere. Many modellers use these size distributions as initial emission size distributions. Thereafter in the far wake region, the rate of evolution is much slower because vehicle–produced turbulence decays with the increasing distance from the tailpipe and mixing is mainly dominated by atmospheric turbulence (Baker, 2001; Eskridge and Hunt, 1979).

2.2 Street canyons

The emitted parcel of exhaust is further spread within the street canyon. This spread is of particular interest because it occurs in traffic locations where people daily spend relevant time, and where regulatory monitoring stations observe the air quality. Direction of the flow is controlled by numerous factors: (i) geometry and aspect ratio (average building height, H, to width, W, ratio) that classify them into regular, deep, avenue, symmetric or non–symmetric canyons; Vardoulakis et al., 2003), (ii) urban roughness elements within the canyon (trees, balconies, slanted roofs, etc.; Gayev and Savory, 1999), (iii) street orientation (Hoydysh and Dabberdt, 1998; Vardoulakis et al., 2003), and (iv) the synoptic wind conditions (Britter and Hanna, 2003). Depending on the above–roof wind speed (U_r; also called synoptic wind or free–stream velocity), the flow can be: (i) neutral when U_r <1.5 m s^{-1} and atmospheric stability is neutral, (ii) perpendicular or near–perpendicular when U_r >1.5 m s^{-1} blowing at an angle of more than 30° to the street axis, and (iii) parallel or near–

parallel when U_r >1.5 m s^{-1} blowing from all other directions (Vardoulakis et al., 2003). In case of regular street canyons (i.e. $H/W \approx 1$), the typical recirculating velocities are ≈ 0.33–0.50 U_r and the turbulence levels are ≈ 0.10 U_r (Britter and Hanna, 2003).

The *mixing* of the parcel within a canyon will depend on the ventilation flux of air from the street canyon (Barlow et al., 2004; Caton et al., 2003), turbulence produced by the wind (De Paul and Sheih, 1986), traffic (Solazzo et al., 2007, 2008) and atmospheric instability (Xie et al., 2005). The shape and strength of the wind vortices might also be affected by the atmospheric stability and other thermal effects induced by the differential heating of the walls and/or the bottom of the canyon (Kim and Baik, 2001; Sini et al., 1996). Wind–and traffic–produced turbulences (hereafter referred as WPT and TPT, respectively) are generally considered to be the main mixing mechanisms at this scale. During calm wind conditions (e.g. when U_r is below about 1.5 m s^{-1}), the mixing of the parcel will be dominated by the TPT and atmospheric stability conditions while the WPT will dominate the mixing during larger wind speeds (De Paul and Sheih, 1986; Di Sabatino et al., 2003; Kastner-Klein et al., 2003; Solazzo et al., 2007). The magnitude of mechanical mixing increases with the increase in wind speed and surface roughness. Solar radiation heating the building walls in a street canyon might generate upward buoyancy forces (Kim and Baik, 2001). However, laboratory, computational and field studies show only a small effect, which is unlikely to be operationally important in most scenarios because the physical width of the free convective boundary layer on the heated wall is small compared with the scale of the mechanically driven motion (Kovar-Panskus et al., 2002; Louka et al., 2002).

2.3 Neighbourhood scale

After the street scale, the parcel of exhaust can be assumed to be advected in the neighbourhood through a network of streets, over and around several buildings (see Table 1). The flow at this scale is more complex than in street canyons. This is due to the interactions of the flow around several buildings and streets (Belcher, 2005). There are two main characteristics: (i) the flow is assumed to have a long fetch over a statistically homogeneous surface, and some quasi–equilibrium flow is established, and (ii) the flow is assumed to have developed as a result of a change from one to another region (Smits and Wood, 1985). Besides the factors playing a role in street scale mixing, the neighbourhood scale is further affected by turbulence generated from the interaction of flows coming from several sets of buildings and streets. Detailed information on this scale can be seen elsewhere (Belcher, 2005; Britter et al., 2002; Britter and Hanna, 2003; Coceal and Belcher, 2005; Grimmond and Oke, 1999; Louka et al., 2000).

2.4 City scale

Further, the advection of a polluted air parcel can be extended to *city scale* (see Table 1). This scale is composed of several neighbourhoods, and generally represents the diameter of an urban area. This area can be distinguished from its surroundings by its relatively large obstacles (buildings and other structures), the infusion of heat, moisture from anthropogenic activities, the large heat storage capacity of concrete and other building materials, and open spaces such as car parks. The city scale can include variations in urban building types and spacing, and primarily concerns the atmospheric boundary layer (ABL) above the average building height (H). To characterise the ABL, which is almost always turbulent having a logarithmic wind profile (Raupach et al., 1980; Rotach, 1993a, 1993b), it is important to comprehend the complex flows and turbulent mixing processes at this scale (Fig. 1). The ABL consists of three major sub–layers: (i) the urban canopy sub–layer where the flow at a specific point is directly affected by local obstacles, (ii) the roughness sub–layer where the flow is still adjusting to the effects of many obstacles, and (iii) the inertial sub–layer where the boundary layer has adapted to the integrated effect of the underlying urban surface (Britter and Hanna, 2003). The roughness sub–layer can extend up to $\approx 2H$. It is generally assumed that a pollutant plume can extend vertically up to $\approx 2H$ over the surface layer and that there is no need to account for the specific effects around individual buildings. Consequently, pollutant concentration fields can be determined by using standard approaches that apply to a general ABL (Di Sabatino, 2005). The mixing of pollutants within the city is greatly influenced by the complex orography (i.e. surface roughness) of the city and the TPT.

2.5 Road tunnels

The flow and mixing characteristics in road tunnels are entirely different than other urban scales, and hence affect the transformation of nanoparticles diversely (see Section 4). Unlike other urban scales, the factors governing the flow inside the tunnels include vertical and horizontal aspirators transporting ‘clean’ air from the outside and pushing it into the tunnel (called as ventilation speed), exhaust fans for discharging the ‘dirty’ air outside the tunnel and the movement of the vehicles taking the air in longitudinal direction out of the tunnel with their wakes (Bellasio, 1997; Cheng et al., 2010; El-Fadel and Hashisho, 2001). The main flow inside the tunnel is induced by the piston effect of the moving vehicles (Bari and Naser, 2010). The effect of atmospheric turbulence (WPT and TCT) and the meteorological conditions (synoptic wind speed and direction) on flow and mixing is insignificant compared with the TPT during normal operational conditions. The flow is generally turbulent and the mixing is intense within the tunnels due to the effect of the TPT in a confined environment along with the buoyancy effects generated by the intake of ‘clean’ air and discharge of ‘dirty’ air.

3. Overview of dispersion models

Several simple to complex models addressing dispersion of gaseous pollutants and particulate matter (on a mass basis) at different urban scales are currently available. These may include simple box models, Lagrangian or Eulerian models, Gaussian models, and computational fluid dynamics (CFD) based models. This section provides a brief overview of studies covering dispersion models but does not go into the details of individual models. However, a brief discussion on the challenges associated with the adaptation of gaseous dispersion models to nanoparticle predictions on a number basis is presented.

Numerous studies in the past have covered dispersion models that address wind flow and pollutant dispersions at vehicle wake, roadside locations, intersections, street canyons, neighbourhood and city scales. A recent review by Seigneur (2009) mostly focused on measurement techniques and ambient measurements of the physical and chemical characteristics of ultrafine particles, besides synthesising some model studies on the evolution of particles in vehicle exhaust plumes. Carpentieri et al. (2010) reviewed various models and techniques used for the dispersion of nanoparticles in the vehicle wake. Milionis and Davies (1994) analysed theoretical aspects, advantages and disadvantages of regression and stochastic models for air pollution studies Sharma and Khare (2001) reviewed commonly used analytical models for the dispersion of vehicle exhaust emissions near roadways, intersections and in street canyons. Later, Sharma et al. (2004) reviewed the philosophy and basic features of commonly used highway dispersion models, together with statistical analysis tools to evaluate the performance of these models. Gokhale and Khare (2004) reviewed various deterministic, stochastic and hybrid (the combination of the former two) vehicular exhaust emission models for traffic intersections and urban roadways. Vardoulakis et al. (2003) presented a comprehensive review on dispersion models for computing wind flows and transport of gaseous and particulate pollutants in street canyons. The same group also reviewed sensitivity and uncertainty involved in street scale dispersion models (Vardoulakis et al., 2002). Likewise, Li et al (2006) discussed the various CFD modelling approaches for determining wind flow and pollutant transfer within the street canyons. Holmes and Morawska (2006) reviewed several simple and complex models covering a wide range of urban scales for the dispersion of particulate matter. A recent study by Holmes et al. (2009) presented a summary discussion of the activities, findings and recommendations of the US EPA funded National Research Councils Committee on Regulatory Environmental Models for assessing practice, pitfalls and prospects of various computational models used for regulatory purposes. A comprehensive list of various dispersion models can be found on Model Documentation System of European Topic Centre on Air and Climate Change (MDS, 2010). Furthermore, a recent COST Action exercise presents various model evaluation and quality assurance case studies (COST732, 2005-2009, 2010).

There are currently very few models which are especially designed to predict particle number concentrations by taking into account the particle dynamics. A summary of these models is presented in Table 2. Several models (Holmes and Morawska, 2006) state that they include 'aerosol dynamics' but these generally predict various fractions of particulate matter on a mass basis, not on a number basis. Theoretically, any gaseous dispersion model should be able to predict number concentrations of inert particles but this is usually not the case at all urban scales (see Table 2). Chemical and physical processes associated with atmospheric particles show a non–linear dependency on their sizes that varies over a broad range. Moreover, the effects of these processes also differ at different urban scales. As discussed in Section 2, complex flow and mixing due to intricate networks of streets and buildings, synoptic scale winds, surface heating and various pollution sources such as moving traffic in urban areas makes prediction of nanoparticle number and size distributions even more challenging (Britter and Hanna, 2003). Nevertheless, gaseous dispersion models can still be modified by integrating the particle dynamics module in them, since the fundamentals for flow and pollutant dispersion predictions in a particular urban setting remains the same. In this case, the most challenging task is to identify the role of sinks and transformation processes and their appropriate treatment at various urban scales (see Section 4). If mitigation policies for nanoparticles on a number basis are adopted in the future, performance evaluation of such modified, new or existing models against measured data in different operational conditions will be required.

4. Relevance of particle dynamics in dispersion modelling

In the beginning of this chapter we give a brief overview of the various transformation processes acting on the particle number concentrations. A detailed description and mathematical formulation of the different processes can not be given here and is treated in several textbooks (e.g. Jacobson 2005; Seinfeld and Pandis, 1998; Hinds, 1999). Table 3 summarises the importance of transformation process at different urban scales and their impacts on total number (and volume) concentrations. Such information is essential since an inadequate treatment of these processes in dispersion modelling may result in uncertainties in the predictions of nanoparticles (see Section 5). Therefore, the effect of various processes at each urban scale is explained separately. Only the processes which are shown as important in Table 3 are included in discussions presented in Sections 4.2–4.5. In the final section 4.6 of this chapter, a few examples for studies comparing several dispersion models for nanoparticles are presented.

