UNIVERSITY OF BIRMINGHAM

University of Birmingham
Research at Birmingham

Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements

Pant, P.; Harrison, R.M.

Document Version
Early version, also known as pre-print

Link to publication on Research at Birmingham portal

1

Estimation of the Contribution of Road Traffic

Emissions to Particulate Matter Concentrations

from Field Measurements: A Review

5

Pallavi Pant and Roy M. Harrison[*][1]

7

Division of Environmental Health & Risk Management,

School of Geography, Earth & Environmental Sciences

University of Birmingham, Edgbaston

Birmingham B15 2TT

United Kingdom

13

[*] 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

14 **ABSTRACT**

15 Road traffic is one of the main sources of particulate matter in the atmosphere. Despite its

16 importance, there are significant challenges in quantitative evaluation of its contribution to airborne

17 concentrations. This article first reviews the nature of the particle emissions from road vehicles

18 including both exhaust and non-exhaust (abrasion and resuspension sources). It then briefly

19 reviews the various methods available for quantification of the road traffic contribution. This

20 includes tunnel/roadway measurements, twin site studies, use of vehicle-specific tracers and other

21 methods. Finally, the application of receptor modelling methods is briefly described. Based on the

22 review, it can be concluded that while traffic emissions continue to contribute substantially to

23 primary PM emissions in urban areas, quantitative knowledge of the contribution, especially of non-

24 exhaust emissions to PM concentrations remain inadquate.

25

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

27

2

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]

78   3.   What have we learnt so far?

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

80

81   The literature search for this review was primarily performed using Web of Knowledge, Science

82   Direct and Google Scholar using various combinations of keywords such as traffic emissions, non-

83   exhaust/exhaust emissions, resuspension, road dust, traffic PM etc. However, the authors

84   acknowledge that not all articles published on the topic have been included in the review.

85   **2.   NON-EXHAUST EMISSIONS**

86   Non-exhaust PM comprises the various emissions that do not derive from the tailpipe of a vehicle

87   including particles generated due to brake and tyre wear, road surface abrasion, wear and

88   tear/corrosion of other vehicle components such as the clutch, and re-suspension of road surface

89   dusts (Table 1). Key reasons for needing to understand non-exhaust emissions include their inherent

90   toxicity including their tendency to act as carriers of heavy metals and carcinogenic components

91   (Adachi and Tainosho, 2004; Hjortenkrans et al., 2007; Johansson et al., 2009; Amato et al., 2011a)

92   and their contribution to exceedences of air quality guidelines and standards (Amato et al., 2011a;

93   Denier van der Gon, 2013).

94   Harrison et al. (2012b) showed that the size distributions of trace metals were indicative of particle

95   sources. Keuken et al. (2010) concluded that re-suspension of accumulated PM and road wear

96   related particles are the primary contributors to non-exhaust emissions, and tyre wear and brake

97   wear contribute to the fine and coarse fraction respectively. However, Narvaez et al. (2008)

98   reported that although a majority of abrasion particles are in the coarse fraction, abrasion can

99   contribute significantly to the fine fraction of PM. In Taiwan, EFs for elements such as Ba and Se

100   were found to be higher in the $PM_{2.5-10}$ fraction indicating mechanical friction and brake wear as

101   potential sources (Chiang and Huang, 2009). Road traffic was found to be a key contributor to

102   fine/ultrafine and nano mode particles of Ba, Zn and Pb (Lin et al., 2005) and in Barcelona (Spain),

103   62-96% of elements including Cu, Sb, Ba, Mn and Zn were found to be present in the PM_{1-10}

104   fraction (Perez et al., 2010). A detailed review of the nanoparticle emissions from non-exhaust

105   sources including traffic and non-traffic sources is presented in Kumar et al. (2013).

106   Non-exhaust emissions are typically characterized by trace metals (e.g. Cu, Zn, Ba, Sb, Mn)

107   although organic markers (e.g. polycyclic aromatic hydrocarbons (PAHs), n-alkanes) have been

108   used in some cases (Wahlin et al., 2006; Amato et al., 2009; El Haddad et al., 2009; Gietl et al.,

109   2010; Oliviera et al., 2011; Kwon and Castaldi, 2012). A list of key tracers used for source

110   characterization of non-exhaust emissions is presented in Table 2. However, emissions of trace

111   metal markers are reported to vary with the fleet composition, with higher emissions reported for

112   some of the elements for heavy duty vehicles (HDVs) (Grieshop et al., 2006; Mancilla and

113   Mendoza, 2012). In addition, the profile of trace metal concentrations in non-exhaust particulate

114   matter is unique for every region and varies based on parameters such as traffic volume and pattern,

115   vehicle fleet characteristics, driving and traffic patterns and climate and geology of the region

116   (Omstedt et al., 2005; Amato et al., 2011a,b; Duong and Lee, 2011). Another important aspect is the

117   variability of tyre and brake composition depending on the manufacturer which makes it very

118   difficult to ascertain fleet-wide composition other than from environmental measurements

119   (Canepari et al., 2008; Gietl et al., 2010; Denier van der Gon et al., 2013).

120
121   Several factors are reported to affect non-exhaust emissions including increase in vehicle speed

122   (Chen et al., 2006; Gustafsson et al., 2008; Hussein et al., 2008; Mathissen et al., 2011) although

123   Keuken et al. (2010) reported a non-linear relationship between traffic volume and non-exhaust

124   emissions. Metal emissions due to road dust show a low correlation to metal emissions due to

125   abrasion/brake wear and combustion (Johansson et al., 2009). One of the major problems in

126   analysis of non-exhaust PM using field data has been the difficulty in distinguishing between wear

127   and tear emissions and road dust since the chemical composition is often very similar (Bukowiecki

128   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

7

154 (Adachi and Tainosho, 2004; Hjortenkrans et al., 2007; Ondracek et al., 2011). The concentration of

155 Zn was found to be approximately 15 times higher in tyres compared to brakes while concentrations

156 of other heavy metals such as copper (Cu) and barium (Ba) were higher for brake materials in

157 Massachusetts (USA) (Apaegyei et al., 2011). Differences have also been observed between tyre

158 tread composition (primarily Zn) and tyre wear (Al, Si, Ca, Fe, Zn, Ti) (Adachi and Tainosho,

159 2004). However, the spatial and temporal trend for Zn was found to be different from other roadside

160 tracers in New York (USA), and a very small portion of airborne Zn concentration was attributed to

161 tyre wear (Peltier et al., 2011). However, Zn is emitted from brake wear, motor oil and other

162 sources and cannot be used as the only tracer for tyre wear (Lough et al., 2005). Benzothiazoles are

163 also used as markers for tyre wear, particularly benzothiazole (BT), 2-hydroxy benzothiazole

164 (HOBT), 2-(4-morpholinyl)benzothiazole (24MoBT) and N-cyclohexyl-2-benzothiazolamine

165 (NCBA) (Kumata et al., 2002; Allen et al., 2006; Wik and Dave, 2009). Among the PAHs, pyrene,

166 benzo(ghi)perylene, fluoranthene and phenanthrene are known to be emitted from tyres

167 (Boonyatumanond et al., 2007; Kwon and Castaldi, 2012). Aatmeeyata and Sharma (2010) reported

168 that small cars tested in India emitted 378 ng of total PAH/tyre/km while in the USA, 200 µg/g of

169 PAHs were found in tyre wear with pyrene, fluoranthene and phenanthrene being present in the

170 highest concentrations (Rogge et al., 1993). Tyre tread wear has also been found to contain an

171 average of 0.53 µg/g of dibenzopyrenes, indicating that tyre dust may be significant source of

172 dibenzopyrenes in the environment (Sadiktis et al., 2012).

173
174 Average tread wear for tyres is reported to be between 0.006-0.009 g/km based on the road, tyre and

175 vehicle conditions (Rogge et al., 1993) and tyres can lose up to 10% of their mass during their

176 lifetime (Milani et al., 2004). Speed has been reported as an influential parameter for particle mass

177 and number concentrations for tyre wear emissions (Gustafsson et al., 2008; Mathissen et al., 2011).

178 Other important parameters for tyre tread wear include road surface type, and tyre and driving

179 conditions (Allen et al., 2006; Gustafsson et al., 2008). For example, asphalt surfaces have been

180   reported to cause less tyre wear than concrete pavements and in Arizona, USA, the emission rate for

181   tyre wear was found to be 1.4-2 times lower for asphalt pavement compared to concrete pavement

182   (Allen et al., 2006). The type of tyre also impacts the magnitude of tyre wear emissions and studded

183   tyres are known to cause more emissions compared with summer and friction tyres (Kupiainen et al.,

184   2005; Gustafsson et al., 2008; Hussein et al., 2008; Schaap et al., 2009). Unimodal (70-90 nm) and

185   bimodal (< 10 and 30-60 nm) peaks in the nano size range have been reported for tyre particles by

186   number under low and high speed conditions respectively (Mathissen et al., 2011).

187

188   2.2   Brake Wear

189   Brake wear, including abrasion of brake lining material and brake discs, caused by grinding of

190   brake pad constituents (coarse range particles) or volatilization and condensation of brake pad

191   materials (fine range particles), is known to release PM directly into the atmosphere and to

192   contribute to the trace metal concentration in airborne PM, particularly less than 10 µm (Garg et al.,

193   2000; Blau and Meyer, 2003; Ingo et al., 2004; Wahlin et al., 2006; Gietl et al., 2010; Varrica et al.,

194   2012). Key components of brake pads include fillers, frictional additives, reinforcing fibres and

195   binder (Chan and Stachowiak, 2004) and the key chemical species used include sulphides of metal,

196   abrasives (e.g. silica), barium silicate/sulphate (particularly in brake linings) and other metallic

197   particles (as filler material), carbon fibres and lubricant (e.g. graphite) (Ingo et al., 2004; Dongarra

198   et al., 2009). Chan and Stachowiak (2004) present a detailed review of the chemical constituents of

199   brake pads. Garg et al. (2000) observed an average mass median diameter (median particle

200   diameter based on mass) of 1.49 µm for brake wear particles using tests on a brake dynamometer

201   whereas Sanders et al. (2003) reported a mass median diameter of 6 µm for brake debris generated

202   during urban driving conditions. Higher brake wear related emissions have been reported during

203   rush hour . A higher number of particles are released in the braking phase as compared to the

204   acceleration phase (Hussein et al., 2008; Mathissen et al., 2011) and various studies have

205   corroborated this observations including Greishop et al. (2006) who reported a higher particle

9

206 number concentration during rush hour where the traffic often operates in the stop-and-go mode and

207 Abu-Allaban et al. (2003) who observed higher contribution of brake wear at freeway exit sites

208 compared to other types of roadside sites (Abu-Allaban et al., 2003).

209 Passenger cars have been estimated to emit nearly 44 g/car/year brake dust (Iijima et al., 2007).

210 Sanders et al. (2003) reported high concentrations of Fe, Ba and Cu in brake lining wear using

211 dynamometer and track tests whereas Adachi and Tainosho (2004) reported Fe to be the most

212 abundant metal in brake dust along with other metals such as Ba, Cu, Sb, Zr and Zn, and

213 Hjortenkrans et al. (2007) reported Cu, Zn and Sb to be present in brake wear emissions. Varrica et

214 al. (2012) undertook a detailed study on Sb in brake dust and reported the most commonly released

215 forms of Sb due to brake abrasion to be Sb (III) and Sb (V). However, brake pads are often found to

216 have different composition based on the brand, particularly with respect to Cu and Sb (Hjortenkrans

217 et al., 2007; Canepari et al., 2008). Polyalkylene glycol ethers are also reported to be present in

218 brake wear particles with small concentrations of n-alkanes and n-alkanoic acids (Rogge et al.,

219 1993).

220
221 Iijima et al. (2007) reported a unimodal number-based distribution for brake abrasion dust with the

222 mode at 1-2 µm while the mass-based distribution had a peak at 3-6 µm. EFs for brake wear have

223 been reported by several studies including Garg et al. (2000) who reported a brake wear EF of 3-9

224 mg/km for light duty gasoline vehicles (LDGV) and Abu-Allaban et al. (2003) who reported an EF

225 of 0-80 mg/km. Wahlin et al. (2006), on the other hand, reported a Cu emission factor of 0.7± 0.2

226 mg/km/vehicle in Copenhagen.

227 Sternbeck et al. (2002) proposed the ratio of Cu:Sb (4.6 ± 2.3) as characteristic of brake wear

228 particles and Table 3 lists the Cu: Sb ratios as reported in the literature. Differences in the ratios can

229 be attributed to the difference in the brake pad composition, and contributions of metals from other

230 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

256 aggregate using the *sandpaper effect*, where the aggregates used to prevent skidding generate PM_{10}

257 particles and lead to further particle emissions from the pavement.

258

259 Pristane, phytane, hopanes, steranes in addition to unresolved complex mixture (UCM) were

260 reported in road dust particles which were found to be enriched in biogenic component by Omar et

261 al. (2007). High concentrations of phenanthrene, fluoranthene and pyrene along with high

262 molecular weight (4-7 ring) PAHs were reported in road dust samples in Taiwan (Fang et al., 2004),

263 China (Liu et al., 2007; Han et al., 2009) and Egypt (Hassanien and Abdel-Latif, 2008) respectively.

264 The spatial distribution of PAHs was reported to be affected by wind direction Liu et al. (2007) and

265 total organic carbon content by Liu et al. (2007) and Han et al. (2009). Mono- (α- and β-glucose)

266 and disaccharides (sucrose and mycose) were used as markers for road dust re-suspension for

267 source apportionment by Omar et al. (2007).

