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Pant, P.; Harrison, R.M.

DOI:
10.1016/j.atmosenv.2013.04.028

Document Version
Early version, also known as pre-print

Citation for published version (Harvard):
Pant, P & Harrison, RM 2013, 'Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements: A review', *Atmospheric Environment*, vol. 77, pp. 78-97. https://doi.org/10.1016/j.atmosenv.2013.04.028

Estimation of the Contribution of Road Traffic Emissions to Particulate Matter Concentrations from Field Measurements: A Review

Pallavi Pant and Roy M. Harrison[*1]

Division of Environmental Health & Risk Management,
School of Geography, Earth & Environmental Sciences
University of Birmingham, Edgbaston
Birmingham B15 2TT
United Kingdom

[*] Corresponding Author: Tel: 00 44 121 414 3494; Fax: 00 44 121 414 3709;
Email: r.m.harrison@bham.ac.uk

[1] Also at: Centre of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah (21589) Saudi Arabia

ABSTRACT

Road traffic is one of the main sources of particulate matter in the atmosphere. Despite its importance, there are significant challenges in quantitative evaluation of its contribution to airborne concentrations. This article first reviews the nature of the particle emissions from road vehicles including both exhaust and non-exhaust (abrasion and resuspension sources). It then briefly reviews the various methods available for quantification of the road traffic contribution. This includes tunnel/roadway measurements, twin site studies, use of vehicle-specific tracers and other methods. Finally, the application of receptor modelling methods is briefly described. Based on the review, it can be concluded that while traffic emissions continue to contribute substantially to primary PM emissions in urban areas, quantitative knowledge of the contribution, especially of non-exhaust emissions to PM concentrations remain inadquate.

Keywords: Particulate matter; road traffic; exhaust emissions; non-exhaust emissions

1. INTRODUCTION

Emissions due to road traffic are known to make a large contribution to total particulate matter (PM) concentrations in urban areas and exposure to PM from vehicular emissions has been demonstrated to have detrimental impacts on human health (Mauderly, 1994; Buckeridge et al., 2002; Fan et al., 2006; HEI, 2010; Masiol et al., 2012; Rissler et al., 2012). An improved understanding of the characteristics of emissions from different traffic-related sources is therefore vital for conducting source apportionment and health effects studies (Schauer et al., 2006). PM emissions from road vehicles include emissions from the tailpipe (exhaust emissions) and emissions due to wear and tear of vehicle parts such as brake, tyre and clutch and re-suspension of dust (non-exhaust emissions). Non-exhaust emissions contribute mainly to the coarse mode of PM ($PM_{2.5-10}$) while exhaust emissions contribute predominantly to fine PM (aerodynamic diameter < 2.5 μm) (Abu-Allaban et al., 2003; Tervahattu et al., 2006; Thorpe et al., 2007; Kam et al., 2012) but this is a far from clear distinction. A number of different properties of traffic emissions are studied including physical shape and structure, particle size distributions, chemical composition, and temporal and spatial variation (Watson and Chow, 2007). Most of the research as well as policy action in the last few decades has largely focused on exhaust emissions, and stringent regulations and technological upgrades have resulted in a decline of the percentage contribution of vehicle tailpipe emissions to total ambient PM concentrations (Allen et al., 2006; Thorpe and Harrison, 2008; Mathissen et al., 2011). As a result, the contribution of non-exhaust PM is becoming more important although detailed information on non-exhaust PM emissions is relatively scarce (Kuhlbusch et al., 2009; Amato et al., 2012; Harrison et al., 2011; 2012b; Denier van der Gon et al., 2013). It has been shown that even with zero tail-pipe emissions, traffic will continue to contribute to fine and ultrafine particles through non-exhaust emissions (Dahl et al., 2006; Kumar et al., 2013) and it is estimated that nearly 90% of the total emissions from road traffic will come from non-exhaust sources by the end of the decade (Rexeis and Hausberger, 2009).

In order to understand the contribution of road traffic emissions to the environment, it is important to understand various aspects including sources, chemical characteristics and quantitative contributions. In the last three decades, a significant amount of research has been conducted on characterization and analysis of exhaust emissions and as a result, there is a very strong body of knowledge in the field. Non-exhaust emissions, however, are becoming more important now, and further research is anticipated in this field in the coming years. This review aims to reconcile the various studies that have been reported around this issue, and tease out some common characteristics, and highlight areas with future research potential.

This paper reviews the current information on primary road traffic emissions including exhaust and non-exhaust emissions. Key characteristics of the major sources are discussed. Various methodologies used for measurement and analysis of such emissions are also discussed with a brief discussion on the merits and shortcomings of each method. In the final section, various source apportionment techniques useful for the estimation of traffic source contributions are discussed together with a summary of some of the reported studies. A number of in-depth review articles have been published in the last few years on various issues related to road traffic emissions on topics such as properties and characteristics of non-exhaust emissions (Thorpe and Harrison, 2008), toxicological impacts of tyre wear emissions (Wik and Dave, 2009), chemical composition of brake pads (Chan and Stachowiak, 2004), development of emission factors (Franco et al., 2013), nanoparticle emissions from non-exhaust sources (Kumar et. al., 2013). However, each of these reviews focuses on one or a few specific aspects of road traffic emissions. This review builds on the information provided in those review articles, and repetition is avoided. In summary, the key questions that we hope to address through this review article are as follows:

1. What are the key characteristics/sources/contributions of road traffic emissions? [What and how much do we know about each of the road traffic emission sources?]
2. How can such emissions be characterized? [Various methods, pros and cons of the methods]

3. What have we learnt so far?

4. What are the key issues that are not resolved yet?

The literature search for this review was primarily performed using Web of Knowledge, Science Direct and Google Scholar using various combinations of keywords such as traffic emissions, non-exhaust/exhaust emissions, resuspension, road dust, traffic PM etc. However, the authors acknowledge that not all articles published on the topic have been included in the review.

2. NON-EXHAUST EMISSIONS

Non-exhaust PM comprises the various emissions that do not derive from the tailpipe of a vehicle including particles generated due to brake and tyre wear, road surface abrasion, wear and tear/corrosion of other vehicle components such as the clutch, and re-suspension of road surface dusts (Table 1). Key reasons for needing to understand non-exhaust emissions include their inherent toxicity including their tendency to act as carriers of heavy metals and carcinogenic components (Adachi and Tainosho, 2004; Hjortenkrans et al., 2007; Johansson et al., 2009; Amato et al., 2011a) and their contribution to exceedences of air quality guidelines and standards (Amato et al., 2011a; Denier van der Gon, 2013).

Harrison et al. (2012b) showed that the size distributions of trace metals were indicative of particle sources. Keuken et al. (2010) concluded that re-suspension of accumulated PM and road wear related particles are the primary contributors to non-exhaust emissions, and tyre wear and brake wear contribute to the fine and coarse fraction respectively. However, Narvaez et al. (2008) reported that although a majority of abrasion particles are in the coarse fraction, abrasion can contribute significantly to the fine fraction of PM. In Taiwan, EFs for elements such as Ba and Se were found to be higher in the $PM_{2.5-10}$ fraction indicating mechanical friction and brake wear as potential sources (Chiang and Huang, 2009). Road traffic was found to be a key contributor to fine/ultrafine and nano mode particles of Ba, Zn and Pb (Lin et al., 2005) and in Barcelona (Spain),

62-96% of elements including Cu, Sb, Ba, Mn and Zn were found to be present in the PM_{1-10} fraction (Perez et al., 2010). A detailed review of the nanoparticle emissions from non-exhaust sources including traffic and non-traffic sources is presented in Kumar et al. (2013).

Non-exhaust emissions are typically characterized by trace metals (e.g. Cu, Zn, Ba, Sb, Mn) although organic markers (e.g. polycyclic aromatic hydrocarbons (PAHs), n-alkanes) have been used in some cases (Wahlin et al., 2006; Amato et al., 2009; El Haddad et al., 2009; Gietl et al., 2010; Oliviera et al., 2011; Kwon and Castaldi, 2012). A list of key tracers used for source characterization of non-exhaust emissions is presented in Table 2. However, emissions of trace metal markers are reported to vary with the fleet composition, with higher emissions reported for some of the elements for heavy duty vehicles (HDVs) (Grieshop et al., 2006; Mancilla and Mendoza, 2012). In addition, the profile of trace metal concentrations in non-exhaust particulate matter is unique for every region and varies based on parameters such as traffic volume and pattern, vehicle fleet characteristics, driving and traffic patterns and climate and geology of the region (Omstedt et al., 2005; Amato et al., 2011a,b; Duong and Lee, 2011). Another important aspect is the variability of tyre and brake composition depending on the manufacturer which makes it very difficult to ascertain fleet-wide composition other than from environmental measurements (Canepari et al., 2008; Gietl et al., 2010; Denier van der Gon et al., 2013).

Several factors are reported to affect non-exhaust emissions including increase in vehicle speed (Chen et al., 2006; Gustafsson et al., 2008; Hussein et al., 2008; Mathissen et al., 2011) although Keuken et al. (2010) reported a non-linear relationship between traffic volume and non-exhaust emissions. Metal emissions due to road dust show a low correlation to metal emissions due to abrasion/brake wear and combustion (Johansson et al., 2009). One of the major problems in analysis of non-exhaust PM using field data has been the difficulty in distinguishing between wear and tear emissions and road dust since the chemical composition is often very similar (Bukowiecki et al., 2010). This may, in any case, not be a clear distinction as wear emissions may deposit to the

129 road surface, only to be re-suspended subsequently. Gehrig et al. (2010) used mobile load
130 simulators to compare the contributions of road abrasion and resuspension emissions, and
131 concluded that particle emissions due to abrasion are a function of the state of the pavement
132 whereas resuspension is normally higher, and increases with higher dust loads on the road. Thorpe
133 and Harrison (2008) present a more elaborate account of sources and properties of non-exhaust
134 emissions.

135 Mixed results have been obtained for the impact of street washing activities on PM and while
136 Amato et al. (2009) observed a reduction, albeit statistically non-significant, in the concentration of
137 trace metals known to be contributed by non-exhaust traffic sources in Spain, Keuken et al. (2010)
138 reported a lack of significant reduction in the non-exhaust emissions after street sweeping or
139 washing in the Netherlands.

140 **2.1 Tyre Wear**

141 Tyre wear particles are generated either by shear forces between the tyre tread and the road surface
142 and are predominantly coarse ($PM_{2.5-10}$) or by volatilization (Rogge et al., 1993; Kupiainen et al.,
143 2005; Allen et al., 2006; Thorpe and Harrison, 2008; Aatmeeyata and Sharma, 2010). Tyre wear
144 particles are reported to be generally elongated with rough surfaces based on microscopic analysis
145 (Gunawardana et al., 2011). Tyre tread, a source of airborne particles, contains natural rubber
146 copolymers such as styrene-butadiene rubber and polyisoprene rubber, and zinc (Zn) is added to
147 tyre tread as zinc oxide and organozinc compounds to facilitate the vulcanization process. Passenger
148 car tyres in EU are known to contain nearly 1% zinc oxide, and rubber, metals and carbon black
149 make up typically 47%, 16.5% and 21.5% respectively (Milani et al., 2004). The key tracer
150 components of tyre wear include n-alkanes, n-alkanoic acids, PAHs, benzothiazoles and trace
151 metals (Rogge et al., 1993; Reddy et al., 1997; Camatani et al., 2001; Adachi and Tainosho, 2004;
152 Boulter et al., 2005; Aatmeeyata and Sharma, 2010). Zn is reported to be nearly 1% by weight in
153 rubber tyres (Councell et al., 2004) and tyre wear has been reported to be a significant source of Zn

(Adachi and Tainosho, 2004; Hjortenkrans et al., 2007; Ondracek et al., 2011). The concentration of Zn was found to be approximately 15 times higher in tyres compared to brakes while concentrations of other heavy metals such as copper (Cu) and barium (Ba) were higher for brake materials in Massachusetts (USA) (Apaegyei et al., 2011). Differences have also been observed between tyre tread composition (primarily Zn) and tyre wear (Al, Si, Ca, Fe, Zn, Ti) (Adachi and Tainosho, 2004). However, the spatial and temporal trend for Zn was found to be different from other roadside tracers in New York (USA), and a very small portion of airborne Zn concentration was attributed to tyre wear (Peltier et al., 2011). However, Zn is emitted from brake wear, motor oil and other sources and cannot be used as the only tracer for tyre wear (Lough et al., 2005). Benzothiazoles are also used as markers for tyre wear, particularly benzothiazole (BT), 2-hydroxy benzothiazole (HOBT), 2-(4-morpholinyl)benzothiazole (24MoBT) and N-cyclohexyl-2-benzothiazolamine (NCBA) (Kumata et al., 2002; Allen et al., 2006; Wik and Dave, 2009). Among the PAHs, pyrene, benzo(ghi)perylene, fluoranthene and phenanthrene are known to be emitted from tyres (Boonyatumanond et al., 2007; Kwon and Castaldi, 2012). Aatmeeyata and Sharma (2010) reported that small cars tested in India emitted 378 ng of total PAH/tyre/km while in the USA, 200 µg/g of PAHs were found in tyre wear with pyrene, fluoranthene and phenanthrene being present in the highest concentrations (Rogge et al., 1993). Tyre tread wear has also been found to contain an average of 0.53 µg/g of dibenzopyrenes, indicating that tyre dust may be significant source of dibenzopyrenes in the environment (Sadiktis et al., 2012).