4.1 Main underlining principles behind transformation and removal processes

Emission from the traffic source contributes to a broad number and size distributions which generally have three distinct modes: nucleation, Aitken and accumulation, and coarse (Kumar et al., 2010c). Each mode has its distinct characteristics and changes both temporally and spatially due to the influence of various processes. *Dilution* is a key process that supersedes and/or induces other processes to act and alter the number and size distributions. Modelling studies agree that both dilution and emissions need to be modelled in great detail before particle dynamics are considered at all urban scales (Gidhagen et al. 2004a; Jacobson and Seinfeld, 2004; Ketzel and Berkowicz, 2004).

Homogeneous nucleation forms new particles (initial size around 1.5–2 nm) through gas–to–particle conversion (Kulmala et al., 2004; Wehner and Wiedensohler, 2003). This occurs as a regional event in preferably clean air masses (not discussed in this paper) and due to rapid dilution near the pollution sources. Nucleation and condensation of sulphuric–acid and semi–volatile organic substances are responsible for the formation of new liquid particles in vehicle exhaust during the first milliseconds of dilution (Kittelson, 1998; Shi et al., 1999). This process exhibits varying effects at different urban scales (Table 3) that needs to be modelled appropriately. Since nucleation is happening directly after the exhaust is released into the ambient air and very likely not later in the dilution process, it is possible and often necessary to regard nucleation as part of the emissions and the corresponding 'effective' vehicle emission factor for particle numbers. Often these 'effective' vehicle emission factors are observed to be dependent on ambient temperature (with higher values at lower temperatures) (Olivares et al., 2007) and on sulphur content in the fuel (higher values at higher sulphur content) (Wåhlin, 2009). Arnold et al. (2006) measured gas–phase sulphuric acid concentration in the exhaust under some driving conditions. Their results indicate that number concentration of nucleation mode particles increased as an increasing sulphuric acid concentration. These dependencies give a strong indication that nucleation plays a major role in the emission process. On the other hand, some measurements indicate that the exhaust includes nucleation mode particles that have a nonvolatile core (for example, oxidized metals or pyrolysed hydrocarbons) formed before the dilution process (e.g. Sakurai et al, 2003; Rönkkö et al., 2007). These core particles grow by condensation of semi–volatile material, mainly hydrocarbons, during dilution and cooling.

Coagulation is the process in which particles collide due to their random (Brownian) motion and coalesce to form larger particles and agglomerates which are made up of several particles. Brownian motion is enhanced by Van der Waals forces, viscous forces, and fractal geometry of aggregates. Van der Waals forces are the result of the formation of momentary dipoles in uncharged, nonpolar molecules (Seinfeld and Pandis, 2006). They enhance the coagulation rate of small particles whereas they are weakest in the continuum regime (Jacobson and Seinfeld, 2004). For small particles, fractal geometry enhances the coagulation kernel with increasing size of the colliding particle. Coagulation is especially efficient between particles of different sizes, with smaller particles having high mobility and larger particles providing a large cross–section. The coagulation process reduces the number of (mainly) the smaller particles while preserving the total mass. However, coagulation modifies the particle number size distribution, and internally mixes particles of different original composition over the population (Jacobson, 2002). Neglecting coagulation in models will over–predict the nanoparticle number concentration even in the cases if particles below 10 nm are not included in the simulations. When this process takes place between solid particles, the process is sometimes called agglomeration and the resulting particle clusters are known as agglomerates (Hinds, 1999).

Condensation is a 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. Condensation causes an increase in the volume of particles but does not change number concentrations. Condensation and nucleation are often competing processes since both involve condensable gas species that either can condense on pre–existing particles or form new nucleating particles (e.g. Jacobson et al., 2002; Pirjola et al., 2004; Kulmala et al., 2004 and references therein). Smaller concentrations of pre–existing particles favour both the production of new particles and their growth to detectable sizes (Kulmala et al., 2004). Conversely, high concentrations of pre–existing particles promote the condensation of the semi–volatile vapours and disfavour both the growth of fresh nuclei and their survival from high coagulation scavenging (Kerminen et al., 2004).

Evaporation is the reverse process compared to condensation, which reduces the volume concentration of particles. It occurs when molecules on a particle surface change to the gas phase and diffuse away from the surface driven by the lower vapour pressure in the air (Jacobson, 2005). Ultrafine particles evaporate faster than coarse particles due to the Kelvin effect (Hinds, 1999; Fushimi et al., 2008), and lose more volume because of their volatile nature (Kittelson et al., 2004). Semi–volatile organics evaporate almost immediately from liquid particles that are composed of unburned fuel, unburned lubricating oil and sulphate, and form near the tailpipe by nucleation and condensation during initial dilution and cooling (Jacobson et al., 2005). It is not just the heating which evaporates the volatile material from the ultrafine particles, a low carbon number (high volatility) of organic compounds (Sakurai et al., 2003) and dilution of volatile gases can also cause the particles in the ultrafine size range to shrink by evaporation (Zhang et al., 2004). Kuhn et al. (2005) studied the volatility of both outdoor and indoor particles. They heated the particles to 60°C and did not observe significant losses in number or volume concentrations but these particles shrank to approximately half of their original size at 130°C where they attained their non–volatile core. Evaporation seems to be important during periods of high ambient temperature and for the spatial scales dominated by high dilution (e.g. wake regions;

see Section 4) and fresh emissions (e.g. roadside). It should also be noted that partial evaporation increases the rate of coagulation by increasing the diffusion coefficient of the remaining particles (Jacobson et al., 2005).

Dry deposition removes particles through deposition to air–surface interfaces (Seinfeld and Pandis, 2006). This process is mainly driven by Brownian diffusion and inertial impaction. The former is more effective for nucleation mode particles due to their larger diffusion coefficient and the latter is only important for particles larger than 100 nm in turbulent flow conditions (Lee and Gieseke, 1994). Dry deposition is therefore an important process to consider in dispersion modelling at all scales. A review of various size resolved particle dry deposition schemes for air quality and climate models can be found in Petroff and Zhang (2010).

Wet deposition removes particles by precipitation (Laakso et al., 2003). This can occur by two processes: nucleation scavenging (rainout) and aerosol–hydrometeor coagulation (washout). Washout is due to coagulation of precipitation hydrometeors with interstitial and below–cloud aerosols whereas rainout is due to the removal of precipitation and incorporated aerosols (Jacobson, 2003). Based on the model simulations, Jacobson (2003) concluded that washout appears to play a substantial role in controlling aerosol number globally. On the other hand, rainout is an ‘episodic’ process which is more relevant to the removal of coarse particles (aerosol mass) but does not show countable effects on the removal of ultrafine particles (<100 nm). For example, Andronache (2005) reported that ultrafine particles formed in the ABL (see Fig. 1) by the nucleation process need to grow to a diameter of ~100 nm to become activated as cloud droplets. By considering a typical growth rate of about 5 nm h^{-1} (Kulmala et al., 2004), the time required to reach to about 100 nm is approximately 2–3 days. If precipitation occurs, most ultrafine particles are too small to become cloud droplets, and only a few particles are removed by this scavenging process (Andronache, 2005).

4.2 Transformation processes in the vehicle wake

The flow and dispersion in the vehicle wake has been discussed in Section 2.1. The intensity of turbulence and dilution in wake regions can be described by the dilution ratios, being the ratio of the volume of a polluted air parcel after dilution to original volume before. For instance, Zhang and Wexler (2004) reported that dilution ratio can reach up to 1000:1 in 1–3 s after the release of emissions. Kittelson (1998) found about the same dilution ratio occurring in 1–2 s behind the tailpipe during their field study. These numbers can be related to the near wake region though these studies did not distinguish wake regions explicitly. These observations also indicate that the time scales for evolution in the near–wake are substantially smaller, and hence capturing them *experimentally* is highly challenging due to a limited sampling frequency of available particle spectrometers (Kumar et al., 2010c). Therefore, computational studies could be useful if used to gain detailed insight into near–wake processes (Albriet et al., 2010; Chan et al., 2010). Conversely, the case for the far–wake region is relatively less complicated where the particle processes may last up to 10’s of seconds, depending on the local geometry of urban settings and the atmospheric conditions (Kumar et al., 2009c; Zhang and Wexler, 2004).

Irrespective of any wake region, a common finding from the studies are that the dilution is the most important parameter (Jacobson and Seinfeld, 2004; Uhrner et al., 2007; Zhang and Wexler, 2004; Pohjola et al., 2003; 2007) and should be considered appropriately in dispersion models. This is followed by the nucleation, condensation and deposition of nanoparticles. On a short time scale coagulation of particles above 10 nm is too slow to substantially affect the number concentrations. Summary of their importance and effect on ToN concentrations are presented in Table 3 while below some references from the literature are discussed.

4.2.1 Role of transformation and removal processes in near– and far–wake

Kumar et al. (2009c) measured number and size distributions in the wake of a moving vehicle. They found that dilution was so quick in the near–wake of a moving vehicle that the competing effects of the transformation processes were nearly over within 1 s after emission. Further extension of these experiments included both ground–fixed and on–board measurements of particles in the 5–560 nm size range in the wake of a moving diesel car running at 4 different speeds (Carpentieri and Kumar, 2011). Up to four sub–stages of particle evolution were observed during various experimental runs. Each sub–stage showed distinct evolution patterns of particle size distributions. In line with the previous results, dilution was found to be a dominant process throughout all the evolution stages. Dilution generally follows the "power law" (i.e. increasing with distance (x) in the vehicle wake) depending on traffic type and conditions. Approximate maximum value of dilution profile, D(x), immediately after the emissions can be estimated using the following equations: $17.6x^{1.3}$ (Zhang and Wexler, 2004) or $7.01x^{0.955}$ (Kittelson et al., 1988). Zhang and Wexler (2004) modelled the exhaust emissions coming from different type of engines and examined the effect of dilution, nucleation, condensation and coagulation processes on the different size distributions between the tailpipe and road. Uhrner at al. (2007) performed a detailed CFD calculation of particle formation by nucleation and dispersion directly in the vehicle wake and compared them to on–road measurements. Both studies concluded that new particle formation through sulphuric–acid triggered nucleation and growth of these particles is the dominant particle production mechanism which depends on the amount of condensable materials remaining in the gas phase at the exit of tailpipe. Ketzel et al. (2007) also observed that particle number emissions can double when temperature decreases from +20 to +5 °C due to particle formation in the immediate exhaust plume. Because the nucleation process depends on ambient temperature, it might be responsible for the observed temperature dependence of the emission factors. This is then followed by the condensation of organic compounds, resulting in the rapid growth of nucleation mode particles and relatively slow growth of accumulation mode particles.