268 Han et al. (2007) analysed suspended dust in Beijing and reported concentrations of elements such

269 as Ca, S, Cu, Zn, Ni, Pb, and Cd to be much higher than the crustal abundances. Cu, Zn, Ni, and Pb

270 were attributed to traffic emissions together with coal burning, and ions such as Ca^{2+}, SO_4^{2-}, Cl^-,

271 NO_3^-, K^+, and Na^+ were also found to be present in re-suspended dust. Luo et al. (2011) observed

272 that the concentrations of trace metals contributed by vehicle exhaust and tyre abrasion, i.e. Pb, Zn

273 and Cu have been reported to be much higher in roadside samples than the concentrations of these

274 elements in background soils in China. Wei and Yang (2010) analysed urban soil, urban road dust

275 and agricultural soil in Chinese cities and found relatively higher concentrations of Cd, Cu, Pb and

276 Zn in urban soil samples and Cu, Cr, Pb, Zn, Ni and Cd in urban road dusts. The high

277 concentrations of metals were attributed to anthropogenic sources - road traffic and industrial

278 emissions. Higher concentration levels for several trace elements were reported for roadside dust

279 compared to dust collected at a distance of 100 m in Bosnia, and particularly high concentrations

280 were reported for Hg and Co and were attributed to vehicle exhaust and brake pads respectively.

281 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

17

410   Similar observations regarding mass-based size distributions for trace metals were reported by

411   Samara and Voutsa (2005) for a traffic site in Greece. Metals such as Ba, Pb and Zn were found to

412   exhibit bimodal distributions whereas some others such as Cr and V showed trimodal distributions

413   in Southern Taiwan (Lin et al., 2005). The presence of an Aitken mode in the ultrafine range may

414   be indicative of a contribution from vehicular emissions, both diesel and gasoline. Hays et al. (2011)

415   reported erratic mass size distributions for various elements including Ni, Cu, Cd, and Zn and

416   attributed this to a range of emission sources near highways in North Carolina (USA). A peak was

417   observed at 80 nm for soot particles in a street canyon corresponding to diesel engine emissions in

418   Leipzig, Germany and the concentration of soot particles was reported to be influenced by traffic

419   volume (Rose et al., 2006)

420

421   **4.2   Number-Size Distributions**

422   Ultrafine particles are characterized as particles with diameter less than 100 nm diameter and are

423   typically measured as particle number concentration (PNC) (Charron and Harrison, 2009). Such

424   particles may have a short lifetime and as a result, the spatial variability of the particles differs

425   based on proximity to the emission source (Puustinen et al., 2007; Cyrys et al., 2008; Charron and

426   Harrison, 2009). Traffic has been reported as one of the major sources of UFPs (Abu-Allaban et al.,

427   2002; Nanzetta and Holmen, 2004; Westerdahl et al., 2009) and particle number is linked to fresh

428   vehicle exhaust emissions (Rodriguez et al., 2007). Number size distributions are heavily influenced

429   by proximity to traffic (Rose et al., 2006; Kerminen et al., 2007; Beddows et al., 2009; Keuken et

430   al., 2012). As a consequence of this, concentrations of organic compounds have also been reported

431   to be highly variable for near-highway sites (Sun et al., 2012. Highest PNC has been reported

432   during morning rush hours in Germany (Wehner et al., 2002) and Spain (Perez et al., 2010). A

433   significant difference has also been reported between weekdays and weekends, highlighting the

434   influence of traffic (Morawska et al., 2002; Wehner et al., 2002; Van Poppel et al., 2012). A

435   significant difference was observed in the particle number size distribution in the Kingsway tunnel

18

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

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

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

488 For traffic emissions, distance from the road is also reported to cause changes in PNC due to

489 processes other than dilution (Zhu et al., 2006). This feature is of considerable interest; particularly

490 from an epidemiological perspective as this would mean that exposure from near-roadway sites can

491 be much different from other sites. HEI (2010) reported the key exposure zone for traffic emissions

492 to be within 300-500 m of a roadway. In Los Angeles, both PNC and size distribution changed with

493 an increase in distance from the road, particularly for particles with a diameter less than 30 nm and

494 it has been suggested that this drastic change in PNC is caused by coagulation and atmospheric

495 dilution (Zhu et al., 2002a). Padro-Martinez et al. (2012) found PNC to be at least two times higher

496 at 0-50 m from the highway compared to further distances while Zhu et al. (2002b) observed the

497 PNC to be 25 times higher near the freeway compared to background locations. Rose et al. (2006)

498 also observed a decrease in soot particle number concentration with increasing distance from the

499 street canyon which was attributed to coagulation processes. Kittleson et al. (2004) observed a

500 decline in the nuclei mode particles as the distance from the highway increased and recently,

501 Canagaratna et al. (2010) also reported a decrease in PNC with increasing distance away from the

502 roadway. Dilution processes will inevitably cause a reduction in PNC with distance from the road,

503 as they do for other traffic-generated pollutants (e.g. NO_x, BC, etc.). Coagulation causes an

504 increase in particle size, and a decrease in number, but is unlikely to be a significant influence close

505 to the road due to an insufficient number concentration to drive the process, and short travel times.

506 An observed increase in particle diameters with distance from a road is most probably the result of

507 mixing of traffic emissions with a coarser background aerosol leading to a large modal diameter

508 (Shi et al., 1999). The nucleation mode traffic particles are semi-volatile and this can lead to shifts

509 in size distribution (typically to smaller sizes) and particle loss due to evaporation (Dall'Osto et al.,

510 2011; Harrison et al., 2012a).

511
512 Charron and Harrison (2003) reported that increasing wind speeds and lower temperatures can

513 increase the abundance of smaller nucleation mode particles (diameter of sub-30 nm) and this could

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

540   for PNC and PM_{10} concentration. Similarly, Padro-Martinez et al. (2012) also reported a similar

541   spatial trend for PNC, NO_x, particulate PAHs and BC in the USA while a different trend was

542   reported for $PM_{2.5}$.

543

544   Thus, key factors that influence the particle number concentration measurements include wind

545   speed, traffic density and the distance from source at which the measurements are made. Nanzetta

546   and Holmen (2004) observed differences in the PNC across different seasons and concluded that

547   results for one season cannot be used for generalization of PNC.

548

549   **5.   METHODOLOGIES FOR ASSESSMENT OF TRAFFIC EMISSIONS**

550   There are several ways of evaluating emissions from road vehicles including tests with

551   dynamometers, measurements in tunnels/highways and near roadside measurements and road

552   simulator tests (Figure 1) (Mclaren et al., 1996; Phuleria et al., 2007; Handler et al., 2008;

553   Gustafsson et al., 2008; El Haddad et al., 2009; Yan et al., 2009; Sjodin et al., 2010). Table 5

554   presents a qualitative description of measurements using dynamometers under lab conditions and

555   tunnel/roadway/twin-site studies under ambient conditions. In recent years, significant differences

556   have been observed between laboratory-tested and real-world mixed source traffic emissions

557   (Gertler et al., 2002; Yan et al., 2009; Ancelet et al., 2011). Near roadside/kerbside, traffic tunnel

558   and highway measurements are reported to be more realistic since they represent mixed-fleet

559   emissions under real-world driving conditions (Phuleria et al., 2007). Beddows and Harrison (2008)

560   compared particle number EFs measured in dynamometer studies with those estimated in roadside

561   studies, finding generally good agreement overall. However, Canagaratna et al. (2010) reported the

562   EFs calculated using on-road measurements to be nearly 50% lower than EFs measured using a

563   dynamometer.

564

565 An emission factor is defined as "the mass (or number) of pollutant released per unit time/distance

566 travelled or mass of fuel used" and typically increases with vehicle speed for LDVs as well as

567 HDVs (Jones and Harrison, 2006). Characterization of emissions from road vehicles includes

568 analysis of size distributions and chemical composition (organic, inorganic and trace metal) and can

569 lead to generation of EFs. EFs typically depend on conditions at the sampling site, traffic intensity

570 and modal shares of HDVs and LDVs (Staehelin et al., 1995; Imhof et al., 2005a). A recent review

571 article on development of emission factors delves into this issue in greater detail (Franco et al.,

572 2013).

573
574 EFs can be measured using a number of different methods including dynamometer studies (Oanh et

575 al., 2010; Chiang et al., 2012), roadway tunnel studies (Gertler et al., 2002; Handler et al., 2008;

576 Mancilla and Mendoza, 2012), on-road chase experiments (Wang et al., 2011) and remote sensing

577 (Zhang et al., 1995). Phuleria et al. (2007) reported overall agreement between EFs determined at

578 tunnel and roadway sites in USA.

579
580 Two of the most common methods for EF expression are fuel-based EF (Ban-Weiss et al., 2010;

581 Wang et al., 2011) and vehicle kilometre travelled (VKT)-based calculation (Abu-Allaban et al.,

582 2002; Kittleson et al., 2004; Jones and Harrison, 2006; Amato et al., 2012). Phuleria et al. (2006,

583 2007) and Bukowiecki et al. (2010) describe the methodology of EF derivation in greater detail.

584 Fuel-based EF can be defined as "grams of pollutant emitted for every kilogram of fuel burned" and

585 can be calculated for various pollutants using CO_2 and CO as a measure of fuel burned (Liacos et al.,

586 2012). The concentration of pollutant with respect to the concentration of CO_2 + CO can be used to

587 calculate the fuel burnt using the following formula (Miguel et al., 1998; Liacos et al., 2012):

$$EF_P = 10^6 \times \left(\frac{\Delta P}{\Delta CO2 + \Delta CO} \right) w_c \qquad (1)$$

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

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:

$$EF_x = \frac{\Delta Concentration_x \times D}{n_{total}} \qquad (3)$$

600 where $\Delta Concentration_x$ 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):

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

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

27

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

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

685　sites, the data generated is representative only of that part of the total vehicle fleet that was sampled

686　(Liacos et al., 2012).

687

688　Harrison et al. (2003) concluded that the trace element enrichment observed at roadside sites is

689　contributed largely by wear and tear of tyres and brakes. Significant roadside enrichment has been

690　reported for elements, ions and organic compounds in various cities including London (UK) (Gietl

691　et al., 2010) Los Angeles (USA) (Ntziachristos et al.,20082007), Sicily, Rome and Milan (Italy)

692　(Manno et al., 2006; Canepari et al., 2008; Perrone et al., 2012), Barcelona (Spain) (Amato et al.,

693　2011), Portugal (Oliviera et al., 2011) and Queensland (Australia) (Gunawardana et al., 2011). . In

694　New York City, Fe, Al and Si were found to contribute between 0.5- 1.0 µg/m^3 to the ambient

695　$PM_{2.5}$ loading (Peltier et al., 2011) whereas Lin et al., 2008 reported Ca, Al and Fe to constitute

696　between 65-77% of suspended particles in Taiwan. Nearly 90% of the road traffic emissions of Cu

697　were reported to be due to brake wear in Stockholm (Sweden) (Johansson et al., 2009). Amato et al.

698　(2011b) analyzed roadside PM_{10} enrichment (RE) of trace elements in Barcelona (Spain) reported

699　more than 70% roadside enrichment ratio for key traffic tracers including EC, Fe, Ba, Cu, Sb, Cr

700　and Sn. Gietl et al. (2010) estimated that based upon published emission factors brake dust

701　comprises nearly 13% of the total PM at a roadside site in London (UK) and Amato et al. (2011b)

702　reported approximately 60% enrichment of brake wear particles in a roadside location compared to

703　an urban background location in Barcelona (Spain). Pey et al. (2010) reported that the

704　concentration of EC was found to be 3-8 times higher for the urban site in comparison with the

705　background site in Barcelona (Spain) and Perrone et al. (2012) found EC to be contributing 11-15%

706　of $PM_{2.5}$ in urban areas compared to 3-5% at a rural site and 1-2% at the background site in Milan

707　(Italy). Vertical concentration gradients have also been reported for trace elements with highest

708　concentrations at kerbside as compared with rooftop (Johansson et al., 2009).Analysis of size-

709　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 jth species in the ith sample, f_{pj} is the concentration of

775 the jth species in material emitted by the source p, g_{ip} is the contribution of the pth source to the ith

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

782   In recent years, there have been many studies for source apportionment of road traffic-generated

783   PM, some of which appear in Table 7. There have also been studies that have used both CMB and

784   factor analysis models (Larsen and Baker, 2003; Bullock et al., 2008).

785

786   **6.1   Multivariate Statistical Methods**

787   There are a number of different methods based on factor analysis including PCA, PMF, UNMIX

788   and ME. Such methods do not require a priori information about source emission characteristics and

789   are useful in cases where relevant source profiles are not available (Hopke, 2003; Viana et al.,

790   2008). In the simplest matrix form, the equation 6 can be represented as (Hopke, 2003)

$$X = GF'$$

791   A number of studies have been conducted using the different factor analysis methods and studies in

792   different locations have often reported different magnitudes of exhaust and non-exhaust emissions.

