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1 **Non-Exhaust Vehicle Emissions of Particulate Matter**
2 **and VOC from Road Traffic: A Review**

3
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36 **ABSTRACT**

37 As exhaust emissions of particles and volatile organic compounds (VOC) from road vehicles have
38 progressively come under greater control, non-exhaust emissions have become an increasing
39 proportion of the total emissions, and in many countries now exceed exhaust emissions. Non-
40 exhaust particle emissions arise from abrasion of the brakes and tyres and wear of the road surface,
41 as well as from resuspension of road dusts. The national emissions, particle size distributions and
42 chemical composition of each of these sources is reviewed. Most estimates of airborne
43 concentrations derive from the use of chemical tracers of specific emissions; the tracers and
44 airborne concentrations estimated from their use are considered. Particle size distributions have
45 been measured both in the laboratory and in field studies, and generally show particles to be in both
46 the coarse (PM_{2.5-10}) and fine (PM_{2.5}) fractions, with a larger proportion in the former. The
47 introduction of battery electric vehicles is concluded to have only a small effect on overall road
48 traffic particle emissions. Approaches to numerical modelling of non-exhaust particles in the
49 atmosphere are reviewed. Abatement measures include engineering controls, especially for brake
50 wear, improved materials (e.g. for tyre wear) and road surface cleaning and dust suppressants for
51 resuspension. Emissions from solvents in screen wash and de-icers now dominate VOC emissions
52 from traffic in the UK, and exhibit a very different composition to exhaust VOC emissions. Likely
53 future trends in non-exhaust particle emissions are described.

54

55 **KEYWORDS:** non-exhaust emissions; road traffic; particulate matter, wear, resuspension,
56 emissions, volatile organic compounds

57

58

59 **1. INTRODUCTION**

60 Road traffic has long been viewed as a major contributor to urban air pollution. The advent of
61 three- way catalytic converters on gasoline vehicles has led to a huge reduction in exhaust
62 emissions of carbon monoxide, hydrocarbons and NO_x (Winkler et al., 2018). Reductions in
63 emissions from diesel exhausts came much later, and these remain a major source of particulate
64 matter and NO_x pollution in cities globally (Harrison et al., 2021). Diesel Particle Filters are a
65 highly efficient means of controlling particulate matter emissions from diesels, and the latest
66 particle number emissions standards in Europe require the use of Gasoline Particle Filters.
67 Consequently, particulate matter emissions from traffic exhaust in developed countries, where new
68 vehicles have to adopt these technologies, have declined rapidly, and as a consequence, non-exhaust
69 emissions from road vehicles now widely exceed exhaust emissions (AQEG, 2019). According to
70 estimates reported by OECD (2020), the total amount of non-exhaust particulate matter (PM_{2.5})
71 emitted by passenger vehicles worldwide is likely to rise by 53% by 2030 from a 2017 baseline,
72 mainly due to increased vehicle mileage.

73
74 Non-exhaust emissions of particles arise mainly from four separate sources. Three of these arise
75 from abrasion: brake wear, tyre wear and road surface wear. The fourth, particle resuspension, arises
76 from road dust particles being suspended into the atmosphere by passing traffic. There are also
77 emissions of VOC from use of screenwash, which are rarely considered. All of these emission
78 types are generated by battery-electric vehicles as well as internal combustion engine vehicles, and
79 although their magnitude may change, road traffic will continue to be a source of particulate matter
80 and VOC emissions, even for a fully electric vehicle fleet.

81
82 Measurements of non-exhaust emissions come from both controlled laboratory studies and field
83 experiments. Brake wear, tyre wear and road surface wear can all be evaluated in the laboratory.
84 Brake wear can be determined under highly controlled conditions using a brake dynamometer, and

85 standardised methods allowing comparative studies of materials are under development. Methods
86 for evaluating tyre and road surface wear are more difficult to standardise and there are a range of
87 test methods, with relatively few data available. Such studies are, however, important as they are
88 the basis for deriving the emissions factors which form the building blocks for emissions
89 inventories, and are essential for numerical modelling of airborne concentrations. Resuspension is
90 more difficult to estimate. It is often not included in emissions inventories, and most estimates are
91 based upon algorithms using vehicle mass and road surface dust loading as input data. Non-exhaust
92 particles can also be measured in the atmosphere by use of chemical tracers, and by using receptor
93 modelling methods such as Positive Matrix Factorization. This is challenging and relatively few
94 data exist, although these have increased recently.

95

96 Past reviews of this topic, or specific aspects of it, have previously been published by Thorpe and
97 Harrison (2008), Grigoratos and Martini (2015), Amato et al. (2014) and AQEG (2019). It is
98 however, a topical and rapidly moving field, and hence we feel it valuable to provide a brief review
99 of the latest state of knowledge.

100

101 **2. INVENTORIES OF NON-EXHAUST EMISSIONS**

102 Emission inventories for many countries include estimates of emissions of PM₁₀ and PM_{2.5} from
103 tyre wear, brake wear and road abrasion and, in some cases, for road dust resuspension. The
104 methods used are mainly suited for estimating emissions on a national or regional scale as they
105 simplify the effect of vehicle size, type and technology, driving style and road condition. This is
106 normally done out of necessity when national statistics on vehicle population and mileage and travel
107 on different types of roads at different average speeds are used to estimate emissions on a large
108 scale. These approaches are less well suited to estimating emissions on a particular road section at a
109 particular time where emissions are expected to be highly variable.

110

111 In Europe, countries report emission inventories annually to conform with guidelines set out by the
112 European Environment Agency. This is to ensure consistency and comparability in inventories
113 reported by EU Member States. The EMEP/EEA Emissions Inventory Guidebook (EMEP/EEA,
114 2019) provides methods and emission factors for estimating emissions from tyre wear, brake wear
115 and road abrasion for different vehicle types. Emission factors are in mg/km and correction factors
116 are provided to account for different average speeds for mileages done on urban, rural and highway
117 roads and different vehicle loads in the case of heavy duty vehicles. Emissions from resuspension
118 of previously deposited material are not required to be reported and no method is provided in the
119 Guidebook. This is partly because this would be considered a double-count of an emission for
120 accounting purposes, though it is recognised that this source does have to be considered when
121 considering the full impact of traffic on ambient concentrations of PM in urban environments.

122

123 The emission factors provided in the EEA Guidebook are mostly based on analysis of data from
124 literature sources of nearly 20 years ago on wear rates for tyre and brake material combined with
125 estimates of the quantities that become airborne in different particle size ranges, with some data
126 from direct measurement using a simulated wheel or brake operation in the laboratory. The
127 factors may not well represent emissions from modern vehicle technologies and tyre and brake
128 systems and materials. Countries are permitted to use country-specific factors if these can be
129 justified and take account of national circumstances. For example, the Netherlands and Sweden
130 consider a smaller proportion of tyre wear and brake wear PM emitted in the PM_{2.5} range relative to
131 PM₁₀ compared with the values given in the Guidebook. Sweden and Finland apply a larger
132 emission factor for PM₁₀ from road abrasion to account for the effect of studded tyres relative to
133 non-studded tyres and use a much smaller PM_{2.5}/PM₁₀ ratio for this source to reflect the high
134 proportion of these emissions in the coarse particle range (see for example, Finland's 2020
135 Informative Inventory Report for Transport at <https://www.environment.fi/en->

136 US/Maps_and_statistics/Air_pollutant_emissions/Finnish_air_pollutant_inventory_to_the_CLRTA
137 P). Further details on emission factors used in Europe are given in AQEG (2019).