Pohjola et al. (2003) simulated the time evolution of Aitken and accumulation mode particles emitted by a light duty diesel vehicle in the wake region for 25 s. They found that condensation of an insoluble organic vapour was important if the concentration of the condensable vapour exceeds a value of 10^{10} or 10^{11} molecules cm^{-3} for Aitken and accumulation mode particles, respectively, and that the effect of coagulation was substantial only, if dilution was neglected. Likewise, a recent study by Chan et al. (2010) computationally simulated these processes in the vehicle wake. They concluded that nucleation and coagulation processes were nearly complete within the near–wake regions (i.e. within 0.25–0.5 m, and 4 m, respectively) behind the vehicular exhaust tailpipe, and dilution then spreads the particles which are carried by advection from the near wake to the far wake. In the far–wake region, particles can still grow by condensation but the growth rates decrease with distance away from the tailpipe due to decreasing dilution factor and concentrations of condensable species. Time scales for coagulation are considerably larger compared with dilution (Fig. 3) meaning that under many conditions coagulation is slow enough, compared with dilution, to reduce the ToN concentrations. Based on time scale analysis of Brownian coagulation for a particle size distribution measured on highway, Zhang and Wexler (2004) stated that coagulation plays minor role in the far wake region; however, coagulation will have some effect on particles below 10 nm or under slow dilution conditions. Likewise, Kerminen et al (2007) showed that the slow dilution due to inefficient mixing gives time for many other aerosol processes, such as self– and inter–modal coagulation as well as condensation and evaporation, to become important. Jacobson and Seinfeld (2004) found that dilution is more important than coagulation at reducing the ToN concentrations near the source of emission, but the relative importance of dilution versus coagulation varies with concentration. In any case, coagulation cannot be ignored in the models. Jacobson et al. (2005) found that treating the additional process of evaporation of low–molecular–weight organic vapours from small (<15 nm) liquid particles enhanced their coagulation rates allowing for the full particle evolution to be accounted for.

Potentially relevant removal mechanisms of nanoparticles in the near– and far–wake region are the dry and wet deposition. As discussed in Section 4.1, nucleation scavenging (rainout) can be ignored whereas washout is important removal mechanism at least for city scale modelling. Dry deposition is an important process for both wake regions. Dry deposition can substantially remove particles at the air–road interface (Gidhagen et al., 2004a; Gidhagen et al., 2004b). Fig. 2 shows the size dependent deposition speed using the resistance model (aerodynamic resistance, molecular diffusion, and chemical, biological and physical interactions) suggested by Seinfeld and Pandis (2006) and an alternative description based on several field studies by Shack Jr et al. (1985).

4.3 Street canyons

There is a consensus in the literature that removal processes such as dilution and dry deposition should be considered at street scale modelling (Gidhagen et al., 2004a; Ketzel et al., 2007). Evidence of dilution dominating over other processes is evident from the studies showing high correlation between particle number concentrations and NO*x* which is generally inert at street scale (Jacobson and Seinfeld, 2004; Ketzel et al., 2007). Similarly, deposition onto the road surface and/or walls in the street canyons is strongly influenced by the traffic movement. This can reduce the total number concentrations by about 10–20% in a street canyon (Gidhagen et al., 2004a; Ketzel et al., 2007).

There are two contentions about the role of nucleation, coagulation and condensation at street scale. One that favours their inclusion in dispersion models and the others that does not. Literature (e.g. Gidhagen et al., 2004a; Ketzel and Berkowicz, 2004) shows evidence in support of both these contentions but the underlining principles, as discussed in Section 4.1, remain the same that relevance of these processes may vary at different locations depending on the concentration of the exhaust emissions, the background concentrations (pre–existing particles) and meteorological conditions (Charron and Harrison, 2003; Wehner et al., 2002). Moreover, competing influences of the various processes might cancel each other and in total not affect the net particle number concentrations notably (Kumar et al., 2009a; Kumar et al., 2008c). For example, Vignati et al. (1999) applied a Brownian coagulation–dilution model to a plume emitted from a diesel engine into the street air. The emitted particle size distribution possessed one or two modes covering particles in the size range of 0.002–10 μm. They found that due to rapid dilution only very small particles (<0.002 μm) have a coagulation time scale which is comparable with typical residence times of pollutants in a street. They also found that condensation does not lead to any substantial transformation of the particle size distributions as water vapour condensation on freshly emitted diesel particles led to only a marginal increase in particle size.

One method of determining the relative importance of various processes is a *time scale analysis*, as seen in Fig. 3 for the processes dilution, deposition and coagulations at different urban scales (Ketzel and Berkowicz, 2004). The process with the smallest time scale at a specific spatial scale (exhaust plume, kerbside etc.) is the fastest and most relevant to include in numerical modelling. Dilution is by far the most relevant process at all scales, except for the road tunnel, where dilution is slowed down by the confined environment and acts at similar time scale as deposition and coagulation. At street scale deposition occurs about 10 times slower (i.e. larger time scale) than dilution. Separately, time scales were estimated for a street canyon in Cambridge, UK (Kumar et al., 2008c). The deduced time scales were of the order of 40s for dilution, 30 to 130s for dry deposition on the road surface, and 600 to 2600s for the dry deposition on the street walls, about 10^5s for coagulation, and about 10^4–10^5 for condensation for extreme growth rates at 1 and 20 nm h^{-1}, respectively (Kumar et al., 2008c). Comparison of these estimated time scales shows that dilution is quick and it does not allow other processes (except for dry deposition on the road surface) to alter the size distributions.

In the following we present further observations supporting the view that particle dynamics could be ignored at street scale, especially when either sub–10 nm particles or Van der Waals forces and the effects of fractal geometry on such forces are not considered. As a part of a recent study (Kumar et al., 2009c), the time scales of particle evolution in the wake of a moving vehicle (~ 1s) were compared with the time scales for these particles to reach the kerb side in a street canyons (~45 s). These observations led to the hypothesis that 'the competing influences of transformation processes were nearly over by the time these particles are measured at the road side and ToN can then be assumed to be conserved'. Consistent with this were the observations found during the pseudo–simultaneous measurements of particle number and size distributions where measurements were made at 4 different heights (1 m, 2.25 m, 4.62 m and 7.37 m) of an 11.75 m high regular street canyon (Kumar et al., 2008c). Results indicated that number and size distributions were similar in shape, peaking at about 13.3 nm and 86.6 nm and shifting upwards and downwards at different heights due to the combined effect of dilution and vertical concentration gradient in the canyon (see Fig. 4). Moreover, there was negligible shift in geometric mean diameters of number and size distributions in both modes at each height (i.e. 16.4 ± 0.9 nm for particles in the 10–30 nm range and 64.7 ± 5.1 nm for 30–300 nm range). Similar results were reported by Ketzel and Berkowicz (2004), Gidhagen et al. (2004a) and Pohjola et al. (2003, 2007) on the treatment of street level particle dynamics in dispersion models.

4.4 Neighbourhood and city scales

There are contrasting conclusions on the role of coagulation under typical urban ambient conditions. Few studies show coagulation is too slow to alter the particle size distributions (Gidhagen et al., 2005; Zhang and Wexler, 2002) but others show significant particle growth to the effect of coagulation in urban environments (Wehner et al., 2002) or during episodic conditions (Gidhagen et al., 2003). City scale modelling studies agree with the latter, showing a countable effect of coagulation on number concentrations (Ketzel and Berkowicz, 2005).

Condensation and evaporation do not change the total number concentrations but will alter the size distributions and particle volume. A number of nucleation mechanisms such as the binary water–sulphuric acid nucleation, ternary water–sulphuric acid–ammonia nucleation, ion–induced nucleation or photochemically induced nucleation play a role in the formation of new particles (Kulmala et al., 2004). For urban locations, growth rates are generally between 1 and 10 nm^{-1} and the new particle formation rate is about 100 # cm^{-3} s^{-1} (Kulmala et al., 2004), indicating a notable effect on ToN concentrations. Therefore, most of these processes are taken into account in various urban scale models (e.g. MAT, MATCH) described in Table 2. Differences between measurements and model calculations using MAT (with and without particle processes) were estimated by Ketzel and Berkowicz (2005). They reported that coagulation can remove 10% of the total number concentrations while dry deposition can remove between 50–70% depending on the assumed value of removal velocities. Model calculations using MATCH by Gidhagen et al. (2005) reported losses, as compared with inert treatment, due to dry deposition up to 25% under average meteorological conditions while these can be up to 50% during episodes with low wind speeds and stable conditions. The overall ranges of change including all processes compared with inert treatment can lie between 13 and 23% of loss in ToN concentrations (Ketzel and Berkowicz, 2005).

4.5 Road tunnels

In a confined environment like a road tunnel, particles collide and merge with each other due to an accumulation of particles with the increasing length of the tunnel (Cheng et al., 2010). Thus, studies have identified coagulation and deposition as the most important depletion processes (Gidhagen et al., 2003; Sturm et al., 2003). Both processes show larger influence on smaller particles of less than about 30 nm. Model calculations (MONO32 with the CFD code StarCD) by Gidhagen et al. (2003) showed that combined losses of coagulation and dry deposition onto the tunnel walls, were about 77% and 41% of the ToN concentrations for particles smaller than 10 nm and between 10 and 29 nm, respectively. Due to the presence of precursor gases in high concentrations and intense mixing due to traffic–produced turbulence, road tunnels provide an ideal environment for the formation of nucleation mode particles. However, coagulation is generally found to be fast enough to efficiently remove the smallest particles (Cheng et al., 2010; Gidhagen et al., 2003; Ketzel and Berkowicz, 2004).

4.6 Modelling case studies showing effects of transformation processes on ToN concentrations and size distributions

Two case studies using five different models are included in this section. The first study shows the effect of various transformation processes on number and size distributions of particles at city scale. The other study shows modelled ToN concentrations (assuming the particles as inert tracer) and their comparison with the measurements at street scale.

4.6.1 Modelling of particle size distributions using MAT and AEROFOR model

This section presents a modelling case study that was based on data from the Copenhagen area assuming prevailing westerly wind, using the models AEROFOR and MAT (see Table 2 for model details). The processes included in the modelling are emissions from a near ground source, dilution with background air, deposition, Brownian coagulation and condensation (Ketzel and Berkowicz, 2005; Ketzel et al., 2007). In this work, the size range of particles treated in the models is from 1 nm to 2 μm.

The simulation starts at time = 0 s assuming a background particle size distribution measured at the station Lille Valby located west of Copenhagen, i.e. upwind for the assumed case. The air mass is then transported over an urban traffic–related area source with a constant wind speed. The evolution of the size distribution will be followed by the two models for a certain simulation time, assuming a homogeneous emission density of the area source having a typical size distribution for traffic emitted particles. Model runs are performed with sensitivity analysis for specific processes included or excluded in the simulations. The outputs of the both models are shown in Fig. 5. There are two points to notice: (i) the comparison of results between two models, and (ii) the effect of processes on number and size distributions. Note that the case 'EmDi' represents the reference case considering the particles as inert (i.e. a case without removal or aerodynamic processes). The cases added to EmDi is condensation (+Con), coagulation (+Coa), deposition (+De), deposition and coagulation (+DeCoa), deposition, coagulation and condensation (+DeCoaCon). For the condensation process two different versions are considered that differ in the concentration of the condensing vapour that corresponds to typical urban growth rates of 1 nm/h (...Con1) and 6 nm/h (…Con2).