Average tread wear for tyres is reported to be between 0.006-0.009 g/km based on the road, tyre and vehicle conditions (Rogge et al., 1993) and tyres can lose up to 10% of their mass during their lifetime (Milani et al., 2004). Speed has been reported as an influential parameter for particle mass and number concentrations for tyre wear emissions (Gustafsson et al., 2008; Mathissen et al., 2011). Other important parameters for tyre tread wear include road surface type, and tyre and driving conditions (Allen et al., 2006; Gustafsson et al., 2008). For example, asphalt surfaces have been

reported to cause less tyre wear than concrete pavements and in Arizona, USA, the emission rate for tyre wear was found to be 1.4-2 times lower for asphalt pavement compared to concrete pavement (Allen et al., 2006). The type of tyre also impacts the magnitude of tyre wear emissions and studded tyres are known to cause more emissions compared with summer and friction tyres (Kupiainen et al., 2005; Gustafsson et al., 2008; Hussein et al., 2008; Schaap et al., 2009). Unimodal (70-90 nm) and bimodal (< 10 and 30-60 nm) peaks in the nano size range have been reported for tyre particles by number under low and high speed conditions respectively (Mathissen et al., 2011).

2.2 Brake Wear

Brake wear, including abrasion of brake lining material and brake discs, caused by grinding of brake pad constituents (coarse range particles) or volatilization and condensation of brake pad materials (fine range particles), is known to release PM directly into the atmosphere and to contribute to the trace metal concentration in airborne PM, particularly less than 10 µm (Garg et al., 2000; Blau and Meyer, 2003; Ingo et al., 2004; Wahlin et al., 2006; Gietl et al., 2010; Varrica et al., 2012). Key components of brake pads include fillers, frictional additives, reinforcing fibres and binder (Chan and Stachowiak, 2004) and the key chemical species used include sulphides of metal, abrasives (e.g. silica), barium silicate/sulphate (particularly in brake linings) and other metallic particles (as filler material), carbon fibres and lubricant (e.g. graphite) (Ingo et al., 2004; Dongarra et al., 2009). Chan and Stachowiak (2004) present a detailed review of the chemical constituents of brake pads. Garg et al. (2000) observed an average mass median diameter (median particle diameter based on mass) of 1.49 µm for brake wear particles using tests on a brake dynamometer whereas Sanders et al. (2003) reported a mass median diameter of 6 µm for brake debris generated during urban driving conditions. Higher brake wear related emissions have been reported during rush hour . A higher number of particles are released in the braking phase as compared to the acceleration phase (Hussein et al., 2008; Mathissen et al., 2011) and various studies have corroborated this observations including Greishop et al. (2006) who reported a higher particle

number concentration during rush hour where the traffic often operates in the stop-and-go mode and Abu-Allaban et al. (2003) who observed higher contribution of brake wear at freeway exit sites compared to other types of roadside sites (Abu-Allaban et al., 2003).

Passenger cars have been estimated to emit nearly 44 g/car/year brake dust (Iijima et al., 2007). Sanders et al. (2003) reported high concentrations of Fe, Ba and Cu in brake lining wear using dynamometer and track tests whereas Adachi and Tainosho (2004) reported Fe to be the most abundant metal in brake dust along with other metals such as Ba, Cu, Sb, Zr and Zn, and Hjortenkrans et al. (2007) reported Cu, Zn and Sb to be present in brake wear emissions. Varrica et al. (2012) undertook a detailed study on Sb in brake dust and reported the most commonly released forms of Sb due to brake abrasion to be Sb (III) and Sb (V). However, brake pads are often found to have different composition based on the brand, particularly with respect to Cu and Sb (Hjortenkrans et al., 2007; Canepari et al., 2008). Polyalkylene glycol ethers are also reported to be present in brake wear particles with small concentrations of n-alkanes and n-alkanoic acids (Rogge et al., 1993).

Iijima et al. (2007) reported a unimodal number-based distribution for brake abrasion dust with the mode at 1-2 µm while the mass-based distribution had a peak at 3-6 µm. EFs for brake wear have been reported by several studies including Garg et al. (2000) who reported a brake wear EF of 3-9 mg/km for light duty gasoline vehicles (LDGV) and Abu-Allaban et al. (2003) who reported an EF of 0-80 mg/km. Wahlin et al. (2006), on the other hand, reported a Cu emission factor of 0.7± 0.2 mg/km/vehicle in Copenhagen.

Sternbeck et al. (2002) proposed the ratio of Cu:Sb (4.6 ± 2.3) as characteristic of brake wear particles and Table 3 lists the Cu: Sb ratios as reported in the literature. Differences in the ratios can be attributed to the difference in the brake pad composition, and contributions of metals from other sources. In some cases, concentrations in ambient air can also vary due to the site characteristics.

2.3 Road Dust and Road Surface Wear

Road dust, of which crustal dust is a key component, consists of primarily coarse-sized particles derived from different sources such as traffic, industrial emissions, mineralogical dust etc. (Kupianinen et al., 2005; Tanner et al., 2008). Composition of road dust shows spatial as well as temporal variation and it is often difficult to classify dust into crustal/resuspended/direct emission etc. In Monterrey (Mexico), re-suspended dust was found to be contributing nearly 20-25% to the $PM_{2.5}$ EF (Mancilla and Mendoza, 2012). The amount of re-suspended road dust particles depends on a number of factors including vehicle movement (particularly traffic speed), street maintenance, season and associated meteorological parameters and speed of traffic (Etyemezian et al., 2003; Gertler et al., 2006; Thorpe et al., 2007; Bhaskar and Sharma, 2008; Kaunhaniemi et al., 2011; Laidlaw et al., 2012; Majumdar et al., 2012). Nicholson (1988) presents a detailed review of the mechanisms of resuspension of road dust. Thorpe et al. (2007) reported a strong association between heavy duty traffic and re-suspension in the UK with wind speed not found to be a strong influence. In addition, precipitation was found to have no influence on the amount of re-suspension. In somewhat related research, street-washing has been reported to be ineffective for PM control based on experiments in Spain (Karanasiou et al., 2012). However, in Sweden, road wetness was found to be an important factor in the amount of re-suspension (Omstedt et al., 2005), but this appears related to road sanding and the use of studded tyres in Sweden. It is also important to note that the residence time of PM_{10} on paved roads (travel lanes) has been estimated as a few hours (Etyemezian et al., 2003). In the case of studded tyres, interaction between tyres and the road pavement can generate particles less than 0.1 μm (Gustafsson et al., 2008) and such tyres have been shown to increase PM_{10} emissions by a factor of 1.5 (Tervahattu et al., 2006). The type of pavement is another important factor for road dust emissions. Granite pavements have been reported to emit more PM compared to quartzite pavements (Kupiainen et al., 2005; Gustafsson et al., 2009). Tervahattu et al. (2006) also explained the higher PM_{10} concentration attributed to use of anti-skid

aggregate using the *sandpaper effect*, where the aggregates used to prevent skidding generate PM_{10} particles and lead to further particle emissions from the pavement.

Pristane, phytane, hopanes, steranes in addition to unresolved complex mixture (UCM) were reported in road dust particles which were found to be enriched in biogenic component by Omar et al. (2007). High concentrations of phenanthrene, fluoranthene and pyrene along with high molecular weight (4-7 ring) PAHs were reported in road dust samples in Taiwan (Fang et al., 2004), China (Liu et al., 2007; Han et al., 2009) and Egypt (Hassanien and Abdel-Latif, 2008) respectively. The spatial distribution of PAHs was reported to be affected by wind direction Liu et al. (2007) and total organic carbon content by Liu et al. (2007) and Han et al. (2009). Mono- (α- and β-glucose) and disaccharides (sucrose and mycose) were used as markers for road dust re-suspension for source apportionment by Omar et al. (2007).

Han et al. (2007) analysed suspended dust in Beijing and reported concentrations of elements such as Ca, S, Cu, Zn, Ni, Pb, and Cd to be much higher than the crustal abundances. Cu, Zn, Ni, and Pb were attributed to traffic emissions together with coal burning, and ions such as Ca^{2+}, SO_4^{2-}, Cl^-, NO_3^-, K^+, and Na^+ were also found to be present in re-suspended dust. Luo et al. (2011) observed that the concentrations of trace metals contributed by vehicle exhaust and tyre abrasion, i.e. Pb, Zn and Cu have been reported to be much higher in roadside samples than the concentrations of these elements in background soils in China. Wei and Yang (2010) analysed urban soil, urban road dust and agricultural soil in Chinese cities and found relatively higher concentrations of Cd, Cu, Pb and Zn in urban soil samples and Cu, Cr, Pb, Zn, Ni and Cd in urban road dusts. The high concentrations of metals were attributed to anthropogenic sources - road traffic and industrial emissions. Higher concentration levels for several trace elements were reported for roadside dust compared to dust collected at a distance of 100 m in Bosnia, and particularly high concentrations were reported for Hg and Co and were attributed to vehicle exhaust and brake pads respectively. Elements such as platinum, palladium and rhodium have also been reported to be present in road

dust and are attributed to the catalytic converters (Prichard and Fisher, 2012). Duong and Lee (2011) analyzed heavy metal contamination in road dust in high traffic areas in Korea and concluded that the concentrations of heavy metals are much higher in high traffic areas in relation to a background site. Their analysis also confirmed a high degree of correlation between the concentration of heavy metals at a specific location and the traffic throughput in the area. Martuzevicius et al. (2011) analysed street dust and reported that it can be a major source for particle-bound PAHs together with other PM. Han et al. (2009) reported a high correlation between PAHs and OC in road dust, and traffic emissions were identified as a major contributor to PAHs in road dust using diagnostic ratio analysis. Mathematical models have been proposed for estimation of PM emissions due to road dust although they do not include emissions due to wear of brake/tyre materials (Omstedt et al., 2005; Ketzel et al., 2007; Berger and Denby, 2011; Kaunhaniemi et al., 2011).

Aatmeeyata et al. (2009) reported a bimodal number and mass distribution (0.3 μm and 4-5 μm) for PM_{10} generated due to surface (concrete pavement) and tyre wear . Duong and Lee (2011) reported a multi-modal distribution for road dust particles (based on weight) collected from the roadside with a majority of the particles between 180-850 μm in Ulsan, Republic of Korea. Chen et al. (2006) reported a bimodal (5-10 μm and > 30 μm) mass size distribution for road dust particles in Beijing (China).