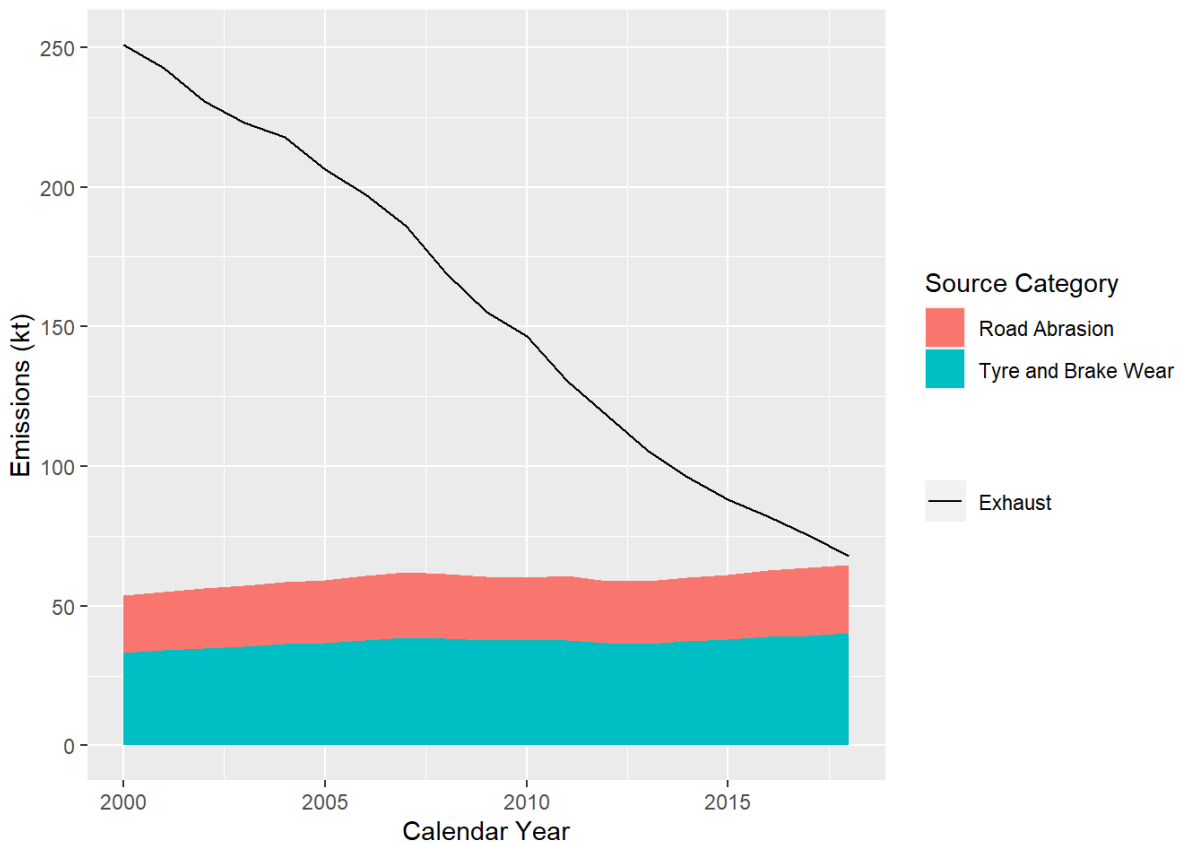
138

139 Without any regulation on non-exhaust emissions or understanding of how modern vehicle
140 technologies have changed emissions, the emission factors used in Europe remain constant over
141 time. As a consequence, all inventories show emissions from these sources increasing over time as
142 the numbers of vehicles and kilometres travelled have increased. In contrast, exhaust emissions in
143 most countries have been decreasing as newer vehicles meeting tighter emission standards enter the
144 fleet, particularly in recent years as increasing numbers of diesel vehicles are fitted with particulate
145 filters.

146

147 Figure 1 shows the trend in exhaust and non-exhaust emissions of $PM_{2.5}$ for all EU28 countries
148 combined from 2000-2018 taken from inventories officially submitted by the Parties to the UNECE
149 Convention on Long-range Transboundary Air Pollution (EEA, 2021). Inventory data submitted
150 by countries combine emissions from tyre and brake wear in a single category, but road abrasion
151 emissions are reported separately. Whilst there are some differences between individual countries,
152 most are showing the same general trends as Figure 1. For most countries, each of the three non-
153 exhaust sources contribute roughly similar amounts and there is no dominant source, though
154 countries such as Sweden and Finland have a much greater contribution of road abrasion to the
155 PM_{10} inventory.

156



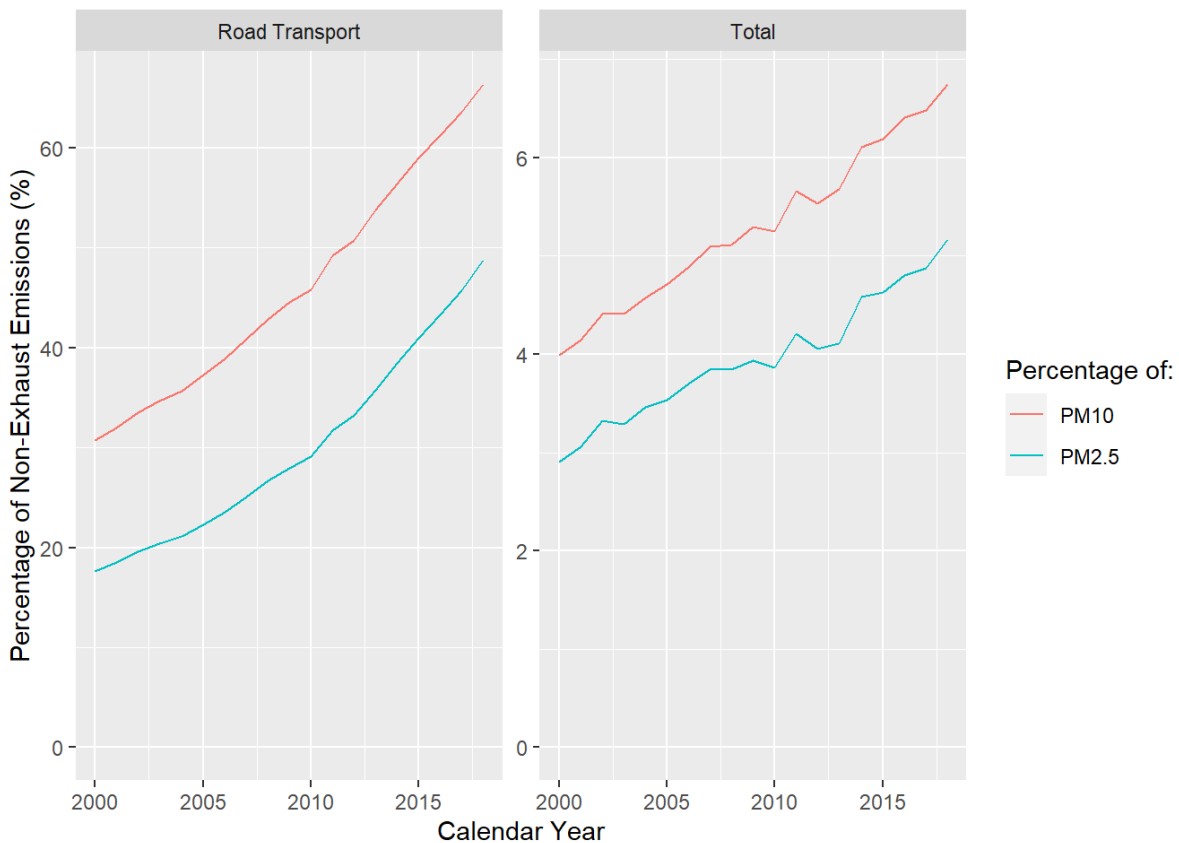
157

158 **Figure 1:** Total exhaust and non-exhaust road transport emissions of PM_{2.5} from EU28 countries
 159 from emission inventories reported by Parties under the Convention on Long-range Transboundary
 160 Air Pollution in 2020, covering years up to 2018 (EEA, 2021).

