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Review

Ultrafine particles in cities



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ABSTRACT

Ultrafine particles (UFPs; diameter less than 100 nm) are ubiquitous in urban air, and an acknowledged risk to human health. Globally, the major source for urban outdoor UFP concentrations is motor traffic. Ongoing trends towards urbanisation and expansion of road traffic are anticipated to further increase population exposure to UFPs. Numerous experimental studies have characterised UFPs in individual cities, but an integrated evaluation of emissions and population exposure is still lacking. Our analysis suggests that the average exposure to outdoor UFPs in Asian cities is about four-times larger than that in European cities but impacts on human health are largely unknown. This article reviews some fundamental drivers of UFP emissions and dispersion, and highlights unresolved challenges, as well as recommendations to ensure sustainable urban development whilst minimising any possible adverse health impacts.

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1. Introduction

Whilst cities are facing challenges in addressing the problem of conventional air pollutants that are part of current regulatory frameworks, the emergence of unregulated pollutants, such as airborne ultrafine particles (UFPs; diameter less than 100 nm), has added an additional

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dimension to this already complex problem. UFPs differ from larger-sized particles in their potential for lung deposition and translocation to other parts of the body (HEI, 2013). There are suggestive hypotheses that these particles may have a greater potential for adverse health impacts compared with their larger counterparts (WHO, 2013). Long-term exposure studies would be required to confirm these hypotheses, but these are currently unavailable (Heal et al., 2012; HEI, 2013; Rückerl et al., 2011). Because of their negligible mass compared with larger-sized particles, UFPs are preferentially evaluated through measurements of particle number concentration (PNC) (Harrison et al., 2000; Kumar et al., 2010a). What makes them distinct from other pollutants is their dynamic nature and it is this on-going transformation of their physical and chemical properties, including number and size distributions, that contribute to substantial temporal (Sabaliauskas et al., 2012) and spatial variability (Heal et al., 2012), which increases with decreasing particle size (Birmili et al., 2013; Costabile et al., 2009). Knowledge of this variability is the key in characterising human exposure and designing monitoring strategies for both developed and developing cities.

The variability of ambient UFP concentrations depends on the dispersion conditions governed by current wind speed and direction, temperature inversions and the “breathability” of cities (Buccolieri et al., 2010). The “breathability” depends on the urban morphology, which dictates the exchange of polluted air within the urban canopy layer (UCL, the lowest portion of the urban boundary layer, UBL, extending to about twice the average building height) with the cleaner air above. Densely packed high-rise buildings and diverse meteorological conditions in cities play a key role in limiting this air exchange and hence, the dispersion of UFPs, further elevating their concentrations. Ever-growing numbers of on-road vehicles in cities worldwide emit UFPs within the UCL, whilst other anthropogenic emission sources, such as power plants, usually emit them at a higher level. Those emitted within the UCL spread laterally by dispersion and affect inhabitants downstream, whilst those released from tall stacks can be re-entrained into the UCL through vertical exchange. Combustion is, in general, a direct source of UFPs, but also of precursors for secondary particle formation. The latter may occur via photochemical processes and the condensation of semi-volatile vapours (or very low volatility vapours produced in photochemical reactions). Compared to new particle formation within the UCL, combustion emissions to the higher level of the UBL efficiently lead to new particle formation due to the smaller surface area of pre-existing particles that results in much lower coagulation/condensation sinks for nano-sized particles and vapours (Kulmala et al., 2000; O’Dowd et al., 2002).

During the last decade, a number of experimental and numerical studies have advanced understanding of the emission, formation, dispersion, exposure and health effects of UFPs. Most of these studies were conducted in European cities (Kumar et al., 2013b), with only a handful in emerging Asian cities where a majority of the world’s urban population resides (Kumar et al., 2013a). The primary reason for such a lack of attention in Asian cities is their focus on complying with regulations for criteria pollutants, the concentrations of which often exceed the regulatory limits by orders of magnitude (Sharma et al., 2013). Since there are no air quality regulations for UFPs, they have not received due attention of regulatory authorities (Kumar et al., 2011c). Taking advantage of existing knowledge, the focus of this paper is on UFP emissions from road vehicles, a dominant source that contributes up to 90% of total PNCs at busy roadsides (Kumar et al., 2010a). For the first time, we present a critical assessment of the spatial variability in emission levels, concentrations and exposure, in addition to discussing the impact of new particle formation on PNC in a range of city environments. Discussion is then extended to present unresolved challenges that need to be tackled, together with regulatory concerns and directions for future research to guide the sustainable development of cities.

2. Particle number emissions across countries

The estimation of particle number (PN) emissions helps to assess the contribution of various sources. In turn, this information is vital to understand their overall impact on human health and environment, and thus, to design effective mitigation strategies. Knowledge of PN emission factors for different types of vehicles and fuels under varying driving conditions is an important input for such computations. Numerous research studies have reported emission factors (Kumar et al., 2011b) that researchers have used to develop PN emission inventories. Consequently, consolidated emission factor data bases (e.g. COPERT, PARTICULATES, TRANSPHORM) have also become available for constructing PN emission inventories in Europe (Kumala et al., 2011; Paasonen et al., 2013; Reddington et al., 2011) and the UK (AEA, 2010). Currently, most of the available emission factors are for vehicle fleets of European cities. Outside the European Union (EU), PN emission inventories are only available for Brisbane, Australia (Keogh et al., 2009) and Delhi, India (Kumar et al., 2011a). This clearly indicates a major research gap and the need to quantify PN emission factors for cities elsewhere in the world, so that local PN emission inventories can be constructed and their impact on human health and global climate assessed.