793   In Taiwan, gasoline, diesel and fuel oil emissions together with dust and brake wear were found to

794   be key contributors to traffic-generated PM using PCA (Lin et al., 2005). Using COPREM, Wahlin

795   et al. (2006) apportioned the local traffic emissions to exhaust, road dust, brake wear and winter

796   salting in Copenhagen (Denmark). Amato et al. (2009) analysed road dust in Barcelona (Spain)

797   using ME-2, and re-suspension was found to contribute 37%, 15% and 3% of total traffic emissions

798   respectively in PM_{10}, $PM_{2.5}$ and PM_1 respectively. Other identified sources included secondary

799   aerosols and a soil source. Sjodin et al. (2010) found road wear to be the most important source of

800   PM_{10} emissions in streets as well as background locations in Sweden, which is probably a reflection

801   of road sanding and use of studded tyres. Using PMF, three distinct traffic sources including vehicle

802   abrasion (36%), re-suspension (43%) and fuel combustion (21%) were resolved for traffic sources

803   using trace metals in Nice (France) from data collected in a road tunnel (Fabretti et al., 2009). Based

804   on PMF analysis in Switzerland, Bukowiecki et al. (2010) reported 21%, 38% and 41% of the

805   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

831   by Cheng et al. (2011) in Hong Kong where traffic emissions and re-suspended dust emissions were

832   found to contribute 38% and 5% using PMF.

833   Dreyfus et al. (2009) conducted PMF analysis with time-resolved organic molecular marker data

834   and identified diesel and road dust/car emissions factors which accounted for 2/3rd of the total OC

835   concentration. Receptor modelling analysis for PM$_{2.5}$ was conducted using PMF and UNMIX, and

836   PMF distinguished between diesel and gasoline engine emissions using temperature resolved

837   carbon fractions (Sahu et al., 2011). Pey et al. (2009) performed source apportionment using PCA

838   and regression with a large dataset including information on particle number concentration; size

839   distribution and chemical composition for an urban background site in Barcelona, and road traffic

840   was identified as the most significant source (52- 86%) for aerosols in the sub-micrometer range.

841   Liu et al. (2007) attributed the PAHs in road dust in Shanghai (China) to traffic emissions and coal

842   combustion using principal component analysis (PCA) and compositional analysis. Using the same

843   technique, Han et al. (2009) attributed PAHs in road dust in Anshan (China) to emissions from

844   traffic, coal combustion, steel industry and cooking. Using MLR, Boonyatumanond et al. (2007)

845   reported tyres, diesel vehicle soot and used engine oil to be the key sources for PAHs found in street

846   dust in Bangkok (Thailand). Hassanien and Abdel-Latif (2008) attributed PAHs in road dust in

847   Cairo (Egypt) to emissions from traffic, industry and incomplete combustion of waste based on

848   correlation analysis. Teixeira et al. (2012) reported vehicular emissions and coal and wood

849   combustion to be the key sources for PAHs in Porte Alegre (Brazil) using PCA.

850   Other approaches have also been used for estimation of source contributions. Recently, Harrison et

851   al. (2012b) used field data for estimating the contribution of brake wear, tyre wear and road dust

852   using size distributions of Ba, Zn and Si as the tracers for the sources respectively. By merging size

853   distributions measured with a Scanning Mobility Particle Sizer (SMPS) and an Aerodynamic

854   Particle Sizer (APS), Harrison et al. (2011) were able to generate continuous particle size

855   distributions for a roadside site from 15 nm – 10 µm diameter. By applying PMF, they were able to

35

856 separately identify nucleation mode and accumulation mode exhaust particles as well as brake dust

857 and re-suspension particles, and several particle classes arising from the urban background.

858

859 **7.2   Chemical Mass Balance (CMB) Model**

860 The CMB model uses the ambient measurement data for chemical species together with the

861 associated uncertainty and source profiles for different sources as inputs and the output consists of

862 estimates of contribution of each source to the total mass. The CMB model has several assumptions

863 including (Watson et al., 2002):

864 • chemical species do not react with one another and the species composition does not change

865   between the source(s) and the receptor(s)

866 • number of species are more than the number of sources

867 • all of the potential sources are included the model

868 • source profiles are not collinear

869 • random and non-correlated uncertainties

870
871 It is important to note that while the multivariate models do not require any information on source

872 emission composition, the CMB model requires the species abundances (in the form of source

873 profiles) in each source as a model input. Source profiles are "the mass abundances, i.e. fraction of

874 total mass of chemical species in source emissions, and such profiles are generally representative of

875 source categories rather than individual emitters" (Watson et al., 2002). There are several ways of

876 preparing source profiles for road vehicles including tests with chassis dynamometers,

877 measurements in tunnels/ highways and near roadside measurements (Phuleria et al., 2007; Handler

878 et al., 2008; Yan et al., 2009) as discussed in section 6. Source profiles are used for quantification

879 of source contributions to particulate matter using the chemical mass balance (CMB) model as well

880 as to compare and validate results obtained from factor analysis models. The CMB model relies to a

881 large extent on the accuracy of the source profiles used as an input. The typical components of any

882 source profile are found to be more-or-less similar although the mass abundances vary depending

883 on location and emitter characteristics. Thus, different combinations of source profiles can provide

884 statistically valid yet completely different solutions and selection of a source profile comparable to

885 ambient data is one of the key prerequisites for a good CMB solution (Robinson et al., 2006).

886 Absence of locally-representative source profiles and inter-region variability can result in errors in

887 estimation of source contributions from the CMB model (Yan et al., 2009; Rutter et al., 2011).

888 Significant differences have also been reported between real-world emissions and laboratory-tested

889 emissions (Yan et al., 2009; Ancelet et al., 2011). Analysis of vehicular source profiles in Bangkok,

890 Thailand led to the conclusion that while source profiles for different vehicle categories vary

891 significantly, ranges of the light duty and heavy duty average source profiles overlap and can be

892 represented by a single composite profile (Oanh et al., 2010). Fraser et al. (2003) and Fujita et al.

893 (2007) used the CMB model to analyse the apportionment of PM mass between diesel and gasoline

894 vehicles. While high molecular weight (4-7 ring) PAHs (coronene, benzo(ghi)perylene), hopanes

895 and steranes were found to be influential in case of gasoline engine emissions, 2-4 ring methylated

896 PAHs and EC were reported to be key species in the case of diesel engine emissions. Hanedar et al.

897 (2008) used the CMB model to resolve the sources of PAHs in Istanbul (Turkey) and identified

898 vehicle emissions (gasoline and diesel), natural gas combustion, coal and wood burning as major

899 sources with vehicular emissions contributing more than 50% at both urban and rural location.

900 Perrone et al. (2012) used CMB analysis to resolve PM sources in Milan (Italy) and reported traffic

901 emissions to contribute between 17-24% to $PM_{2.5}$ mass in an urban location compared to 7-9% at a

902 rural site and El Haddad et al. (2011) reported vehicular emissions to contribute 17% to the PM

903 mass in Marseille (France). In Detroit (USA), traffic emissions and road dust emissions were

904 reported to contribute 24-36% and 3-5% respectively of mass using CMB analysis of $PM_{2.5}$ data.

905
906 In recent years, there have been several studies focusing on characterization of trace metals and/or

907 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

934 only from road vehicles but also from other sources. However, significant gaps still exist in our

935 understanding of traffic emissions, particularly:

936 1. Characterization and quantification of different non-exhaust sources.

937 2. Impacts of non-exhaust emissions upon human health: While there are many studies reporting

938   links between exposure to air pollution and adverse health impacts, detailed information on

939   the components that contribute to toxicity is missing (Kelly and Fussell, 2012).

940 3. Aerosol chemistry in high-traffic environments, and the evolution of particles emitted from

941   vehicles

942 Further, most of the reported analyses have been carried out in the USA or Europe, and there is a

943 lack of reliable information on traffic emissions in areas with high population density in Africa,

944 Asia and South America. The field offers much scope for future research , including the

945 development of enhanced methods for quantification of non-exhaust contributions to airborne

946 concentrations.

947

REFERENCES

Aatmeeyata, K. D. S. and Sharma, M. (2009). Traffic generated non-exhaust particulate emissions from concrete pavement: A mass and particle size study for two-wheelers and small cars. Atmospheric Environment, 43(35), 5691-5697.

Aatmeeyata, K. D. S., and Sharma, M. (2010). Polycyclic aromatic hydrocarbons, elemental and organic carbon emissions from tire-wear. Science of the Total Environment, 408(20), 4563-4568.

Abu-Allaban, M., Coulomb, W., Gertler, A.W., Gillies, J., Pierson, W.R., Rogers, C.F., Sagebiel, J.C. and Tarnay, L. (2002). Exhaust particle size distribution measurements at the Tuscarora Mountain Tunnel. Aerosol Science and Technology, 36(6), 771-789.

Abu-Allaban, M., Gillies, J.A., Gertler, A.W., Clayton, R. and Proffitt, D. (2003). Tailpipe, resuspended road dust, and brake wear emission factors from on-road vehicles. Atmospheric Environment, 37, 5283-5293.

Adachi, K., and Tainosho, Y. (2004). Characterization of heavy metal particles embedded in tire dust. Environment International, 30(8), 1009-1017.

Aldabe, J., Elustondo, D., Santamaría, C., Lasheras, E., Pandolfi, M., Alastuey, A. and Querol, X. (2011). Chemical characterisation and source apportionment of PM2.5 and PM10 at rural, urban and traffic sites in Navarra (North of Spain). Atmospheric Research, 102(1-2), 191-205.

Allen, A. G., Nemitz, E., Shi, J. P., Harrison, R. M., and Greenwood, J. C. (2001). Size distributions of trace metals in atmospheric aerosols in the United Kingdom. Atmospheric Environment, 35(27), 4581-4591.

Allen, J. O., Alexandrova, O. and Kaloush, K.E. (2006). Tire wear emissions for asphalt rubber and portland cement concrete pavement surfaces: Arizona State University. Report submitted to Arizona Department of Transportation (Contract KR-04-0720-TRN).

Almeida, S. M., Pio, C. A., Freitas, M. C., Reis, M. A., and Trancoso, M. A. (2006). Source apportionment of atmospheric urban aerosol based on weekdays/weekend variability: evaluation of road re-suspended dust contribution. Atmospheric Environment, 40(11), 2058-2067.

Amato, F., Pandolfi, M., Viana, M., Querol, X., Alastuey, A., and Moreno, T. (2009). Spatial and chemical patterns of PM10 in road dust deposited in urban environment. Atmospheric Environment, 43(9), 1650-1659.

Amato, F., Pandolfi, M., Moreno, T., Furger, M., Pey, J., Alastuey, A., Bukowiecki, N., Prevot, A.S.H., Baltensperger, U. and Querol, X. (2011a). Sources and variability of inhalable road dust particles in three European cities. Atmospheric Environment, 45(37), 6777-6787.

Amato, F., Viana, M., Richard, A., Furger, M., Prévôt, A. S. H., Nava, S., Lucarelli, F., Bukowiecki, N., Alastuey, A., Reche, C., Moreno, T., Pandolfi, M., Pey, J. and Querol, X. (2011b). Size and time-resolved roadside enrichment of atmospheric particulate pollutants. Atmospheric Chemistry and Physics, 11(6), 2917-2931.

Amato, F., Karanasiou, A., Moreno, T., Alastuey, A., Orza, J.A.G., Lumbreras, J., Borge, R., Boldo, E., Linares. C. and Querol, X. (2012). Emission factors from road dust resuspension in a Mediterranean freeway. Atmospheric Environment, 61, 580-587.

40

Ancelet, T., Davy, P. K., Trompetter, W. J., Markwitz, A., and Weatherburn, D. C. (2011). Carbonaceous aerosols in an urban tunnel. Atmospheric Environment, 45(26), 4463-4469.

Apeagyei, E., Bank, M. S. and Spengler, J. D. (2011). Distribution of heavy metals in road dust along an urban-rural gradient in Massachusetts. Atmospheric Environment, 45(13), 2310-2323.

Ban-Weiss, G.A., Lunden, M.M, Kirchstetter, T.W. and Harley, R.A. (2010). Size-resolved particle number and volume emission factors for gasoline and diesel motor vehicles. Aerosol Science, 41, 5-12.

Barmpadimos, I., Nufer, M., Oderbolz, D. C., Keller, J., Aksoyoglu, S., Hueglin, C., Baltensperger, U. and Prevot, A.S.H. (2011). The weekly cycle of ambient concentrations and traffic emissions of coarse (PM10–PM2.5) atmospheric particles. Atmospheric Environment, 45(27), 4580-4590.

Beddows, D. C. S. and Harrison, R. M. (2008). Comparison of average particle number emission factors for heavy and light duty vehicles derived from rolling chassis dynamometer and field studies. Atmospheric Environment, 42, 7954-7966.

Beddows, D. C. S., Dall'Osto, M. and Harrison, R. M. (2009). Cluster analysis of rural, urban and curbside atmospheric particle size data. Environmental Science and Technology, 43, 4694-4700.

Belis, C.A., Karagulian, F., Larsen, B.R. and Hopke, P.K. (2013). Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe. Atmospheric Environment, 69, 94-108.

Berger, J. and Denby, B. (2011). A generalized model for traffic induced road dust emissions. Model description and evaluation. Atmospheric Environment, 45, 3692-3703.

Bhaskar, V.S. and Sharma, M. (2008). Assessment of fugitive road dust emissions in Kanpur, India. Transportation Research Part D, 13 , 400-403.