Considering the complexity of the modelled cases, results shown in Fig. 5 between both models indicate a good agreement. Both models treat aerosol dynamics in a similar way, except the dry deposition which has different parameterisation in AEROFOR. Consequently, effect of dry deposition on the size distributions by the AEROFOR is so small that we cannot distinguish the case EmDi (not shown in the graphs) from the case EmDi+dep. This is also the reason why the curves for EmDi+coa and EmDi+DeCoa overlap in Figs. 5b, d. Coagulation is less efficient in reducing ToN concentrations at the start of the simulations, when concentrations are lower as compared to the end of the simulations (Figs. 5a, b). Larger deviations between the two models were observed for the simulations including treatment of condensation (see Figs. 5c, d). This is expected since the chemical composition and properties of the aerosol are different in the two models.

4.6.2 Modelling of ToN concentrations using CFD, OSPM and a modified Box model

ToN concentrations in the 10–300 nm size range were modelled using a simple modelling approach (modified Box model, including vertical variation), the operational street pollution model (OSPM) and the CFD code FLUENT (Kumar et al., 2009b). All models neglected the particle dynamics. CFD simulations were carried out using a standard κ–ε turbulence closure scheme (κ is turbulent kinetic energy and ε is dissipation rate of kinetic energy; Vardoulakis et al., 2011) in a simplified geometry of our previously studied street canyon which has height (H) to width (W) ratio of about unity (i.e. H = W = 11.6 m). ToN concentrations were measured pseudo–simultaneously on the leeward side of the canyon at four different heights i.e. z = 1.00 m, 2.25 m, 4.67 m and 7.37 m (Kumar et al., 2008c). These measurements were made continuously using a differential mobility spectrometer (DMS500) in the 5–2500 nm size range at a sampling frequency of 0.5Hz (Kumar et al., 2008a; Kumar et al., 2008c). Measured ToN concentrations at different heights were compared with the modelled concentrations from all three models (see Fig. 6 of Kumar et al., 2009b). The values of correlation coefficients (R) for OSPM, CFD and modified Box model at z = 1.00 m were 0.84, 0.80 and 0.80, at z = 2.25 m were 0.85, 0.90 and 0.90, at z = 4.67 m were 0.75, 0.69 and 0.70, and at z = 7.37 m were 0.74, 0.69 and 0.71, respectively. Each model showed good agreement with measurements with values of R that ranged between 0.7 and 0.9. The values of R were relatively larger for the OSPM at all heights (except for z = 2.25 m) than for the Box and CFD models. The OSPM consistently under–predicted the ToN concentrations while the other two models over–predicted the concentrations in most cases. Furthermore, ToN concentrations predicted by the Box and CFD models were generally in closer agreement compared with the OSPM results. This could be because the OSPM explicitly takes into account the turbulence created by the traffic (Vardoulakis et al., 2007), which was not the case for other two models. Furthermore, differences between the modelled results and measurements can be attributed to a large uncertainty in the particle number emission factors (PNEF) as discussed in Section 5.2. Overall, the modelled and measured concentrations were found to agree within a factor of two to three, which is a fairly good agreement for ToN concentrations. These observations support to a certain extent the hypothesis presented in Section 4.3 that the role of particle dynamics may be ignored for street scale modelling when particles down to 1 nm size are not considered, although it can be argued that inclusion of particle dynamics might have marginally improved the modelled results. Gidhagen et al. (2004a) simulated the particle dynamics in a street canyon in Stockholm using the combined results of MONO32 and CFD model StarCD (see Table 2 for details) and found about 10–30% larger losses in ToN concentrations as compared with inert treatment during low wind conditions. However, this effect would be much smaller during average wind conditions due to the efficient ventilation of the canyon (Section 2) and less ToN concentrations. Moreover, this potential improvement in modelled ToN concentrations would come at the cost of complex modelling and heavier input information requirements.

5. Uncertainties in dispersion modelling of nanoparticles

Particle dispersion models suffer from the similar uncertainties associated with the gaseous dispersion models in addition to the uncertainties related to the particle dynamics. All models represent simplifications of various processes and such simplifications can produce mainly two types of uncertainties, (i) structural, and (ii) parametric, in addition to inherent uncertainties related to stochastic processes (e.g. turbulence) in the atmosphere (Holmes et al., 2009; Vardoulakis et al., 2002). Stochastic processes play an important role in the dispersion of nanoparticles, e.g. atmospheric turbulence (see Section 2) leads to their dilution that in turn can change the number and size distributions of nanoparticles, as described in Section 4. However, stochastic fluctuations cannot be accurately modelled, although they are approximated in semi–empirical and CFD models. The following two subsections focus on the structural and parametric uncertainties associated with the dispersion modelling of nanoparticles.

5.1 Structural uncertainty

Structural uncertainties are due to the fundamental representation of the physical and chemical processes considered in the model structure. COST Action732 (2005-2009) suggest a method for inter–comparing the structural uncertainty of gaseous dispersion models by comparing the modelled results with the observations. These also apply for the particle dispersion models and include frequently used testing parameters: (i) FB (fractional bias), (ii) FAC2 (fractions of predictions within a factor of 2), (iii) NMSE (normalised mean square error), (iv) MG (Geometric mean), and (v) VG (geometric variance). Inappropriate treatment of the sinks and transformation processes at various urban scales in nanoparticle dispersion models is one predominant reason for additional structural uncertainties than those in gaseous dispersion models (see Section 4). However, the degree of this type of uncertainty will vary at different urban scales depending on the relevance of particle transformation processes. One way of reducing such type of uncertainty is by introducing more physically realistic and computationally efficient algorithms for various relations and nanoparticles (Vardoulakis et al., 2002). Uncertainty exists in some numerical techniques (e.g. conservative versus non–conservative schemes) and particle size distribution representations by moment methods (modal and monodisperse) and by sectional methods.

5.2 Parametric uncertainty

Parametric uncertainty is due to the use of uncertain input values (e.g. wind speed and direction, traffic volume, number emission factors) for model calculations. This may be due to the lack of representative datasets (e.g. local meteorological data), uncertainties in data (e.g. instrument calibration, unsteadiness in the measurement conditions) or limited knowledge of key parameters (e.g. emission factors). For example, Lohmeyer et al. (2002) reported that predictions of gaseous pollutants from different models can vary up to a factor of four for identical situations, depending on the quality of input information. However, such comparative studies are not currently available for nanoparticle dispersion models, although they are essential for improving dispersion models that can be used for developing mitigation policies.

It is recognised that the main source of parametric uncertainty in dispersion models lies with the PNEFs (e.g. Holmes and Morawska, 2006). The PNEFs are directly proportional to the predicted particle number concentrations, therefore any inaccuracy in their estimation would directly lead to a similar degree of inaccuracy in the model predictions. Number and size distributions of particles change rapidly after the emissions exit the tailpipe due to rapid dilution similar to gaseous pollutants. However unlike gaseous pollutants, the particle number concentration flux can not be conserved due to secondary particle formation e.g. by dilution induced nucleation (Wehner et al., 2009). Therefore, there are no standard databases available for routine use. Moreover, studies on emissions from specific types of vehicles under controlled conditions (e.g. constant speed or load) provide very limited information on the PNEFs for different types of vehicles or a composite fleet of vehicles under real-world urban driving conditions.

PNEF studies use several methods for their estimations. Most commonly used methods involve (i) road side measurements of air pollutants and accounting for dispersion by use of models, so-called 'inverse modelling technique' or by use other pollutants with known emissions as NO_x or CO_2 so-called 'tracer method' (Corsmeier et al., 2005; Gidhagen et al., 2004a; Gidhagen et al., 2004b; Gidhagen et al., 2003; Gramotnev et al., 2003a; Gramotnev et al., 2004; Hueglin et al., 2006; Imhof et al., 2005a; Imhof et al., 2005c; Jamriska and Morawska, 2001; Jones and Harrison, 2006; Keogh et al., 2009; Ketzel et al., 2003; Kittelson et al., 2004; Kumar et al., 2008b; Kumar et al., 2008c; Morawska et al., 2005; Rijkeboer et al., 2005; Zhang et al., 2005; Zhu and Hinds, 2005), (ii) motorway tunnel measurements (Abu-Allaban et al., 2002; Imhof et al., 2005b; Kirchstetter et al., 2002; Kristensson et al., 2004), (iii) measurements directly through chassis dynamometer tests at the exit of tailpipe (Dahl et al., 2006; Jayaratne et al., 2009; Morawska et al., 1998; Prati and Costagliola, 2008; Ristovski et al., 2005; Ristovski et al., 2002; Ristovski et al., 2004), or (iv) measurements in the exhaust plume of individual cars under real-world conditions by remote sensing or so-called car-chasing (Hak et al., 2009; Vogt et al., 2003; Wehner et al., 2009). While the first methods (i+ii) provide data for a mixture of many vehicles (vehicle fleet) the latter methods (iii+iv) give results for a limited number of individual cars. A comprehensive summary of numerous PNEF studies, categorised by vehicle types, is presented in Table 4.

The chassis dynamometer method measures PNEFs close to the source. The other methods, such as inverse modelling techniques, estimate PNEFs using ambient concentrations close to the receptor but away from the source. The *source* and *receptor* based PNEFs are different from each other because of the unequal treatment of the transformation processes in dispersion models. Whether it is appropriate to choose *source* specific PNEFs for modelling purposes as opposed to the receptor specific PNEFs is highly debatable. Source based estimates of the PNEFs can be more appropriate because these accurately represent the emission strength of a vehicle. However, such measurements leave the transformation processes occurring between the tailpipe and the receptor location to be accounted by the dispersion models which have their own limitations in treating the particle dynamics. On the other hand, receptor based estimates of the PNEFs are back–calculated from the roadside ambient measurements which have already undergone natural transformation processes by the time they reach the measurement site. These estimates are based on realistic nanoparticle concentrations but may not represent the actual emission strength of a vehicle or a traffic fleet since effects of transformation processes in inverse modelling methods are generally ignored. Therefore adequate treatment of particle dynamics is essential in nanoparticle dispersion models that are used for back–calculating PNEFs (Wehner et al., 2009).

Other factors influencing the estimates of PNEFs include vehicle type, speed, load and driving conditions, lower and upper cut–off values of the particle size range considered, and sulphur content in the fuel. Typical driving conditions in urban environments represent varying vehicle speed, with stop–start or acceleration–deceleration conditions, leading to a considerable variability in particle emissions from vehicles and uncertainty in PNEF estimates (Kittelson et al., 2004). Vehicle speeds are generally expected to be less than 60 km h^{-1} in typical driving conditions in the urban areas. A closer inspection of the summary of several studies presented in

Table 4 indicates up to an order of magnitude difference in the PNEFs for a mixed traffic fleet under near–identical conditions.