Particles smaller than 300 nm diameter contribute over 99% of total PN emissions (Kumar et al., 2009a). Unlike the lower cut-off size, any upper cut-off size over 300 nm does not influence PN estimates greatly. By combining knowledge on the prevailing engine technology with the amount of fuel consumption, Paasonen et al. (2013) compiled a PN emission inventory for the 28 EU countries (EU28; see Section S1), covering the 3–1000 nm size range. Fig. 1 presents the PN emissions from road traffic and other sources in EU28 for the year 2010, and their significant linear correlation with the countries’ populations. According to the study, road transport contributed over 60% of the total PN emissions, followed by non-road transport (~19% to total PN emissions; including national ship traffic) and domestic combustion (~13%). About 84% of the total PN emissions were found to be within the UFP size band, which is within the range of the percentages observed by individual urban monitoring studies in London (Dall’Osto et al., 2011), Cambridge (Kumar et al., 2008a), Brisbane (Meja et al., 2008), Dresden (Birmili et al., 2013), Delhi (Monkkonen et al., 2005a) and Beijing (Wu et al., 2008). In Europe, the contribution from road traffic varied from ~32% of total PN emissions in Greece to ~97% in Luxembourg. France, Spain, Germany, Italy, UK and Poland are the top six PN emitters in the EU28 and together, their road traffic contributes nearly 3/4 (~72%) of the total traffic-induced PN emissions in the EU28.

Comparison of the annual PN emission estimates for Brisbane (1.08×10^{25}) and Delhi (1.37×10^{25}) shows that emissions from traffic alone were 1.4% and 1.8% of the EU28’s traffic-derived PN emissions, respectively, and this share increased to 5.0 and 6.3% when the top six PN emitting countries were excluded from calculations. Interestingly, these statistics change when per capita traffic-related PN emissions are estimated for each European country and compared with each other, as well as with a more densely populated megacity, Delhi (Fig. S1). Total PN emissions from road traffic in Delhi were 79% higher than those in Brisbane, but the trend for per capita emissions was the opposite, being 559% higher in the latter than Delhi (Keogh et al., 2009; Kumar et al., 2011a). With the exception of France, none of the top six traffic-related PN emitting countries remained on the list, which included Luxembourg, Cyprus, Ireland, Austria, Portugal and France (Fig. S1). All of the European countries emitted between 1.2 and 18.3 times more than Delhi, with the exception of Romania, whose traffic-related per capita PN emissions were ~0.67-fold lower.

In 2010, there were 21 cities worldwide with a population of at least 10 million or more (called megacities), ten of which were in Asian countries (Kumar et al., 2013a). Delhi is one of these Asian megacities and road traffic in Delhi alone contributed ~32-fold higher PN emissions than Malta, the European country with the lowest PN emissions, and

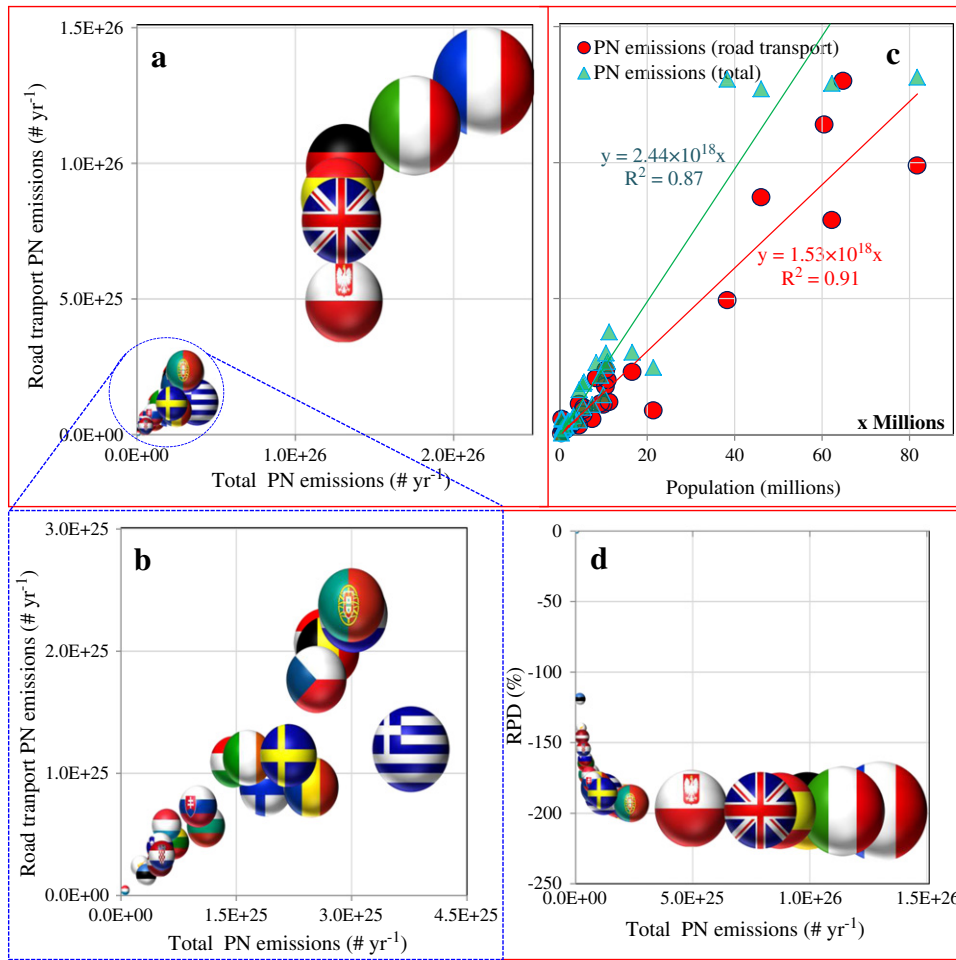


Fig. 1. (a) Three-dimensional bubble map showing total and road-traffic induced PN emissions in EU28 for 2010, (b) an enlarged version of parts of (a), (c) correlations between total and traffic derived PN emissions in each of the EU28 countries during 2010 with their respective population, and (d) RPD values based on the annual transport emissions of PN in various European countries, using Malta (with the smallest PN emissions) as the reference country. The details of estimating the RPD values can be seen in Section S2 and the explanation of their variation between 0 and –200% is available in Section 2. The PN emission data is taken from Paasonen et al. (2013), as explained in Section S1. Each bubble represents one European country and the size of each bubble corresponds to UFP (diameter less than 100 nm) emissions in that country, starting from the smallest bubble for Malta ($6.24 \times 10^{23} \text{ yr}^{-1}$) to the largest bubble for France ($1.35 \times 10^{26} \text{ yr}^{-1}$).