Birmili, W., Allen, A. G., Bary, F., and Harrison, R. M. (2006). Trace Metal Concentrations and Water Solubility in Size-Fractionated Atmospheric Particles and Influence of Road Traffic. Environmental Science and Technology, 40(4), 1144-1153.

Blau, P.J . and Meyer, H. M. III (2003). Characteristics of wear particles produced during friction tests of conventional and unconventional disc brake materials. Wear, 255, 1261-1269.

Boonyatumanond, R., Murakami, M., Wattayakorn, G. Togo, A. and Takeda, H. (2007). Sources of polycyclic aromatic hydrocarbons (PAHs) in street dust in a tropical Asian mega-city, Bangkok, Thailand. Science of the Total Environment, 384, 420-432.

Boulter, P. G., Thorpe, A. J., Harrison, R. M. and Allen, A. G. (2005). Road vehicle non-exhaust particulate matter: Final report on emission modelling. Published Project Report PPR110. Available at http://uk-air.defra.gov.uk/reports/cat15/0706061624_Report2__Emission_modelling.PDF.

Brook, J. R., Graham, L., Charland, J. P., Cheng, Y., Fan, X., Lu, G., Li, S.M., Lillyman, C., MacDonald, P., Caravaggio, G. and MacPhee, J.A. (2007). Investigation of the motor vehicle exhaust contribution to primary fine particle organic carbon in urban air. Atmospheric Environment, 41(1), 119-135.

Buckeridge, D.L., Glazier, R., Harvey, B.J., Escobar, M., Amrhein, C. and Frank, J. (2002). Effect of motor vehicle emissions on respiratory health in an urban area. Environmental Health Perspectives, 110(3), 293-300.

Bukowiecki, N., Lienemann, P., Hill, M., Furger, M., Richard, A., Amato, F., Prevot, A.S.H., Baltensperger, U., Buchmann, B. and Gehrig, R. (2010). PM10 emission factors for non-exhaust particles generated by road traffic in an urban street canyon and along a freeway in Switzerland. Atmospheric Environment, 44(19), 2330-2340.

Bullock, K.R., Duvall, R.M., Norris, G.A., McDow, S.R. and Hays, M.D. (2008). Evaluation of the CMB and PMF models using organic molecular markers in fine particulate matter collected during the Pittsburgh Air Quality Study. Atmospheric Environmet, 42, 6897-6904.

Caceres, D., Adonis, M., Gil, L., and Miguel, A. H. (1998). Exhaust PAH in Chilean vehicles: Levels, source signatures and receptor modeling. Journal of Aerosol Science, 29, Supplement 2(0), S741-S742.

Camatani, M., Crosta, G. F., Dolukhanyan, T., Sung, C., Giuliani, G., Corbetta, G. M., Cencetti, S. and Regazzoni, C. (2001). Microcharacterization and identification of tire debris in heterogenous laboratory and environmental specimens. Materials Characterization, 46, 271-283.

Canagaratna, M. R, Onasch, T. B., Wood, E. C., Herndon, S. C., Jayne, J. T., Cross, E. S., Miake-Lye, R. C., Kolb, C. E. and Worsnop, D. R. (2010). Evolution of vehicle exhaust particles in the Atmosphere. Journal of Air and Waste Management Association, 60, 1192-1203.

Canepari, S., Perrino, C., Olivieri, F., and Astolfi, M. L. (2008). Characterisation of the traffic sources of PM through size-segregated sampling, sequential leaching and ICP analysis. Atmospheric Environment, 42(35), 8161-8175.

Cass, G. R. (1998). Organic molecular tracers for particulate air pollution sources. Trends in Analytical Chemistry, 17(6), 356-366.

Chan, D. and Stachowiak, G. W. (2004). Review of automotive brake friction materials. Proceedings of the Institution of Mechanical Engineers, Part D: Journal of Automobile Engineering, 218 (9), 953-966.

Chan, T. W. and Mozurkewich, M. (2007). Application of absolute principal component analysis to size distribution data: identification of particle origins. Atmospheric Chemistry and Physics, 7, 887-897.

Chang, T. Y., Modzelewski, S. W., Norbeck, J. M., and Pierson, W. R. (1981). Tunnel air quality and vehicle emissions. Atmospheric Environment, 15(6), 1011-1016.

Charron, A. and Harrison, R. M. (2003). Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere. Atmospheric Environment, 37(29), 4109-4119.

Charron, A. and Harrison, R. M. (2009). Atmospheric Nanoparticles. Environmental and Human Health Impacts of Nanotechnology, John Wiley and Sons, Ltd, 63-209.

Chellam, S., Kulkarni, P. and Fraser, M. P. (2005). Emissions of organic compounds and trace metals in fine particulate matter from motor vehicles: a tunnel study in Houston, Texas. Journal of Air and Waste Management Association, 55, 60-72.

1103 Chen, J., Zheng, H., Wang, W., Liu, H., Lu, L., Bao, L. and Ren, L. (2006). Resuspension method
1104 for road surface dust collection and aerodynamic size distribution characterization. China
1105 Particuology, 4(6), 300-303.

1107 Chen, F., Hu, W. and Zhong, Q. (2013). Emissions of particle-phase polycyclic aromatic
1108 hydrocarbons (PAHs) in the Fu Gui-Shan Tunnel of Nanjing, China. Atmospheric Research, 124,
1109 53-60.

1111 Cheng, Y., Lee, S. C., Ho, K. F., Chow, J. C., Watson, J. G., Louie, P. K. K., Cao, J. J. and Hai, X.
1112 (2010). Chemically-speciated on-road PM2.5 motor vehicle emission factors in Hong Kong.
1113 Science of the Total Environment, 408(7), 1621-1627.

1115 Cheng, Y., Zou, S. C., Lee, S. C., Chow, J. C., Ho, K. F., Watson, J. G., Han, Y. M., Zhang, R. J.,
1116 Zhang, F., Yau, P.S., Huang, Y., Bai, Y. and Wu, W. J. (2011). Characteristics and source
1117 apportionment of PM1 emissions at a roadside station. Journal of Hazardous Materials, 195, 82-91.

1119 Chiang, H.-L. and Huang, Y.-S. (2009). Particulate matter emissions from on-road vehicles in a
1120 freeway tunnel study. Atmospheric Environment, 43, 4014-4022.

1122 Chiang, H.-L., Lai, Y.-M. and Chang, S.-Y. (2012). Pollutant constituents of exhaust emitted from
1123 light-duty diesel vehicles. Atmospheric Environment, 47, 399-406.

1125 Chirico, R., Prevot A. S. H., DeCarlo, P. F., Heringa, M. F., Richter, R., Weingartner, E. and
1126 Baltensperger, U. (2011). Aerosol and trace vehicle emission factors measured in a tunnel using an
1127 Aerosol Mass Spectrometer and other on-line instrumentation. Atmospheric Environment, 45,
1128 2182-2192.

1130 Choi, J.-K., Heo, J.-B., Ban, S.-J., Yi, S.-M. and Zoh, K.-D. (2012). Chemical characteristics of
1131 PM2.5 aerosol in Incheon, Korea. Atmospheric Environment, 60, 583-592.

1133 Councell, T. B., Duckenfield, K. U., Landa, E. R. and Callender, E. (2004). Tire wear particles as a
1134 source of Zn to the environment. Environmental Science and Technology, 38(15), 4206-4214.

1136 Crawford, J., Chambers, S., Cohen, D. D., Dyer, L., Wang, T. and Zahorowski, W. (2007).
1137 Receptor modelling using Positive Matrix Factorisation, back trajectories and Radon-222.
1138 Atmospheric Environment, 41(32), 6823-6837.

1140 Cyrys, J., Pitz, M., Heinrich, J., Wichmann, H. E. and Peters, A. (2008). Spatial and temporal
1141 variation of particle number concentration in Augsburg, Germany. Science of the Total
1142 Environment, 401(1–3), 168-175.

1144 Dahl, A., Gharibi, A., Swietlicki, E., Gudmundsson, A., Bohgard, M., Ljungman, A., Blomqvist, G.
1145 and Gustafsson, M. (2006). Traffic-generated emissions of ultrafine particles from pavement–tire
1146 interface. Atmospheric Environment, 40(7), 1314-1323.

1148 Dall'Osto, M., Thorpe, A., Beddows, D. C. S., Harrison, R. M., Barlow, J. F., Dunbar, T., Williams,
1149 P. I. and Coe, H. (2011). Remarkable dynamics of nanoparticles in the urban atmosphere.
1150 Atmospheric Physics and Chemistry, 11, 6623-6637.

Denier van der Gon, H., Gerlofs-Nijland, M.E., Gehrig, R., Gustafsson, M., Janssen, N., Harrison, R.M., Hulskotte, J., Johansson, C., Jozwicka, M., Keuken, M., Krijgsheld, K., Ntziachristos, L., Riediker, M. and Cassee, F.R. (2013). The Policy Relevance of Wear Emissions from Road Transport, Now and in the Future- An International Workshop Report and Consensus Statement. Journal of the Air and Waste Management Association, 63(2), 136-149.

Dongarra, G., Manno, E. and Varrica, D. (2009). Possible markers of traffic-related emissions. Environmental Monitoring and Assessment, 154, 117-125.

Dreyfus, M. A., Adou, K., Zucker, S. M. and Johnston, M. V. (2009). Organic aerosol source apportionment from highly time-resolved molecular composition measurements. Atmospheric Environment, 43(18), 2901-2910.

Duong, T. T. and Lee, B. K. (2011). Determining contamination level of heavy metals in road dust from busy traffic areas with different characteristics. Journal of Environmental Management, 92(3), 554-562.

El Haddad, I., Marchand, N., Dron, J., Temime-Roussel, B., Quivet, E., Wortham, H., Jaffrezo, J.L., Baduel, C., Voisin, D., Besombes, J.L. and Gille, G. (2009). Comprehensive primary particulate organic characterization of vehicular exhaust emissions in France. Atmospheric Environment, 43(39), 6190-6198.

El Haddad, I., Marchand, N., Wortham, H., Piot, C., Besombes, J.-L., Cozic, J., Chauvel, C., Armengaud, A., Robin, D. and Jaffrezo, J.-L. (2011). Primary sources of PM2.5 organic aerosol in an industrial Mediterranean city, Marseille. Atmospheric Chemistry and Physics, 11, 2039- 2058.

Etyemezian, V., Kuhns, H., Gillies, J., Chow, J., Hendrickson, K., McGown, M. and Pitchford, M. (2003). Vehicle-based road dust emissions measurement (III): effect of speed, traffic volume, location, and season on PM10 road dust emissions in the Treasure Valley, ID. Atmospheric Environment, 37, 4583-4593.

Fabretti, J.-F., Sauret, N., Gal, J.-F., Maria, P.-C. and Schärer, U. (2009). Elemental characterization and source identification of PM2.5 using Positive Matrix Factorization: The Malraux road tunnel, Nice, France. Atmospheric Research, 94(2), 320-329.

Fan, Y., Meng, Q., Weisel, C., Shalat, S.,Laumbach, R., Ohman-Strickland, P., Black, K., Rodriguez, M. and Bonanno, L. (2006). Acute Short-Term Exposures to PM2.5 Generated by Vehicular Emissions and Cardiopulmonary Effects in Older Adults. Epidemiology, 17(6), S213-S214.

Fang, G.-C., Chang, C.-N., Wu, Y.-S., Fu, P. P.-C., Yan, I.-L. and Chen, M.-H. (2004). Characterization, identification of ambient air and road dust polycyclic aromatic hydrocarbons in central Taiwan, Taichung. Science of the Total Environment, 327, 135-146.

Franco, V., Kousoulidou, M., Muntean, M., Ntziachristos, L., Hausberger, S. and Dilara, P. (2013). Road vehicle emission factors development: a review. Atmospheric Environment, 70, 84-97.

Fraser, M.P., Cass, G.R. and Simoneit, B.R.T. (1998). Gas-Phase and Particle-Phase Organic Compounds Emitted From Motor Vehicle Traffic in a Los Angeles Roadway Tunnel. Environmental Science and Technology, 32, 2051-2060.

Fraser, M.P., Buzcu, B., Yue, Z.W., Mcgaughey, G.R., Desai, N.R., Allen, D.T., Seila, R.L., Lonneman, W.A. and Harley, R.A.(2003). Separation of Fine Particulate Matter Emitted from Gasoline and Diesel Vehicles Using Chemical Mass Balancing Techniques. Environmental Science and Technology, 37, 3904-3909.

Fujita, E. M., Campbell, D. E. and Zielinska, B. (2006). Chemical analysis of lubrication oil samples from a study to characterize exhaust emissions from light-duty gasoline vehicles in the Kansas City metropolitan area (Final Report).

Fujita, E. M., Campbell, D. E., Arnott, W. P., Chow, J. C. and Zielinska, B. (2007). Evalautions of the Chemical Mass Balance Method for Determining Contributions of Gasoline and Diesel Exhaust to Ambient Carbonaceous Aerosols. Journal of Air and Waste Management Association, 57, 721-740.

Fuller, G. W., Carslaw, D. C. and Lodge, , H. W. (2002). An empirical approach for the prediction of daily mean PM10 concentrations. Atmospheric Environment, 36, 1431-1441.