3. EXHAUST EMISSIONS

Motor vehicles are an important source for carbonaceous aerosols particularly for the particles in the fine size range (aerodynamic diameter < 2.5 μm) (El Haddad et al., 2009; Kam et al., 2012; Keuken et al., 2012). PM emissions from vehicles depend on the engine type and age and maintenance and contain carbon in the form of OC and EC with smaller amounts of trace metals and ions (Brook et al., 2007; Robert et al., 2007a; Fulper et al., 2010; Peltier et al., 2011). For example, Robert et al. (2007a,b) reported that that emission rates for ultrafine and fine particles for heavy-duty diesel vehicles (HDDVs) are one order of magnitude higher than LDGVs. Similarly,

307 Chen et al. (2013) reported 4-times higher diesel EFs compared to gasoline EFs in a tunnel
308 environment. Higher emissions from the high-emitting (smoker) vehicles have previously been
309 attributed to lubricating oil (Fujita et al., 2006). Emissions from diesel and gasoline vehicles are
310 different in terms of composition as diesel engines emit both a greater mass of PM and a larger
311 number of ultrafine particles (UFPs) compared to gasoline vehicles (Rose et al., 2006). Gasoline
312 engines are known to release a higher fraction of OC while diesel engines emit more EC (Watson et
313 al., 1994; Weingartner et al., 1997; Ntziachristos et al., 2007) although Sodeman et al. (2005)
314 demonstrated a dominance of EC in the ultrafine range in gasoline vehicles. Zhu et al. (2010)
315 reported $PM_{2.5}$ emissions from diesel and gasoline vehicles to be rich in different fraction of EC and
316 OC; while EC2 and OC2 fraction were higher in case of diesel vehicles, EC3 and OC3 were higher
317 in case of gasoline vehicle emissions. Vehicles are also a major source of n-alkanes, and diesel
318 engines are known to emit more n-alkanes compared to gasoline engines (Rogge et al., 1993).
319 Results from tunnel studies and chassis dynamometer analyses have shown that lower molecular
320 weight PAHs such as phenanthrene and fluoranthene are characteristic of diesel-derived aerosols
321 whereas gasoline engines contributed more to higher molecular weight PAHs such as
322 benzo(ghi)perylene (Smith and Harrison, 1996). PAH emissions from gasoline engines are reported
323 to be one order of magnitude higher than the diesel engine emissions integrated across the U.S.
324 vehicle fleet (Lough et al., 2005). A detailed description of diesel particulate matter is presented in
325 Maricq (2007). The situation in relation to exhaust emissions from traffic is constantly changing as
326 abatement technologies develop. In particular the adoption of diesel particle filters is greatly
327 reducing mass emissions.

328 Both trace elements and organic compounds are used as source markers for vehicle emissions.
329 However, since the removal of lead from gasoline, trace metals have proved far less useful as a
330 tracer of engine exhaust (Harrison et al., 2003). Organic compounds released from vehicles are
331 particularly useful markers for conducting receptor modelling analyses since such markers help in
332 distinguishing the vehicular emissions from other sources. The most commonly used molecular

markers for vehicular emissions are hopanes and steranes and PAHs such as benzo(ghi)perylene which are attributed to lubricating oil (Rogge et al., 1993; Schauer et al., 1996; Cass, 1998; Lough et al., 2007) and due to their source, the distribution of hopane emissions from vehicles is suggested to be independent of fleet composition (He et al., 2008). It has also been suggested that hopanes and steranes (derived from lubricating oil) and PAHs (reactions at high temperature and fuel combustion) are indicative of different processes during the organic carbon formation in vehicle engines (Lee et al., 1995; Fujita et al., 2006; Riddle et al., 2007; Hanedar et al., 2008).

A number of PAHs have been reported to be present in vehicle exhaust including benzo(e)pyrene, benzo(a)pyrene, indeno(1,2,3-cd) pyrene, coronene and benzo(ghi)perylene (Rogge et al., 1993; Lough et al., 2007; Riddle et al., 2007). However, Gao et al. (2011) attributed indeno(1,2,3-cd)pyrene and benzo(ghi)perylene to biomass combustion in Guangzhou (China). PAHs are found predominantly in the smallest size fraction (<0.4 μm), and most of the high molecular weight PAHs are found in the fine particle fraction (Hien et al., 2007). Diagnostic ratio analysis including the ratio between concentrations of indeno(1,2,3-cd) pyrene to the sum of indeno(1,2,3-cd) pyrene and benzo(ghi)perylene is also used for distinguishing between gasoline and diesel emissions (Chellam et al., 2005; He et al., 2006; Shen et al., 2010; Ancelet et al., 2011; Kim et al., 2012). Ratios between methylphenanthrenes/phenanthrene has also been used for estimation of the contribution of traffic to PAH concentrations in ambient air (Lim et al., 1999). However, it is important to remember that PAHs can be altered by atmospheric chemical reactions (Wu et al., 2007). Also, emission profiles for PAHs are difficult to generalize since they vary across the different vehicle classes (Miguel et al., 1998).Congener profiles of PAH from road traffic have been derived from tunnel studies (e.g. Smith and Harrison, 1996) and from roadside concentrations from which the local background is subtracted (Mari et al., 2010). Nielsen (1996) also prepared a traffic PAH profile for Copenhagen (Denmark) using a roadside study.

359 Elemental markers which have been used for vehicular emissions include Cu, Mn, Fe, Zn, Ba, Sn,
360 Ni, Mo and Sb (Lough et al., 2005; Almeida et al., 2006; Birmili et al., 2006; Crawford et al., 2007;
361 Dongarra et al., 2009; Fabretti et al., 2009; Gietl et al., 2010; Amato et al., 2011a,b). Metals can be
362 emitted from various exhaust-related sources including fuel and lubricant combustion, catalytic
363 converters, particulate filters and engine corrosion (Lough et al., 2005; Pulles et al., 2012; Sysalova
364 et al., 2012; Varrica et al., 2012) but many of these appear most likely to arise from non-exhaust
365 sources. Ni and V have also been reported to be present in emissions due to oil combustion (Pey et
366 al.,, 2010). Cheng et al. (2010) reported higher emission rates for elements such as Br, Ba, Sb and V
367 for gasoline and liquefied petroleum gas (LPG) engines compared to diesel engines in Hong Kong
368 and indicated potential use of these species as markers for gasoline/LPG engines. Ba, Cd, Zn, Sb
369 and V in nanoparticles have been reported to be strongly associated with diesel fuel whereas Cu,
370 Mn and Sr in the particles <0.1 μm have been found to be associated with gasoline (Lin et al., 2005).
371 Metal concentrations from diesel and gasoline vehicles are reported to vary over two orders of
372 magnitude (Pulles et al., 2012). In a traffic emissions study in New York (USA), Ba, Br and black
373 carbon (BC) were used as tracers for gasoline/diesel combustion (Peltier et al., 2011). However, the
374 levels of trace elements emitted in the exhaust are very low, and great care is needed to distinguish
375 them from non-exhaust traffic emissions and other sources. In addition, different authors have
376 attributed metals to different sources which renders precise source attribution with trace metals
377 alone very difficult unless detailed local information on source particle composition is available (e.g.
378 Pant and Harrison, 2012).

379 **4. PARTICLE SIZE DISTRIBUTIONS**

380 The size distribution of a particular element or compound not only influences the potential health
381 impact (in terms of respiratory deposition – Harrison et al., 2010), but also influences the extent of
382 atmospheric dispersion (Allen et al., 2001). Size distributions can be measured in terms of mass, number
383 or surface area (Harrison et al., 2000). A number of factors can influence the size distribution

384 including, but not limited to vehicle fleet mix, road type and grade, meteorological conditions (e.g.
385 season type) and distance from road (Zhu et al., 2006; Beddows et al., 2009; Hays et al, 2011; Song
386 and Gao, 2011). For example, differences were observed in the modal structure of particles close to
387 the freeway and at a crossroad in Prague and the difference was attributed to the higher
388 concentration of UFPs in the freeway due to the high volume of traffic (Ondracek et al., 2011).
389 However, in a study in the USA, Padro-Martinez et al. (2012) concluded that distance from the
390 highway does not impact the particle size distribution. Many other studies (e.g. Shi et al., 1999)
391 have reported the opposite. The behaviour of nanoparticles after emission is discussed later in this
392 section. Size distributions can be useful in ascertaining the sources of exhaust and non-exhaust
393 particle emissions. For example, sampling on a busy London (UK) highway, Harrison et al. (2011)
394 used Positive Matrix Factorization (PMF) to disaggregate the particle size distribution which
395 demonstrated a nucleation mode centered on around 20 nm and a solid particle mode at 50-60 nm in
396 the number distribution. However, in using such data, it should be borne in mind that the size
397 distributions measured by cascade impactors may be modified from those in ambient air as an
398 artefact caused by the semi-volatile nature of the PAHs, hopanes and steranes.

399

400 **4.1 Mass-Size Distributions**

401 Based on size distribution analysis for trace metals found in atmospheric aerosols at urban and
402 background sites in the UK, it was concluded that there are three key categories; metals with most
403 of the mass in the accumulation (fine) mode (e.g. Cs, Sn, Pb) with an additional minor mode, metals
404 with mass distributed between fine, coarse and intermediate mode (e.g. Ni, Zn, Cu, Mn) and metals
405 with most of the mass in the coarse range (Fe, Ba, Sr) (Allen et al., 2001; Birmili et al., 2006). Song
406 and Gao (2011) reported similar results for size-segregated PM at a highway site in the USA with a
407 bimodal mass-based distribution with the two peaks at 0.32-0.56 μm and 3.2-5.6 μm Table 4
408 summarizes some of the published data for traffic-related elements.

409

Similar observations regarding mass-based size distributions for trace metals were reported by Samara and Voutsa (2005) for a traffic site in Greece. Metals such as Ba, Pb and Zn were found to exhibit bimodal distributions whereas some others such as Cr and V showed trimodal distributions in Southern Taiwan (Lin et al., 2005). The presence of an Aitken mode in the ultrafine range may be indicative of a contribution from vehicular emissions, both diesel and gasoline. Hays et al. (2011) reported erratic mass size distributions for various elements including Ni, Cu, Cd, and Zn and attributed this to a range of emission sources near highways in North Carolina (USA). A peak was observed at 80 nm for soot particles in a street canyon corresponding to diesel engine emissions in Leipzig, Germany and the concentration of soot particles was reported to be influenced by traffic volume (Rose et al., 2006)

4.2 Number-Size Distributions

Ultrafine particles are characterized as particles with diameter less than 100 nm diameter and are typically measured as particle number concentration (PNC) (Charron and Harrison, 2009). Such particles may have a short lifetime and as a result, the spatial variability of the particles differs based on proximity to the emission source (Puustinen et al., 2007; Cyrys et al., 2008; Charron and Harrison, 2009). Traffic has been reported as one of the major sources of UFPs (Abu-Allaban et al., 2002; Nanzetta and Holmen, 2004; Westerdahl et al., 2009) and particle number is linked to fresh vehicle exhaust emissions (Rodriguez et al., 2007). Number size distributions are heavily influenced by proximity to traffic (Rose et al., 2006; Kerminen et al., 2007; Beddows et al., 2009; Keuken et al., 2012). As a consequence of this, concentrations of organic compounds have also been reported to be highly variable for near-highway sites (Sun et al., 2012. Highest PNC has been reported during morning rush hours in Germany (Wehner et al., 2002) and Spain (Perez et al., 2010). A significant difference has also been reported between weekdays and weekends, highlighting the influence of traffic (Morawska et al., 2002; Wehner et al., 2002; Van Poppel et al., 2012). A significant difference was observed in the particle number size distribution in the Kingsway tunnel

436 in the UK (peak at 30-40 nm) and Plabutsch tunnel in Austria (80-100 nm) and this was attributed
437 to the differences in traffic intensity and the fleet composition, i.e. proportion of HDVs and LDVs
438 (Imhof et al., 2005a). It is important to note that although PNC is heavily influenced by road traffic,
439 it is also influenced by other sources including atmospheric nucleation processes (Imhof et al.,
440 2005a; Reche et al., 2011).