161

162 Figure 2 shows non-exhaust emissions of PM_{2.5} and PM₁₀ for EU28 countries combined as a
 163 percentage share of total road transport and of all national primary emissions. These trends imply
 164 that the share of road transport emissions from non-exhaust sources has been increasing with time,
 165 rising from 18% in 2000 to 49% in 2018 in the case of PM_{2.5} and 31% to 66% in the case of PM₁₀.
 166 As a share of all primary emissions in the EU28, non-exhaust emissions increase from 2.9% in 2000
 167 to 5.2% in 2018 in the case of PM_{2.5} and 4.0% to 6.7% in the case of PM₁₀.

168



169

170 **Figure 2:** Percentage share of total non-exhaust emissions of PM_{2.5} and PM₁₀ for EU28 countries
 171 combined, as a percentage share of total road transport and of all national primary emissions (shown
 172 in the figure marked ‘Total’). Based on data from EEA (2021).
 173

174 However, the range in the contribution made by non-exhaust emissions to total road transport
 175 emissions between countries is very wide, according to the official inventories of EU28 Member
 176 States (EEA (2021), these include the UK for the purpose of these comparisons). For PM_{2.5}, the
 177 range in 2018 varies from 11% to 87% and may reflect the different transport activities in each
 178 country as well as country-specific circumstances, such as the use of studded tyres in Sweden or it
 179 may reflect the use of different emission factors. The range in the PM_{2.5} share in emissions is
 180 smaller for the largest European countries (UK, France, Germany, Spain, Italy), from 42% to 67%
 181 of all road transport emissions in 2018.

182

183 The National Emissions Inventory for the U.S. includes emissions of PM₁₀ and PM_{2.5} from tyre and
 184 brake wear and resuspension. Emissions from tyre and brake wear are included with exhaust

185 emissions in the ‘On-road – Mobile’ category and cannot be viewed separately; emissions from
186 resuspension are reported separately under ‘Paved road dust’.

187

188 Tyre and brake wear emissions are estimated by the USEPA MOVES tool (USEPA, 2020a). Tyre
189 wear emissions in MOVES are derived from wear rates correlated with average vehicle speed,
190 similar to the approach used in Europe. It assumes that 8% of tyre wear is emitted as PM₁₀, 1.5% as
191 PM_{2.5}. This leads to an average PM₁₀ factor for passenger cars of 5 mg/km which is comparable to
192 the values used in Europe, but the average value for trucks is lower than European values at less
193 than 20 mg/km. This may be due to different assumptions about vehicle sizes and average speed
194 and how tyre wear emissions are correlated. The US approach implies a much smaller tyre wear
195 PM_{2.5}/PM₁₀ ratio of 0.15 on average (USEPA, 2020a), compared with 0.7 in Europe (EMEP/EEA,
196 2019).

197

198 MOVES uses g/hour emission rates for brake wear based on brake dynamometer and wind tunnel
199 measurements at different deceleration rates for US-specific brake materials (USEPA, 2020a).
200 These are combined with estimates of the amount of braking done at different deceleration levels
201 for different real-world vehicle operating modes defined in terms of vehicle specific power, a more
202 detailed approach than the Guidebook approach used in Europe. The average brake wear emission
203 factor for PM₁₀ calculated in MOVES for passenger cars in the US inventory is quoted as 14 mg/km
204 which is higher than the UK average of 7mg/km derived from the UK’s national inventory (NAEI,
205 2020) using factors from EMEP/EEA (2019), but MOVES implies a much smaller brake wear
206 PM_{2.5}/PM₁₀ ratio of 0.12 on average (USEPA, 2020a), compared with 0.4 in Europe (EMEP/EEA,
207 2019).

208

209 Emissions from resuspension in the U.S. inventory, categorised under the term ‘paved road dust’
210 are calculated in a method described in the USEPA’s AP-42 compilation manual (USEPA, 2011).

211 A near-linear empirical equation is used that relates an emission factor in g/km to vehicle mass and
212 silt loading of the road surface. A correction is made for the number of precipitation days in the
213 model period. Road surface and environmental conditions are highly variable and the factors from
214 this approach are highly uncertain and not necessarily universal as it applies only to the range of
215 variables from which the equation was derived. The applicability to other conditions of vehicle
216 speed, mass and road conditions may be limited. AP-42 provides an alternative equation for
217 unpaved roads.

218

219 Comparing non-exhaust emissions from on-line national inventory data sources for the U.S. and
220 Europe is difficult on a like-for-like basis because the scope of coverage and accessibility of data
221 are different. Europe shows tyre and brake wear emissions (combined) and road abrasion emissions
222 separately, but excludes resuspension; the U.S. includes resuspension (paved road dust), but
223 combines tyre and brake wear with exhaust emissions. A report by Panko et al. (2013) states that
224 according to the USEPA, tyre wear contributed 15% of total vehicle emissions of PM₁₀ in 2008
225 which compares with a value of 17% in the UK for the same year (NAEI, 2020). On-line inventory
226 data are available for the State of California where emissions for all sources can be seen with tyre
227 and brake wear and resuspension (paved road dust) viewed separately. Combining data extracted
228 from the CARB mobile emissions data source EMFAC (CARB, 2017) and the emissions tool
229 CEPAM (CARB, 2016) for all other sources in 2018, the percentage contributions of tyre and brake
230 wear and resuspension relative to all road transport emissions and as a percentage of all California
231 emissions can be derived.

232

233 Table 1 compares these contributions for the UK, Europe, California and the U.S. as far as possible
234 on a like-for-like basis. A comparison between the UK, Europe and California excludes
235 resuspension as this is not covered in European inventories. Only emissions for resuspension can
236 be shown for the U.S. The table shows comparisons in contributions as a percentage of all road

237 transport emissions in the country/region and as a percentage of all primary PM emissions for the
 238 year 2018, apart from the U.S. where the most recent data is for 2017.

239
 240 **Table 1:** Contribution of non-exhaust sources to road transport and all primary sources of PM
 241 emissions in Europe and the U.S. These are according to current estimates from emission
 242 inventories for the respective regions, with comparisons done on a like-for-like basis, as far as is
 243 possible. Data are derived for the UK from NAEI (2020), for EU28 from EEA (2021), for
 244 California from CARB (2016) and CARB (2017) and for the USA from USEPA (2020a).
 245

				PM_{2.5}	PM₁₀
UK	2018	Tyre, brake, road abrasion	as % of all road transport emissions (exc resuspension)	67%	79%
		Tyre, brake, road abrasion	as % of all primary source emissions (exc resuspension)	8%	9%
EU28	2018	Tyre, brake, road abrasion	as % of all road transport emissions (exc resuspension)	49%	66%
		Tyre, brake, road abrasion	as % of all primary source emissions (exc resuspension)	5%	7%
California	2018	Tyre & brake	as % of all road transport emissions (exc resuspension)	71%	85%
		Tyre & brake	as % of all primary source emissions (exc resuspension)	7%	5%

246

				PM_{2.5}	PM₁₀
USA	2017	Resuspension	as % of all road transport emissions	65%	79%
		Resuspension	as % of all primary source emissions	4%	5%
California	2018	Resuspension	as % of all road transport emissions	46%	72%
		Resuspension	as % of all primary source emissions	7%	11%

247
 248 Emissions as a fraction of all primary emissions in the country/region will reflect the quantity of
 249 emissions from non-transport sources which can be quite variable. Even so, it is clear to see that
 250 resuspension alone makes a large contribution to area-wide emissions from road transport of PM_{2.5}
 251 and PM₁₀ in the U.S., according to the inventories. In Europe, where emissions from resuspension
 252 are excluded from inventories, it is clear how even emissions from the other non-exhaust sources
 253 make large contributions to total road transport emissions of PM, often more than 50% in 2018, and
 254 this is consistent with the situation in California where the contributions from tyre and brake wear
 255 may be even greater. The inventory for California suggests that when all non-exhaust sources are

256 accounted for, including resuspension, they contribute to 96% of all emissions of PM₁₀ from road
257 traffic and 15% of primary PM₁₀ emissions from all sources in 2018 (CARB, 2016, 2017).