Fulper, C.R., Kishan, S., Baldauf, R.W., Sabisch, R., Warila, J., Fujita, E.M., Scarbro, C., Crews, W.S., Snow, R., Gabele, P., Sanots, R., Tierney, E. and Cantrell, B. (2010). Methods of Characterizing the Distribution of Exhaust Emissions from Light-Duty, Gasoline-Powered Motor Vehicles in the U.S. Fleet. Journal of Air and Waste Management Association, 60, 1376-1387.

Funasaka, K., Miyazaki, T., Kawaraya, T., Tsuruho, K. and Mizuno, T. (1998). Characteristics of particulates and gaseous pollutants in a highway tunnel. Environmental Pollution, 102(2–3), 171-176.

Furusjo, E., Sternbeck, J. and Cousins, A.P. (2007). PM10 source characterization at urban and highway roadside locations. Science of the Total Environment, 387, 206-219.

Fushimi, A., Hasegawa, S., Takahashi, K., Fujitani, Y., Tanabe, K. and Kobayashi, S. (2008). Atmospheric fate of nuclei-mode particles estimated from the number concentrations and chemical composition of particles measured at roadside and background sites. Atmospheric Environment, 42(5), 949-959.

Gao, B., Yu, J.-Z., Li, S.-X., Ding, X., He, Q.-F. and Wang, X.-M. (2011). Roadside and rooftop measurements of polycyclic aromatic hydrocarbons in PM2.5 in urban Guangzhou: Evaluation of vehicular and regional combustion source contributions. Atmospheric Environment, 45(39), 7184-7191.

Garg, B. D., Cadle, S. H., Mulawa, P. A., Groblicki, P. J., Laroo, C. and Parr, G. A. (2000). Brake Wear Particulate Matter Emissions. Environmental Science and Technology, 34(21), 4463-4469.

Gehrig, R., Hill, M., Buchmann, B., Imhof, D., Weingartner, E. and Baltensperger, U. (2004). Separate determination of PM10 emission factors of road traffic for tailpipe emissions and emissions from abrasion and resuspension processes. International Journal of Environment and Pollution, 22(3): 312-325.

Gehrig, M., Zeyer, K., Bukowiecki, N., Lienemann, P., Poulikakos, L.D., Furger, M. and Buchmann, B. (2010). Mobile load simulators- A tool to distinguish between the emissions due to abrasion and resuspension of PM10 from road surfaces. Atmospheric Environment, 44, 4937-4943.

Gertler, A.W. and Pierson, W.R. (1996). Recent measurements of mobile source emission factors in North American tunnels. The Science of the Total Environment, 189/190, 107-113.

Gertler, A.W., Gillies, J.A., Pierson, W.R., Rogers, C.F., Sagebiel, J.C., Abu-Allaban, M., Coulombe, W., Tarnay, L. and Cahill, T.A. (2002). Real-World Particulate Matter and Gaseous Emissions from Motor Vehicles in a Highway Tunnel. Research Report 107, Health Effects Institute.

Gertler, A., Kuhns, H., Abu-Allaban, M., Damm, C., Gilies, J., Etyemezian, V., Clayton, R. and Proffitt, D. (2006). A case study of the impact of winter road sand/salt and street sweeping on road dust re-entrainment. Atmospheric Environment, 40, 5976-5985.

Gietl, J. K., Lawrence, R., Thorpe, A. J. and Harrison, R. M. (2010). Identification of brake wear particles and derivation of a quantitative tracer for brake dust at a major road. Atmospheric Environment, 44(2), 141-146.

Gillies, J.A., Gertler, A.W., Sagebiel, J.C. and Dippel, W.A. (2001). On-Road Particulate Matter (PM2.5 and PM10) Emissions in the Sepulveda Tunnel, Los Angeles, California. Environmental Science and Technology, 35, 1054-1063.

Grieshop, A. P., Lipsky, E. M., Pekney, N. J., Takahama, S. and Robinson, A. L. (2006). Fine particle emission factors from vehicles in a highway tunnel: Effects of fleet composition and season. Atmospheric Environment, 40, 287-298.

Gunawardana, C., Goonetilleke, A., Egodawatta, P., Dawes, L. and Kokot, S. (2011). Source characterisation of road dust based on chemical and mineralogical composition. Chemosphere, 87(2), 163-170.

Gustafsson, M., Blomqvist, G., Gudmundsson, A., Dahl, A., Swietlicki, E., Bohgard, M., Lindbom, J. and Ljungman, A. (2008). Properties and toxicological effects of particles from the interaction between tyres, road pavement and winter traction material. The Science of the Total Environment, 393(2-3), 226-240.

Gustafsson, M., Blomqvist, A., Gudmundsson, A., Dahl, A., Jonsson, P. and Swietlicki, E. (2009). Factors influencing PM10 emissions from road pavement wear. Atmospheric Environment, 49, 226-240.

Han, L., Zhuang, G., Cheng, S., Wang, Y. and Li, J. (2007). Characteristics of re-suspended road dust and its impact on the atmospheric environment in Beijing. Atmospheric Environment, 41(35), 7485-7499.

Han, B., Bai, Z., Guo, G., Wang, F., Li, F., Liu, Q., Ji, Y., Li, X. and Hu, Y. (2009). Characterization of PM(10) fraction of road dust for polycyclic aromatic hydrocarbons (PAHs) from Anshan, China. Journal of hazardous materials, 170(2-3), 934-940.

Handler, M., Puls, C., Zbiral, J., Marr, I, Puxbaum, H. and Limbeck, A. (2008). Size and composition of particulate emissions from motor vehicles in the Kaisermühlen-Tunnel, Vienna. Atmospheric Environment, 42(9), 2173-2186.

Hanedar, A., Alp, K., Kaynak, B., Baek, J., Avsar, E. and Odman, M.T. (2008). Concentrations and sources of PAHs at three stations in Istanbul, Turkey. Atmospheric Research, 99, 391-399.

Harris, S. J. and Maricq, M. M. (2001). Signature size distributions for diesel and gasoline engine exhaust particulate matter. Journal of Aerosol Science, 32(6), 749-764.

Harrison, R. M., Deacon A. R. and Jones, M. R. (1997). Sources and processes affecting concentrations of PM10 and PM2.5 Particulate Matter in Birmingham (U.K.). Atmospheric Environment, 31, 4103-4117.

Harrison, R.M., Shi, J.P., Xi, S., Khan, A., Mark, D., Kinnersley, R. and Yin, J. (2000). Measurement of number, mass and size distribution of particles in the atmosphere, Phil. Trans. R. Soc. Lond., A, 358, 2567-2580.

Harrison, R. M., Tilling, R., Romero, M.S.C., Harrad, S. and Jarvis, K. (2003). A study of trace metals and polycyclic aromatic hydrocarbons in the roadside environment. Atmospheric Environment, 37, 2391-2402.

Harrison, R. M., Jones, A. M. and Barrowcliffe, R. (2004). Field study of the influence of meteorological factors and traffic volumes upon suspended particle mass at urban roadside sites of different geometry. Atmospheric Environment, 38, 6361–6369.

Harrison, R. M. (2009). Airborne Particulate Matter from Road Traffic: Current Status of Knowledge and Research Challenges Paper presented at the ETTAO09. In 17th Transport and Air Pollution Symposium - 3rd Environment and Transport Symposium.

Harrison, R. M., Giorio, C., Beddows, D. C. and Dall'Osto, M. (2010). Size distribution of airborne particles controls outcomes of epidemiological studies. Science of the Total Environment, 409, 289-293.

Harrison, R.M., Beddows, D.C.S. and Dall'Osto, M., (2011). PMF analysis of wide-range particle size spectra collected on a major highway. Environmental Science and Technology, 45, 5522-5528.

Harrison, R. M., Dall'Osto, M., Beddows, D. C. S., Thorpe, A. J., Bloss, W. J., Allan, J. D., Coe, H., Dorsey, J. R., Gallagher, M., Martin, C., Whitehead, J., Williams, P. I., Jones, R. L., Langridge, J. M., Benton, A. K., Ball, S. M., Langford, B., Hewitt, C. N., Davison, B., Martin, D., Petersson, K., Henshaw, S. J., White, I. R., Shallcross, D. E., Barlow, J. F., Dunbar, T., Davies, F., Nemitz, E., Phillips, G. J., Helfter, C., Di Marco, C. F. and Smith, S. (2012a). Atmospheric chemistry and physics in the atmosphere of a developed megacity (London): An overview of the REPARTEE experiment and its conclusions. Atmospheric Chemistry and Physics, 12, 3065-3114.

Harrison, R. M., Jones, A. M., Gietl, J., Yin, J. and Green, D. C. (2012b). Estimation of the contributions of brake dust, tire wear, and resuspension to nonexhaust traffic particles derived from atmospheric measurements. Environmental Science and Technology, 46, 6523-6529.

Hassanien, M. A. and Abdel-Latif, N. M. (2008). Polycyclic aromatic hydrocarbons in road dust over Greater Cairo, Egypt. Journal of Hazardous Materials, 151, 247-254.

Hays, M. D., Cho, S.-H., Baldauf, R., Schauer, J. J. and Shafer, M. (2011). Particle size distributions of metal and non-metal elements in an urban near-highway environment. Atmospheric Environment, 45(4), 925-934.

He, L.-Y., Hu, M., Huang, X.-F., Zhang, Y.-H., Yu, B.-D. and Liu, D.-Q. (2006). Chemical characterization of fine particles from on-road vehicles in the Wutong tunnel in Shenzhen, China. Chemosphere, 62(12), 1565-1573.

He, L.-Y., Hu, M., Zhang, Y.-H., Huang, X.-F. and Yao, T.-T. (2008). Fine Particle Emissions from On-Road Vehicles in the Zhujiang Tunnel, China. Environmental Science and Technology, 42(12), 4461-4466.

HEI Panel on the Health Effects of Traffic-Related Air Pollution (2010). Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects. HEI Special Report, 17, Health Effects Institute, Boston, MA.

Hien, T. T., Thanh, L. T., Kameda, T., Takenaka, N. and Bandow, H. (2007). Distribution characteristics of polycyclic aromatic hydrocarbons with particle size in urban aerosols at the roadside in Ho Chi Minh City, Vietnam. Atmospheric Environment, 41(8), 1575-1586.

Hildemann, L. M., Markowski, G. R., Jones, M. C. and Cass, G. R. (1991). Submicrometer aerosol mass distributions of emissions from boilers, fireplaces, automobiles, diesel trucks and meat cooking operatios. Aerosol Science and Technology, 14(1), 138-152.

Hjortenkrans, D. S. T., Bergbäck, B. G. and Häggerud, A. V. (2007). Metal Emissions from Brake Linings and Tires: Case Studies of Stockholm, Sweden 1995/1998 and 2005. Environmental Science and Technology, 41(15), 5224-5230.

Hopke P. K. (2003). Recent developments in receptor modelling. Journal of Chemometrics, 17, 255-265.

Huang, X.-F., Chen, D.-L., Lan, Z.-J., Feng, N., He, L.-Y., Yu, G.-H. and Luan, S.-J. (2012). Characterization of organic aerosol in fine particles in a mega-city of South China: Molecular composition, seasonal variation, and size distribution. Atmospheric Research 114-115, 28-37.

Hussein, T., Johansson, C., Karlsson, H. and Hansson, H.-C. (2008). Factors affecting non-tailpipe aerosol particle emissions from paved roads: On-road measurements in Stockholm, Sweden. Atmospheric Environment, 42(4), 688-702.

Iijima, A., Sato, K., Yano, K., Tago, H., Kato, M., Kimura, H. and Furuta, N. (2007). Particle size and composition distribution analysis of automotive brake abrasion dusts for the evaluation of antimony sources of airborne particulate matter. Atmospheric Environment, 41(23), 4908-4919.

Imhof, D., Weingartner, E., Prevot, A.S.H., Ordonez, C., Kurtenbach, R., Weisen, P., Rodler, J., Sturm, P., McCrae, I., Sjodin, A. and Baltensperger, U. (2005a). Aerosol and NOx emissions factors and submicron particle size distributions in two road tunnels with different traffic regimes. Atmospheric Chemistry and Physics Discussions, 5, 5127-5166.

Imhof, D., Weingartner, E., Ordonez, C., Gehrig, R., Hill, M., Buchmann, B. and Baltensperger, U. (2005b). Real-world emission factors of fine and ultrafine aerosol particles for different traffic situations in Switzerland. Environmental Science and Technology, 39(21), 5127-5166.

Ingo, G. M., D'Uffizi, M., Falso, G., Bultrini, G. and Padeletti, G. (2004). Thermal and microchemical investigation of automotive brake pad wear residues. Thermochimica Acta, 418, 61-68.

Jamriska, M., Morawska, L., Thomas, S. and He, C. (2004). Diesel Bus Emissions Measured in A Tunnel Study. Environmental Science and Technology, 38, 6701-6709.

Jayaratne, E. R., Wang, L., Heuff, D., Morawska, L. and Ferreiera, L. (2009). Increase in particle number emissions from motor vehicles due to interruption of steady traffic flow. Transportation Research Part D, 14, 521-526.

Johansson, C., Norman, M. and Burman, L. (2009). Road traffic emission factors for heavy metals. Atmospheric Environment, 43(31), 4681-4688.

Jones, A. M. and Harrison, R. M. (2006). Estimation of emisison factors of particle number and mass fractions from traffic at a site where mean vehicle speeds vary over short distances. Atmospheric Environment, 35, 7125-7137.