441
442 The typical particle number distribution of a vehicle has 3 modes: nucleation mode (particles < 50
443 nm), consisting of particles formed due to condensation of exhaust particles; accumulation mode
444 (50-300 nm with a peak at ~ 80-100 nm) consisting of soot particles, and a coarse mode (1µm-
445 10µm) consisting of particles generated due to abrasion and wear and tear of brake pads and tyres
446 and re-suspension of road dust (Kittleson et al., 2004; Imhof et al., 2005a). The number size
447 distribution for diesel engine emissions has been reported as lognormal with mean diameter
448 between 60-120 nm whereas gasoline engine emissions have asymmetric distributions with a mean
449 diameter between 40-80 nm (Harris and Maricq, 2001). PNC shows higher variability compared to
450 particle mass (Weijers et al., 2004). While nuclei mode particles only constitute about 10% of the
451 total particle mass, they contribute more than 90% to PNC (Kittleson et al., 2004). Fushimi et al.
452 (2008) reported the presence of a nuclei mode at 20 nm in the number distribution for PM at
453 roadside while no such mode was observed at the background site in Kawasaki, Japan. PNC was
454 reported to be significantly higher in a street canyon compared to an urban background location in a
455 study in Leipzig, Germany (Wehner et al., 2002).

456
457 The particle number distribution for vehicular exhaust emissions measured in ambient air is
458 typically bimodal with one peak at 10-40 nm and another one at 50-90 nm (Charron and Harrison,
459 2009). Gasoline exhaust particles fall in the size range of 20-60 nm whereas diesel exhaust particles
460 are in the size range of 20-130 nm (accumulation mode between 50-90 nm and a smaller nucleation
461 mode) (Zhu et al., 2002b; Rissler et al., 2012). Such bimodal distributions have been reported for
462 vehicular emissions in many parts of the world including Korea (30-50 nm and 100-400 nm for

463 diesel engines) (Lim et al., 2008), Czech Republic (25-30 and 60-70 nm) (Ondracek et al., 2011)
464 and United Kingdom (13.3 nm and 86.6 nm) (Kumar et al., 2008). On the other hand, multi-modal
465 distributions have been reported in several cities and the larger number of modes has been attributed
466 to mixing of traffic and background aerosol (Ondracek et al., 2011). Lingard et al. (2006) reported
467 a four-mode particle number size distribution for an urban roadside site in Leeds (UK) with
468 particles in modes II and III to be highest during the rush hours indicating a traffic source. The four
469 modes were classified as sub-11 nm nucleation mode, super-11 nm nucleation mode, Aitken mode
470 and accumulation mode and were attributed to photolysis, diesel engine emissions and secondary
471 aerosols (Lingard et al., 2006). Rosenbohm et al. (2006) observed a bimodal (20 nm and 80 nm)
472 size distribution at a site downwind of the motorway whereas a unimodal distribution (80 nm) was
473 observed for an upwind site. El Haddad et al. (2009) reported a single mode (50-90 nm) distribution
474 for PM in a road tunnel in Marseille, France and Abu-Allaban et al. (2002) observed the highest
475 PNC in the nucleation mode (11-17 nm) in a roadway tunnel in USA.. A bimodal (11 nm and 80 nm)
476 distribution was reported for $PM_{0.5}$ emissions during a vehicle chase experiment (Wang et al., 2011).
477 Although the magnitude of PNC emissions from a gasoline spark ignition engine exhaust is smaller
478 than from a diesel engine, emissions from gasoline engines are comparable to diesel engines in the
479 cases of acceleration and freeway cruising (Kittleson et al., 2004). Klems et al., (2010) also reported
480 higher PNC at the time of acceleration at traffic lights.

481 Significant differences have been reported between dynamometer and real-world studies on PNC. A
482 mean diameter of 15 nm for PM emissions was measured in Leipzig, Germany in a street canyon
483 though some lab-studies on gasoline and diesel engines have shown mean diameters between 40-70
484 nm (Wehner et al., 2002). It is important for dynamometer studies to reflect adequately the dilution
485 properties of the atmosphere or they may suppress the nucleation mode which is formed during the
486 initial dilution of the exhaust plume (Shi and Harrison, 1999).

487

For traffic emissions, distance from the road is also reported to cause changes in PNC due to processes other than dilution (Zhu et al., 2006). This feature is of considerable interest; particularly from an epidemiological perspective as this would mean that exposure from near-roadway sites can be much different from other sites. HEI (2010) reported the key exposure zone for traffic emissions to be within 300-500 m of a roadway. In Los Angeles, both PNC and size distribution changed with an increase in distance from the road, particularly for particles with a diameter less than 30 nm and it has been suggested that this drastic change in PNC is caused by coagulation and atmospheric dilution (Zhu et al., 2002a). Padro-Martinez et al. (2012) found PNC to be at least two times higher at 0-50 m from the highway compared to further distances while Zhu et al. (2002b) observed the PNC to be 25 times higher near the freeway compared to background locations. Rose et al. (2006) also observed a decrease in soot particle number concentration with increasing distance from the street canyon which was attributed to coagulation processes. Kittleson et al. (2004) observed a decline in the nuclei mode particles as the distance from the highway increased and recently, Canagaratna et al. (2010) also reported a decrease in PNC with increasing distance away from the roadway. Dilution processes will inevitably cause a reduction in PNC with distance from the road, as they do for other traffic-generated pollutants (e.g. NO_x, BC, etc.). Coagulation causes an increase in particle size, and a decrease in number, but is unlikely to be a significant influence close to the road due to an insufficient number concentration to drive the process, and short travel times. An observed increase in particle diameters with distance from a road is most probably the result of mixing of traffic emissions with a coarser background aerosol leading to a large modal diameter (Shi et al., 1999). The nucleation mode traffic particles are semi-volatile and this can lead to shifts in size distribution (typically to smaller sizes) and particle loss due to evaporation (Dall'Osto et al., 2011; Harrison et al., 2012a).

Charron and Harrison (2003) reported that increasing wind speeds and lower temperatures can increase the abundance of smaller nucleation mode particles (diameter of sub-30 nm) and this could

514 be due to the greater dilution and lower condensation sink. Ruellan and Cachier (2001) also
515 reported an influence of wind speed and boundary layer height on PNC. Kumar et al. (2008)
516 reported traffic volume and wind speed as the key variables influencing PNC in street canyon
517 conditions and they observed a linear dependence of PNC on traffic volume with a higher
518 correlation in the case of smaller particles (diameter < 300 nm) and a weaker correlation in the case
519 of larger particles for which other sources are large relative contributors. Highest PNC was
520 observed for calm winds and lowest for wind speeds above 1.6 m/s in Somerville (USA) (Padro-
521 Martinez et al., 2012). PNC was found to be correlated with traffic intensity in France (Ruellan and
522 Cachier, 2001; El Haddad et al., 2009; Perez et al., 2010) and in Los Angeles, PNC was reported to
523 be influenced by traffic density and wind speed and direction (Zhu et al., 2002b). In a study in
524 London, Milan and Barcelona, Rodriguez et al. (2007) concluded that vehicular exhaust emissions
525 largely influence ultrafine particles and found a unimodal distribution for particle number with a
526 mode at 37 nm. Kittleson et al. (2004) observed a direct influence of increasing vehicle speed on
527 nuclei-mode particles. Decreasing the engine load from 100% to about 60% was reported to
528 decrease the PNC as well as EC and OC emissions from diesel vehicles (Lim et al., 2008).
529 Morawska et al. (2002), on the other hand, indicated a 70% increase in PNC with a 50% increase in
530 traffic flow. While roadside PNC measurements have indicated an influence of temperature on the
531 concentration, similar studies in laboratory conditions using chassis dynamometers have not shown
532 such influence (Abu-Allaban et al., 2002; Kittleson et al., 2004). Abu-Allaban et al. (2002) reported
533 an inverse relationship between ambient air temperature and the diameter of nucleation mode
534 particles. Using modelling analysis, Jayaratne et al. (2009) observed an increase in PNC at a
535 signalized pedestrian crossing due to the stop and go nature of traffic at the signal and the increase
536 was found to be higher for HDVs compared to LDVs. This is similar to the observations from Abu-
537 Allaban et al. (2002) and Wang et al. (2010) who observed higher concentrations arising from
538 HDVs compared to LDVs in USA and Denmark respectively. Rosenbohm et al. (2006) reported a
539 correlation between PNC and NO in Heidelberg (Germany) although no correlation was observed

for PNC and PM_{10} concentration. Similarly, Padro-Martinez et al. (2012) also reported a similar spatial trend for PNC, NO_x, particulate PAHs and BC in the USA while a different trend was reported for $PM_{2.5}$.

Thus, key factors that influence the particle number concentration measurements include wind speed, traffic density and the distance from source at which the measurements are made. Nanzetta and Holmen (2004) observed differences in the PNC across different seasons and concluded that results for one season cannot be used for generalization of PNC.

5. METHODOLOGIES FOR ASSESSMENT OF TRAFFIC EMISSIONS

There are several ways of evaluating emissions from road vehicles including tests with dynamometers, measurements in tunnels/highways and near roadside measurements and road simulator tests (Figure 1) (Mclaren et al., 1996; Phuleria et al., 2007; Handler et al., 2008; Gustafsson et al., 2008; El Haddad et al., 2009; Yan et al., 2009; Sjodin et al., 2010). Table 5 presents a qualitative description of measurements using dynamometers under lab conditions and tunnel/roadway/twin-site studies under ambient conditions. In recent years, significant differences have been observed between laboratory-tested and real-world mixed source traffic emissions (Gertler et al., 2002; Yan et al., 2009; Ancelet et al., 2011). Near roadside/kerbside, traffic tunnel and highway measurements are reported to be more realistic since they represent mixed-fleet emissions under real-world driving conditions (Phuleria et al., 2007). Beddows and Harrison (2008) compared particle number EFs measured in dynamometer studies with those estimated in roadside studies, finding generally good agreement overall. However, Canagaratna et al. (2010) reported the EFs calculated using on-road measurements to be nearly 50% lower than EFs measured using a dynamometer.

An emission factor is defined as "the mass (or number) of pollutant released per unit time/distance travelled or mass of fuel used" and typically increases with vehicle speed for LDVs as well as HDVs (Jones and Harrison, 2006). Characterization of emissions from road vehicles includes analysis of size distributions and chemical composition (organic, inorganic and trace metal) and can lead to generation of EFs. EFs typically depend on conditions at the sampling site, traffic intensity and modal shares of HDVs and LDVs (Staehelin et al., 1995; Imhof et al., 2005a). A recent review article on development of emission factors delves into this issue in greater detail (Franco et al., 2013).

EFs can be measured using a number of different methods including dynamometer studies (Oanh et al., 2010; Chiang et al., 2012), roadway tunnel studies (Gertler et al., 2002; Handler et al., 2008; Mancilla and Mendoza, 2012), on-road chase experiments (Wang et al., 2011) and remote sensing (Zhang et al., 1995). Phuleria et al. (2007) reported overall agreement between EFs determined at tunnel and roadway sites in USA.

Two of the most common methods for EF expression are fuel-based EF (Ban-Weiss et al., 2010; Wang et al., 2011) and vehicle kilometre travelled (VKT)-based calculation (Abu-Allaban et al., 2002; Kittleson et al., 2004; Jones and Harrison, 2006; Amato et al., 2012). Phuleria et al. (2006, 2007) and Bukowiecki et al. (2010) describe the methodology of EF derivation in greater detail. Fuel-based EF can be defined as "grams of pollutant emitted for every kilogram of fuel burned" and can be calculated for various pollutants using CO_2 and CO as a measure of fuel burned (Liacos et al., 2012). The concentration of pollutant with respect to the concentration of CO_2 + CO can be used to calculate the fuel burnt using the following formula (Miguel et al., 1998; Liacos et al., 2012):

$$EF_{\mathrm{P}} = 10^{6} \times \left(\frac{\Delta P}{\Delta CO2 + \Delta CO}\right) w_{\mathrm{c}} \qquad (1)$$

588 where EF_P is the emission factor for the pollutant (µg/kg fuel burned), ΔP (ng/m^3), ΔCO_2 and ΔCO
589 (µgC/m^3) are the increments over background concentration and w_c is the fraction of carbon weight
590 in the fuel.

591
592 One of the other most commonly used methods is the derivation of PM EFs using NO_x EFs, which
593 are calculated as described in the following equation (Gehrig et al., 2004; Imhof et al., 2005b). It is
594 assumed in this case that both NO_x and PM have similar dilution.