258

259 There are a few reported estimates of non-exhaust emissions from road transport in China. Zhang
260 et al. (2020) recently conducted a comprehensive study combining tunnel measurements, laboratory
261 dynamometer and resuspension experiments and chemical mass balance modelling to estimate road
262 dust resuspension, tyre wear and brake wear emissions in four Chinese megacities. The
263 experiments yielded emission factors for PM_{2.5} not dissimilar to the factors used in Europe for tyre
264 and brake wear, but the authors acknowledged their factors could be underestimates, not typical of
265 most traffic in China because of the smooth traffic flow and low amount of braking in the tunnels.
266 The authors also concluded a total inventory for non-exhaust emissions of PM_{2.5} in China of 11.4
267 ktonnes based on measurements done in 2017/18, of which 8.1 ktonnes were from resuspension
268 (Zhang et al, 2020). Although it is not clear whether this value only applied to light duty traffic, the
269 value seems low when one considers the tyre and brake wear and road abrasion emissions alone in
270 the UK were 8.7 ktonnes in 2019 (NAEI, 2020), emissions from resuspension on paved roads were
271 209 ktonnes in the U.S. in 2017 (USEPA, 2017) as well as the fact this would be small relative to
272 estimates of total road transport emissions of PM₁₀ in China ranging from 40 – 400 ktonnes in 2008,
273 depending on inventory source (Saikawa et al., 2017).

274

275 An air pollutant emissions inventory is available for road transport in 2013 in Australia (Smit,
276 2014), developed using the same method as used in Europe, from the EMEP/EEA Emissions
277 Inventory Guidebook. The inventory suggests non-exhaust emissions were responsible for 26% of
278 all road transport emissions of PM_{2.5}, similar to the contributions in Europe for 2013 (EEA, 2021).

279

280 The OECD has published a highly detailed review of non-exhaust emissions of PM which considers
281 the causes, consequences and policy responses to tackle these emissions globally (OECD, 2020).

282 The report considered the limitations of emission inventories, including the inclusion or otherwise
283 of resuspension, and reviews inventories for different regions of the world, mainly Europe and the
284 U.S., but also in parts of Latin America. Inventories for Mexico, Bogota in Colombia, and Santiago
285 in Chile, all highlight the importance of road dust resuspension with contributions to total primary
286 emissions from all sources ranging from 16% to 54% (PM₁₀) and 9% to 56% (PM_{2.5}) in these
287 regions. The contribution of tyre and brake wear emissions to total emissions in Bogota was much
288 less than that, at 1.4% for PM₁₀ and 0.8% for PM_{2.5}.

289

290 In a review of tyre wear and its impact on the environment, Kole et al. (2017) estimated the global
291 emission of tyre wear to be 5.9 million tonnes per year. On the assumption that 3-7% of this
292 becomes airborne PM_{2.5} emissions (Kole et al., 2017; Grigoratos and Martini; 2014), a global
293 emission rate of 175-410 ktonnes PM_{2.5} from tyre wear is implied.

294

295 The OECD conclude that globally in 2014 at least 50% of total traffic emissions of PM₁₀ and 5% of
296 primary PM₁₀ from all sources were non-exhaust emissions, even when resuspension is excluded.
297 Corresponding figures for PM_{2.5} are 34% and 4%, respectively. When resuspension is taken into
298 account, non-exhaust emissions represent 95% of all road traffic emissions and 15% of all source
299 emissions, as stated earlier for California.

300

301 Non-exhaust sources do not only contribute to airborne PM₁₀ and PM_{2.5}, but also various metals and
302 polyaromatic hydrocarbons (AQEG, 2019). Emissions are generally related to the composition of
303 materials used in tyres and brake linings. Inventories include emissions of several different metals
304 and PAH species based on factors from fairly old literature sources, though some countries use rates
305 based on more recent composition studies. Some countries take account of the recent prohibition of
306 “PAH-rich” extender oils used in tyre production leading to a reduction in PAH emissions from tyre
307 wear (AQEG, 2019).

308 There are large uncertainties in emission inventories for non-exhaust sources. This is in part due to
309 the difficulty in measuring emissions, but also because of the high variability in emissions and the
310 factors which influence them. These are referred to in the studies mentioned above and in
311 Guidebooks and reports describing inventory methodologies. Emissions depend on driving style,
312 speed and the amount of acceleration and deceleration and cornering, weight and load of the
313 vehicle, the design of the brake system, type and composition of tyre and brake materials, the
314 condition and roughness of the road surface and ambient temperature, precipitation and humidity.
315 Inventories necessarily approximate and average out their effects or parameterise them in fairly
316 simple ways. Emissions from brake wear are not continuous but are inevitably greatest where
317 braking events are most significant. Methods have been developed to map brake wear emissions at
318 high spatial resolution where there are areas of high braking intensity according to changes in
319 vehicle specific power based on real-world vehicle measurements data associated with different
320 road situations (FAT, 2019). However, these methods have not yet been applied to inventories.

321

322 **3. PHYSICAL PROPERTIES OF NON-EXHAUST EMISSIONS**

323 **3.1 Brake Wear**

324 Brake wear particles arise from wear of both the brake lining (pad) and the rotor (disc) or drum.
325 There are three major types of brake lining, referred to as non-asbestos organic (NAO), low metallic
326 (LM) and semi-metallic (SM) (Kukutschova and Filip, 2018), each with a different composition and
327 favoured in different markets. Ceramic linings are also used on some of the more expensive
328 vehicles (Ostermeyer and Muller, 2008). The lining is made up from four main components:
329 binders, fibres, fillers and friction modifiers, and according to Roubicek et al. (2008), more than
330 3000 materials are used across brands. This is reflected in a very wide range of chemical
331 composition exemplified by the study of Hulskotte et al. (2014). The compositional differences
332 affect the rate of particle emissions (Kim et al., 2020), and most probably the physical properties of
333 emissions, but this has not been systematically studied. At normal brake operating temperatures

334 (<150°C), brake wear particles are formed by abrasion due to friction between the pad and rotor or
335 drum. They are irregular in shape with strong angular features, and contain multiple elements in the
336 same particle (Wahlstrom et al., 2010a). Exceptionally, at higher temperatures, ultrafine particles
337 are formed, and appear broadly similar in shape to the larger particles (Wahlstrom et al.,
338 2010a). Many studies have been conducted under controlled conditions, giving a range of results for
339 median diameters, which may arise from different designs of measurement system, or different
340 brake materials. In tests of various brake linings including both NAO and SM types using a brake
341 dynamometer, Garg et al. (2000) reported highly varied particle mass size spectra, which were
342 attributed by Sanders et al. (2003) in part to particle losses in the sampling system. Garg et al.
343 (2000) reported that between 7 – 48% of the particles became airborne, and at a brake temperature
344 of 100°C, of the particles sampled by a MOUDI impactor, 80%, 68% and 26% of mass was below
345 10 µm, 2.5 µm and 0.1 µm respectively, with a mass median diameter of 2.49 +/- 3.47 µm. Sanders
346 et al. (2003) measured from NAO, LM and SM types on a brake dynamometer and found particle
347 mass size spectra with medians at around 5 -6 µm and modes at 3-4 µm, despite the use of different
348 braking scenarios. Iijima et al. (2008) studied NAO linings and reported a mass concentration mode
349 at 3 - 6 µm. In tests of NAO linings, Hagino et al. (2016) found 2 – 21% of brake wear particles
350 became airborne, and particle mass was predominantly in the 0.8 – 2.2 µm size range. Various
351 brake pad and disc materials were tested by Wahlstrom et al. (2009; 2010b) in a laboratory pin-on-
352 disc rig, who found a mode in the mass distribution around 3 µm.