Jones, A. M., Yin, J. and Harrison, R. M. (2008). The weekday-weekend difference and the estimation of the non-vehicle contributions to the urban increment of airborne particulate matter. Atmospheric Environment, 42, 4467-4479.

Kam, W., Liacos, J. W., Schauer, J. J., Delfino, R. J. and Sioutas, C. (2012). Size-segregated composition of particulate matter (PM) in major roadways and surface streets. Atmospheric Environment, 55, 90-97.

Karanasiou, A., Moreno, T., Amato, F., Lumbreras, J., Narros, A., Borge, R., Tobias, A., Boldo, E., Linares, C., Pey, J., Reche, C., Alastuey, A. and Querol, X. (2011). Road dust contribution to PM levels – Evaluation of the effectiveness of street washing activities by means of Positive Matrix Factorization. Atmospheric Environment, 45(13), 2193-2201.

Karanasiou, A., Moreno, T., Amato, F., Tobias, A., Boldo, E., Linares, C., Lumbreras, J., Borge, R., Alastuey, A. and Querol, X. (2012). Variation of PM2.5 concentrations in relation to street washing activities. Atmospheric Environment, 54, 465- 469.

Kaunhaniemi, M., Kukkonen, J., Harkonen, J., Nikmo, J., Kangas, L., Omstedt, G., Ketzel, M., Kousa, A., Haakana, M. and Karppinen, A. (2011). Evalaution of a road dust suspension model for predicting the concentrations of PM10 in a street canyon. Atmospheric Environment, 45, 3646-3654.

Kelly, F.J. and Fussell, J.C. (2012). Size, source and chemical composition as determinats of toxicity attributable to ambient particulate matter. Atmospheric Environment, 60, 504- 526.

Kerminen, V.-M., Pakkanen, T. A., Mäkelä, T., Hillamo, R. E., Sillanpää, M., Rönkkö, T., Virtanen, A., Keskinen, J., Pirjola, L., Hussein, T. and Hameri, K. (2007). Development of particle number size distribution near a major road in Helsinki during an episodic inversion situation. Atmospheric Environment, 41(8), 1759-1767.

Ketzel, M.G., Omstedt, G., Johansson, C., During, I., Pohjola, M., Oettl, D., Gidhagen, L., Wahlin, P., Lohmeyer, A. and Berkowicz (2007). Estimation and validation of PM2.5/PM10 exhaust and non-exhaust emission factors for practical street pollution modelling. Atmospheric Environment, 41, 9370-9385.

Keuken, M., Denier van der Gon, H. and van der Valk, K. (2010). Non-exhaust emissions of PM and the efficiency of emission reduction by road sweeping and washing in the Netherlands. The Science of the Total Environment, 408(20), 4591-4599.

Keuken, M., Henzing, B., Zandveld, P., van den Elshout, S. and Karl, M. (2012). Dispersion of particle numbers and elemental carbon from road traffic, a harbour and an airstrip in the Netherlands. Atmospheric Environment, 54, 320-327.

Kim, J. Y., Lee, J. Y., Kim, Y. P., Lee, S. B., Jin, H. C. and Bae, G. N. (2012). Seasonal characteristics of the gaseous and particulate PAHs at a roadside station in Seoul, Korea. Atmospheric Research, 116, 142-150.

Kittleson, D. B., Watts, W. F. and Johnson, J.P. (2004). Nanoparticle emisisons on Minnesota highways. Atmospheric Environment, 38, 9-19.

Klems, J. P., Pennington, M. R., Zordan, C. A. and Johnston, M. V. (2010). Ultrafine particles near a roadway intersection: Origin and apportionment of fast changes in concentration. Environmental Science and Technology, 44(20), 7903-7907.

Kuhlbusch, T.A.J., John, A.C. and Quass, U. (2009). Sources and source contributions to fine particles. Biomarkers, 14(1), 23- 28.

Kumar, P., Fennell, P., Langley, D. and Britter, R. (2008). Pseudo-simultaneous measurements for the vertical variation of coarse, fine and ultrafine particles in an urban street canyon. Atmospheric Environment, 42(18), 4304-4319.

Kumar, P., Pirjola, L., Ketzel, M. and Harrison, R.M. (2013). Nanoparticle emissions from 11 non-vehicle exhaust sources- A review. Atmospheric Environment, 67, 252-277.

Kumata, H., Yamada, J., Masuda, K., Takada, H., Sato, Y., Sakural, T. and Fujiwara, K. (2002). Benzothiazolamines as tire-derived molecular markers: sorptive behavious in street runoff and application to source apportioning. Environmental Science and Technology, 36(4), 702-708.

Kupiainen, K. J., Tervahattu, H., Räisänen, M., Mäkelä, T., Aurela, M. and Hillamo, R. (2005). Size and Composition of Airborne Particles from Pavement Wear, Tires, and Traction Sanding. Environmental Science and Technology, 39(3), 699-706.

Kwon, E.E. and Castaldi, M.J. (2012). Mechanistic understanding of polycyclic aromatic hydrocarbons (PAHs) from the therman degradation of tires under various oxygen concentration atmospheres. Environmental Science and Technology, 46(23), 12921- 6.

Laidlaw, M. A. S., Zahran, S., Mielke, H. W., Taylor, M. P. and Filippelli, G. M. (2012). Re-suspension of lead contaminated urban soil as a dominant source of atmospheric lead in Birmingham, Chicago, Detroit and Pittsburgh, USA. Atmospheric Environment, 49, 302-310.

Landis, M. S., Lewis, C. W., Stevens, R. K., Keeler, G. J., Dvonch, J. T. and Tremblay, R. T. (2007). Ft. McHenry tunnel study: Source profiles and mercury emissions from diesel and gasoline powered vehicles. Atmospheric Environment, 41(38), 8711-8724.

Larsen, R.K. and Baker, J.E. (2003). Source apportionment of polycyclic aromatic hydrocarbons in the urban atmosphere: A comparison of three methods. Environmental Science and Technology, 37(9), 1873-1881.

Lee, W.-J., Wang, Y.-F., Lin, T.-C., Chen, Y.-Y., Lin, W.-C., Ku, C.-C. and Cheng, J.-T. (1995). PAH characteristics in the ambient air of traffic-source. The Science of the Total Environment, 159, 185-200.

Liacos, J. W., Kam, W., Delfino, R. J., Schauer, J. J. and Sioutas, C. (2012). Characterization of organic, metal and trace element PM2.5 species and derivation of freeway-based emission rates in Los Angeles, CA. Science of the total Environment, 435-436, 5283-5293.

Lim, L.H., Harrison, R.M. and Harrad, S. (1999). The contribution of traffic to atmospheric concentrations of polyaromatic hydrocarbons. Environmental Science and Technology, 33, 3538-3542.

Lim, J., Yu, L.E.. Kostetski, Yu. Yu., Lim, C., Ryu, J. and Kim, J. (2008). Effects of driving conditions on diesel exhaust particles. Journal of Air and Waste Management Association, 58, 1077-1085.

Lin, C.-C., Chen, S.-J., Huang, K.-L., Hwang, W.-I., Chang-Chien, G.-P. and Lin, W.-Y. (2005). Characteristics of Metals in Nano/Ultrafine/Fine/Coarse Particles Collected Beside a Heavily Trafficked Road. Environmental Science and Technology, 39(21), 8113-8122.

Lin, H.-Y., Hung, C.-H., Yuan, C.-S., Chen, C.-J., Chen, W.-C., Chiang, S.-W. and Wang, C., J.-L. (2008). Characteristics and source identification of roadside suspended particles. Transportation Research Part D: Transport and Environment, 13(8), 531-538.

Lingard, J. J. N., Agus, E. L., Young, D. T., Andrews, G. E. and Tomlin, A. S. (2006). Observations of urban airborne particle number concentrations during rush-hour conditions: analysis of the number based size distributions and modal parameters. Journal of Environmental Monitoring, 8(12), 1203-1218.

Liu, M., Cheng, S.B., Ou, D.N., Hou, L.J., Gao, L., Wang, L.L., Xie, Y.S., Yang, Y., Xu, S.Y. (2007). Characterization, identification of road dust PAHs in central Shanghai areas, China. Atmospheric Environment, 41, 8785-8795.

Lough, G. C., Schauer, J. J., Park, J.-S., Shafer, M. M., DeMinter, J. T. and Weinstein, J. P. (2005). Emissions of Metals Associated with Motor Vehicle Roadways. Environmental Science and Technology, 39(3), 826-836.

Lough, G. C., Christensen, C. G., Schauer, J. J., Tortorelli, J., Mani, E., Lawson, D. R., Clark, N. N. and Gabela, P. A. (2007). Development of molecular marker source profiles for emissions from on-road gasoline and diesel vehicle fleets. Journal of the Air and Waste Management Association, 57(10), 1190-1199.

Luo, X. S., Yu, S., Zhu, Y. G. and Li, X. D. (2011). Trace metal contamination in urban soils of China. The Science of the Total Environment, 421-422, 17-30.

Majumdar, D., Rajaram, B., Meshram, S. and Chalapati Rao, C.V. (2012). PAHs in Road Dust: Ubiquity, Fate, and Summary of Available Data. Critical Reviews in Environmental Science and Technology, 42(12), 1191-1232.

Mancilla, Y. and Mendoza, A. (2012). A tunnel study to characterize PM2.5 emissions from Gasoline-powered vehicles in Monterrey, Mexico. Atmospheric Environment, 59, 449-460.

Manno, E., Varrica, D. and Dongarrà, G. (2006). Metal distribution in road dust samples collected in an urban area close to a petrochemical plant at Gela, Sicily. Atmospheric Environment, 40(30), 5929-5941.

Mari, M., Harrison, R.M., Schuhmacher, M., Domingo, J.L. and Pongpiachan S. (2010). Inferences over the sources and processes affecting polycyclic aromatic hydrocarbons in the atmosphere derived from measured data. Science of the Total Environment, 408, 2387-2393.

Martuzevicius, D., Kliucininkas, L., Prasauskas, T., Krugly, E., Kauneliene, V. and Strandberg, B. (2011). Resuspension of particulate matter and PAHs from street dust. Atmospheric Environment, 45(2), 310-317.

Mathissen, M., Scheer, V., Vogt, R. and Benter, T. (2011). Investigation on the potential generation of ultrafine particles from the tire–road interface. Atmospheric Environment, 45(34), 6172-6179.

Maricq, M. M. (2007). Chemical characterization of particulate emissions from diesel engines: A review. Journal of Aerosol Science, 38(11), 1079-1118.

Masiol, M., Hofer, A., Squizzato, S., Piazza, R., Rampazzo, G. and Pavoni, B. (2012). Carcinogenic and mutagenic risk associated to airborne particle-phase polycyclic aromatic hydrocarbons: A source apportionment. Atmospheric Environment, 60, 375- 382.

Mauderly, J.L. (1994). Toxicological and epidemiological evidence for health risks from inhaled engine emissions. Environmental Health Perspectives, 102, 165-171.

Mclaren, R., Gertler, A.W., Wittorff, D.N., Belzer, W., Dann, T. and Singleton, D.L. (1996). Real-World Measurements of Exhaust and Evaporative Emissions in the Cassiar Tunnel Predicted by Chemical Mass Balance Modeling. Environmental Science and Technology, 30, 3001-3009.

Miguel, A. H., Kirchstetter, T. W., Harley, R. A. and Hering, S. V. (1998). On-Road Emissions of Particulate Polycyclic Aromatic Hydrocarbons and Black Carbon from Gasoline and Diesel Vehicles. Environmental Science and Technology, 32(4), 450-455.

Milani, M., Pucillo, F.P., Ballerini, M., Camatani, M., Gualtieri, M. and Martino, S. (2004). First evidence of tyre debris characterization at the nanoscale by focused ion beam. Materials Characterization, 52, 283-288.

Morawska, L., Jayaratne, E.R., Mengersen, K., Jamriska, M. and Thomas, S. (2002). Differences in airborne particle and gaseous concentrations in urban air between weekdays and weekends. Atmospheric Environment, 36, 4375-4383.

Nanzetta, K. M. and Holmen, B. A. (2004). Roadside Particle Number Distributions and Relationships between Number Concentrations, Meteorology, and Traffic along a Northern California Freeway. Journal of Air and Waste Management Association, 54, 540-554.

Narvaez, R. F., Hoepner, L., Chillrud, S. N., Yan, B., Garfinkel, R., Whyatt, R., Camann, D., Perera, F. P., Kinney, P. L. and Miller, R. L. (2008). Spatial and Temporal Trends of Polycyclic Aromatic Hydrocarbons and Other Traffic-Related Airborne Pollutants in New York City. Environmental Science and Technology, 42, 7330-7335.

Nicholson, K.W. (1988). A review of particle resuspension. Atmospheric Environment (1967), 22(12), 2639-2651.

Nielsen, T. (1996). Traffic contribution of polycyclic aromatic hydrocarbons in the centre of a large city. Atmospheric Environment, 30(20), 3481-3490.

Ntziachristos, L., Ning, Z., Geller, M.D., Sheesley, R.J., Schauer, J.J. and Sioutas, C. (2007). Fine, ultrafine and nanoparticle trace element compositions near a major freeway with a high heavy-duty diesel fraction. Atmospheric Environment, 41, 5864-5696.