595 $$D = \frac{N_{\mathrm{LDV}} \times EF\ NOx_{\mathrm{LDV}} + N_{\mathrm{HDV}} \times EF\ NOx_{\mathrm{HDV}}}{\Delta NOx} \qquad (2)$$

596 where D is the dilution rate (m^2 h^{-1}), n is the number of vehicles (h^{-1}), and EF NO_x is the emission
597 factor for NOx in mg/km.

598
599 Using the dilution rate, the EF (per hour) can be calculated per vehicle based on:

$$EF_{\mathrm{x}} = \frac{\Delta Concentration_{\mathrm{x}} \times D}{n_{\mathrm{total}}} \qquad (3)$$

600 where $\Delta\mathrm{Concentration}_x$ is the difference in concentration of species x between roadside and
601 background and n is the total number of vehicles crossing the sampling point per hour. The
602 respective contribution of HDVs and LDVs can then be estimated with multiple linear regression
603 (MLR) (Gertler and Pierson, 1996; Imhof et al., 2005b). Table 6 presents some EFs reported for
604 PM_{10}, $PM_{2.5}$ and particle number concentration in the literature.

605

606 **6.1 Tunnel/Roadway Measurements**

607 EF calculations from tunnel studies are typically based on calculation of the difference in
608 concentration between entrance and exit (Weingartner et al., 1997; Abu-Allaban et al., 2002), and
609 Gertler and Pierson (1996) describe the methodology for calculation of emission factors using
610 tunnel measurements. Chang et al. (1981) developed a model relating air quality in the tunnel to

611 vehicular emissions which can also be used for calculation of emission factors using tunnel
612 measurements. For the tunnel studies, "the velocity and concentration are assumed to be the same
613 across the tunnel's cross-section and the difference between the influx and outflux is assumed to be
614 the sum of emission rates of vehicles in the tunnel" (Rogak et al., 1998). The following formula is
615 typically used for calculation of the EF (Gertler and Pierson, 1996; He et al., 2008):

$$EF = \frac{C_{\text{out}}V_{\text{out}} - C_{\text{in}}V_{\text{in}}}{NL} \qquad (4)$$

616 where C is the concentration of the pollutant, V is the volume of air calculated using duration of
617 sampling, cross-sectional area of tunnel and wind speed; N is the number of vehicles and; L is the
618 distance between sampling locations at inlet and outlet of the tunnel.
619
620 Several studies have been conducted for measurement of PM emissions in roadway tunnels in
621 different parts of the world including the USA (Fraser et al., 1998; Rogak et al., 1998; Gillies et al.,
622 2001; Abu-Allaban et al., 2002; Gertler et al., 2002), China (He et al., 2006; He et al., 2008; Chiang
623 and Huang, 2009), Europe (Stechmann and Dannecker, 1990; Smith and Harrison, 1996;
624 Weingartner et al., 1997; Imhof et al., 2005a; Handler et al., 2008; El Haddad et al., 2009; Oliviera
625 et al. 2011), New Zealand (Ancelet et al., 2011), Japan (Funasaka et al., 1998), Taiwan (Chiang and
626 Huang, 2009), Chile (Caceres et al., 1998) and Mexico (Mancilla and Mendoza, 2012).
627
628 Although tunnel studies provide an opportunity to conduct measurements on a mixed vehicle fleet,
629 it is important to remember that factors such as variations in speed, aerodynamic conditions in the
630 tunnel and the fleet characteristics (i.e. proportion of HDVs and LDVs) can cause variability in
631 measurements (Rogak et al., 1998; He et al., 2008). In addition, vehicles in the tunnel are often
632 driving at a steady speed which does not happen under other road conditions where traffic follows a
633 stop-and-go pattern (Gertler et al., 2002). Chirico et al. (2011) have shown that the distribution of
634 organic compounds between particles and vapour is heavily affected by the high PM concentrations

635 in a road tunnel, and thus may influence the estimate of emission factors for semi-volatile
636 components.

637
638 PM_3 (particles with diameter < 3 μm) was analysed in the Gubrist Tunnel in Zurich, Switzerland
639 and it was concluded that most of the particles are emitted from the tailpipe with a small
640 contribution from tyre wear and road dust (Weingartner et al., 1997). While BC accounted for 31%
641 of the total emissions, particulate PAHs accounted for 0.86% of PM_3 (Weingartner et al., 1997). In
642 New Zealand, PAHs were reported to contribute nearly 0.10% of the total PM mass with a higher
643 proportion of low molecular weight PAHs indicating a higher contribution of diesel emissions
644 (Ancelet et al., 2011). In roadway tunnel studies in China, EC was found to contribute 63% and 45%
645 to PM mass whereas OC contributed 34% and 31% (He et al., 2006; He et al., 2008). Ancelet et al.
646 (2011), however, found OC to contribute between 32-46% to PM mass. Gillies et al. (2001) and He
647 et al. (2008) have attributed higher $PM_{2.5}$ emissions to HDVs compared to light duty vehicles
648 (LDVs) in USA and China respectively. Caceres et al. (1998) prepared a PAH source signature for
649 traffic using tunnel measurements in Santiago de Chile whereas Smith and Harrison (1996)
650 prepared a source profile fingerprint using tunnel measurements for traffic in Birmingham, UK.
651 Funasaka et al. (1998) analysed emissions in a traffic tunnel in Osaka, Japan and EC and NO_x (NO
652 as the main component) were found to be correlated with diesel traffic volume whereas OC did not
653 show good correlation. In addition, the concentration of EC and SPM was observed to increase
654 linearly with an increase in traffic volume. In Lisbon, (Portugal), nearly 84% of the total PAH mass
655 in a road tunnel was reported to be less than 0.49 μm diameter by Oliviera et al. (2011).

656 Concentrations of trace elements were not found to be correlated with EC concentration in Zhujiang
657 Tunnel (China) and the authors concluded non-exhaust sources to be contributing to the trace
658 element mass (He et al., 2008). A similar conclusion was reported by Funasaka et al. (1998) and
659 Ancelet et al. (2011) who attributed coarse particles in a road tunnel in Osaka, Japan and inorganic
660 elements such as Fe, Zn etc. respectively to non-exhaust emissions, primarily re-suspension of road

661 dust. It should be noted, however, that the contributors to road dust in a tunnel will differ from
662 those in a normal roadway environment and that both chemical composition and emission factors
663 will be different. Partitioning of the semi-volatile components between particles and vapour can
664 also be influenced by the high concentrations present (Chirico et al., 2011).

665

666 **6.2 Twin-Site Studies**

667 A number of papers have reported the estimation of the contribution of traffic emissions to total PM
668 using twin-site studies in recent years (Yan et al., 2009; Bukowiecki et al., 2010; Gietl et al., 2010;
669 Oliviera et al., 2010; Pey et al., 2010). With the assumption that all sources other than traffic
670 (including any local or regional sources) have the same impact at both roadside and background
671 sites, the increment at the roadside site obtained using the following equation is used as a local
672 traffic increment estimate (Harrison, 2009; Yan et al., 2009; Wang et al., 2010).

673 $$Concentration\ of\ X_{\text{traffic}} = Concentrationof\ X_{\text{roadside}} - Concentration\ of\ X_{\text{background}} \quad (5)$$

674 Typically, the difference between rural and urban concentration observation provides an estimate
675 for the urban increment while the difference between roadside and urban background concentration
676 observations provides an estimate for the traffic increment (Harrison, 2009). However, results from
677 such studies can be influenced by street geometry. Harrison et al. (2004) observed different
678 behaviour for particle mass based on the site geometry, where the traffic contribution increases for
679 larger daily background mass concentrations at open sites, whereas the magnitude of the traffic
680 mass increment is independent of daily background concentration for enclosed sites, thus making
681 selection of a suitable sampling location critical for coherent results. They also found the pattern of
682 air circulation to be an important determinant of ambient PM concentration at an enclosed street site.
683 Roadway studies are useful in providing a reasonable estimate of pollutant concentrations that
684 commuters are exposed to (Phuleria et al., 2007) but if the sampling is carried out only at roadside

sites, the data generated is representative only of that part of the total vehicle fleet that was sampled (Liacos et al., 2012).

Harrison et al. (2003) concluded that the trace element enrichment observed at roadside sites is contributed largely by wear and tear of tyres and brakes. Significant roadside enrichment has been reported for elements, ions and organic compounds in various cities including London (UK) (Gietl et al., 2010) Los Angeles (USA) (Ntziachristos et al.,~~2008~~2007), Sicily, Rome and Milan (Italy) (Manno et al., 2006; Canepari et al., 2008; Perrone et al., 2012), Barcelona (Spain) (Amato et al., 2011), Portugal (Oliviera et al., 2011) and Queensland (Australia) (Gunawardana et al., 2011). . In New York City, Fe, Al and Si were found to contribute between 0.5- 1.0 µg/m^3 to the ambient $PM_{2.5}$ loading (Peltier et al., 2011) whereas Lin et al., 2008 reported Ca, Al and Fe to constitute between 65-77% of suspended particles in Taiwan. Nearly 90% of the road traffic emissions of Cu were reported to be due to brake wear in Stockholm (Sweden) (Johansson et al., 2009). Amato et al. (2011b) analyzed roadside PM_{10} enrichment (RE) of trace elements in Barcelona (Spain) reported more than 70% roadside enrichment ratio for key traffic tracers including EC, Fe, Ba, Cu, Sb, Cr and Sn. Gietl et al. (2010) estimated that based upon published emission factors brake dust comprises nearly 13% of the total PM at a roadside site in London (UK) and Amato et al. (2011b) reported approximately 60% enrichment of brake wear particles in a roadside location compared to an urban background location in Barcelona (Spain). Pey et al. (2010) reported that the concentration of EC was found to be 3-8 times higher for the urban site in comparison with the background site in Barcelona (Spain) and Perrone et al. (2012) found EC to be contributing 11-15% of $PM_{2.5}$ in urban areas compared to 3-5% at a rural site and 1-2% at the background site in Milan (Italy). Vertical concentration gradients have also been reported for trace elements with highest concentrations at kerbside as compared with rooftop (Johansson et al., 2009).Analysis of size-resolved $PM_{2.5}$ at a roadside site in Beijing revealed that re-suspended dust and construction dust

710 contribute 32.7-50.4% of total $PM_{2.5}$ (Song et al., 2012) and a reduction was observed in
711 concentrations of species associated with traffic during the 2008 Olympic Games).

712
713 Comparatively higher concentrations of hopanes, steranes, n-alkanes and PAHs have been reported
714 for highway/near-roadway sites compared to other sites (Yan et al., 2009; Aldabe et al., 2011;
715 Peltier et al., 2011; Perrone et al., 2012). Yan et al. (2009) analysed $PM_{2.5}$ concentrations at urban
716 and rural sites and observed a higher concentration for EC and primary organic compounds released
717 by motor vehicle exhaust such as hopanes and steranes. They found organic matter to be
718 contributing between 51-72% to PM2.5 mass at the urban site compared with 40-54% at the rural
719 site. Aldabe et al. (2011) reported a concentration gradient for total carbon with the highest
720 concentration at the roadside site and lowest concentration at a rural site, and a decreasing trend of
721 OC concentration was observed in Prague with a decrease in traffic intensity (Ondracek et al., 2011).
722 Gao et al. (2011) also reported higher concentrations for EC and hopanes at the roadside site
723 compared to a rooftop site, highlighting the contribution of traffic emissions. Jones and Harrison
724 (2006) used ratios of concentration to NO_x for determination of EFs for road traffic or particle
725 number and mass metrics with the assumption that particle dispersion is similar to dispersion of
726 NO_x in the atmosphere. The EFs that they obtained from this study for particle mass and number
727 (Table 6) were comparable with emission factors calculated using other methods such as
728 dynamometer and tunnel/roadway studies. Liacos et al. (2012) observed higher concentrations of
729 trace metals using on-road measurements and postulated that studies using sampling from near
730 roadside conditions might not always capture the pollutant concentrations that drivers are exposed
731 to (Liacos et al., 2012).