353

354 Brake wear size distributions have also been estimated from field measurements. In a receptor
355 modelling study, Wahlin et al. (2006) apportioned a range of elements to the brakes source:
356 chromium (Cr), iron (Fe), copper (Cu), zinc (Zn), zirconium (Zr), molybdenum (Mo), tin (Sn),
357 antimony (Sb), barium (Ba) and lead (Pb), and reported that particles created by brake abrasion had
358 mass median aerodynamic diameters around 2.8 µm. In a Positive Matrix Factorization (PMF)
359 study of wide range particle size spectra at a roadside site, Harrison et al. (2011) identified a source

360 of airborne particles which had a mode at 3 μm diameter and was associated with traffic-influenced
361 wind directions and heavy duty vehicle numbers which they concluded was caused by brake wear.
362 Similarly, analysing metals data from the same site (Marylebone Road, London), Harrison et al.
363 (2012) report a mass modal diameter of 3 μm for Ba, used as a tracer of brake wear. Similar modes
364 were seen for other elements such as Fe, Cu and Sb, associated with brake wear.

365

366 Sanders et al. (2003) in dynamometer and test track studies reported considerable difference in total
367 brake wear rates with low metallic (LM) pads emitting 3-4 times the material of semi-metallic (SM)
368 and non-asbestos organic (NAO) linings, but a consistent presence of Fe, Cu and Ba and a mass
369 median diameter of ca. 6 μm . Working in road tunnels in the United States, Lough et al. (2005)
370 measured mass modes at 3-6 μm diameter for brake-associated metals (Cu, Ba, Fe, Sb). According
371 to Gietl et al. (2010), the dominant mass mode of iron, copper, barium and antimony found at
372 Marylebone Road, London was between 1.2 and 7.2 μm aerodynamic diameter, and peaked at
373 around 3 μm . In analysis of antimony mass size spectra, Iijima et al. (2009) identified a bimodal
374 structure and attributed a mode at 3.6-5.2 μm as being due to brake dust. In the vast majority of
375 these studies, the modal diameter of brake wear particles is close to 2.5 μm , the diameter
376 conventionally taken in regulations as the boundary between fine and coarse particles. The
377 consequence is that when apportioning particulate matter mass into fine ($\text{PM}_{2.5}$) and coarse ($\text{PM}_{2.5-10}$)
378 fractions, significant quantities of brake wear particulate matter is recorded in both fractions.
379 Minor differences in experimental conditions can therefore result in substantial differences in the
380 $\text{PM}_{2.5}:\text{PM}_{2.5-10}$ ratio as seen in studies above (Garg et al., 2000; Iijima et al., 2008). Nosko and
381 Olofsson (2017) report brake wear particles to have an effective density ($0.75 \pm 0.2 \text{ g cm}^{-3}$), far
382 lower than the bulk density of the constituent material, and concluded that the particles are porous.

383

384 There has been much recent interest in emissions of nanoparticles from brake wear. Since these are
385 very small (defined as $<100\text{nm}$ diameter), when present they typically account for a very large

386 number of the particles, but very little mass. In an early study of the wear of brake components,
387 Roubicek et al. (2008) identified the emission of nanoparticles, using emission of CO₂ from the
388 brake pad as a measure of wear. Using a laboratory pin-on-disc apparatus, Wahlstrom et al. (2009;
389 2010b) found modes in the number distribution of particles emitted from various brake pad and disc
390 materials of around 350 nm and 550 nm, with a tail extending to much smaller sizes. As the
391 nanoparticle fraction is likely to arise from vaporisation-condensation of metals, rather than
392 abrasion, it is unsurprising that subsequent research has shown that emissions increase sharply
393 above a critical temperature (160-190°C) (Mathissen et al., 2018; Gonet and Maher, 2019). It is
394 reported that this temperature is not normally achieved during on-road vehicle usage (Farwick zum
395 Hagen et al., 2019a,b). In a pin-on-disc study, such particles could be as small as 1.3-10 nm
396 diameter (Nosko et al., 2017). Consistent with this, Mathissen et al. (2011) identified a unimodal
397 particle number size distribution near the disc brake, with a maximum at 11 nm.

398

399 In measurements within a road tunnel, Lough et al. (2005) observed a sub-micron mode in
400 atmospheric measurements of Cu, Fe, Pb and Ca, while Gietl et al. (2010) reported a secondary
401 mass mode of Cu and Sb in roadside tests at a diameter of 0.2 – 0.4 µm. These appear likely to be
402 associated with brake wear, although other sources, such as combusted lubricating oil may also
403 contribute for some elements such as Ca and Zn which are present in oil additives. Iijima et al.
404 (2008) reported a number concentration mode at 0.8 µm in brake dust spectra, but in a subsequent
405 paper, Iijima et al. (2009) suggest that a mode in the Sb size spectrum at 0.5 – 0.7 µm may have
406 been due to fly ash from waste incineration.

407

408 **3.2 Tyre Wear**

409 There are limited data on tyre wear particle emissions when compared with brake wear. Abrasion
410 occurs due to shear and friction forces arising from the contact between the vehicle wheel and the
411 road surface, and there is observational evidence of particle emissions containing fragments of road

412 surface material embedded in tyre wear particles, referred to as Tyre Road Wear Particles (TRWP).
413 The rate of tyre wear is affected by road surface roughness by factors of up to two to three
414 (Kennedy et al., 2002), and increased emissions may occur when the road surface is unpaved, or
415 especially when studded tyres are in use (Gustafsson et al., 2009). The latter are used to give grip in
416 icy conditions, and work has been carried out in Scandinavia (Johansson et al. 2012; Denby et al.,
417 2013) to predict the emissions from roads subject to damage from studded tyres and the addition of
418 grit to aid traction during winter. This will include both tyre and road surface wear.

419

420 Electron microscopy can reveal the composition and morphology of tyre dust particles. Studies by
421 Kreider et al. (2010) and Panko et al. (2013) have shown from on-road studies the association of
422 tyre rubber internally mixed with other constituents such as road surface wear, road dust or other
423 traffic-related sources within individual particles, known as Tyre Road Wear Particles (TRWP).
424 Aerosol Time-of-Flight Mass Spectrometry (ATOFMS) is an alternative form of single particle
425 analysis and has also shown an association of crustal elements with particles derived from tyre dust
426 (Dall'Osto et al., 2014). The particles are typically elongated with sausage-like shapes (Baensch-
427 Baltruschat et al., 2020; Klockner et al., 2021).

428

429 The majority of the mass of tyre wear particles is relatively large (Nielsen et al., 2003; Adachi and
430 Tainosho, 2004; Kreider et al., 2010) including elongated particles in excess of 100 μm in size, with
431 metal and mineral particles embedded within them. Kennedy et al. (2002) estimated that 80% of
432 tyre wear debris is deposited on the carriageway surface, with only 20% becoming airborne (as
433 TSP). According to ten Broeke et al. (2008), 5% of tyre wear occurs in the PM_{10} fraction.
434 Pischinger and Rauterberg-Wulff (1999) used elemental carbon and low volatility organic carbon to
435 distinguish between diesel exhaust and tyre debris in PM_{10} and $\text{PM}_{2.5}$ samples collected in a
436 highway tunnel, and report tyre wear particles only in the coarse fraction ($\text{PM}_{2.5-10}$).