~11% of the traffic-related emissions in France, the country with the largest PN emissions in Europe. Compared to Delhi, ten major Asian megacities have similar or worse traffic emission conditions, as demonstrated by recent health studies (Gurjar et al., 2010). Assuming that daily per capita PN emissions from road traffic in these cities (Fig. S2) are similar to those in Delhi (1.70×10^{15}) (Kumar et al., 2011a), their road transport PN emissions are expected to be ~14% of those in the whole EU28.

We derived the relative percentage differences (RPDs; Section S2) for facilitating a good graphical and symmetrical representation, in order to assess the variability in emissions between different countries against a reference country, which was chosen to be Malta (Fig. S3). The RPD is calculated by dividing the absolute difference between the two values (i.e. difference between the reference category and each category in the classifying variable) by the average of the same two values, and then multiplying the resulting values by 100 (as detailed in Section S2). Numeric values for the RPD can therefore vary between –200 and +200%; although not directly applicable to the present case, a value within $\pm 30\%$ is generally considered as acceptable from the air quality modelling perspective. In this case, the lowest annual PN emissions for Malta are used as a reference value that gives a negative value of the numerator term in Eq. (S1) and therefore the values of the RPD vary between 0 and –200%. As expected, reasonably large RPD values of –199% were obtained, which did not change much (–193%) after excluding the six highest PN emitting countries.

However, these six countries, which also had the highest RPD values, showed very little variability, ranging from –198% to –199%. These values were closely followed by Delhi (–188%) and Brisbane (–185%). Such variability was expected, given that PN emissions are the result of many diverse factors, such as the number and type of vehicles, fuel quality, travel behaviour and the distance travelled by each vehicle, as discussed in Section 3.

3. Spatial variability of UFP concentrations across cities

Ambient particle mass concentrations (PMCs) are usually dominated by particles above 100 nm. In the absence of strong local sources, PMCs show only minor spatial variability within a city environment (Birmili et al., 2013). However, the spatial inhomogeneity of UFPs remains a key challenge for the assessment and control of these particles. This inhomogeneity is generally largest close to roadsides and is largely due to high PNCs in the UFP size range. This is reflected by multiple-site intra-city PNC monitoring studies, which show up to an order of magnitude difference with respect to minimum average PNCs. For instance, spatial differences in average PNCs between sites within a city were found to vary from a factor of ~2 in Cassino, Italy (Buonanno et al., 2011a) and Antwerp, Belgium (Mishra et al., 2012) up to ~8 in Brisbane, Australia (Mejia et al., 2008) and ~9 in Dresden, Germany (Birmili et al., 2013). Mobile measurements of the UFPs, generally carried out for the assessment of personal exposure, also reveal a marked spatial variability

in the UFP concentrations at different locations within busy areas of cities. For instance, results of the DAPPLE (Dispersion of Air Pollution and Penetration into the Local Environment) experiments highlighted a noticeable difference ($33,162\text{--}163,110\text{ cm}^{-3}$) in the minimum and maximum exposure concentrations of UFPs between the kerbside and building side locations in central London (Kaur et al., 2005). As also shown by other studies (Fujitani et al., 2012; Pirjola et al., 2006; Zhu et al., 2002), the findings of the DAPPLE experiments indicated a striking decline in UFP concentrations with increasing distance away from the road, besides showing over a factor of ~4 variation ($29,951\text{--}131,415\text{ cm}^{-3}$) in the minimum and maximum UFP concentrations present during the measurements in the different modes of transport (Kaur et al., 2006). In fact, a summary of many such recent studies suggests that mean UFP concentrations can range by an order of magnitude, with the lowest and the highest UFP concentrations of $\sim 34,000 \pm 18,000\text{ cm}^{-3}$ and $\sim 300,000 \pm 260,000\text{ cm}^{-3}$ experienced during cycling and in automobiles during tunnel travel in the range, respectively (Knibbs et al., 2011).

As for PNC variability across the cities, we analysed the PNC data measured over 40 different roadside locations in numerous cities within and outside Europe (Fig. 2). The RPD values were derived against London as a reference site and showed large overall variability in the range – 172% to 102% (Fig. 3a). In absolute terms, cross-comparison of average PNC data suggests large differences, although these variations

are smallest for European cities compared with Asian or US cities. In fact, the data suggested a difference of ~6 and ~19 times between the average PNCs within the EU and Asian cities, respectively. These differences increased up to ~26-fold between Asian (Delhi; with the largest PNCs) and European (Essen; the smallest PNCs) cities. It is worth noting that short-term (1 s average) peak PNCs over few tens of seconds have been found to be over an order of magnitude larger compared with long-term (hourly) averages along the roadsides (Kumar et al., 2008b). This means that the differences in PNCs can further increase if peak concentrations are taken into consideration, suggesting further challenges for relevant regulatory designs.

This large spatial variability is a product of numerous factors affecting the emission and dispersion. Some of this variability is due to differences in experimental methods. The lower cut-off size for the PNC measurement, for example, varied between 3 and 25 nm in numerous studies, as seen in Table S1 and this can account for up to ~35% of the total PNCs in roadside environments (Kumar et al., 2009a). Distance of the measurement location away from the road is another consideration. For the unobstructed topographic settings, studies have found an exponential decay in PNCs with perpendicular distance away from the road, meaning that PNC levels can decrease by up to ~40% of their kerbside level within a distance of 10 m (Fujitani et al., 2012; Zhu et al., 2002). Some of the spatial variations may be caused by seasonal (e.g. temperature inversion) effects that have been found to significantly increase