732

733 **6.3 NO_x as Tracer**

734 With the assumption that road traffic is the primary urban source for NO_x in the urban atmosphere,
735 Harrison et al. (1997) used regression analysis between particle mass fractions and NO_x to obtain

736 contributions from the traffic source for Birmingham (UK) with a total of 32% of the fine PM
737 concentration attributed to vehicular exhaust emissions for the winter season. The method is very
738 simple, using the assumption that the regression intercept in PM at zero NO_x is due to non-traffic
739 sources, with the traffic contribution estimated by difference from the measured mean concentration.
740 The same approach was also used by Fuller et al. (2002) who estimated PM_{10} and $PM_{2.5}$
741 concentrations using NO_x and PM emissions for London (UK) and derived a typical PM_{10}: NO_x
742 ratio of 0.045± 0.003. Lim et al. (1999) also used this approach to determine the contribution of
743 PAHs to traffic in Birmingham and concluded that traffic contributed 80-82% and 61-67% of PAH
744 concentrations in the city centre and an urban background site (university campus) respectively.
745 Thorpe et al. (2007) used this method to calculate re-suspension EFs. The PM_{10} emission factor was
746 calculated using NO_x as a tracer and coarse particle source strength and abrasion emissions were
747 subtracted from total PM_{10} emission to a obtain a re-suspension emission factor. However, it is
748 important to note that this method is only useful if the predominant source for local ground-level
749 NO_x is traffic, unless used in a twin-site approach.

750

751 **6.4 Other Methods**

752 Several studies have reported lower levels of vehicular emissions on weekends (Rose et al., 2006;
753 Barmpadimos et al., 2011). Analysis of the weekly cycle of coarse PM concentrations (2.5-10 μm)
754 with the weekly PM coarse traffic emission was carried out in Zurich (Switzerland) and it was
755 estimated that nearly 70% and 53% of the ambient coarse PM mass is contributed by traffic on
756 weekdays and weekends respectively (Barmapadimos et al., 2011). With the assumption that the
757 difference in PM concentrations between weekdays and weekends at an urban background site is
758 due to traffic movement, it is possible to calculate the traffic increment to urban PM concentrations,
759 although the results show large uncertainties (Jones et al., 2008).

760 Vehicle-chase experiments have also been used where vehicles are chased for a specific period of
761 time to collect pollutant concentrations allowing data to be collected for real-world conditions of
762 dilution (Canagaratna et al. 2010; Wang et al., 2011). However, the EFs are only valid for the
763 specific sampling conditions and there is no control over driving conditions (Wang et al., 2011).

764 **6. RECEPTOR MODELLING OF TRAFFIC PM**

765 Watson and Chow (2007) describe receptor models as models that "interpret measurements of
766 physical and chemical properties taken at different times and places to infer the possible sources of
767 excessive concentrations and to quantify the contributions from those sources". A number of
768 receptor models are used for source apportionment including the Chemical Mass Balance (CMB)
769 model, statistical models such as PCA and PMF, Multilinear Engine (ME), Constrained Physical
770 Receptor Model (COPREM) and UNMIX. With the assumption that the relative concentrations of
771 chemical species are preserved between sources and receptors, receptor models use the principle of
772 mass conservation for apportionment of PM mass to different air pollution sources. Thus, the
773 concentration of a species measured in a particular sample can be described as (Hopke, 2003):

$$X_{ij} = \sum_{p=1}^{p} g_{ip} f_{pj} + e_{ij} \qquad (6)$$

774 where X_{ij} is the measured concentration of the *j*th species in the *i*th sample, f_{pj} is the concentration of
775 the *j*th species in material emitted by the source *p*, g_{ip} is the contribution of the *p*th source to the *i*th
776 sample and e_{ij} is the portion of the measurement that cannot be fitted by the model. Different
777 receptor models use different approaches to solve this equation, for e.g., the CMB model uses the
778 effective-variance least squares method whereas UNMIX uses eigenvector analysis. Concentrations
779 of trace elements (e.g. Si, Fe, Cu), ions (e.g. SO_4^{2-}), EC/OC and organic compounds (e.g. PAHs,
780 hopanes, alkanoic acids) are used as inputs for the models.

781

In recent years, there have been many studies for source apportionment of road traffic-generated PM, some of which appear in Table 7. There have also been studies that have used both CMB and factor analysis models (Larsen and Baker, 2003; Bullock et al., 2008).

6.1 Multivariate Statistical Methods

There are a number of different methods based on factor analysis including PCA, PMF, UNMIX and ME. Such methods do not require a priori information about source emission characteristics and are useful in cases where relevant source profiles are not available (Hopke, 2003; Viana et al., 2008). In the simplest matrix form, the equation 6 can be represented as (Hopke, 2003)

$$X = GF'$$

A number of studies have been conducted using the different factor analysis methods and studies in different locations have often reported different magnitudes of exhaust and non-exhaust emissions. In Taiwan, gasoline, diesel and fuel oil emissions together with dust and brake wear were found to be key contributors to traffic-generated PM using PCA (Lin et al., 2005). Using COPREM, Wahlin et al. (2006) apportioned the local traffic emissions to exhaust, road dust, brake wear and winter salting in Copenhagen (Denmark). Amato et al. (2009) analysed road dust in Barcelona (Spain) using ME-2, and re-suspension was found to contribute 37%, 15% and 3% of total traffic emissions respectively in PM_{10}, $PM_{2.5}$ and PM_1 respectively. Other identified sources included secondary aerosols and a soil source. Sjodin et al. (2010) found road wear to be the most important source of PM_{10} emissions in streets as well as background locations in Sweden, which is probably a reflection of road sanding and use of studded tyres. Using PMF, three distinct traffic sources including vehicle abrasion (36%), re-suspension (43%) and fuel combustion (21%) were resolved for traffic sources using trace metals in Nice (France) from data collected in a road tunnel (Fabretti et al., 2009). Based on PMF analysis in Switzerland, Bukowiecki et al. (2010) reported 21%, 38% and 41% of the traffic emissions to be contributed by brake wear, re-suspended road dust and exhaust emissions

806 respectively for street canyons. For freeways, while the contribution of exhaust emissions remained
807 the same, the contribution of brake wear was much less at 3% whereas re-suspended road dust
808 contributed as much as 56%. Movement of traffic also contributes to re-suspension of road dust and
809 re-suspended road dust has been estimated to contribute as much as 38% of the total PM in a street
810 canyon and 56% on a highway (Bukowiecki et al., 2010; Laidlaw et al., 2012). Analysis of PM in
811 Madrid (Spain) using PMF estimated that nearly 29% of the PM_{10} in the city is contributed by road
812 dust while another 31% is contributed by direct vehicular emission (Karanasiou et al., 2011). In
813 Oporto (Portugal), exhaust emissions and re-suspension due to traffic were found to contribute 12-
814 55% of the coarse PM and 14-66% of the fine PM based on PCA- multiple regression analysis and
815 inter-site mass balance analysis (Oliviera et al., 2010). They used inter-site mass balance for
816 separating the traffic increment in the road dust and concluded that nearly 30% of the fine fraction
817 and 50-70% of the coarse fraction PM is contributed by road dust. They also used a roadside impact
818 factor (defined as the fractional difference between the concentrations at the roadside and urban
819 background) and the highest factors were observed for Cu, Zr, Fe, Cr and Sn indicating a traffic
820 source for these metals. Thorpe et al. (2007) reported re-suspended road dust, abrasion source
821 emissions and exhaust and fine-range abrasion emissions to contribute 20-22%, 19% and 60% to
822 roadside incremental PM_{10} concentration respectively in a major highway in London. Four major
823 sources for road surface dust particles of diameter less than 10 μm including motor exhaust, brake
824 and tyre wear and road surface wear together with background urban dust were reported for
825 European cities with an increase in the concentrations of trace metals and OC/EC with an increase
826 in traffic volume (Amato et al., 2011a). Song and Gao (2011) identified three distinct sources of
827 trace metals at a highway site in New Jersey (USA) using factor analysis: brake wear and fuel
828 combustion, primary fuel combustion and tyre wear and fuel combustion. PCA analysis was used
829 for determining $PM_{2.5}$ sources in Incheon (Korea) and motor vehicles/sea salt and soil emissions
830 were found to contribute 39% and 7% respectively (Choi et al., 2012). Similar results were obtained

by Cheng et al. (2011) in Hong Kong where traffic emissions and re-suspended dust emissions were found to contribute 38% and 5% using PMF.

Dreyfus et al. (2009) conducted PMF analysis with time-resolved organic molecular marker data and identified diesel and road dust/car emissions factors which accounted for 2/3rd of the total OC concentration. Receptor modelling analysis for $PM_{2.5}$ was conducted using PMF and UNMIX, and PMF distinguished between diesel and gasoline engine emissions using temperature resolved carbon fractions (Sahu et al., 2011). Pey et al. (2009) performed source apportionment using PCA and regression with a large dataset including information on particle number concentration; size distribution and chemical composition for an urban background site in Barcelona, and road traffic was identified as the most significant source (52- 86%) for aerosols in the sub-micrometer range.

Liu et al. (2007) attributed the PAHs in road dust in Shanghai (China) to traffic emissions and coal combustion using principal component analysis (PCA) and compositional analysis. Using the same technique, Han et al. (2009) attributed PAHs in road dust in Anshan (China) to emissions from traffic, coal combustion, steel industry and cooking. Using MLR, Boonyatumanond et al. (2007) reported tyres, diesel vehicle soot and used engine oil to be the key sources for PAHs found in street dust in Bangkok (Thailand). Hassanien and Abdel-Latif (2008) attributed PAHs in road dust in Cairo (Egypt) to emissions from traffic, industry and incomplete combustion of waste based on correlation analysis. Teixeira et al. (2012) reported vehicular emissions and coal and wood combustion to be the key sources for PAHs in Porte Alegre (Brazil) using PCA.

Other approaches have also been used for estimation of source contributions. Recently, Harrison et al. (2012b) used field data for estimating the contribution of brake wear, tyre wear and road dust using size distributions of Ba, Zn and Si as the tracers for the sources respectively. By merging size distributions measured with a Scanning Mobility Particle Sizer (SMPS) and an Aerodynamic Particle Sizer (APS), Harrison et al. (2011) were able to generate continuous particle size distributions for a roadside site from 15 nm – 10 μm diameter. By applying PMF, they were able to

separately identify nucleation mode and accumulation mode exhaust particles as well as brake dust and re-suspension particles, and several particle classes arising from the urban background.

7.2 Chemical Mass Balance (CMB) Model

The CMB model uses the ambient measurement data for chemical species together with the associated uncertainty and source profiles for different sources as inputs and the output consists of estimates of contribution of each source to the total mass. The CMB model has several assumptions including (Watson et al., 2002):

- chemical species do not react with one another and the species composition does not change between the source(s) and the receptor(s)
- number of species are more than the number of sources
- all of the potential sources are included the model
- source profiles are not collinear
- random and non-correlated uncertainties

It is important to note that while the multivariate models do not require any information on source emission composition, the CMB model requires the species abundances (in the form of source profiles) in each source as a model input. Source profiles are "the mass abundances, i.e. fraction of total mass of chemical species in source emissions, and such profiles are generally representative of source categories rather than individual emitters" (Watson et al., 2002). There are several ways of preparing source profiles for road vehicles including tests with chassis dynamometers, measurements in tunnels/ highways and near roadside measurements (Phuleria et al., 2007; Handler et al., 2008; Yan et al., 2009) as discussed in section 6. Source profiles are used for quantification of source contributions to particulate matter using the chemical mass balance (CMB) model as well as to compare and validate results obtained from factor analysis models. The CMB model relies to a large extent on the accuracy of the source profiles used as an input. The typical components of any

source profile are found to be more-or-less similar although the mass abundances vary depending on location and emitter characteristics. Thus, different combinations of source profiles can provide statistically valid yet completely different solutions and selection of a source profile comparable to ambient data is one of the key prerequisites for a good CMB solution (Robinson et al., 2006). Absence of locally-representative source profiles and inter-region variability can result in errors in estimation of source contributions from the CMB model (Yan et al., 2009; Rutter et al., 2011). Significant differences have also been reported between real-world emissions and laboratory-tested emissions (Yan et al., 2009; Ancelet et al., 2011). Analysis of vehicular source profiles in Bangkok, Thailand led to the conclusion that while source profiles for different vehicle categories vary significantly, ranges of the light duty and heavy duty average source profiles overlap and can be represented by a single composite profile (Oanh et al., 2010). Fraser et al. (2003) and Fujita et al. (2007) used the CMB model to analyse the apportionment of PM mass between diesel and gasoline vehicles. While high molecular weight (4-7 ring) PAHs (coronene, benzo(ghi)perylene), hopanes and steranes were found to be influential in case of gasoline engine emissions, 2-4 ring methylated PAHs and EC were reported to be key species in the case of diesel engine emissions. Hanedar et al. (2008) used the CMB model to resolve the sources of PAHs in Istanbul (Turkey) and identified vehicle emissions (gasoline and diesel), natural gas combustion, coal and wood burning as major sources with vehicular emissions contributing more than 50% at both urban and rural location. Perrone et al. (2012) used CMB analysis to resolve PM sources in Milan (Italy) and reported traffic emissions to contribute between 17-24% to $PM_{2.5}$ mass in an urban location compared to 7-9% at a rural site and El Haddad et al. (2011) reported vehicular emissions to contribute 17% to the PM mass in Marseille (France). In Detroit (USA), traffic emissions and road dust emissions were reported to contribute 24-36% and 3-5% respectively of mass using CMB analysis of $PM_{2.5}$ data.