If we look at the different vehicle types independently (Table 4), it can be seen that emissions of petrol vehicles are much more engine load and vehicle speed dependent compared with diesel vehicles (Kittelson et al., 2004). However, the PNEFs from light duty diesel vehicles (especially cars and buses) are expected to be relatively consistent (i.e. within a factor of 3) compared with heavy duty vehicles that can have up to an order of magnitude larger than light duty petrol or diesel vehicles (Imhof et al., 2005c; Prati and Costagliola, 2008). These observations reflect the large uncertainty in PNEFs meaning that modelled results are likely to be affected to a similar degree irrespective of the accuracy of the dispersion model.

Sulphur content of the fuel also plays a major role in the formation of nanoparticles and consequently influences the PNEFs. For example, the sulphur content in Danish fuels was reduced twice from 500 ppm to 50 ppm in 2000 and to 10 ppm in 2005; in both cases changes in the emitted size distribution of particles were observed. Studies by Wåhlin et al. (2001) and Wåhlin (2009) indicate about 27% reduction in average particle number concentration from the period 2002–2004 to the period 2005–2007 during their kerbside study at a busy street in Copenhagen. Most of these reductions were in the ultrafine size range and more particularly in particles below 30 nm. Similar observations were found by Ristovski et al. (2006) in their chassis dynamometer study on low and ultra low sulphur buses in Brisbane, Australia.

6. Conclusions and future research challenges

This article discusses various aspects related to dispersion modelling of nanoparticles at five spatial scales: vehicle wake, street canyon, neighbourhood, city and road tunnel. Key flow and mixing characteristics at these scales are discussed. Currently available particle dispersion models, their capabilities and limitations are briefly examined. The relevance of transformation processes such as dilution, emission, nucleation, coagulation, condensation, evaporation, dry and wet depositions at the selected five scales are critically assessed and suggestions for their adequate treatment in dispersion models are outlined. Furthermore, the impact of structural and parametric uncertainties on modelled particle number concentrations is critically discussed.

Each spatial scale has distinct flow and mixing characteristics which are complex and thus difficult to generalise (Section 2). Several aerosol dispersion models are currently available for covering the discussed spatial scales (Section 3) but most of them are mainly used for research purposes and are not available commercially for regulatory use. Moreover, the models treating the particle dynamics in detail are complex, resource intensive and require a great amount of additional input information (e.g. type and concentration of condensable species or size dependent chemical composition of nanoparticles), which is not readily available for routine use. It is therefore necessary to identify the relevant key transformation processes at different spatial scales for reducing complexity in model structure and the amount of input information required. The discussion presented in Section 4 indicated that, irrespective of any spatial scale, emission and dilution are crucial processes that need to be modelled in detail before considering the aerosol dynamics. Dilution is also the fastest process at any spatial scale, except for road tunnel environment where dilution is impeded by limited air flow (Ketzel and Berkowicz, 2004). However, sink processes such as coagulation and dry deposition play an important role in road tunnel modelling (Sturm et al., 2003). Generally, other transformation processes become progressively slower with time after emissions are released. For instance, at urban rooftop and city scale the dilution is sufficiently slow to allow coagulation, deposition and condensation to alter the size distribution; these processes need to be considered in urban scale dispersion models (Table 3). Under some conditions (e.g. rapid dilution, initial particles larger than 10 nm), particle dynamics may be disregarded for street scale modelling because the competing influences of these processes have negligible net effects on ToN concentrations (Section 4.6).

Aerosol dispersion models are affected by similar uncertainties (structural and parametric) to those of gaseous dispersion models (Vardoulakis et al., 2002) in addition to the uncertainties caused by inappropriate treatment of particle transformation processes. One of the major parametric uncertainties in ToN emission modelling originates in the estimation of PNEFs (Section 5.2). More coherence in their estimation methods is required for obtaining consistent emission factors and reducing the differences between modelled and measured ToN concentrations.

Research questions that need further attention related to the dispersion modelling of particle number concentrations include: (i) what prediction accuracy should be acceptable for modelling purposes at various spatial scale (e.g. ±10% of measured data, within a factor of 2 or more), and (ii) which of the uncertainties play a dominant role in the model prediction (e.g. the representation of certain transformation processes, or the inputs provided). Since limited information is available on scientific evaluation, verifications and validations of particle dispersion models, any of the above questions can not be answered precisely. The review qualitatively assesses the relative importance of various transformation processes. Further work can include harmonisation of various model outputs through their inter–comparison under a range of data sets for quantitatively evaluating and establishing the relative importance of particle transformation processes at different urban scales. However, the major practical constraint remains the easy accessibility of currently available particle dispersion models which are mainly used for research purposes by individual groups.

The greater number of long–term nanoparticle measurements (including number and size distributions) and the easy accessibility of that data to the scientific community would help to evaluate the performance of particle dispersion models, and to reduce structural and parametric uncertainties. Moreover, there is a need for establishing a standard measurement methodology for the PNEFs that can be readily used for the dispersion modelling of nanoparticles. Developing the capabilities of existing particle dispersion models through comprehensive performance evaluation, and reducing uncertainties by appropriately treating particle dynamics at different urban scales and providing accurate input information are essential steps for accurately predicting ToN concentrations and developing mitigation policies for urban areas.

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Figure Captions

Fig. 1. Schematic diagram of the flow through and over an urban area (Grimmond and Oke, 1999). Also are shown various layers in the ABL and horizontally spatially averaged mean velocity profile (Bottema, 1997).

Fig. 2. Size dependent deposition speed of particles onto the surfaces calculated using two different equations within a range of typical urban parameters; figure adapted from Ketzel and Berkowicz (2004).

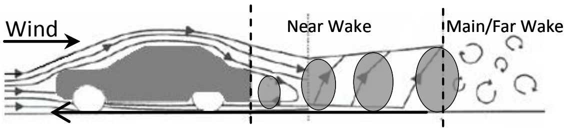
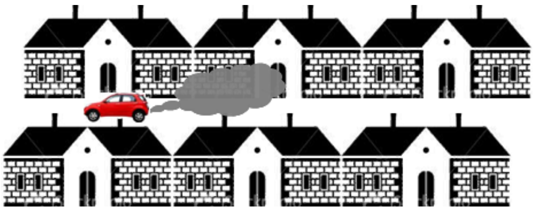
Fig. 3. Time scales for dilution, coagulation and deposition at different spatial scales. For each concentration level the process with the smallest time scale is the most relevant one. For the dilution of the exhaust plume the time scale is very different for a road tunnel and the ambient atmosphere (e.g. street); figure adapted from Ketzel and Berkowicz (2004).

Fig. 4. Measured and corrected (for losses in sampling tubes) particle number distributions at (a) z/H = 0.09, (b) z/H = 0.19, (c) z/H = 0.40 and (d) z/H = 0.64 of an 11.75 m high (H) street canyon; figure adapted from Kumar et al. (2008c). Dotted lines represent mode fitting curves to corrected particle number distributions. Error bars show the standard deviation of hourly averaged particle number distributions at each height; only positive error bars are plotted for the clarity of the figures.

Fig. 5. Inter–comparison of modelled results from the MAT (a, c) and AEROFOR (b, d) for the test case in Copenhagen; figure adapted from Ketzel et al. (2007). Figs. a and b show time dependence of the ToN concentration at ground level; results are from 5 simulations including different aerosol dynamics processes. Figs. c and d show size distribution calculated with different particle dynamics processes considered. Note that two different growth rates (GR) considered for the simulation with condensation; these are 1 and 6 nm h^{-1} for cases ending with letters Con1 and Con2, respectively. For comparison the measured size distributions at Lilly Valby (near–city) and urban rooftop of the H.C. Orsted Institute (HCOE). Simulation time was 12000 s.

List of Tables

Table 1. Description of flow and mixing characteristics at various urban scales (Belcher, 2005; Britter and Hanna, 2003; Hucho, 1987; Hunt et al., 1992). The abbreviations indicate as follows NW (near wake), MW (main/far wake), WPT (wind produced turbulence), TPT (traffic produced turbulence), and TCT (thermally produced turbulence).

Urban scales	Key flow and mixing features
 Vehicle wake (L_v ~ 0–20 m)	• Flow is more turbulent in NW region compared with MW region. • TPT is intense in NW and dominate the mixing • Atmospheric turbulence (WPT and TCT) lead the further mixing in MW region
 Street scale (L_s ~ 100–200 m)	• Flow features and mixing determined by surface roughness below building height • Internal mixing layers below or above the buildings are of order of building or street dimensions • TPT dominate the mixing over the WPT during calm wind conditions • TCT may play role in mixing during hot sunny weather and light winds
 Neighbourhood scale (L_N ~1–2 km)	• Flow and mixing above and between buildings is determined by the turbulence generated by interaction of flows from adjacent canyons, surface and wall roughness, WPT, TPT and TCT. • A growing internal layer of flow can be seen above buildings • Average geometrical features dominate mean flow and mixing
 City scale (L_C ~10~20 km)	• Flow and mixing over buildings is relatively less complex than within the city due to orographic (i.e. surface roughness elements) effects • Mixing below the building heights in the ABL is dominated by surface roughness and the TPT; the atmospheric turbulence (i.e. WPT and TCT) plays a key role in flows over urban canopy

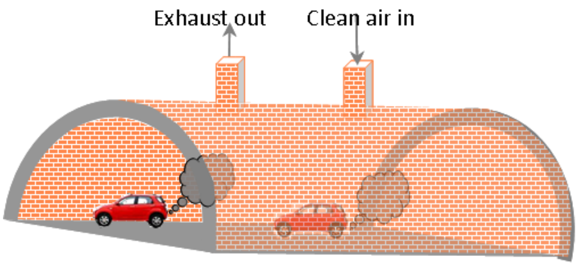
 Road tunnels (L_T ~50–5000 m)	• Flow is generally very turbulent and the mixing is dominated by the TPT and the so–called piston effect generating a mean flow in driving direction of the traffic • Effect of atmospheric turbulence on the mixing of pollutants is negligible

Table 2. Examples of few urban scale dispersion models that address particle number concentrations.