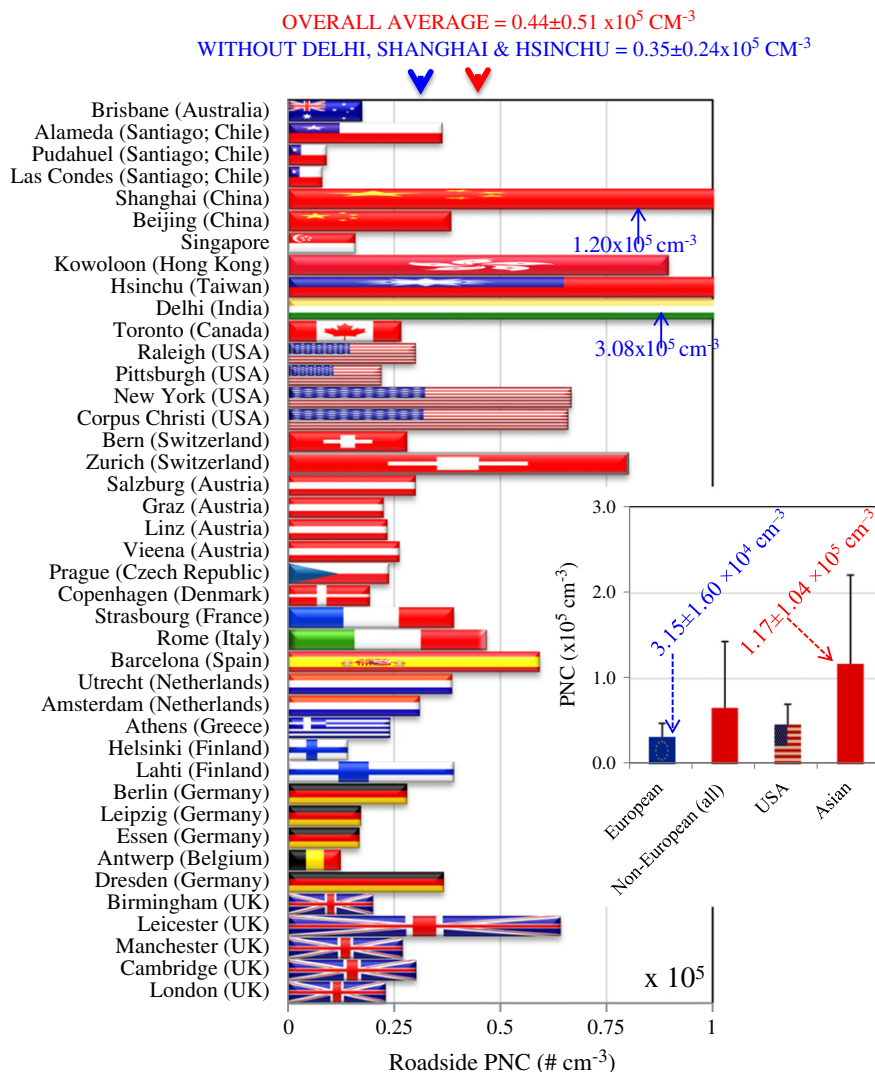


Fig. 2. Typical measured PNC levels at the roadside in 42 different cities. Details of the source studies are presented in Table S1.

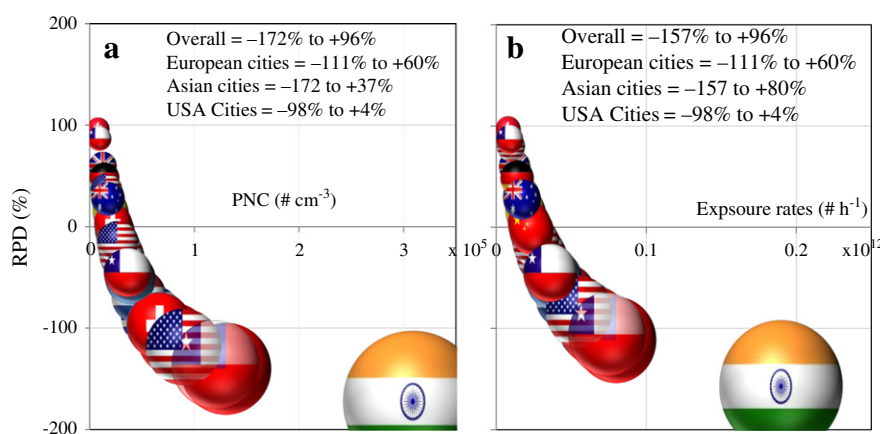


Fig. 3. RPD values for the (a) roadside PNCs in various cities, using PNC values for London as a reference, and (b) exposure to roadside PNCs recorded at various city locations, using the exposure for London as a reference. The size of each bubble is based on the PNC/exposure at each location, and the flags represent the country in which each city is located, as noted in Table S1.

the PNCs during cold months (Buonanno et al., 2013). The average PNCs have generally been found up to ~300% higher during winter compared with summer under identical traffic emission conditions (Fujitani et al., 2012; Pirjola et al., 2006; Sabaliauskas et al., 2012). In summary, nearly 400% of variability can be expected among the PNC values presented in Table S1, based on the above factors alone. The rest of the observed variability can be attributed to other local factors including traffic volume, fuel type, urban morphology, climate, dispersion conditions specific to individual sampling locations, and uncertainty in the measured data due to manual (e.g. data collection and handling) and mechanical (e.g. instrument calibration) errors, which are difficult to generalise.

The overall levels as well as inter-city differences in PNC notably decreased when restricting the analysis to European data. The EU is currently the only region worldwide where PN emission standards for vehicles are in place (EC, 2008). Euro 5 and Euro 6 standards have set a PN emission limit of 6.0×10^{11} solid particles per km for compression ignition diesel vehicles (EC, 2008). Euro 6 standard also places a similar limit (6.0×10^{11} solid particles per km) for direct injection gasoline vehicles, which is however exempted to 6.0×10^{12} solid particles per km for the first three years of its implementation date in September 2014 (EC, 2008). It is worth noting that two types of particles are present in vehicle exhaust – the semi-volatile nucleated particles and the larger particles with a graphitic core (Harrison et al., 2011) – but the Euro 5/6 emission limitation refers to non-volatile particles that are larger than 23 nm after heating at 300 °C.

Both gasoline (petrol) and diesel engines emit UFPs, but diesel engines are considered to be more important because of substantially higher emission factors (number emitted per vehicle-km) (Beddows and Harrison, 2008). Whilst relatively little work has been conducted into the detailed characterisation of UFP from gasoline engines, the major difference in the case of conventional (port injection) engines appears to be a much lower concentration of solid soot-mode carbonaceous particles relative to the smaller semi-volatile nucleation particles deriving from condensation of high molecular weight hydrocarbon material derived mainly from unburned engine oil (Eastwood, 2008; Harris and Maricq, 2001). In the case of the less common gasoline direct injection engine, UFP emissions are much more similar to those from a diesel engine. The semi-volatile compounds within the nucleation mode particles tend to evaporate as the particles disperse away from the point of emission and concentrations of vapour are reduced. On a timescale of tens of minutes, it has been suggested that the particles can reduce in diameter by a factor of more than two (Dall'Osto et al., 2011).