In recent years, there have been several studies focusing on characterization of trace metals and/or organic marker compounds in PM contributed by road traffic emissions and preparation of mixed

traffic source profiles (El Haddad et al., 2009; Fabretti et al., 2009; Yan et al., 2009; Ancelet et al., 2011). El Haddad et al. (2009) prepared a source profile for the vehicle fleet in France using tunnel measurements and nearly 70 organic markers, whereas Fabretti et al. (2009) used trace element marker data from tunnel-based measurements to derive chemical fingerprints for different traffic-related sources including combustion, re-suspended dust and abrasion-related particles. Tunnel studies were also used in USA for preparation of source profiles for gasoline and diesel engines using inorganic and organic markers (Phuleria et al., 2006; Landis et al., 2007). Yin et al. (2010) used organic molecular source tracers to provide separate quantification of gasoline, diesel and smoker vehicle emissions at two UK sites. Recently, an attempt has also been made to characterize the contribution of lubricating oil and fuel to the PM emissions (Sonntag et al., 2012).

7. CONCLUSIONS

Road traffic frequently makes a very significant contribution (5-80% depending on site and location) to airborne concentrations of PM, whether expressed in terms of particle mass or number concentration (Thorpe and Harrison, 2008; Belis et al., 2013). Particles arise not only from the engine exhaust but also from abrasion of tyres, road surface and brake components as well as from resuspension from the road surface. The sizes of particles emitted from these sources differ considerably and size association may be a useful guide to particle source, as is chemical composition. Fleet mix, type of road, traffic characteristic and fuel type are the factors that impact the emission type and the relative contributions from exhaust and non-exhaust emissions. The determination of particle mass and number increments due to traffic emissions can be based on a number of methods including measurements in well controlled environments such as a road tunnel, or twin site studies such as a highway and urban background (allowing for subtraction of the background contribution), as well as using vehicle-specific tracers and the more sophisticated receptor modelling methods based on multi-variate statistics or Chemical Mass Balance which have been used in many studies to estimate the contributions to particulate matter concentrations, not

only from road vehicles but also from other sources. However, significant gaps still exist in our understanding of traffic emissions, particularly:

1. Characterization and quantification of different non-exhaust sources.
2. Impacts of non-exhaust emissions upon human health: While there are many studies reporting links between exposure to air pollution and adverse health impacts, detailed information on the components that contribute to toxicity is missing (Kelly and Fussell, 2012).
3. Aerosol chemistry in high-traffic environments, and the evolution of particles emitted from vehicles

Further, most of the reported analyses have been carried out in the USA or Europe, and there is a lack of reliable information on traffic emissions in areas with high population density in Africa, Asia and South America. The field offers much scope for future research , including the development of enhanced methods for quantification of non-exhaust contributions to airborne concentrations.

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TABLE LEGENDS

Table 1: Mechanisms for non-exhaust particle emission (Rogge et al., 1993; Boulter et al., 2005; Wahlin et al., 2006; Thorpe et al. 2007; Amato et al., 2009; Harrison, 2009; Gietl et al., 2010; Barmapadimos et al., 2011; Hays et al., 2011; Denier van der Gon, 2013).

Table 2: Key tracers used for non-exhaust PM.

Table 3: Ratio of Cu to Sb characteristic of brake wear particles in ambient PM reported in the literature.

Table 4: (a) Results from mass size distribution analysis for traffic-related PM as reported in the literature (in µm); (b) Results from mass size distribution analysis for traffic-related elements as reported in the literature (in µm).

Table 5: Comparison between dynamometer and tunnel/roadway measurements (Allen et al., 2001; Imhof et al., 2005a; Phuleria et al., 2006; Phuleria et al., 2007; Handler et al., 2008; He et al., 2008; El Haddad et al., 2009; Sanchez-Ccoyllo et al., 2009; Franco et al., 2013).

Table 6: Emission factors as reported in the literature (selected studies).

Table 7: Source apportionment analyses for road traffic-generated PM (selected studies).

FIGURE LEGEND

Figure 1: Methods for direct road traffic emissions analyses.

Table 1: Mechanisms for non-exhaust particle emission **(**Rogge et al., 1993; Boulter et al., 2005; Wahlin et al., 2006; Thorpe et al. 2007; Amato et al., 2009; Harrison, 2009; Gietl et al., 2010; Barmapadimos et al., 2011; Hays et al., 2011; Denier van der Gon, 2013)

Emission type	**Mechanism**	**Includes**
Direct	Abrasion and wear and tear	Tyre, brake, clutch, road surface
	Corrosion	Vehicle, street furniture
Indirect	Re-suspension (due to tyre shear, wind and vehicle turbulence)	

Table 2: Key tracers used for non-exhaust PM

Reference	**Brake wear**	**Tyre wear**	**Re-suspension**
Adachi and Tainosho (2004)	Fe, Ba, Cu, Sb, Zr	Zn	-
Schauer et al. (2006)	Fe, Cu, Ba		-
Grieshop et al. (2006)	Cu, Sb, Ba and Ga		
Wahlin et al. (2006)	Cr, Fe, Cu, Zn, Zr, Mo, Sn, Sb, Ba and Pb	Al, Si, K, Ca, Ti, Mn, Fe, Zn and Sr (together with road dust)	
Tanner et al. (2008)	Cu, Cd	Zn	
Canepari et al. (2008)	Ba, Fe, Sb, Sr	-	-
Harrison (2009)	Ba, Cu	-	Al
Dongarra et al. (2009)	Cu, Mo, Sb	-	
Fabretti et al. (2009)	Cu, Zn, Sb, Sn (vehicular abrasion)	-	Rb, Sr, Mn, Fe, As
Keuken et al. (2010)	Cu	Zn	-
Bukowiecki et al. (2010)	Fe, Cu, Zn, Zr, Mo, Sn, Sb and Ba		
Pey et al. (2010)	-	-	Fe, Ca, Sb, Sn, Cu, Zn
Perez et al. (2010)	Sb, Cu, Ni, Sn (wear of brake, tire and other parts)		Fe
Amato et al. (2011a)	Fe, Cu, Zn, Cr, Sn, Sb	OC, S, Zn	Al, Ca, Fe, V
Apeagyei et al. (2011)	Fe, Ti, Cu, Ba	Zn, Ca, W, K, Fe, Ti, Cr, Mo	-
Duong and Lee (2011)	Ni, Cu	Zn	
Ondracek et al. (2011)	Cu, Ba, Fe, Zn	-	
Song and Gao (2011)	Sb, Cu, Fe, Pb	Zn, Co	
Sahu et al. (2011)	Zn (brake and tyre wear)	-	
Peltier et al. (2011)	-	-	Al, Si, Ti, Fe
Harrison et al. (2012b)	Ba, Cu, Fe, Sb	Zn	Si, Al

Table 3: Ratio of Cu to Sb characteristic of brake wear particles in ambient PM reported in the literature

Reference	**Location**	**Cu:Sb ratio**
Stechmann and Dannecker (1990)	Hamburg, Germany	5.6
Adachi and Tainosho (2004)	Kobe, Japan	1.3
Lin et al. (2005)	Taiwan	9.1±1.79 (coarse fraction) 2.17±0.83 (fine fraction)
Wahlin et al. (2006)	Copenhagen, Denmark	4.4±0.3
Hjortenkrans et al. (2007)	Stockholm , Sweden	5.3
Fabretti et al. (2009)	Nice, France	7.0±1.3
Dongarra et al. (2009)	Palermo, Italy	4.9 (coarse fraction) 2.5 (fine fraction)
Amato et al. (2011b)	Barcelona, Spain	8.0 ±1.5 (roadside) 6.8 ± 1.9 (urban)
Bukowiecki et al. (2010)	Zürich-Weststrasse, Switzerland	6.2 (coarse fraction)
Pey et al. (2010)	Barcelona, Spain	9 (coarse fraction) 10 (fine fraction)
Gietl et al. (2010)	London, UK	9.1

Table 4: (a) Results from mass size distribution analysis for traffic-related PM as reported in the literature (in µm)

Reference	Location	Site type	Component	Distribution type	Primary peak
Allen et al. (2001)	USA	Tunnel	PM_{10}	Unimodal	0.1-0.18
Hildemann et al.(1991)	USA	Freeway	PM	Trimodal	Ultrafine (<0.2), fine (0.2-1) and coarse (1-10)
Robert et al. (2007)	USA	Dynamometer	$PM_{1.8}$ (HDGV)	Unimodal	0.1-0.18
			$PM_{1.8}$ (LDGV)	Unimodal	0.1-0.18

(b) Results from mass size distribution analysis for traffic-related elements as reported in the literature (in µm)

Reference	Location	Metal(s)	Distribution type	Primary peak	Secondary peak
Stechmann and Dannecker (1990)	Germany	Cu, Ba	Unimodal	2.5	
Funasaka et al. (1998)	Japan (tunnel)	EC	Unimodal		
		OC	Bimodal	1	2-10
Lin et al., 2005	Taiwan	Si, Al, Fe, Ca	Bimodal	3.2- 5.6	Nano size range
		Ba	Bimodal	1.0-1.8	0.18-0.32
		Pb	Bimodal	3.2- 5.6	0.32- 0.56
		Zn	Bimodal	1.0-1.8	0.32-0.56
		Mn, Cu, Cd, Sr	Bimodal		
		Cr, V	Trimodal		
Lough et al. (2005)	USA	Cu	Bimodal	3.0	0.1
Wahlin et al. (2006)	Denmark	Ba, Sb	Unimodal	3	
		Cu	Bimodal	3	0.1
Hien et al. (2007)	Vietnam	PAHs in $PM_{2.5}$	Bimodal	Coarse and fine	
Riddle et al. (2007)	USA	Hopanes and steranes in $PM_{1.8}$	Unimodal	0.10-0.18	0.32 (vehicles without catalysts
Wang et al. (2009)	China	n-Alkanes, PAHs and Hopanes	Unimodal	0.7-1.1	
Gietl et al. (2010)	UK	Ba, Fe	Unimodal	1.2- 7.2	
		Cu, Sb	Bimodal	1.2- 7.2	0.2- 0.4
Huang et al. (2012)	China	n-Alkanes	Unimodal	0.32-0.56	
		PAHs	Unimodal	0.56-1.0	
		Hopanes	Unimodal	0.56-1.0	3.2 (in some cases)
Ondráček et al. (2011)	Czech Republic (freeway)	Fe, Cu, Mn, Zn	Unimodal	2.5	
		Si, Al, Ca and coarse mode K	Unimodal	5	
Song and Gao (~~2012~~2011)	USA	Al, Fe, Cu, Sb, Sc, Mn	Bimodal	3.2- 5.6	1.0-1.8
		Cd	Bimodal	0.18- 0.56	3.2- 5.6
		Pb, Zn	Trimodal	0.32-0.56	1.0-1.8, 3.2- 5.6
		Ni, V	Bimodal	0.18-0.32	3.2- 5.6