Model	Remarks	Source / Example
OSPM (Operational Street Pollution Model)	• Predicts particle *number* concentrations at street scale, but note the OSPM completely neglects particle dynamics	(Berkowicz, 2000)
UHMA (University of Helsinki Multi Component Aerosol Model)	• Provides size segregated predictions for number and size distributions between 0.7 nm and 2 μm, with a focus on new particle formation and growth in the atmosphere.	(Korhonen et al., 2004)
MAT (Multi-plume Aerosol dynamics and Transport)	• Predicts particle number size distribution in urban environments. • It uses a novel multi-plume scheme for vertical dispersion and routines of the sectional aerodynamics models AERO3 (Vignati et al., 1999).	(Ketzel and Berkowicz, 2005)
MONO32 (Multimono)	• Simplified version of the Lagrangian type atmospheric chemistry and aerosol dynamics box model MULTIMONO which refers to multi component condensation of different vapours; each size section is assumed to be monodisperse. • Can predict the particles between 1 nm and 2.5 μm using optional number of size fractions. Implemented into the EMEP model.	(Pirjola and Kulmala, 2000; Pirjola et al., 2003; Pohjola et al., 2003)
AEROFOR/AEROFOR2 (Model for Aerosol Formation and dynamics)	• Lagrangian type sectional box model which includes gas–phase chemical reactions together with aerosol dynamics; predicts number and size distributions of particles, AEROFOR2 also composition size distribution.	(Pirjola, 1999; Pirjola and Kulmala, 2001)
MATCH (Multi–scale Atmospheric Transport and Chemistry)	• Eulerian grid-point model which describes the physical and chemical processes that govern emissions, atmospheric transport and dispersion, chemical transformation, wet and dry deposition of pollutants; particle dynamics modules for predicting number and size distributions was included.	(Gidhagen et al., 2005; Robertson et al., 1999)
GATOR–GCMM (Gas, Aerosol, Transport, Radiaition, General circulation and mesoscale meteorological)	• GATOR-GCMM is derived from GATORG (global) and GATORM (regional) models; this is a single, unified model and can be switched to run global, regional or nested mode with/without gases, aerosols, radiation, meteorology, transport, deposition, cloud physics, surface processes, etc.	(Jacobson, 1997; Jacobson, 2001) and references therein

	• The model is capable of treating nearly all the size and composition resolved aerosol processes (emissions, nucleation, coagulation, condensation, dry deposition and sedimentation).	
ADCHEM (Aerosol Dynamics, gas and particle phase CHEMistry)	• Unlike Lagrangian box–models (0–space dimensions), the ADCEHM treats both vertical and horizontal dispersion perpendicular to an air mass trajectory (2–space dimension). • The model is suitable for local to regional scales to predict number and size distributions in the 1.5 to 2500 nm; this treats Brownian coagulation, dry and wet depositions, in – cloud processing, condensation, evaporation, primary particle emissions and homogeneous nucleation.	(Roldin et al., 2010a; Roldin et al., 2010b; Wang et al., 2010b)
CFD based models (e.g. StarCD, MISKAM and FLUENT codes) using RANS or LES techniques	• Either simulates the number and size distributions separately or the codes for simulation of particle dynamics are coupled with the CFD models.	(Carpentieri et al., 2010; Chan et al., 2010; Gidhagen et al., 2004a; Gidhagen et al., 2003; Kumar et al., 2009b)

Table 3. Summary of the importance of various transformation processes at various urban scales for consideration in dispersion models (Ketzel and Berkowicz, 2004; Kumar et al., 2010c). Symbols +, – and 0 denotes gain, loss and no effect of the transformation processes on ToN concentrations, respectively. Acronyms I, V and n stand for important, very important and not important (can be ignored), respectively.

Transformation processes	Effects on concentrations		Vehicle wake		Street canyons	Neighb–ourhood	City	Tunnel
	number	volume	near	far				
Emissions	+	+	V	V	V	V	V	V
Nucleation	+	+	V	I	I[*]	I[*]	I	I
Dilution	+/–	+/–	V	V	V	V	V	V
Coagulation	–	0	n[***]	n[***]	n[**]	n[**]	I	V
Condensation	0	+	V	I	n[**]	n[**]	I	I
Evaporation	0/–	–	I	V	I	I	n	I
Dry deposition	–	–	V	V	I	I	I	V
Wet deposition	–	–	n	n	n	n	I	n

[*]Important near the source (i.e. vehicle tail pipe); probably not important later though will depend on the background concentrations, dilution and other meteorological parameters (i.e. wind speed, direction, temperature, solar radiation).
[**] Depending on the background concentrations, fresh emissions and meteorological parameters; relevant especially for sub–10 nm particles.
[** *] Important when very small particles <10 nm are considered.

Table 4. Summary of PNEFs for various types of road vehicles at different speeds.

PNEF (average ± standard deviation × 10^{14} # veh^{-1} km^{-1})	Size range covered (nm)	Average vehicle speed ($km\ h^{-1}$)	Instruments used	Location	Author (year)
		Mixed vehicle fleet (range ~10^{14})			
2.15 ± 0.05	10–700	90–110	DMPS	Highway, Copenhagen, Denmark	Wang et al. (2010a)
1.87 ± 0.03	10–700	40–50	DMPS	Urban street, Copenhagen, Denmark	Wang et al. (2010a)
1.33 ± 0.5	10–300	20–30	DMS500	Cambridge street, UK	Kumar et al. (2009b)
1.57 ± 0.76	5–1000	~30	DMS500	Cambridge street, UK	Kumar et al. (2008b)
7.9	7–3000	86 (HDVs), 116 (cars)	CPC	Haerkingen, Switzerland	Hueglin et al. (2006)
1.8 4.7	30–10000 10–400	112 (HDVs), 86 (LDVs)	SMPS–OPC SMPS	Bundesautobahn motorway, Germany	Corsmeier et al. (2005)
1.11 ± 0.90 0.57 ± 0.28	15–700 15–700	100 <60 (stop–start)	SMPS SMPS	Brisbane roads, Australia	Morawska et al. (2005)
3.9 11.7 13.5	>7 >7 >7	50 100 120	CPC, SMPS CPC, SMPS CPC, SMPS	Zurich roads, Switzerland	Imhof et al. (2005a)
0.96–4.7	6–220	85	-	Los Angeles motorway, USA	Zhang et al. (2005)
5.2	>6	96.6	CPC	Freeway Los Angeles, USA	Zhu and Hinds (2005)
2.8 (± 23%) HDVs (18.1%) 0.23 (± 24%) HDVs (2.7%)	15–700 15–700	100 100	SMPS–CPC	Motorway, Brisbane Australia	Gramotnev et al. (2004)
1.5 ± 0.08 1.26 ± 0.10	18–700 18–700	80 64	SMPS SMPS	Plabutsch tunnel, Austria Kingsway tunnel, UK	Imhof et al. (2005b)
1.8 ± 0.42	30–10000	120	ELPI	Bundesautobahn motorway, Germany	Imhof et al. (2005c)
2.7 ± 1.1 3.3 ± 1.4 5.4 ± 3.4 11 ± 5.7 4.6 ± 1.9	30–900 30–900 30–900 30–900 30–900	70 75 80 85 70–90	DMPS DMPS DMPS DMPS DMPS	Stockholm Tunnel, Sweden	Kristensson et al. (2004)
0.87–2.73 1.93–9.94	8–300 3–1000	8–80 8–80	SMPS CPC	Minnesota roadway, USA	Kittelson et al. (2004)
2.8 ± 0.5	10–700	40–50	CPC, DMPS	Copenhagen roads, Denmark	Ketzel et al. (2003)

	10–500		SMPS	Tuscarora Mountain Tunnel, Pennsylvania, USA	Gertler et al. (2002)
1.75 (Standard error 67.6%)	17–890	100	SMPS	On–road model estimation, Queensland Australia	Jamriska and Morawska (2001)
Heavy duty vehicles, HDVs (range ~10^{14}–10^{15})					
17.5 ± 0.68	10–700	90–110	DMPS	Highway, Copenhagen, Denmark	Wang et al. (2010a)
22.06 ± 1.28	10–700	40–50	DMPS	Urban street, Copenhagen, Denmark	Wang et al. (2010a)
7.06 ± 1.81	>7	39.5	CPC	Marylebone road, London, UK	Beddows and Harrison (2008)
6.67± 0.91	11–437	<50	SMPS	London roads, UK	Jones and Harrison (2006)
7.8	30–10000	85.8 ± 7.5	SMPS–OPC	Motorway, Germany	Corsmeier et al. (2005)
55 69 73	>7 >7 >7	50 100 120	SMPS–CPC SMPS–CPC SMPS–CPC	Zurich roads, Switzerland	Imhof et al. (2005a)
7.79 ± 6.32	30–10000	86	ELPI	Bundesautobahn motorway, Germany	Imhof et al. (2005c)
3.23 ± 9.9	7–270	-	SMPS–CPC	Caldecott Tunnel Berkeley, USA	Geller et al. (2005)
3.9	7–450	40	DMPS–CPC	Roadside Stockholm, Sweden	Gidhagen et al. (2004a)
52	3–450	100–120	DMPS–CPC	Highway, Stockholm, Sweden	Gidhagen et al. (2004b)
73.3	3–900	48–85	DMPS	Road tunnel, Stockholm, Sweden	Gidhagen et al. (2003)
2.8 (standard deviation 10–15%)	15–700	–	SMPS	Motorway, Brisbane Australia	Gramotnev et al. (2003b)
2.1–3.1 (HDV>64%) 0.51–0.54 (HDV 13–15%)	10–400 10–400	<90 <90	SMPS SMPS	Tuscarora mountain tunnel, Pennsylvania, USA	Abu-Allaban et al. (2002)
Light duty vehicles, LDVs (range ~10^{13}–10^{14})					
0.81 ± 0.07	10–700	90–110	DMPS	Highway, Copenhagen, Denmark	Wang et al. (2010a)

1.01 ± 0.06	10–700	40–50	DMPS	Urban street, Copenhagen, Denmark	Wang et al. (2010a)
0.63 ± 0.16	>7	39.5	CPC	Marylebone road, London, UK	Beddows and Harrison (2008)
0.60 ± 0.16	11–437	<50	SMPS	London roadside, UK	Jones and Harrison (2006)
0.0018 (Euro–4 with DPF)	7–10,000	50–120	ELPI	Dynamometer, Napoli, Italy	Prati and Costagliola (2008)
1.2	30–10000	111.5 ± 16	SMPS–OPC	Motorway, Germany	Corsmeier et al. (2005)
0.8 3.2 6.9	>7 >7 >7	50 100 120	SMPS–CPC SMPS–CPC SMPS–CPC	Zurich roads, Switzerland	Imhof et al. (2005a)
1.22 ± 0.49	30–10000	113	ELPI	Bundesautobahn motorway, Germany	Imhof et al. (2005c)
2.2 ± 1.24	7–270	-		Caldecott Tunnel Berkeley, USA	Geller et al. (2005)
1.4	3–450	100–120	DMPS–CPC	Highway, Stockholm, Sweden	Gidhagen et al. (2004b)
1.74 10.1	3–900 3–900	48 85	DMPS	Road tunnel, Stockholm, Sweden	Gidhagen et al. (2003)
Cars (petrol–fuelled) (range ~10^{12}–10^{14})					
0.03–1.3	>10	50, 70	CPC	Gothenberg road, Sweden	Hak et al. (2009)
~0.017 ~0.018 ~0.074	7–400 7–400 7–400	30 95 150	SMPS SMPS SMPS	Leipzig road, Germany	Wehner et al. (2009)
0.0234 (Euro–2 and Euro–3)	7–10,000	50–120	ELPI	Dynamometer, Napoli, Italy	Prati and Costagliola (2008)
0.17–0.45	11–450	<50	SMPS	London, UK	Jones and Harrison (2006)
0.189 ± 0.34 0.218 ± 0.06	15–700 15–700	100 <60 (stop–start)	SMPS SMPS	Brisbane, Australia	Morawska et al. (2005)
Cars (diesel–fuelled) (range ~10^{14})					
1.4 –1.8	>10	50, 70	CPC	Gothenberg roads, Sweden	Hak et al. (2009)
~1.6 (Urban roads) ~0.6 (low engine load) ~4.2 (high engine load) ~1.1 (low engine load) ~1.3 (medium engine load)	7–400 7–400 7–400 7–400 7–400	30 105 105 120 149	SMPS SMPS SMPS SMPS SMPS	Leipzig urban roads and freeways, Germany	Wehner et al. (2009)