437

438 Laboratory studies have examined the airborne particles arising from tyre wear. Dominant mass
439 modal diameters of tyre emission particles have been found at 2 μm (Sjodin et al., 2010) and at 2 –
440 5 μm (with an average size of 3.8 μm) by Raisenen et al. (2003). Kupiainen (2007) observed a mass
441 size spectrum with mode at 5 – 8 μm when operating a friction tyre on the rig used by Gustafsson et
442 al. (2009). Raisenen et al. (2003) found that the particle size distribution was not dependent on
443 asphalt or aggregate mineralogy. In tests on a laboratory rig and in on-road tests, Kreider et al.
444 (2010) found particle volume size spectra with modes at 50 μm to 100 μm , and bimodal particle
445 number concentrations with modes at 25 μm and 5 μm in the on-road data. They do not directly
446 report the fraction of particles below 10 μm or 2.5 μm , but it can be inferred from the volume size
447 spectra which show it to be very small. Park et al. (2018) working on a tyre wear simulator
448 observed a mass modal diameter of around 2 μm , which broadened under harsh friction conditions.
449 Kim and Lee (2018) found tyre wear modal diameters of 3 and 4 μm in unimodal distributions,
450 which is close to the mode in Zn concentrations attributed primarily to tyre wear measured on
451 Marylebone Road, London by Harrison et al. (2012).

452

453 Tyre wear is also able to generate nanoparticle emissions (<100 nm diameter). Mathissen et al.
454 (2011) measured particle number concentrations in the 6 – 562 nm size range near the contact point
455 on the road surface of a car travelling on a test track. This revealed a mode in the number size
456 distribution at 30 – 60 nm (mobility diameter) during acceleration, braking and extreme cornering.
457 During full lock braking, smoke was visible behind the tyre and the modal diameter increased to 60
458 – 70 nm. There was a general relationship observed between particle number concentration and
459 speed. An ultrafine (nanoparticle) mode in the tyre wear size spectrum has also been reported by
460 Kwak et al. (2014), Kim and Lee (2018), Foitzik et al. (2018) and Park et al. (2017). When
461 measuring ultrafine particle number emissions of studded and winter tyres on a laboratory rig, Dahl
462 et al. (2006), found that particle number emissions in the 15 – 700 nm size range increased with
463 vehicle speed, and that the smallest particles are volatile. This implies that the nanoparticle fraction

464 arises from a vapourisation/ condensation process involving oils used in the tyre, in contrast to the
465 larger particles which arise from abrasion (Kwak et al., 2014). This is consistent with a threshold
466 tyre tread temperature of 160°C for generation of this mode reported by Park et al. (2017), with
467 increased nanoparticle generation as the temperature increased.

468

469 **3.3 Road Surface Wear**

470 As explained above, road surface wear particles are normally emitted internally mixed in Tyre Road
471 Wear Particles (TRWP) and hence particle size spectra measured for tyre wear under field or
472 realistic laboratory conditions are likely to be a good representation for road surface wear.

473 However, there have been studies with a mobile load simulator and a road simulator. Gehrig et al.
474 (2010) operated a mobile load simulator and found road wear particles to have a main mode at 6 – 7
475 μm , and a smaller shoulder at ca. 12 μm . Operating a road simulator and studded tyres, Gustafsson
476 and Johansson (in Gustafsson, 2018) reported a mode at 5-6 μm , with particle mass orders of
477 magnitude greater than the mobile load simulator with conventional tyres. The main constituent of
478 road surfaces is mineral material (Gustafsson, 2018) which is unlikely to be differentiated from
479 resuspension on the basis of its chemical composition.

480

481 **3.4 Resuspension Particles**

482 A number of studies have sought sample resuspension by placing a sample inlet behind a tyre on a
483 moving vehicle. These have found a mode at 3 – 5 μm , regardless of whether a standard (summer)
484 or studded tyre was used (Hussein et al., 2008). Lee et al. (2013) report a number mode at 800 nm
485 with a concentration that did not vary with vehicle speed, while the mass mode at 2 – 3 μm saw a
486 slight increase in concentration with vehicle speed. A larger mass mode in the 6 – 10 μm diameter
487 range was found from sampling in a road tunnel by Lough et al. (2005) for the geological elements
488 magnesium (Mg) and calcium (Ca), most likely attributable to resuspension, or possibly road
489 surface wear. Harrison et al. (2011) applied Positive Matrix Factorization to wide range particle size

490 spectra measured on Marylebone Road, London, and identified a factor with a mode at diameter
491 $>10\ \mu\text{m}$ which occurred at traffic-influenced wind directions, and during the morning rush hour
492 when Heavy Duty Vehicle flow was greatest. Using Si as a tracer, Harrison et al. (2012) identified
493 the contribution of resuspension emissions on Marylebone Road, London, finding a mass modal
494 diameter of around $5\ \mu\text{m}$, coarser than the brake or tyre wear, extending to sizes beyond $10\ \mu\text{m}$.

495

496 **4. CHEMICAL COMPOSITION OF NEE**

497 **4.1 Brake Wear Particles**

498 Brake linings of frictional braking systems can be described as metallic, semi-metallic and non-
499 asbestos organic (NAO) according to the proportions of metallic and mineral and/or organic
500 material present (Chan and Stachowiak, 2004; Sanders et al., 2003). Across these classifications, the
501 chemical components in the brake linings typically derive from the following five categories of
502 function, listed here in decreasing order of contribution by mass (Grigoratos and Martini, 2015;
503 Thorpe and Harrison, 2008).

- 504 • Fillers (15-70%): cheaper materials to provide bulk and which include minerals such as barite
505 (BaSO_4), calcite (CaCO_3) and mica (silicates), and particles of rubber.
- 506 • Binders (20-40%): to maintain structural integrity of the brake linings under mechanical and
507 thermal stress and which typically comprise phenolic resins.
- 508 • Fibres (6-35%): to provide mechanical strength and which may be composed of various metals,
509 carbon, glass, Kevlar and, to a lesser extent, ceramic fibres.
- 510 • Friction modifiers or lubricants (5-29%): to stabilise the frictional properties and which are
511 typically composed of graphite, metal sulphides such as antimony trisulphide (Sb_2S_3), and
512 metal oxides.
- 513 • Abrasives (up to $\sim 10\%$): to increase friction and which may comprise a variety of substances
514 including aluminium oxide, iron oxides, silicon oxides (quartz), zirconium oxide and zirconium
515 silicate.

516 The broad range and quantities of chemical components listed above reflects the wide variety of
517 formulations used in brake linings in practice (Blau, 2001; Chan and Stachowiak, 2004), which is
518 also driven by commercial confidentiality. The plethora of materials used in brake liners means that
519 particles emitted from the braking process likewise contain a multitude of chemical components
520 (Grigoratos and Martini, 2015; Kukutschová et al., 2011). In addition, because of the wide variation
521 in resistance to mechanical disintegration and volatilisation across the chemical constituents, the
522 composition of brake-wear particles significantly differs from that of the original lining material
523 (Kukutschová et al., 2011; Österle et al., 2001). A substantial increase in ultrafine particle number
524 emissions when localised brake temperatures exceed $\sim 170^{\circ}\text{C}$, which is attributable to evaporation
525 and condensation of brake binder material (Perricone et al., 2018), may not contribute significantly
526 to mass of brake-wear emissions.

527

528 As the elemental content of brake pads is extremely variable (Hulskotte et al., 2014), almost the
529 complete suite of metallic elements (plus Sb and As) have been detected in brake linings and in
530 brake dust (Hulskotte et al., 2014; Grigoratos and Martini, 2015; Thorpe and Harrison, 2008).
531 However, certain elements stand out in brake-wear particles, either because they are in greater
532 abundance or because brake wear is a dominant source of these particles in ambient air and
533 therefore they act as useful tracers for brake-wear emissions. The four elements most readily
534 identified in brake-wear particles are Fe, Cu, Ba and Sb, which also tend to correlate well with each
535 other (e.g. Gietl et al., 2010; Lawrence et al., 2013) indicating a common source. Formerly, Pb was
536 also a significant component of brake wear but this is no longer the case due to the replacement of
537 Pb in modern linings (Grigoratos and Martini, 2015).