Relatively high PNCs, and hence high RPD values, were seen for Asian cities (Fig. 3). These indicate relatively higher levels of sulphur in diesel fuel promoting sulphuric acid induced nucleation (Kulmala

et al., 2004), use of inferior vehicle technology (Liu et al., 2013) and densely built environments adversely affecting the dispersion of UFPs (Kumar et al., 2013a).

4. UFP exposure across cities

Knowledge of inhalation dosimetry is important to establish links between exposure and health effects. Exposure to high PNCs is known to aggravate existing disease, due to the efficient alveolar deposition of nano-sized particles and their potential to enter the pulmonary vascular space (Shah et al., 2008). The extent of respiratory deposition (i.e. the sum of alveolar, tracheobronchial and extrathoracic regions) is dominated by the number of sub-100 nm particles. For instance, the respiratory deposited fraction decreases in power form, from ~91% for 5 nm particles to ~63% for 30 nm, ~23% for 100 nm and only ~13% for 300 nm particles (ICRP, 1994). This clearly suggests that the knowledge of particle size distributions, which can change dramatically in urban environments (Dall'Osto et al., 2011), is important for the accurate estimation of dose rates.

The strong size-dependence of deposition implies that the exposure is *not* directly proportional to PNC, despite total PNC being identical at different locations. We have demonstrated this by calculating the *average* roadside PNCs for Asian and European environments, which shows a ratio of ~3.7 (Fig. 2). Respiratory deposition rates are computed by using the methodology presented in Section S3, where we assumed the typical size distribution in Delhi (Monkkonen et al., 2005a) and Cambridge (Kumar et al., 2008b) as representative of Asian and European cities, respectively. The respective average dose rates come out at $\sim 7.20 \times 10^{10}$ and 3.12×10^{10} particles per hour, which have a ratio of ~2.3 compared with ~3.7 for the average PNCs. This reduction is mainly due to the large percentage of particles in the nucleation mode (<30 nm) in European cities compared with dominance of accumulation mode particles (~30–300 nm) in Asian cities. These exposures are, of course, sensitive to the selected size distributions, but represent typical examples to demonstrate the significance of knowledge of size distributions.

In order to assess the relative differences in exposure between individual cities, we moved from a continental to a city scale by assuming a fixed deposition fraction of 0.41 for Asian and 0.66 for European and other cities (Section S3), the latter being identical to that used by urban exposure assessment studies in European city environments (Int Panis et al., 2010). The RPD values for exposure, using London as a reference, varied between +102% and -157% for the whole dataset (Fig. 3b). In absolute terms, as expected from the PNCs, differences between the average exposure within the European and Asian cities were found to be ~6 and ~19 times, respectively (Fig. 3b). These

differences were up to ~15 fold between Asian (Delhi; with the largest rates) and European (Essen; the smallest rates) cities. It is worth noting that these estimates are based on long-term average PNCs and the consideration of short-term average PNCs might further increase these differences. Furthermore, the spatial representativeness of exposure in individual cities will be greatly affected by their spread and land use. For instance, cities with a larger area but less built land cover (e.g. Brisbane compared to Delhi, where ~21% of land area is covered by ~1749 km road length per 100 km² (Kumar et al., 2011a)) will provide larger spacing for emissions to disperse before they reach human receptors and hence, relatively lower exposure.

Although indoor and transport micro-environments are not the focus of this article, consideration of UFP exposure in these environments is equally important for the assessment of daily exposure doses of the UFPs for urban dwellers. For non-smoking type indoor environments in the USA, studies have noted relative contribution to the typical 24-h daily exposures in the outdoor, indoor and in-vehicle environments as 36, 47 and 17%, respectively (Wallace and Ott, 2011). This contribution was doubled for indoor (77%), nearly halved for outdoor (17%) and became one-third for in-vehicle (6%) for the smoking indoor environments (Wallace and Ott, 2011). The exposure contribution from outdoor sources is expected to decrease in rural areas and increase in polluted urban areas due to a greater density of road vehicles. For example, the above figures for the non-smoking case were found to be reversed in a Los Angeles based study, where ~46, 36 and 18% of total UFP exposure was found to occur in outdoor, in-vehicle and indoor environments, respectively (Fruin et al., 2008). In terms of comparison of the daily overall exposure doses in the European and Australian population, recent studies suggest that the Italian population experiences significantly higher daily UFP exposure doses than the Australian population, mainly because of the much higher level of PNC exposure during eating, cooking and transportation activities (Buonanno et al., 2011b; Buonanno et al., 2012b).

Recent estimates of the mortality burden based on long-term exposure to PM_{2.5} mass concentrations suggest an average loss of 7–8 months in life expectancy for UK residents and about £20 billion per year equivalent health costs (Defra, 2008). An equivalent estimate for PN exposure is unavailable and hence, the contribution of UFPs to these effects is unclear. However, time-series epidemiological studies show that particle number and mass metrics are predictive of different health outcomes (Atkinson et al., 2010) and the PMCs do not describe well the exposure to UFPs (Harrison et al., 2010).

Some preliminary estimates can be made using only the mortality estimates available for Delhi (Kumar et al., 2011a), showing about 1900 deaths per million due to traffic-related PN exposure in 2010. Similar or worse air pollution conditions can be expected in Asia (Gurjar et al., 2010) and assuming similar mortality rates in Asian megacities to those in Delhi, rough estimates suggest ~0.31 million deaths per year.