Table 5: Comparison between dynamometer and tunnel/roadway measurements (Allen et al., 2001; Imhof et al., 2005a; Phuleria et al., 2006; Phuleria et al., 2007; Handler et al., 2008; He et al., 2008; El Haddad et al., 2009; Sanchez-Ccoyllo et al., 2009; Franco et al., 2013)

	Dynamometer measurements	**Tunnel/Roadway measurements**
Testing conditions	Precise and controlled; different test cycles and driving conditions can be analysed; can include cold-start emissions	Ambient conditions- cannot be physically controlled; cannot include cold-start emissions Better known boundary conditions and dilution effects In some cases, atmospheric conditions in tunnel may not be the same as ambient environment
Representativeness for the vehicle fleet	Low, since the tests are conducted on a sub-set of the in-use fleet and variations in engine type, vehicle age and maintenance and mixing of emissions from different vehicles etc are not accounted for. Such studies do not account for particle aging effects	High, since the measurements are made in ambient environment and for in-use mixed fleet
Emission type accounted for	Exhaust	Exhaust and non-exhaust

Table 6: Emission factors as reported in the literature (selected studies)

- Particle Number

Reference	City, Country	Study type	Particle number EF
Gertler et al. (2002)	USA	Tunnel (mixed LDV and HDV)	5x 10^{12} particles/veh/km (low HDV) 2-3 x 10^{13} particles/veh/km (high HDV)
Abu-Allaban et al. (2002)	USA	Tunnel	5.2- 5.4 x 10^{13} particles/veh/km (low HD fraction) 2.1- 3.1x 10^{14} particles/veh/km (high HD fraction)
Jamriska et al. (2004)	Australia	Tunnel (diesel powered buses)	3.11 ± 2.41 x 10^{14} particles/km
Kittleson et al. (2004)	USA	Roadside (particles > 3 nm)	1.9- 9.9 x 10^{14} particles/km (gasoline-dominant fleet)
Jones and Harrison (2006)	UK	Roadside	5.84 x 10^{13} particles/veh/km 6.36 x 10^{14} particles/veh/km (LDV)
Westerdahl et al. (2009)	China	Roadside- LDV (prevalent during daytime)	1.8 x 10^{15} particles/kg fuel
		Roadside- HDV (prevalent during nighttime)	1.1 x 10^{16} particles/kg fuel
Wang et al. (2009)	Denmark	Highway	(215±15) 10^{12} particles/ veh/km
Ban-Weiss et al. (2010)	USA	Tunnel- LDV	(3.9±1.4) 10^{14} particles/kg fuel
		Tunnel-HDV	(3.3±1.3) 10^{15} particles/kg fuel
Wang et al. (2011)	China	Vehicle chase	7.1 x 10^{16} particles/kg fuel

- Particle Mass

Reference	City, Country	Study type	PM size range	EF
Weingartner et al. (1997)	Switzerland	Tunnel (mixed LDV and HDV with more LDV)	PM_3	310 mg/km (63% vehicles running on diesel)
Gillies et al. (2001)	USA	Tunnel (mixed LDV and HDV)	$PM_{2.5}$	0.052 ± 0.027 g/km
			PM_{10}	0.069 ± 0.030 g/km
Gertler et al. (2002)	USA	Tunnel (mixed LDV and	$PM_{2.5}$	0.062 ± 0.042 g/km

		HDV)	PM_{10}	0.087 ± 0.054 g/km
Jamriska et al. (2004)	Australia	Tunnel (diesel powered buses)	$PM_{2.5}$	583± 451 mg/km
Greishop et al. (2006)	USA	Tunnel	$PM_{2.5}$ (rush hour)	189±23 mg/kg fuel
			$PM_{2.5}$ (mid-day)	158±29 mg/kg fuel
			$PM_{2.5}$ (early morning-HDV)	437±76 mg/kg fuel
Handler et al. (2008)	Austria	Tunnel	TSP	129±45 mg/veh/km
			PM_{10}	62±18 mg/veh/km
			$PM_{2.5}$	26±10 mg/veh/km
Sanchez-Ccoyllo et al., (2009)	Brazil		Coarse	127 mg/km
			Fine	92 mg/km
Wang et al. (2010)	Denmark	Highway	$PM_{2.5}$	29 mg/veh/km
Chiang and Huang (2009)	Taiwan	Freeway tunnel	$PM_{2.5-10}$	18±6.5 mg/veh/km
			$PM_{2.5}$	39±11 mg/veh/km
Cheng et al. (2010)	Hong Kong	Tunnel, roadside sites and urban site	$PM_{2.5}$	257±31 mg/veh/km
Bukowiecki et al. (2010)	Switzerland	Roadside	PM_{10}- LDV	24±8 mg/veh/km
			PM_{10}-HDV	498±86 mg/veh/km
Wang et al. (2011)	China	Vehicle chase	$PM_{0.5}$	2.35 g/kg fuel
Mancilla and Mendoza (2012)	Mexico	Tunnel	PM_{10}	22.8±7.4 mg/veh/km
Chen et al. (2013)	China	Tunnel (mixed with higher proportion of gasoline vehicles)		687 mg/veh/km (summer) 714 mg/veh/km (winter)

- LDV and HDV vehicles (mass)

Reference	**Country**	**Study type**	**PM size range**	**LDV EF**	**HDV EF**
Weingartner et al. (1997)	Switzerland	Tunnel	PM_3	8.53 ± 0.47 mg/km	383.5 ± 10.7 mg/km
Gertler et al. (2002)	USA	Tunnel (mixed LDV and HDV)	$PM_{2.5}$	0.014 ± 0.013 g/km	0.135 ± 0.018 g/km
			PM_{10}	0.010 ± 0.011 g/km	0.181 ± 0.013 g/km
Abu-Allaban et al.	USA	Roadside (LD-Spark	PM10	10-70 mg/km/vehicle	60-570 mg/km

(2003)		ignition and HD-diesel)	$PM_{2.5}$	10-50 mg/km/vehicle	60-480 mg/km
Chiang et al. (2012)	Taiwan	Dynamometer	$PM_{2.5}$	0.172 g/km (diesel)	
Oanh et al. (2010)	Bangkok	Dynamometer	$PM_{2.5}$	230 mg/km	176 mg/km
Jones and Harrison (2006)	UK	Roadside	PM_{10}	0.033±0.006 g/veh/km	0.370±0.032 g/veh/km
			$PM_{2.5}$	0.010±0.004 g/veh/km	0.179±0.022 g/veh/km
Liacos et al. (2012)	USA	Freeway	$PM_{2.5}$	151189.3 mg/km/h (low HDV)	169629.5 mg/km/h (high HDV)

- Non-exhaust emissions

Reference	**City, Country**	**Study type**	**PM size range**	**Road dust**	**Brake wear**	**Tyre wear**
Abu-Allaban et al. (2003)	USA	Roadside (LD-Spark ignition and HD-diesel)	PM_{10} LDV	40-780 mg/km/vehicle	0-80 mg/km/vehicle	-
			PM_{10} HDV	230-7800 mg/km/vehicle	0-610 mg/km/vehicle	
			$PM_{2.5}$ LDV	2-25 mg/km/vehicle	0-5 mg/km/vehicle	-
			$PM_{2.5}$ HDV	15-300 mg/km/vehicle	0-15 mg/km/vehicle	
Bukowiecki et al. (2010)	Switzerland	Roadside	PM10	27 mg/vehicle/km	15 mg/ vehicle /km	
Amato et al. (2012)	Spain	Freeway	PM_{10}	22.7 ± 14.2 mg/vehicle kilometer travelled	-	-

Table 7: Source apportionment analyses for road traffic-generated PM (selected studies)

Reference	Country	Method	Particle type	Sources
Lin et al. (2005)	Taiwan	PCA	Coarse particles	Fugitive dust and brake lining, diesel, fuel oil, gasoline
			Fine particles	Gasoline, diesel, brake lining/tyre wear, fuel oil
			Ultrafine and nano particles	Gasoline, diesel, industry, fuel oil
Furujso et al. (2007)	Stockholm, Sweden	PMF	PM_{10}	Resuspension Vehicle derived (brake wear) Road salt Regional combustion Long range transport
Chan and Mozurkewich (2007)	Ontario, Canada	Absolute PCA	Particle number distribution with trace gas and meteorological data	Common sources across 3 sites: Nucleation particles (photochemically produced), regional pollution factor, boundary layer dynamics factor Specific sources at the three sites: Local industrial emissions (urban) Processed nucleation mode particles (polluted rural) Transported fine particles (downwind Toronto)
Amato et al. (2009)	Barcelona, Spain	PMF2 with ME	PM_{10} and $PM_{2.5}$	Road dust factor accounted for 17% PM10, 8% $PM_{2.5}$ and less than 2% of PM_1. All traffic sources contributed to 46% PM10, 51% $PM_{2.5}$ and 48% of PM_1.
Fabretti et al. (2009)	Nice, France	PMF	$PM_{2.5}$	Vehicle abrasion: Cu, Zn, Sb (36%) Re-suspension: Mn, Fe, As, Rb, Sr (43%) Fuel combustion: V, Ni, Co (21%)
Dreyfus et al. (2009)	Wilmington, USA	PMF	Organic carbon	Diesel factor: Alkyl fragment, benzothiazole, benzenedicarboxylic acids/3,4-dimethoxybenzaldehyde, 3,4-dimethoxybenzoic acid, hopanes Road dust/car emissions: hopanes, benzothiazole and triacontanoic acid
Mancilla and Mendoza (2012)	Monterrey, Mexico	FA	$PM_{2.5}$	Brake emissions Exhaust emissions
Ondracek et al. (2011)	Prague, Czech Republic			Abrasion of vehicle parts: Fe, Cu, Mn, Zn Re-suspension of road dust: Si, Al, Ca Long-range transport/regional background: Ca, K
Song and Gao (2011)	New Jersey, USA	FA	PM	Brake wear and fuel combustion: Fe, Sb, Pb, Cd (~35%) Primary fuel combustion: Cr, Ni, V, Cu (~28.3%) Tyre wear and fuel combustion: Zn, Co (~23.7%)
Yin et al. (2010)	United Kingdom	CMB	$PM_{2.5}$	Vegetative detritus; woodsmoke; natural gas; diesel engines (11.6%); gasoline engines (1.4%); smoking engines (9.7%); coal; dust/soil; secondary organic matter; sea salt; ammonium sulphate; ammonium nitrate

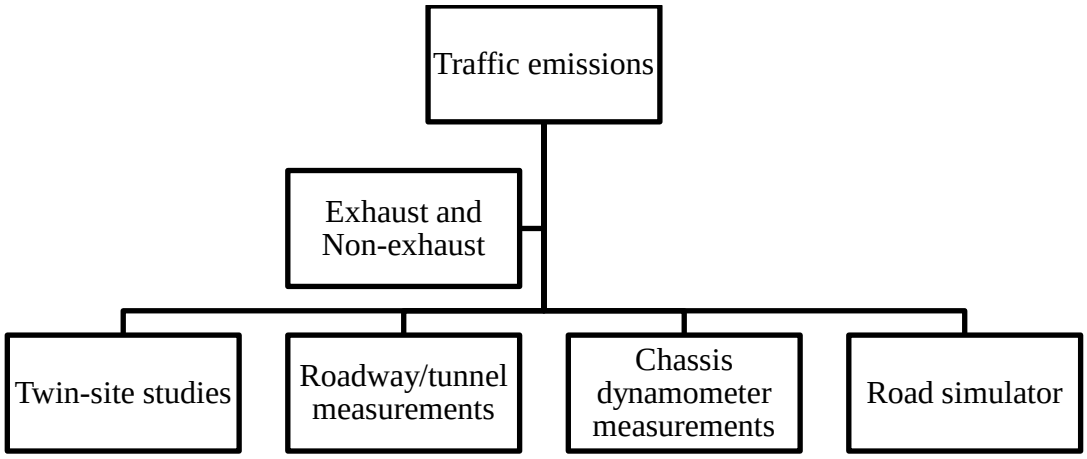

Figure 2: Methods for direct road traffic emissions analyses