Oliveira, C., Pio, C., Caseiro, A., Santos, P., Nunes, T., Mao, H., Luahana, L. and Sokhi, R.S. (2010). Road traffic impact on urban atmospheric aerosol loading at Oporto, Portugal. Atmospheric Environment, 44(26), 3147-3158.

Oliveira, C., Martins, N., Tavares, J., Pio, C., Cerqueira, M., Matos, M., Silva, H., Oliviera, S. and Camoes, F. (2011). Size distribution of polycyclic aromatic hydrocarbons in a roadway tunnel in Lisbon, Portugal. Chemosphere, 83(11), 1588-1596.

Omar, N. Y. M. J., Abas, M. R. B., Rahman, N. A., Tahir, N. M., Rushdi, A. I. and Simoneit, B. R. T. (2007). Levels and distributions of organic source tracers in air and roadside dust particles of Kuala Lumpur, Malaysia. Environmental Geology, 52, 1485-1500.

Omstedt, G., Bringfelt, B., Johansson, C. (2005). A model for vehicle-induced non-tailpipe emissions of particles along Swedish roads. Atmospheric Environment, 39, 6088-6097.

Oanh, N. T. K., Thiansathit, W., Bond, T. C., Subramanian, R., Winijkul, E. and Paw-armart, I. (2010). Compositional characterization of PM2.5 emitted from in-use diesel vehicles. Atmospheric Environment, 44(1), 15-22.

Ondráček, J., Schwarz, J., Ždímal, V., Andělová, L., Vodička, P., Bízek, V., Tsai, C.-J., Chen, S.-C. and Smolik, J. (2011). Contribution of the road traffic to air pollution in the Prague city (busy speedway and suburban crossroads). Atmospheric Environment, 45(29), 5090-5100.

Padro-Martinez, L. T., Patton, A. P., Trull, J. B., Zamore, W., Brugge, D. and Durant, J. L. (2012). Mobile monitoring of particle number concentration and other traffic-related air pollutants in a near-highway neighbourhood over the course of a year. Atmospheric Environment, 61, 253-264.

Pant, P. and Harrison, R. M. (2012). Critical review of receptor modelling for particulate matter: A case study of India, Atmospheric Environment, 49, 1-12.

Peltier, R. E., Cromar, K. R., Ma, Y., Fan, Z.-H. and Lippmann, M. (2011). Spatial and seasonal distribution of aerosol chemical components in New York City: (2) Road dust and other tracers of traffic-generated air pollution. Journal of Exposure Science and Environmental Epidemiology, 21(5), 484-494.

Perez, N., Pey, J., Cusack, M., Reche, C., Querol, X., Alastuey, A. and Viana, M. (2010). Variability of Particle Number, Black Carbon and PM10, PM2.5 and PM1 levels and speciation: Influence of road traffic emissions on urban air quality. Aerosol Science and Technology, 44(7), 487-499.

Perrone, M. G., Larsen, B. R., Ferrero, L., Sangiorgi, G., De Gennaro, G., Udisti, R., Zangranado, R., Gambaro, A. and Bolzacchini, E. (2012). Sources of high PM2.5 concentrations in Milan, Northern Italy: Molecular marker data and CMB modelling. Science of the Total Environment, 414, 343-355.

Pey, J., Querol, X., Alastuey, A., Rodriguez, R., Putuad, J.-P. and Van Dingenen, R. (2009). Source apportionemnt of urban fine and ultra-fine particle number concentration in a Western Mediterranean city. Atmospheric Environment, 43, 4407-4415.

Pey, J., Querol, X. and Alastuey, A. (2010). Discriminating the regional and urban contributions in the North-Western Mediterranean: PM levels and composition. Atmospheric Environment, 44(13), 1587-1596.

Phuleria, H. C., Geller, M. D., Fine, P. M. and Sioutas, C. (2006). Size-Resolved Emissions of Organic Tracers from Light- and Heavy-Duty Vehicles Measured in a California Roadway Tunnel. Environmental Science and Technology, 40(13), 4109-4118.

Phuleria, H. C., Sheesley, R. J., Schauer, J. J., Fine, P. M. and Sioutas, C. (2007). Roadside measurements of size-segregated particulate organic compounds near gasoline and diesel-dominated freeways in Los Angeles, CA. Atmospheric Environment, 41(22), 4653-4671.

Prichard, H.M. and Fisher, P.C. (2012). Identification of Platinum and Palladium Particles Emitted from Vehicles and Dispersed into the Surface Environment. Environmental Science and Technology, 46(6), 3149-3154.

Pulles, T., Denier van der Gon, H., Appelman, W. and Verheul, M. (2012). Emission factors for heavy metals from diesel and petrol used in European vehicles. Atmospheric Environment, 61, 641-651.

Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., ten Brink, H., Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S., Harrison, R., Ayres, J.G., van der Zee, S. and Hok, G. (2007). Spatial variation of particle number and mass over four European cities. Atmospheric Environment, 41(31), 6622-6636.

Reche, C., Querol, X., Alastuey, A., Viana, M., Pey, J., Moreno, T., Rodriguez, S., Gonzalez, Y., Fernandez-Camacho, R., Sanchez de la Campa, A. M., de la Rosa, J., Dall'Osto, M., Prevot, A. S. H., Hueglin, C., Harrison, R.M. and Quincey, P. (2011). New considerations for PM, black carbon and particle number concentration for air quality monitoring across different European cities. Atmospheric Chemistry and Physics, 11, 6207-6227.

Reddy, C. M. and Quinn, J. G. (1997). Environmental Chemistry of Benzothiazoles Derived from Rubber. Environmental Science and Technology, 31, 2847-2853.

Rexeis, M. and Hausberger, S. (2009). Tend of vehicle emission levels until 2020 - Prognosis based on current vehicle measurements and future emission legislation. Atmospheric Environment, 43, 4689-4698.

Riddle, S. G., Robert, M. A., Jakober, C. A., Hannigan, M. P. and Kleeman, M. J. (2007). Size Distribution of Trace Organic Species Emitted from Light-Duty Gasoline Vehicles. Environmental Science and Technology, 41(21), 7464-7471.

Rissler, J., Swietlicki, E., Bengtsson, A., Boman, C., Pagels, J.,Sandstrom, T., Blomberg, A. and Londahl, J. (2012). Experimental determination of deposition of diesel exhaust particles in the human respiratory tract. Journal of Aerosol Science, 48, 18-33.

Robert, M.A., VanBergen, S., Kleeman, M.J. and Jakober, C.A. (2007a). Size and composition distributions of particulate matter emisisons: Part 1- Light-duty diesel vehicles. Journal of Air and Waste Management Association, 57, 1414-1428.

Robert, M. A., Kleeman, M. J. and Jakober, C. A. (2007b). Size and composition distributions of particulate matter emisisons: Part 2- Heavy-duty diesel vehicles. Journal of Air and Waste Management Association, 57, 1429-1438.

Robinson, A. L., Subramanian, R., Donahue, N. M. and Rogge, W. F. (2006). Source Apportionment of Molecular Markers and Organic Aerosol1. Polycyclic Aromatic Hydrocarbons and Methodology for Data Visualization. Environmental Science and Technology, 40(24), 7803-7810.

Rodriguez, S., Van Dingenen, R., Putuad, J.-P., Dell'Acqua, A., Pey, J., Querol, X., Alastuey, A., Chenery, S.,Ho, K.-F., Harrison, R., Tardivo, R., Scarnato, B. and Gemelli, V. (2007). A study on the relationship between mass concentrations, chemistry and number size distribution of urban fine aerosols in Milan, Barcelona and London. Atmospheric Chemistry and Physics, 35, 2217-2232.

Rogak, S. N., Green, S. I. and Pott, U. (1998). Use of Tracer Gas for Direct Calibration of Emission-Factor Measurements in a Traffic Tunnel. Journal of the Air and Waste Management Association, 48(6), 545-552.

Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R. and Simoneit, B. R. T. (1993). Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: roads as sources and sinks. Environmental Science and Technology, 27(9), 1892-1904.

Rose, D., Wehner, B., Ketzel, M., Engler, C., Voigtlander, J., Tuch, T. and Wiedensohler, A. (2006). Atmospheric number size distributions of soot particles and estimation of emission factors. Atmospheric Chemistry and Physics, 6(4), 1021-1031.

Rosenbohm, E., Vogt, R., Scheer, V., Nielsen, O. J., Dreiseidler, A., Baumbach, G., Imhof, D., Baltensperger, U., Fuchs, J. and Jaeschke, W. (2006). Particulate size distributions and mass measured at a motorway during the BAB II campaign. Atmospheric Environment, 37, 5283-5293.

Ruellan, S. and Cachier, H. (2001). Characterisation of fresh particulate vehicular exhausts near a Paris high flow road. Atmospheric Environment, 35(2), 453-468.

Rutter, A. P., Snyder, D. C., Schauer, J. J., Sheesley, R. J., Olson, M. R. and DeMinter, J. (2011). Contributions of resuspended soil and road dust to organic carbon in fine particulate matter in the Midwestern US. Atmospheric Environment, 45(2), 514-518.

Sadiktsis, I., Bergvall, C., Johansson, C. and Westerholm, R. (2012). Automobile Tires—A Potential Source of Highly Carcinogenic Dibenzopyrenes to the Environment. Environmental Science and Technology, 46(6), 3326-3334.

Sahu, M., Hu, S., Ryan, P. H., Le Masters, G., Grinshpun, S. A., Chow, J. C. and Biswas, P. (2011). Chemical compositions and source identification of PM2.5 aerosols for estimation of a diesel source surrogate. Science of the Total Environment, 409(13), 2642-2651.

Samara, C. and Voutsa, D. (2005). Size distribution of airborne particulate matter and associated heavy metals in the roadside environment. Chemosphere, 59, 1197-1206.

Sanchez-Ccoyllo, O. R., Ynoue, R. Y., Martins, L. D., Astolfo, R., Miranda, R. M., Freitas, E. D., Borges, A.S., Fornaro, A., Freitas, H., Moreira, A. and Andrade, M. F. (2009). Vehicular particulate matter emissions in road tunnels in Sao Paulo, Brazil. Environmental Monitoring and Assessment, 149(1-4), 241-249.

Sanders, P. G., Xu, N., Dalka, T. M. and Maricq, M. M. (2003). Airborne brake wear debirs: Size distributions, composition, and a comparison of dynamometer and vehicle tests. Environmental Science and Technology, 37(4), 4060-4069.

Schaap, M., Manders, A.M.M., Hendriks, E.C.J., Cnossen, J.M., Segers, A.J.S., Denier van der Gon, H.A.C., Jozwicka, M., Sauter, F., Velders, G., Matthijsen, J. and Builtjes, P.J.H. (2009). Regional modelling of PM10 over the Netherlands. Technical Report 500099008. Available at http://www.rivm.nl/bibliotheek/rapporten/500099008.pdf [Accessed September 9, 2012].

Schauer, J. J., Rogge, W. F., Hildemann, L. M., Mazurek, M. A., Cass, G. R. and Simoneit, B. R. T. (1996). Source apportionment of airborne particulate matter using organic compounds as tracers. Atmospheric Environment, 30, 3837-3855.

Schauer, J. J., Lough, G. C., Shafer, M. M., Christensen, W. C., Arndt, M. F., DeMinter, J. T. and Park, J.-S. (2006). Characterization of Emissions of Metals Emitted from Motor Vehicles: Health Effects Institute.

Shen, Z., Han, Y., Cao, J., Tian, J., Zhu, C., Liu, S., Liu, P. and Wang, Y. (2010). Characteristics of traffic-related emissions: A case study in roadside ambient air over Xi'an, China. Aerosol and Air Quality Research, 10, 292-300.

Shi, J. P. and Harrison, R. M., (1999). Investigation of ultrafine particle formation during diesel exhaust dilution. Environmental Science and Technology, 33, 3730-3736.

Shi, J. P., Khan, A. A. and Harrison, R. M., (1999). Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. Science of the Total Environment, 235, 51-64.

Sjodin, A., Ferm, M., Bjork, A., Rahmberg, M., Gudmundsson, A., Swietlickli, E., Johansson, C., Gustafsson, M. and Blomqvist, G. (2010). Wear particles from road traffic: a field, laboratory and modelling study. IVL Report B1830. Availabe at: http://www.ivl.se/webdav/files/B-rapporter/B1830.pdf.

Smith, D. J. T. and Harrison, R. M. (1996). Concentrations, trends and vehicle source profile of polynuclear aromatic hydrocarbons in the U.K. atmosphere. Atmospheric Environment, 30(14), 2513-2525.

Sodeman, D. A., Toner, S. M. and Prather, K. A. (2005). Determination of single particle mass spectral signatures from light-duty vehicle emissions. Environmental Science and Technology, 39(12), 4569-4580.

Song, F. and Gao, Y. (2011). Size distributions of trace elements associated with ambient particular matter in the affinity of a major highway in the New Jersey–New York metropolitan area. Atmospheric Environment, 45(37), 6714-6723.

Song, S., Wu, Y., Jiang, J., Yang, L., Cheng, Y. and Hao, J. (2012). Chemical characteristics of size-resolved PM(2.5) at a roadside environment in Beijing, China. Environmental Pollution, 161, 215-221.