5. Role of new particle formation events on PNCs in developing and developed cities

Formation of new nano-sized particles within and above the UCL is significant, but it varies from city to city in terms of frequency and its potential to increase PNCs. For example, in some heavily polluted urban areas like Mexico City or Chinese megacities, it occurs on almost ~50% of days whereas in other cities, the frequency is less than 10% (Kuang et al., 2008; Wu et al., 2007). Less polluted environments are prone to significant secondary formation where the condensation of photochemically-formed low volatility vapours leads to condensational growth and sulphuric-acid induced nucleation (Holmes, 2007). Specific meteorological conditions, including intense solar radiation, low wind speed, and low relative humidity favour secondary formation (Rimnácová et al., 2011). On the other hand, there is always a condensation sink in the form of the surface of pre-existing solid particles, which are usually higher in polluted environments, to inhibit secondary

formation. The integrated effects of new particle formation and the condensation sink determine the net rate of new particle formation in urban areas. As a result, the formation rate of 3 nm particles within the UBL and urban areas can be in the range of 0.01–10 and up to 100 particles cm⁻³ s⁻¹, respectively, with a typical particle growth rate in the range of 1–25 nm h⁻¹, which is usually much higher during summer than winter (Kulmala et al., 2004). The following examples demonstrate the importance of secondary formation in influencing the PNC in developed and developing cities.

The formation rate of 3 nm particles from the observed events varied from 3.3 to 13.9 cm⁻³ s⁻¹ in Delhi, with the growth rate varying from 11.6 to 18.1 nm h⁻¹ (Monkkonen et al., 2005a). Given the very high background particle loading in Delhi's atmosphere (Fig. 2), larger condensation sinks causing more effective suppression of the nucleation process are expected. In the case of another megacity, Beijing, recent studies suggest that the annual average PNCs of nucleation mode (3–20 nm), Aitken mode (20–100 nm), and accumulation mode (0.1–1 µm) particles in ambient air were 9000 cm⁻³, 15,900 cm⁻³, and 7800 cm⁻³, respectively (Wu et al., 2008). These PNCs are generally higher than those in the cities of developed countries, especially for accumulation mode particles (Dall'Osto et al., 2011). The higher concentration of accumulation mode particles results in a higher condensation sink for precursor vapours for new particle formation and, together with strong scavenging of newly formed nanoparticles, this hinders the occurrence of observed new particle formation events. Nevertheless, a high frequency (~40%) of nucleation events was unexpectedly observed in the urban areas of Beijing (Wu et al., 2007). Especially in spring, frequent new particle formation events led to both the highest total PNC and the lowest volume concentration compared to other seasons (Wu et al., 2008). Long-term measurements showed that the range of formation rate spanned from 3.3 to 81.4 cm⁻³ s⁻¹ and the growth rate varied from 0.1 to 11.2 nm h⁻¹. Meanwhile, simultaneous nucleation events were found at both urban and regional background sites, indicating that this is a regional phenomenon in the Beijing area (Wang et al., 2013). This means that a rapid growth of newly formed particles significantly contributed to the nucleation mode and Aitken mode over a large area.

In terms of European cities, a systematic study in Birmingham found new particle formation to be infrequent but not insignificant (Alam et al., 2003; Shi et al., 2001). For instance, nucleation events were observed on ~5% of monitoring days, with typical growth rates of around 4 nm h⁻¹ for particles between 10 and 30 nm (Alam et al., 2003). Ketzel et al. (2004) reported that total PNCs in Copenhagen increased by up to 5–10 times within a few hours, for clean air and high solar radiation conditions. They also observed particle growth rates in the 1–6 nm h⁻¹ range, which were similar to those found at another European suburban background location in Prague, where the average value of particle growth rate was found to be ~5.4 nm h⁻¹ (Rimnácová et al., 2011). Reche et al. (2011) recently demonstrated nucleation as a significant process influencing PNC levels throughout southern Europe. They observed that the occurrence of SO₂ peaks may also contribute to the occurrence of midday nucleation bursts in specific industrial or shipping-influenced areas, although at several central European sites similar levels of SO₂ were recorded without leading to nucleation episodes. This allowed the authors to conclude that the nucleation variability in different European urban environments was not influenced to the same degree by the same emission sources and atmospheric processes. Studies have also found nucleation to be a dominant process in North America. Stanier et al. (2004) found regional-scale new particle formation of UFPs to occur on 30% of the study days in Pittsburgh. The intensity of events differed, sometimes leading to an increase in PNC between 50,000 cm⁻³ up to 150,000 cm⁻³.

As explained above, regional nucleation processes are favoured by high gas-phase precursor concentrations (especially sulphur dioxide, but also oxidisable organic compounds), but disfavoured by a high pre-existing particle surface area that provides a condensation sink for

vapours of low volatility. Since the atmospheres of less developed countries typically show both attributes, it is difficult to predict the likelihood of nucleation frequently influencing PNC. However, the European study of Reche et al. (2011) shows the importance of climate, with southern European cities showing a much greater influence of nucleation processes than those in northern Europe. This suggests that the high sunshine levels in many cities in the developing world may be a key influence in promoting atmospheric oxidation processes to the extent that they overwhelm the condensation sink hence leading to frequent observations of nucleation in many such cities despite high pre-existing particle concentrations.

It is apparent that the condensation sink may be too great in many of the developing Asian cities, due to high particle loadings, to allow much particle formation by these mechanisms, but it is certainly an important mechanism. However, in-depth assessments of long-term continuous observations are needed in order to clarify and quantify the role of regional new particle formation events in affecting PNCs in urban areas. One of the difficulties in performing such quantification is that both “nucleation particles in ambient air” and “nucleation particles in the exhaust plume” tend to exhibit similar size distributions. Most of the studies performed to date have focused on identifying the events with “strong new particle formation rates” (so-called “nucleation events”) in rural areas or downwind of urban emissions (Hamed et al., 2007; Qian et al., 2007; Stanier et al., 2004). However, attempts to quantify day-to-day contributions of the “new particle formation processes” to total urban PNC have not been made.