538

539 For Cu, Ba and Sb, brake wear emission is now their major source into ambient air, away from
540 specific industrial point source emissions (Hjortenkrans et al., 2007). For example, the UK National
541 Atmospheric Emissions Inventory (NAEI) estimates that, in 2016, 90% of UK national emissions of

542 Cu derived from non-exhaust emissions, a percentage that will be higher still in areas with large
543 road traffic activity. This is consistent with the 80-90% estimates made for the UK by Denier van
544 der Gon et al. (2007). Brake-wear emissions are also an important source of atmospheric emissions
545 of Sb (Bukowiecki et al., 2009; Iijima et al., 2008; Varrica et al., 2013): many brake linings contain
546 up to a few % of stibnite (Sb_2S_3), which can be oxidised to Sb_2O_3 during the braking process.
547 The three elements Cu, Ba and Sb, have all been used as chemical tracers for brake wear in source
548 apportionment studies (Alves et al., 2020; Amato et al., 2010a; Charron et al., 2019; Dong et al.,
549 2017; Dongarrà et al., 2008; Gietl et al., 2010; Hicks et al., 2021; Lawrence et al., 2013; Lough et
550 al., 2005; Pant et al., 2015). Where a mass ratio of the element to brake-wear particles can be
551 estimated or assumed, then quantification of that element in ambient air provides a quantitative
552 measure of the contribution of brake-wear emissions at the measurement location. For example,
553 Gietl et al. (2010) used Ba as a tracer for the contribution of brake-wear particles to different size
554 fractions of ambient PM at a roadside and an urban background site in central London. These
555 authors estimated that the mean proportion of Ba in brake wear emissions in PM_{10} (for the UK, at
556 that time) was 1.1%. Hicks et al. (2021) also used Ba as a tracer, applying a scaling factor of 91 to
557 their roadside increment in Ba concentration to derive the contribution to PM_{10} from brake wear.
558 Charron et al. (2019) report that ratios involving copper (Cu/Fe and Cu/Sn) could be used as brake-
559 wear emissions tracers as long as brakes with Cu remain in use.

560

561 Other studies that applied multivariate statistical techniques to a suite of PM elemental composition
562 data have likewise identified Cu, Ba and Sb (together with Fe) as associated with a brake-wear
563 source (e.g. Beddows et al., 2015; Crilley et al., 2017; Visser et al., 2015). (The multiple sources of
564 Fe in airborne PM mean it is not so useful as a source tracer.) In a harmonised study of five major
565 southern European cities, Amato et al. (2016a) identified by PMF a factor showing broadly similar
566 composition within all of the cities, separate to a vehicle exhaust factor, which they attributed to
567 vehicular non-exhaust emissions. The factor showed chemical features typical of brake wear (Cu,

568 Ba and Sb), tyre wear (Zn) and road dust (Si, Al and Ca). Since these elements do not derive
569 exclusively from one source, there are ambiguities and uncertainties in quantification of individual
570 non-exhaust emissions sources by these statistical approaches. It is also not possible unambiguously
571 to distinguish from the composition of ambient PM between direct emissions of brake (and tyre and
572 road) wear particles and the vehicle-induced resuspension or entrainment of brake dust deposited on
573 the road surface or road verges.

574

575 Compared with elemental abundances, there is very limited information concerning the presence of
576 organic compounds in brake wear particles (Grigoratos and Martini, 2015). This reflects a number
577 of factors: organic compounds are in lower abundance than metallic and mineral compounds in
578 brake linings and brake wear; quantification of individual organic species is analytically more
579 challenging than for individual elements; and the existence of elemental markers for brake wear
580 reduces the motivation to undertake speciation of organic compounds. Emissions of organic
581 compounds during braking derive from the high-temperature and high-pressure reactions of
582 graphite and phenolic resin components. Plachá et al. (2017) report BTEX, PAH and other semi-
583 volatile organic compound emissions from a low-metallic brake lining tested using the ISO 26867
584 friction evaluation standard procedure. Rogge et al. (1993) also reported PAHs in brake dust, with
585 their most abundant class of organic compounds being polyalkylene glycol ethers and n-alkanoic
586 acids. However, brake liner formulations will have changed since these earlier measurements were
587 made. In a recent study, Alves et al. (2021) report the presence of aliphatic and aromatic
588 hydrocarbons, alcohols, glycol/glycerol derivatives, plasticisers, sugars, sterols and various phenolic
589 constituents in particles derived from brake wear. Total carbon accounted for 5 – 74 % of PM₁₀
590 mass in tests on a range of pad formulations and braking cycles. A useful organic marker of brake-
591 wear particles is considered unlikely (Grigoratos and Martini, 2015; Thorpe and Harrison, 2008).
592 Non-frictional, regenerative braking systems significantly reduce this source of non-exhaust particle
593 emissions.

594 4.2 Tyre Wear Particles

595 In contrast to brake systems, whose composition is dominated by metals and metal-containing
596 inorganic compounds (such as silicates, sulphides, oxides), tyre composition is dominated by
597 organic materials. However, similar to brake systems, the specific composition of tyres varies
598 widely between vehicle type (e.g. passenger car, heavy-duty vehicle) and manufacturer. The main
599 categories of materials in a tyre by mass are as follows (Baensch-Baltruschat et al., 2020;
600 Grigoratos and Martini, 2014; Sommer et al., 2018).

- 601 • Rubber hydrocarbons (40-60%): natural rubber (polyisoprene $[C_5H_8]_n$) and synthetic rubber,
602 e.g. styrene butadiene rubber (SBR) or butadiene rubber (BR).
- 603 • Filler (20-35%): typically soot/carbon black, silica (SiO_2) and chalk ($CaCO_3$).
- 604 • Softener (15%): hydrocarbon oils and resin.
- 605 • Textile and metal reinforcement (5-10%).
- 606 • Vulcanization agents (2-5%): sulphur and zinc oxide (ZnO).
- 607 • Other additives (5-10%): preservatives (halogenated cyanoalkanes), anti-oxidants (amines,
608 phenols), desiccants (calcium oxides), and plasticizers (aromatic and aliphatic esters).

609
610 Although tyre wear is primarily carbonaceous, a number of inorganic materials are used in tyre
611 manufacture. In particular, ZnO is used as an activator for the vulcanisation process and Zn can
612 account for around 1% of tyre tread material (Smolders and Degryse, 2002; Davis et al., 2001).
613 Tyre wear is therefore a significant source of Zn in the environment (Councell et al., 2004; Adachi
614 and Tainosho, 2004; Blok, 2005). In the UK, the NAEI estimates that 23% of Zn emissions to air in
615 the UK in 2016 derived from tyre wear. This proportion will be considerably higher near roads.
616 Enrichment of Cd in tyre wear has also been observed (Fabretti et al., 2009; Hjortenkrans et al.,
617 2007; Lanzerstorfer, 2018). However, Cd is also a constituent of brake wear (Song and Gao, 2011),
618 although emissions from this latter source may be declining (Hjortenkrans et al., 2007). No Cd was
619 detected in tyre-wear particles by Kreider et al. (2010).