Sonntag, D. B., Bailey, C. R., Fulper, C. R. and Baldauf, R. W. (2012). Contribution of Lubricating Oil to Particulate Matter Emissions from Light-Duty Gasoline Vehicles in Kansas City. Environmental Science and Technology, 46(7), 4191–4199.

Stechmann, H. and Dannecker, W. (1990). Characterization and source analysis of vehicle-generated aerosols. Journal of Aerosol Science, 21, Supplement 1, S287-S290.

Staehelin, J., Schlapfer, K., Burgin, T., Steinemann, U., Schneider, S., Brunner, D., Baumle, M., Meier, M., Zahner, C., Keiser, S., Stahel, W. and Keller, C. (1995). Emission factors from road traffic from a tunnel study (Gubrist tunnel, Switzerland). Part I: concept and first results. Science of the Total Environment, 169, 141-147.

Sternbeck, J., Sjödin, Å. and Andréasson, K. (2002). Metal emissions from road traffic and the influence of resuspension—results from two tunnel studies. Atmospheric Environment, 36(30), 4735-4744.

Sun, Y.L., Zhang, Q., Schwab, J.J., Chen, W.-N., Bae, M.-S., Hung, H.-M., Lin, Y.-C., Ng, N.L., Jayne, J., Massoli, P., Williams, L.R. and Demerjian, K.L. (2012). Characterization of near-highway submicron aerosols in New York City with a high-resolution aerosol mass spectrometer. Atmospheric Chemistry and Physics, 12, 2215- 2227.

Sysalova, J., Sykorova, I., Havelcova, M., Szakova, J., Trejtnarova, H. and Kotlik, B. (2012). Toxicologically important trace elements and organic compounds investigated in size –fractionated urban particulate matter collected near the Prague highway. Science of the total Environment, 437, 127- 136.

Tanner, P.A., Hoi-Ling, M. and Yu, P.K.N. (2008). Fingerprinting metals in street dust in Beijing, Shanghai and Hong Kong. Environmental Science and Technology, 42, 7111-7117.

Teixiera, E.C., Agudelo-Castaneda, D.M., Fachel, J.M.G., Leal, K.L., Garcia, K.O. and Wiegand, F. (2012). Source identification and seasonal variation of polycyclic aromatic hydrocarbons associated with atmospheric fine and coarse particles in the Metropolitan Area of Porto Alegre, RS, Brazil. Atmospheric Research, 118: 390-403.

Tervahattu, H., Kupiainen, K.J., Raisanen, M., Makela, T. and Hillamo, R. (2006). Generation of urban road dust from anti-skid and asphalt concrete aggregates. Journal of Harazdous Materials, 132, 39-46.

Thorpe, A. J., Harrison, R. M., Boulter, P. G. and McCrae, I. S. (2007). Estimation of particle resuspension source strength on a major London Road. Atmospheric Environment, 41, 8007-8020.

Thorpe, A. and Harrison, R. M. (2008). Sources and properties of non-exhaust particulate matter from road traffic: a review. Science of the Total Environment, 400(1-3), 270-282.

Van Poppel, M., Panis, L.I., Govarts, E., Van Houtte, J. and Maenhaut, W. (2012). A comparative study of traffic related air pollution next to a motorway and a motorway flyover. Atmospheric Environment, 60, 132-141

Varrica, D., Bardelli, F., Dongarra, G. and Tamburro, E. (2012). Speciation of Sb in airborne particulate matter, vehicle brake linings and brake pad wear residues. Atmospheric Environment, 64, 18-24.

Viana, M., Kuhlbusch, T.A.J., Querol, X., Alastuey, A., Harrison, R.M., Hopke, P.K., Winiwarter, W., Vallius, M., Szidat, S., Prevot, A.S.H., Hueglin, C., Bloemen, H., Wahlin, P., Vecchi, R., Miranda, A.I., Kasper-Giebl, A., Maenhaut, W. And Hitzenberger, R. (2008). Source apportionment of particulate matter in Europe: A review of methods and results. Aerosol Science, 39, 827-849.

Wahlin, P., Berkowicz, R. and Palmgren, F. (2006). Characterisation of traffic-generated particulate matter in Copenhagen. Atmospheric Environment, 40(12), 2151-2159.

Wang, G., Kawamura, K., Xie, M., Hu, S., Gao, S., Cao, J., An, Z. and Wang, Z. (2009). Size distribution on n-alkanes, PAHs and hopanes and their sources in the urban, mountain and marine atmospheres over East Asia. Atmospheric Chemistry and Physics, 14: 8869-8882.

Wang, F., Ketzel, M., Ellermann, T., Wahlin, P., Jensen, S.S., Fang, D. and Massling, A. (2010). Particle number, particle mass and NOx emission factors at a highway and an urban street in Copenhagen. Atmospheric Chemistry and Physics, 10, 2745-2764.

Wang, X., Westerdahl, D., Wu, Y., Pan, X. and Zhang, K. M. (2011). On-road emission factor distributions of individual diesel vehicles in and around Beijing, China. Atmospheric Environment, 45(2), 503-513.

Watson, J.G., Chow, J.C., Lowenthal, D.H., Pritchett, L.C., Frazier, C.A., Neuroth, G.R. and Robbins, R. (1994). Differences in the carbon composition of source profiles for diesel- and gasoline- powered vehicles. Atmospheric Environment, 28(15), 2493-2505.

Watson, J. G., Zhu, T., Chow, J. C., Engelbrecht, J., Fujita, E. M. and Wilson, W. E. (2002). Receptor modeling application framework for particle source apportionment. Chemosphere, 49(9), 1093-1136.

Watson, J.G. and Chow, J.C. (2007). Receptor models for source apportionment of suspended particles.In Introduction to Environmental Forensics, 2nd Edition, 2, B. Murphy and R. Morrison (Eds.), Academic Press, New York, NY, 279-316.

Wehner, B., Birmili, W., Gnauk, T. and Wiedensohler, A. (2002). Particle number size distributions in a street canyon and their transformation into the urban-air background: measurements and a simple model study. Atmospheric Environment, 36, 2215-2223.

Wei, B. and Yang, L. (2010). A review of heavy metal contaminations in urban soils, urban road dusts and agricultural soils from China. Microchemical Journal, 94(2), 99-107.

Weingartner, E., Keller, C., Stahel, W. A., Burtscher, H. and Baltensperger, U. (1997). Aerosol emission in a road tunnel. Atmospheric Environment, 31(3), 451-462.

Weijers, E. P., Khlystov, A. Y., Kos, G. P. A. and Erisman, J. W. (2004). Variability of particulate matter concentrations along roads and motorways determined by a moving measurement unit. Atmospheric Environment, 38, 2993-3002.

Westerdahl, D., Wang, X., Pan, X. and Zhang, M.K. (2009). Characterization of on-road vehicle emission factors and microenvironmental air quality in Beijing, China. Atmospheric Environment, 43, 697-705.

Wik, A. and Dave, G. (2009). Occurrence and effects of tire wear particles in the environment--a critical review and an initial risk assessment. Environmental pPllution, 157(1), 1-11.

Wu, S.-P., Tao, S., Zhang, Z.-H., Lan, T. and Zuo, Q. (2007). Characterization of TSP-bound n-alkanes and polycyclic aromatic hydrocarbons at rural and urban sites of Tianjin, China. Environmental Pollution, 147(1), 203-210.

Yan, B., Zheng, M., Hu, Y., Ding, X., Sullivan, A. P., Weber, R. J., Baek, J., Edgerton, E. S. and Russell, A.G. (2009). Roadside, Urban, and Rural Comparison of Primary and Secondary Organic Molecular Markers in Ambient PM2.5. Environmental Science and Technology, 43(12), 4287-4293.

Yin, J., Harrison, R. M., Chen, Q., Rutter, A. and Schauer, J. J. (2010). Source apportionment of fine particles at urban background and rural sites in the UK atmosphere. Atmospheric Environment, 44, 841-851.

1930 Zhang, Y., Stedman, D. H., Bishop, G. A., Guenther, P.L. and Beaton, S. P. (1995). Worldwide On-
1931 Road Vehicle Exhaust Emissions Study by Remote Sensing. Environmental Science
1932 andTechnology, 29, 2286-2294.
1933
1934 Zhu, Y., Hinds, W. C., Kim, S. and Sioutas, C. (2002a). Concentration and Size Distribution of
1935 Ultrafine Particles Near a Major Highway. Journal and Air and Waste Management Association, 52,
1936 1032-1042.
1937
1938 Zhu, Y., Hinds, W. C., Kim, S., Shen, S. and Sioutas, C. (2002b). Study of ultrafine particles near a
1939 major highway with heavy-duty diesel traffic. Atmospheric Environment, 36(27), 4323-4335.
1940
1941 Zhu, Y., Kuhn, T., Mayo, P. and Hinds, W.C. (2006). Comparison of daytime and nighttime
1942 concentration profiles and size distributions of ultrafine particles near a major highway.
1943 Environmental Science and Technology, 40, 2531-2536.
1944
1945 Zhu, C.-S., Chen, C.-C., Cao, J.-J, Tsai, C.-J., Chou, C. C.-K., Liu, S.-C. and Roam, G.-D. (2010).
1946 Characterization of carbon fractions for atmospheric fine particles and nanoparticles in a highway
1947 tunnel. Atmospheric Environment, 44, 2668-2673.
1948

1949
1950

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)	

1988

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

1990

1991

1992 **Table 3:** Ratio of Cu to Sb characteristic of brake wear particles in ambient PM reported in the
1993 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

1994

1995

1996 **Table 4: (a)** Results from mass size distribution analysis for traffic-related PM as reported in the
1997 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

1998
1999 **(b)** Results from mass size distribution analysis for traffic-related elements as reported in the
2000 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

2001 **Table 5:** Comparison between dynamometer and tunnel/roadway measurements (Allen et al., 2001;
2002 Imhof et al., 2005a; Phuleria et al., 2006; Phuleria et al., 2007; Handler et al., 2008; He et al., 2008;
2003 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

2004

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)	5×10^{12} particles/veh/km (low HDV) $2\text{-}3 \times 10^{13}$ particles/veh/km (high HDV)
Abu-Allaban et al. (2002)	USA	Tunnel	$5.2\text{-}5.4 \times 10^{13}$ particles/veh/km (low HD fraction) $2.1\text{-}3.1 \times 10^{14}$ particles/veh/km (high HD fraction)
Jamriska et al. (2004)	Australia	Tunnel (diesel powered buses)	$3.11 \pm 2.41 \times 10^{14}$ particles/km
Kittleson et al. (2004)	USA	Roadside (particles > 3 nm)	$1.9\text{-}9.9 \times 10^{14}$ particles/km (gasoline-dominant fleet)
Jones and Harrison (2006)	UK	Roadside	5.84×10^{13} particles/veh/km 6.36×10^{14} particles/veh/km (LDV)
Westerdahl et al. (2009)	China	Roadside- LDV (prevalent during daytime)	1.8×10^{15} particles/kg fuel
		Roadside- HDV (prevalent during nighttime)	1.1×10^{16} particles/kg fuel
Wang et al. (2009)	Denmark	Highway	$(215 \pm 15) \, 10^{12}$ particles/veh/km
Ban-Weiss et al. (2010)	USA	Tunnel- LDV	$(3.9 \pm 1.4) \, 10^{14}$ particles/kg fuel
		Tunnel-HDV	$(3.3 \pm 1.3) \, 10^{15}$ particles/kg fuel
Wang et al. (2011)	China	Vehicle chase	7.1×10^{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$_{2.5 \text{ (rush hour)}}$	189 ± 23 mg/kg fuel
			PM$_{2.5 \text{ (mid-day)}}$	158 ± 29 mg/kg fuel
			PM$_{2.5 \text{ (early morning-HDV)}}$	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
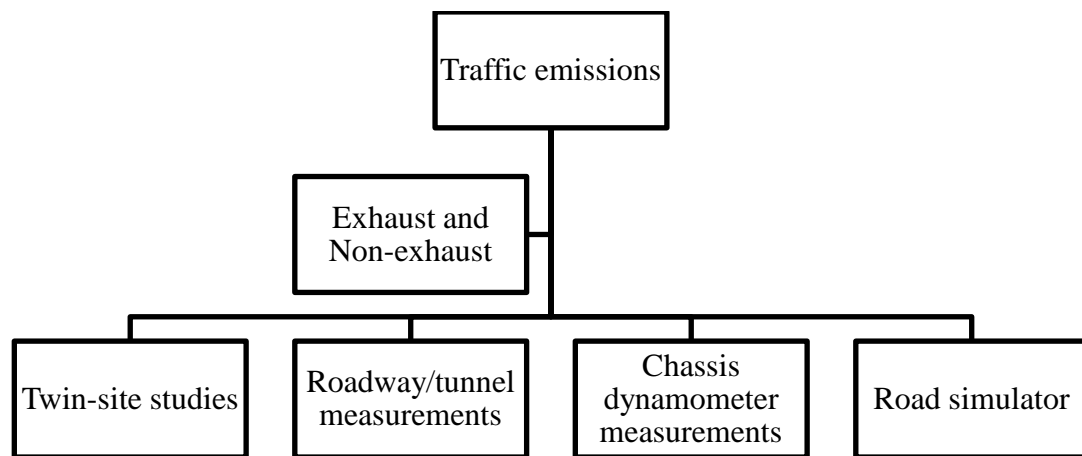
Figure 2: Methods for direct road traffic emissions analyses