~4.4 (high engine load)	7–400	149	SMPS		
1.32 (Euro–3; without DPF)	7–10,000	50–120	ELPI	Dynamometer, Napoli, Italy	Prati and Costagliola (2008)
0.44 0.57 1 (all results for steady state conditions)	8–400 8–400 8–400	50 70 100	SMPS SMPS SMPS	Delft, Netherlands	Rijkeboer et al. (2005)
7.17 ± 2.80 2.04 ± 0.24	15–700 15–700	100 <60 (stop–start)	SMPS SMPS	Brisbane roads, Australia	Morawska et al. (2005)
Buses (diesel–fuelled) (range ~10^{14})					
1.2 (25% engine power) 1.5 (50% engine power) 18 (100% engine power)	5–4000 5–4000 5–4000	60 60 60	SMPS–CPC SMPS–CPC SMPS–CPC	Dynamometer, Brisbane Australia	Jayaratne et al. (2009)
3.11 ± 2.41	17–700	60	SMPS	Woolloongabba Tunnel, Brisbane Australia	Jamriska et al. (2004)
3.87 ± 2.49	8–400	40–80	SMPS	Dynamometer, Brisbane Australia	Ristovski et al. (2002)
1.57	8–304	80	SMPS	Dynamometer, Brisbane Australia	Morawska et al. (1998)
Buses (compressed natural gas; CNG) (range ~10^{08}– 10^{15})					
0.10 (25% engine power) 0.25 (50% engine power) 14 (100% engine power)	5–4000 5–4000 5–4000	60 60 60	SMPS–CPC SMPS–CPC SMPS–CPC	Dynamometer, Brisbane Australia	Jayaratne et al. (2009)
~0.00015–0.0003 (load 6.5 kW) ~0.00006–0.0003 (load 9.5 kW) ~0.000007–0.15 (load 12.5 kW) ~0.05–0.16 (load 18.8 kW) (all results for 6 cylinder SI engine)	5–400 5–400 5–400 5–400	40 60 80 100	SMPS SMPS SMPS SMPS	Dynamometer, Brisbane Australia	Ristovski et al. (2004)
Petrol–fuelled spark ignited vehicles (range ~10^{9}– 10^{13})					
~0.000076–0.15 (load 6.5 kW) ~0.00008–0.15 (load 9.5 kW) ~0.0003–0.1(load 12.5 kW) ~0.2–0.21(load 18.8	5–400 5–400 5–400 5–400	40 60 80 100	SMPS SMPS SMPS SMPS	Dynamometer, Brisbane Australia	Ristovski et al. (2004)

kW) (all results for 6 cylinder SI engine)					
~0.08 ~0.02 ~0.20 ~0.50 (all results for LPG 4 1, 6 cylinder SI engine fuelled by unleaded petrol)	8–400 8–400 8–400 8–400	40 60 80 100	SMPS SMPS SMPS SMPS	Dynamometer, Brisbane Australia	Ristovski et al. (2005)
Light petroleum gas, LPG–fuelled spark ignited vehicles (range ~10^{10}–10^{12})					
~0.0009 ~0.008 ~0.05 ~0.08 (all results for LPG 4 1, 6 cylinder SI engine)	8–400 8–400 8–400 8–400	40 60 80 100	SMPS SMPS SMPS SMPS	Dynamometer, Brisbane Australia	Ristovski et al. (2005)
Vehicles generated through road–tyre interface (range ~10^{11}–10^{12})					
0.0037 0.032 (most particles in 15–50 nm size range)	15–700 15–700	50 70	SMPS–DMA SMPS–DMA	Road simulator, Transport Research Institute, Sweden	Dahl et al. (2006)
Two-wheelers (range ~10^{14}–10^{15})					
0.31 (2–stroke; Euro–1 and Euro–2) 0.10 (4–stroke; Euro–1 and Euro–2)	7–10,000 7–10,000	20–45 20–45	ELPI ELPI	Dynamometer, Napoli, Italy	Prati and Costagliola (2008)
1.9–4 21 11 7.5 (2–stroke motorcycles; values are in # km^{-1}at peaks $dN(d\log D_p)^{-1}$)	8–400 8–400 8–400 8–400	30 50 70 90	SMPS SMPS SMPS SMPS	Delft, Netherlands	Rijkeboer et al. (2005)

DMS = differential mobility spectrometer, SMPS = scanning mobility particle sizer, CPC = condensation particle counter, DMPS = differential mobility particle sizer, EAA = electrical aerosol sizer, UCPC = ultrafine condensation particle counter, ELPI = electrical low pressure impactor

Fig.1.ppt_ABL

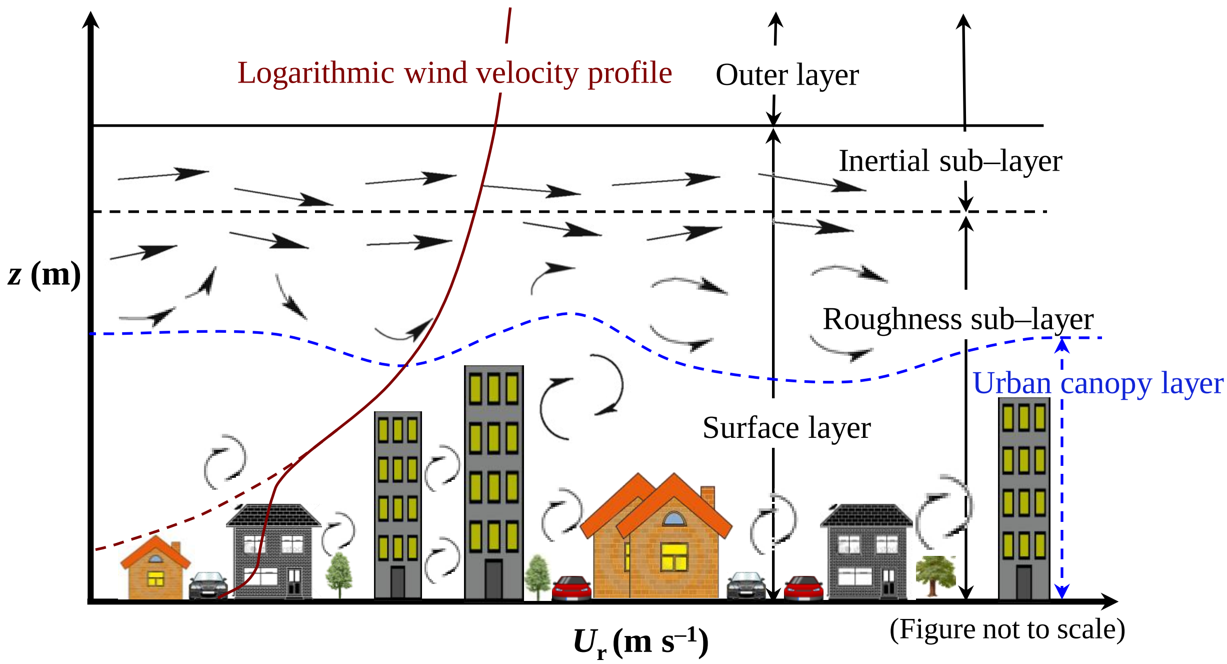

Logarithmic wind velocity profile
Outer layer
Inertial sub–layer
z (m)
Roughness sub–layer
Urban canopy layer
Surface layer
U_r (m s^{-1})
(Figure not to scale)