6. Unresolved challenges

With recent advances in instrumentation and measurement technology, it is now possible to measure atmospheric particles down to 1 nm (Kulmala et al., 2013). The first PN emission inventory in Europe has recently become available, indicating emission levels in European countries (Kumala et al., 2011; Paasonen et al., 2013). The scientific understanding of their measurement, atmospheric formation, dispersion, physical and chemical transformation, environmental and health impacts has improved appreciably over the past decade. However, numerous unresolved technical and practical constraints remain for enforcing any nationwide regulatory framework. For instance, safe levels of PN exposure and the biological mechanisms through which they affect human health are still contentious (HEI, 2013; Peters et al., 1997). A number of instruments are available for these measurements, but due to: (i) their lack of robustness for long-term unattended operation, (ii) high cost for field deployment in sufficient numbers, and (iii) the limited reproducibility of data by different instruments, standard methods to measure airborne UFPs are yet needed to allow scientists to reach a clear consensus (Kumar et al., 2011c). Nucleation mode particles below 30 nm bring additional uncertainty, since these are semi-volatile and form through gas-to-particle conversion (Kulmala et al., 2004), but can contribute up to 40% of total particles by number along busy roadsides (Kumar et al., 2011c). The nucleation rate (i.e. the intensity of gas-to-particle conversion), which partly controls the nucleation mode concentration, is also dependent on temperature. Therefore, similar emissions of precursor vapours for nucleation (e.g. SO₂, some VOCs) may lead to very different concentrations of UFP in different atmospheric conditions. The volatile nature of the nucleation mode particles raises issues in relation to their reliable measurement and remarkable spatio-temporal variability after emission into the atmospheric environment (Dall'Osto et al., 2011).

Densely and unevenly built-up city environments, together with the rapid transformation processes affecting UFPs, further complicate this issue. For instance, the exchange of air inside the urban canopy with the cleaner air above is greatly influenced by the built environment of an individual city (Buccolieri et al., 2010). This contributes to both temporal variability and spatial variability in the concentration of UFPs, which exceeds an order of magnitude within metres of the source,

and reduces by several orders of magnitude within seconds, especially immediately after emission (Carpentieri and Kumar, 2011; Kumar et al., 2009b; Morawska et al., 2009). Dispersion of UFP emissions away from the source depends on synoptic wind conditions, atmospheric stability and the density of the built environment. These, together with the quantity of UFPs emitted at a particular location, play a major role in dictating their concentrations and human exposure. In the EU, PN emissions of solid particles (excluding particles formed in nucleation process) from diesel vehicles are currently regulated through the Euro-5 and Euro-6 vehicle standards (EC, 2008). To meet these standards, the EU undertook widespread deployment of diesel particulate filters (DPFs), in addition to fuel quality improvements in the form of reduced sulphur content. Together, these led to a decrease in UFP levels in European and North American environments, with observational studies in London (Jones et al., 2012), Copenhagen (Wählén, 2009), Toronto (Sabaliauskas et al., 2012) and Los Angeles (Choi et al., 2013; Quiros et al., 2013) supporting this conclusion. However, the case of growing cities elsewhere is different and such actions have yet to find a place in local regulatory planning. Therefore, the concentration levels in Asian cities, for example, are many times higher than those observed in Europe (Fig. 2), as are fuel sulphur concentrations.

Another interesting challenge is emerging due to the widespread deployment of after-treatment systems, such as DPFs, which reduce the soot emissions efficiently. This leads to a lower solid particle surface area available for condensation of gaseous compounds and triggers particle formation through the nucleation. Consequently, DPFs can lead to an increase in PNC, especially when sulphur content is high (Liu et al., 2013), unless the sulphur content in fuels is brought to a minimum level to restrict sulphur-driven nucleation processes. Whilst the sulphur content in fuel varies from below 50 to over 2000 ppm in Asian cities such as in China (Liu et al., 2013) and India (Kumar et al., 2013a), this concentration is much lower in European cities such as in London (<10 ppm) (Jones et al., 2012). This clearly indicates that reductions in sulphur content are key for further reducing PN formation and human exposure by improving air quality in city environments.

An additional option available to urban governments is controlling exposure to UFPs by separating urban residents from high-emission sources. Kumar et al. (2011a) estimated that in 2010 heavy-duty vehicles in Delhi contributed ~4% of vehicle kilometres travelled, but ~65% of PN emissions. Using simple box models, the authors estimated PNCs for roadside areas, ~327,000 cm⁻³, and the city as a whole, ~33,000 cm⁻³. The factor of 10 dilution points to large spatial variations in cities and suggests that efforts could be made to quantify the variation of PNC with distance from road sources and based on this, to separate heavy-duty vehicles from areas where people spend a significant amount of time.

Altogether, these unresolved challenges pose difficulties to national governments worldwide to assess what they can do to improve the urban environment, including fuel substitution, tighter emission standards, location of housing relative to major roads, or the location and the type of industry.

7. The future directions

European cities are currently well placed to make reliable estimates of PN emissions, because of richly available information on vehicle specific emission factors and vehicle fleet inventories (see Section 2). The major challenge remains for Asian cities, which have demonstrably high PN emissions but lack data on UFP emissions. Developing city-specific emission factors for Asian vehicle fleets is a necessary step to construct accurate emission inventories and assess health impacts on the residents of emerging cities.

The UN-ECE Particle Measurement Programme helped establish new systems and protocols for assessing PN emissions from vehicles in Europe (EC, 2008). As a result, mass and number emissions from modern diesel and gasoline direct ignition engines have greatly

reduced. However, the effects of alternative fuels, such as biofuels, on UFP emissions are yet to be understood. Evidence suggests that particulate mass emissions from biodiesel fuelled vehicles have substantially decreased in recent years, but this is likely at the expense of an up to two orders of magnitude increase in PN emissions (Kumar et al., 2010b), mainly due to the reduced number of solid particles in emissions, leading to a considerable increase in nucleation mode particles (Fontaras et al., 2009). The European Directive 2003/30/EC set a legal aim of replacing 20% of conventional fuel in road transportation with alternative fuel by 2020. The impact of this trade-off between increasing PNC emissions and the use of biofuels is hard to predict, given that the toxicity of UFPs produced by bio-fuelled vehicles is unknown.

In many Asian, or other nanoparticle-rich environments, PNCs may increase further as a result of continuously increasing traffic volumes. Vehicle technologies, such as the use of DPFs and reducing the sulphur content in fuel to European levels (<10 ppm) are two factors that can make a difference in the form of decreased human exposure to high UFP levels. These vehicle technologies, originally designed to decrease mass emissions, also seem to be effective in limiting the emission of primary nano-sized particles, and a simultaneous reduction in the sulphur content of fuels will prevent an increase in secondary, nucleation mode particles (Paasonen et al., 2013). As more new vehicles come on the market, the impact of DPFs will start to be seen more clearly, at least in the developed world.