Fig. 2.ppt_Dry Deposition

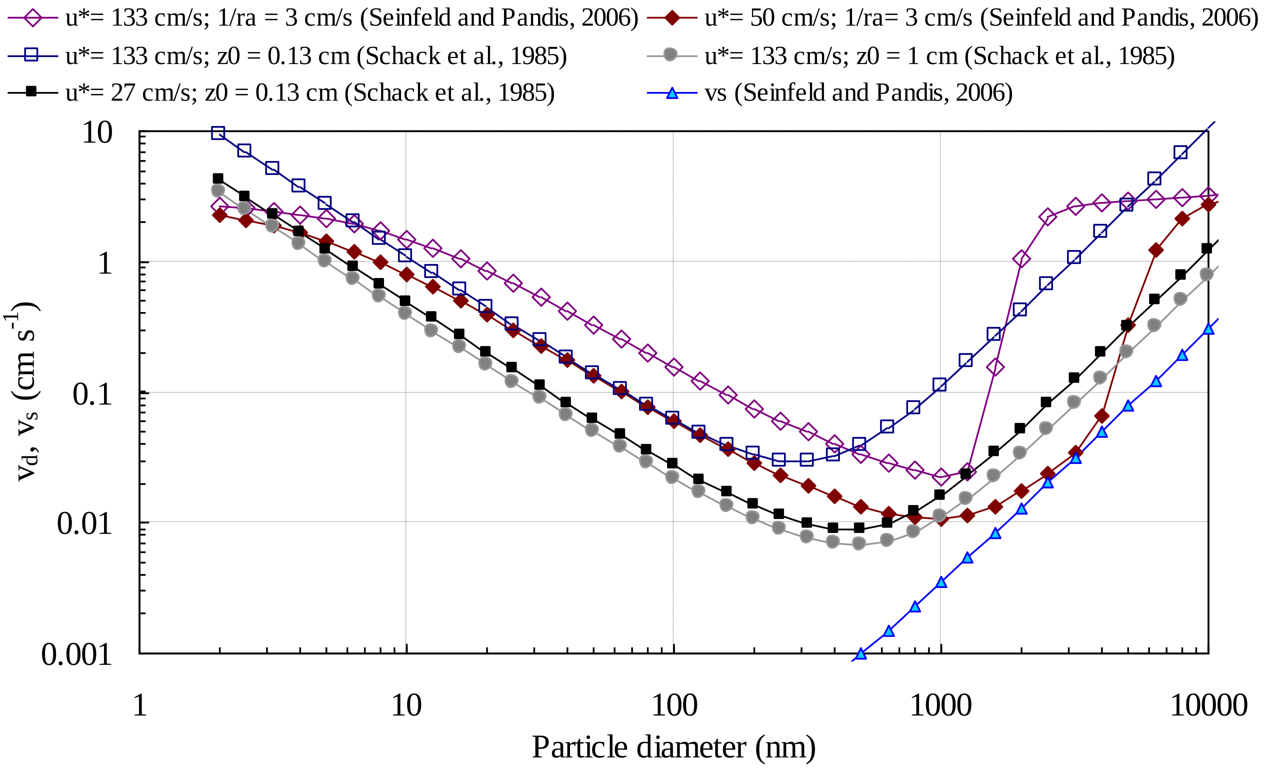


Fig. 3.ppt_Time Scales

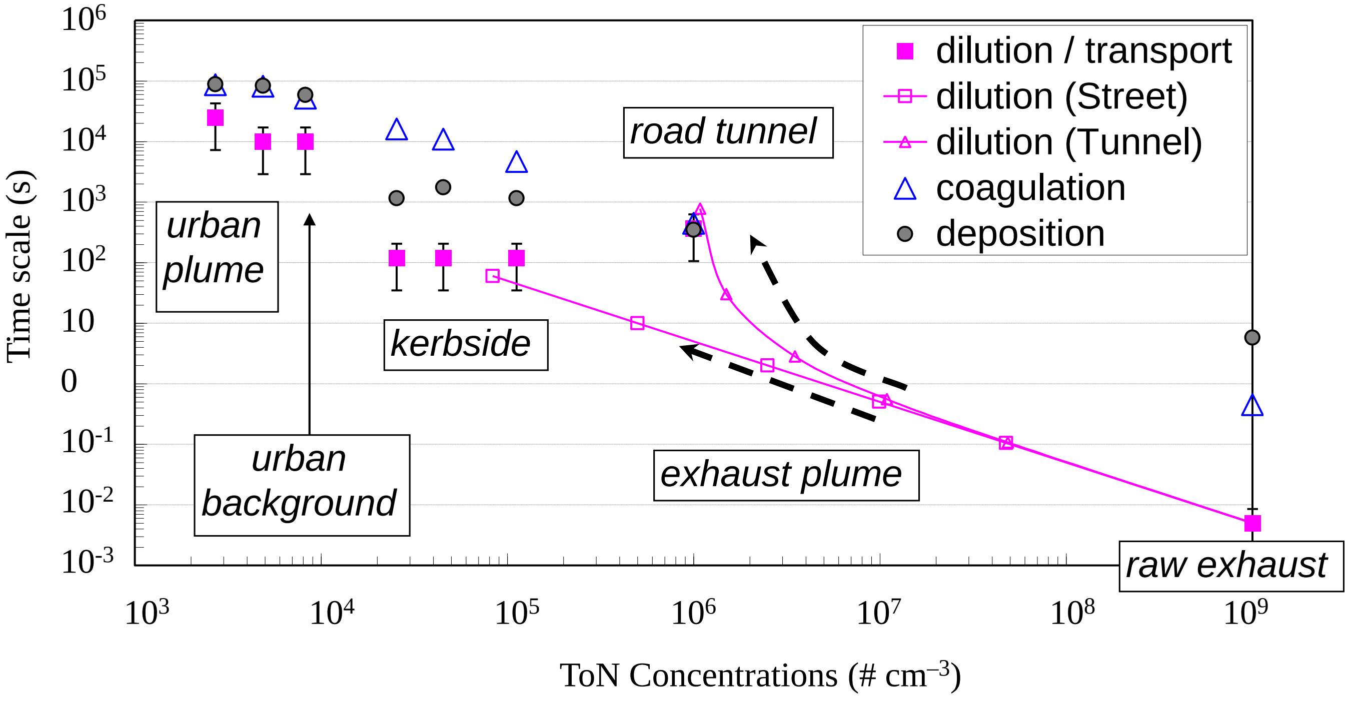


Fig. 4.ppt_PNDs at Different heights

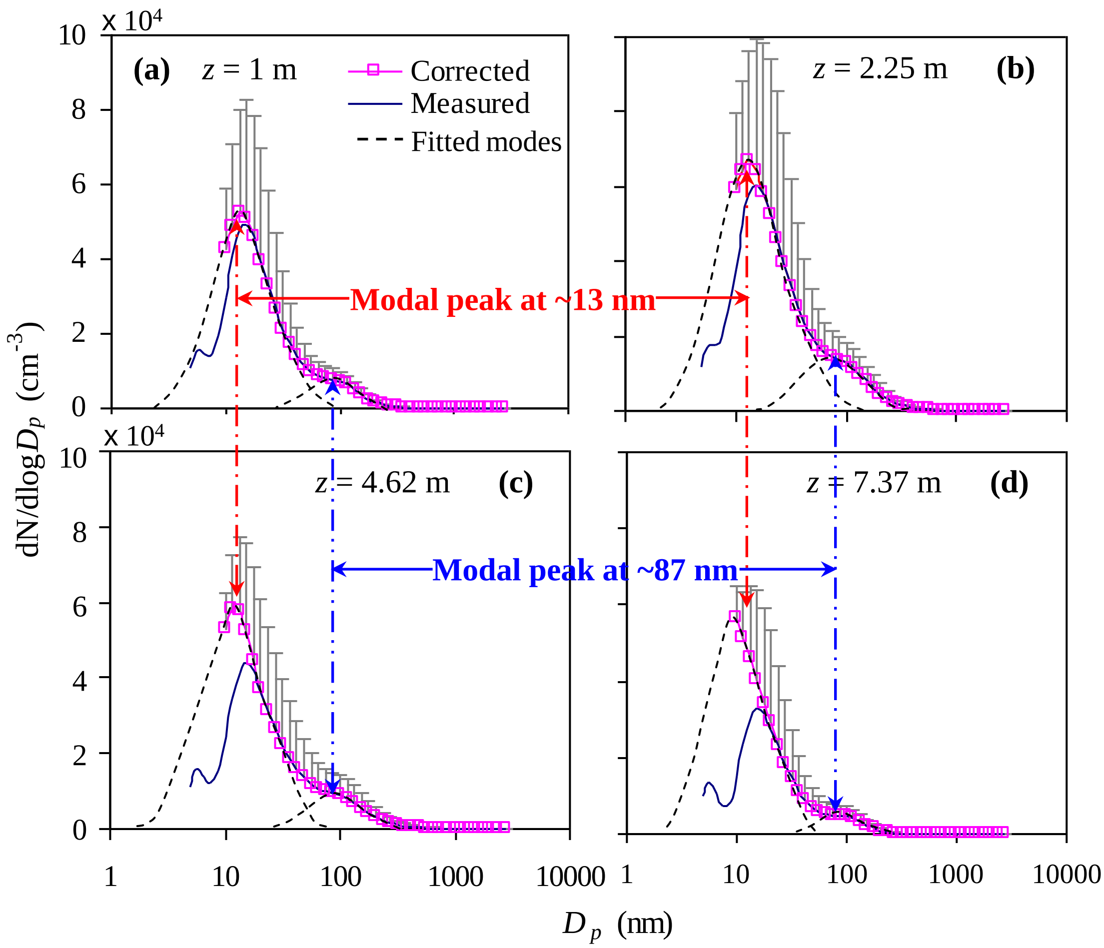
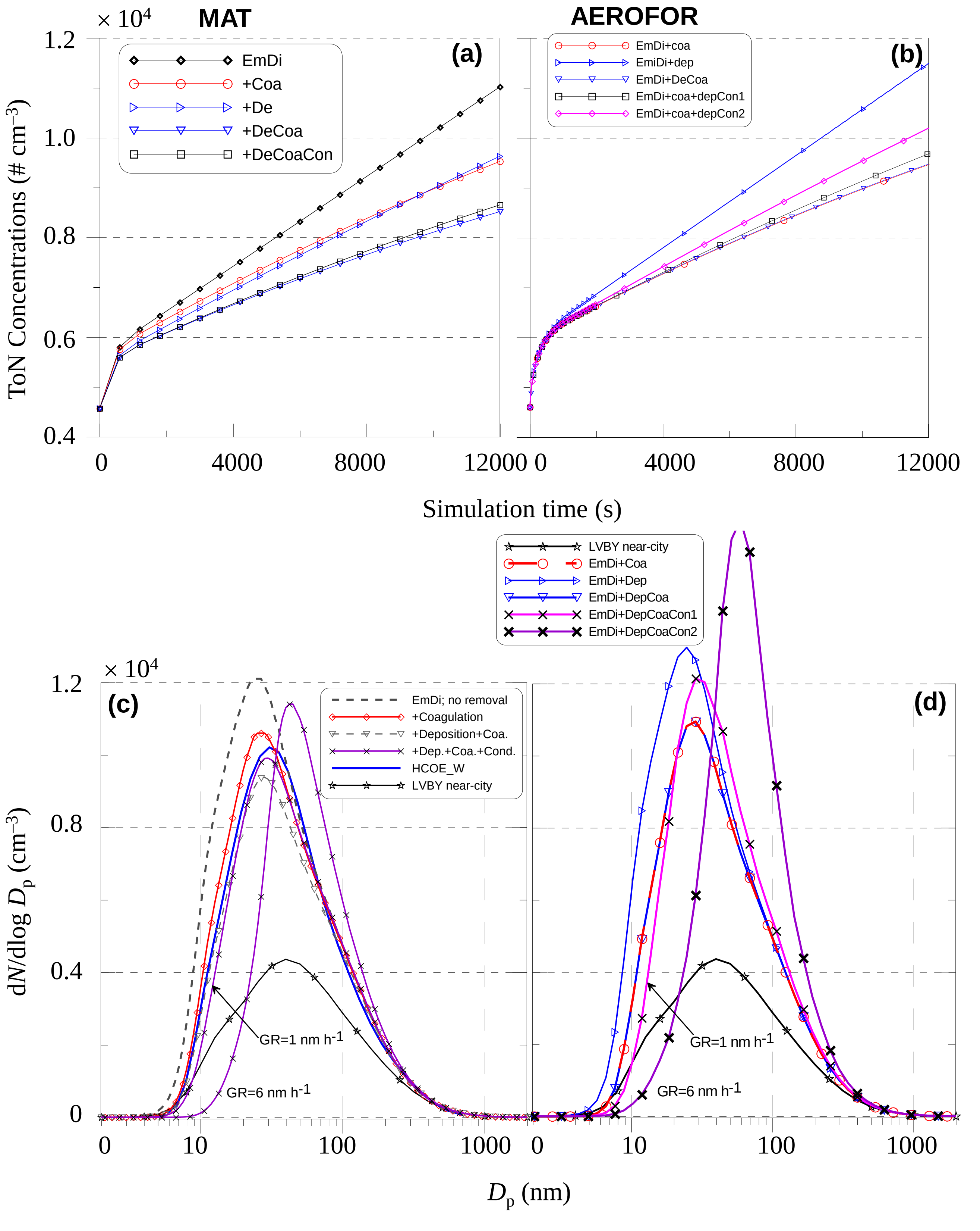


Fig. 5.ppt_Modelling Results



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