Understanding new particle formation in city environments is important to accurately estimate PNC and related exposure. Recently, new instruments have been developed to count these particles down to 1 nm, but their detailed physical and chemical composition still remains unknown (Kulmala et al., 2013). In practice, new instruments such as the particle size magnifier, PSM (Vanhanen et al., 2011), neutral cluster and air ion spectrometer, (N)AIS (Mirme et al., 2007) and chemical ionization with the atmospheric pressure interface time-of-flight mass spectrometer, (CI)-API-ToF (Ehn et al., 2010) should be used simultaneously to obtain sufficient information on new particle formation and subsequent growth. Presently we are missing long-term continuous comprehensive observations in megacities. In addition, the current theories related to nucleation mechanisms have been developed based on observations in clean environments. Whether these can explain the “nanoparticle formation events” that occur in the polluted atmosphere, with abundant potential precursors and strong oxidation capacity (like Beijing), is a key issue to be addressed in further studies.

UFPs in the atmosphere derive from a considerable number of sources, even though road traffic emissions are frequently dominant. To date, there has been relatively little work devoted to disaggregation of the size distribution into source-related components. This can be achieved by the application of multivariate statistical techniques to large datasets of particle number size distribution. Andersen et al. (2008) were able to fit measured Scanning Mobility Particle Sizer (SMPS) size distributions by the sum of four log-normal modes with median diameters at 12, 23, 57 and 212 nm. The data analysed in terms of particle number in each mode were subsequently used in a time series epidemiological study. Harrison et al. (2011) combined SMPS particle size distributions with those measured simultaneously with an Aerodynamic Particle Sizer to create continuous distributions from 15 nm to 10 µm diameter. Application of Positive Matrix Factorization revealed ten source-related components of size distributions measured on Marylebone Road, London. Of these, four were related to traffic on Marylebone Road, arising from semi-volatile engine emissions, solid particle engine emissions, brake wear and resuspension, accounting respectively for 27, 38, 2 and 5% of the total PNC. Such techniques are a critical element of the source apportionment of atmospheric UFP and will find further application in epidemiological studies designed to identify those components of the emissions having the greatest effect upon human health. Such research is difficult to conduct beyond the roadside environment, however, due to the evolution of size

distributions by both particle evaporation (Dall'Osto et al., 2011) and growth (Beddows et al., 2013), and the influence of regional atmospheric nucleation processes leading to new particle formation.

In terms of calculating the number of deaths (total mortality) as a result of exposure to airborne UFPs (as described by particle number count), it is necessary to use an exposure–response relationship that relates a change in particle number count to the number of associated deaths. Whilst these are abundant in the literature for the effects of exposure to PM₁₀ concentrations, they are almost non-existent for particle number. The very few relationships known include that reported by Atkinson et al. (2010) from a time series study conducted in London, by Stölzel et al. (2007) for Erfurt, Germany, and more recently by Meng et al. (2013) for Shenyang, China. These are missing for a broad range of city environments but are vitally important, as they can differ notably for different social conditions (e.g. food, sanitation, social conditions, hospital care) in both developed and developing cities, and can result in huge uncertainty if city/country specific exposure–response functions are not used for mortality estimates.

The scientific and policy focus on traffic-derived UFPs, resulting in tighter PN emission standards in many parts of the world, will lead to a more pronounced contribution from non-vehicle sources (Kumar et al., 2013c). Recent data on PN emissions in Europe support these observations (Fig. S3). For instance, this effect has already been seen in Romania and Finland, where the traffic contribution to PN is only ~36 and 47% of total PN emissions, and domestic combustion and non-road traffic (e.g. ships) contribute ~22 and 26%, respectively. Although it is currently unknown, the contribution from non-vehicle sources (e.g. light petroleum gas, wood and biomass burning for cooking, unregulated small-scale industries, power plants and exhaust-emissions from non-road construction machinery) cannot be neglected when estimating the total PNC in Asian city environments (Monkkonen et al., 2005a; Monkkonen et al., 2005b; Pathak et al., 2012).

Future studies should also consider more extensive personal monitoring of the UFPs in order to reliably estimate the overall daily exposure of individuals whilst in different microenvironments of a city environment. Because of the distinct personal lifestyles of city dwellers, the exposure to UFPs could be entirely different in diverse microenvironments for different age groups, say, of school children compared with working adults. For example, a recent Brisbane-based study found that the proportion of total daily alveolar doses of UFPs for home, school, commuting, and other environments were 55.3, 35.3, 4.5, and 5.0%, respectively (Mazaheri et al., 2014). The outdoor exposure in this study represents the sum of nearly half of the schooling and other activities, which is significantly lower than that (~46% of total exposure) found for adults in outdoor city environments (Fruin et al., 2008). More personal monitoring studies, now nearly non-existent for Asian city environments, are therefore needed so that a database for the variety of diverse city environments can be developed. Such a database can be vital for: (i) apportioning the UFP exposure of different age groups in various microenvironments, (ii) providing necessary information for designing preventative actions to control emissions at the source, receptor and within microenvironments by improving their ventilation and air exchange rates, and (iii) developing an essential tool to identify health risks, set and review air quality standards and evaluate the effectiveness of policy interventions (Buonanno et al., 2012a).

We have not discussed here the emergence of a new risk, the presence of engineered nanomaterials in urban environments (Kumar et al., 2010c), which are suspected to enter urban air in unknown quantities after separating from their original products (Lowry et al., 2012). These are basically the same as UFPs in terms of their size, but are known to deposit in target organs, penetrate cell membranes and lodge in mitochondria, as well as trigger injurious responses (Nel et al., 2006). A thorough health risk assessment of all nanomaterial-integrated products is likely to take decades and this will increase further with the development of the next generation of nanoparticles

(Choi et al., 2009; Kumar et al., 2010c). Their unique, highly reactive physicochemical characteristics are of particular concern in terms of human health (Xia et al., 2009), but very little is currently known about their concentration levels in the European environment, and even lesser for Asian environments (Kumar et al., 2012). A proactive research approach is required to fully reveal their injurious effects and at the same time, targeted efforts are needed to quantify their ambient concentrations by developing instruments that are able to distinguish them from the UFPs produced by other sources, such as traffic.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.envint.2014.01.013>.

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