620 The significant enrichment of Zn in tyre-wear particles compared with crustal dust (e.g. Gustafsson
621 et al., 2008) has led to its routine use as a marker for tyre-wear emissions in the same way as Cu, Sb
622 and Ba are used as markers for brake wear in ambient PM source apportionment studies such as
623 those described above. For example, Hicks et al. (2021) applied a scaling factor of 50 to their
624 roadside increment in Zn concentration (taken from Harrison et al. (2012)) to derive the
625 contribution to PM₁₀ from tyre wear. However, as noted above, these markers are not unambiguous
626 for their respective sources and the suitability of Zn, in particular, has been discussed (Thorpe and
627 Harrison, 2008). Zn also has a significant anthropogenic source from wear of Zn-galvanised
628 materials such as fencing, and a source in biomass burning emissions.

629

630 In respect of other inorganic components in tyres, SiO₂ is commonly used as a reinforcing filler
631 (Rattanasom et al., 2007) and a prevalence of Si-rich particles has been observed from tyre-wear
632 driving simulators (Kim and Lee, 2018; Park et al., 2018). These particles are particularly prevalent
633 in the ultrafine fraction (Park et al., 2018), whose production is substantially enhanced when tyre-
634 tread surface temperatures reached 160°C (Park et al., 2017). Sulphur was identified in the larger
635 particles (Park et al., 2018), but S is not useful as an elemental marker of tyre-wear in ambient PM
636 because of the prevalence of sulphate (SO₄²⁻) from the oxidation of SO₂ emissions. Black carbon is
637 also a significant component of tyre-wear emissions – the EMEP/EEA Emissions Inventory
638 Guidebook applies a BC/PM_{2.5} ratio of 0.15 for tyre wear (EMEP/EEA, 2016) – but the BC
639 emission is both highly uncertain and not a unique source marker for tyre wear.

640

641 Information on the organic compounds in tyre wear particles is sparse because it is laborious to
642 extract, separate and quantify individual organic species by GC-MS, LC-MS and similar off-line
643 techniques (Rogge et al., 1993; Nolte et al., 2002). The focus in this review is identification of
644 potential organic markers for a tyre-wear source. One such class of compounds is benzothiazole
645 (BT) and its derivatives (Rogge et al., 1993; Lin et al., 2010; Zhang et al., 2018), which are used as

646 vulcanization accelerators and UV-light stabilisers for rubbers (Liao et al., 2018). Zhang et al.
647 (2018) quantified benzothiazole and six of its derivatives in tyre-wear particles using solid-liquid
648 extraction and ultra-high performance liquid chromatography-tandem mass spectrometry (UPLC-
649 MS/MS). All seven BTs were found in 17 tyre-wear particle samples from different tyre brands.
650 The mass fractions of all seven BTs summed ranged from 46.93 to 215 $\mu\text{g g}^{-1}$ with an average
651 concentration of 99.32 $\mu\text{g g}^{-1}$. Benzothiazole and 2-hydroxybenzothiazole (2-OH-BT) were the two
652 major compounds, accounting for 56-89% of the total.

653

654 A second class of potential organic markers for tyre wear emissions are hydrogenated resin acids.
655 Resin acids are natural products found in pine tars, which are additives used by manufacturers to
656 give tyres certain desirable properties (Nolte et al., 2002). Some of these resin acids are chemically
657 hydrogenated before being added during the manufacture process, which differentiates them from
658 the natural resin acids that may be present in ambient PM from sources such as wood smoke.
659 Kumata et al. (2011) proposed the following four dihydroresin acids as molecular markers for tyre-
660 wear emissions: 8-isopimaren-18-oic acid (I), 8-pimaren-18-oic acid (II), 13 β (H)-abieten-18-oic
661 acid (III), and 13 α (H)-abiet-8-en-18-oic acid (IV). The absence of I–IV in natural sources and the
662 linear correlations between dihydroresin acids with different skeletons in tyres and in environmental
663 samples demonstrated that I–IV are specific markers for synthetic rubbers. These workers further
664 suggest from the results of photolysis experiments that I–IV can set lower limits for tyre-wear
665 contributions to environmental loads of PM and polycyclic aromatic hydrocarbons (PAHs) with
666 molecular weight ≥ 202 . By comparing III + IV concentrations or (III+IV)/pyrene or
667 (III+IV)/benzo[a]pyrene ratios in tyres and those in environmental matrices, Kumata et al. (2011)
668 estimated the contributions of tyre-wear emissions to airborne PM, pyrene, and benzo[a]pyrene as,
669 respectively, $0.68 \pm 0.54\%$, $6.9 \pm 4.8\%$, and $0.37 \pm 0.18\%$ in roadside PM and $0.83 \pm 0.21\%$, $0.88 \pm$
670 0.52% , and $0.08 \pm 0.06\%$ in rooftop PM.

671

672 Other studies have also highlighted PAHs in tyre-wear emissions. In an experimental simulation of
673 tyre-road interaction, Aatmeeyata and Sharma (2010) observed a linear increase in cumulative
674 emissions of PAH (in particles of diameter $> 10 \mu\text{m}$) over a 14,500 km run of the tyre. The most
675 abundant PAH was pyrene ($30 \pm 4 \text{ mg kg}^{-1}$), followed by benzo[ghi]perylene ($17 \pm 2 \text{ mg kg}^{-1}$).
676 Sadiktsis et al. (2012) analysed for 15 high molecular weight PAH in 8 different tyres, albeit not in
677 tyre-wear emissions. Whilst there was a factor 20 difference in PAH concentration between tyres,
678 the relative abundance of PAHs was quite similar. Almost all (92%) of the total extractable PAH
679 content was attributed to five PAHs: benzo[ghi]perylene, coronene, indeno[1,2,3-cd]pyrene,
680 benzo[e]pyrene, and benzo[a]pyrene. The four dibenzopyrene isomers analysed
681 (dibenzo[a,l]pyrene, dibenzo[a,e]pyrene, dibenzo[a,i]pyrene, dibenzo[a,h]pyrene) constituted $<2\%$
682 of the sum of the 15 PAH determined but represent a potential hazard as they are carcinogenic. In
683 the EU, the concentrations of PAH in tyre-wear emissions should have declined markedly from
684 2010 due to implementation of EU Directive 2005/69/EC that prohibits the use of 'PAH-rich'
685 extender oils in tyres produced after January 2010 (AQEG, 2019). It is also important to note that
686 PAH are emitted to the air from multiple sources other than via tyre-wear particles, in particular for
687 the road environment, in diesel exhaust emissions.

688

689 ATOFMS presents the opportunity to seek m/z distributions unique to a particular source in the
690 mass spectra of individual particles (Dall'Osto et al., 2014). ATOFMS mass spectra of tyre-wear
691 particles derived in a laboratory simulator showed peaks due to sulphur compounds, nitrate, Zn and
692 m/z values >100 attributed to organic polymers for particles in the size range 200–3000 nm.

693 Examination of large datasets of ATOFMS spectra for road dust and PM in ambient air revealed
694 that the majority (95%) of tyre-wear particles present in the road dust samples and atmospheric
695 samples were internally mixed with metals (Li, Na, Ca, Fe, Ti), as well as phosphate.

696

697 The extent to which tyre-wear (and road-wear) particles contribute to the microscopic plastic
698 particles (microplastics) entering the environment depends on the definition of plastic. There is
699 some disagreement on which polymers are “plastics”. The ISO 472:2013 definition of plastic is
700 “material which contains as an essential ingredient a high molecular weight polymer and which, at
701 some stage in its processing into finished products, can be shaped by flow” (Hartmann et al., 2019).
702 Rubbers (and some other elastomers) are excluded from this definition of plastic. However, this
703 definition reflects the historic industrial landscape rather than perspectives about the behaviour of
704 rubber fragments in the environment. Other authors include rubber within their definition of plastics
705 and conclude that tyre-wear is a substantial, and under-reported, source of microplastics into the
706 environment (e.g. Kole et al., 2017; Knight et al., 2020; Sommer et al., 2018). The annual global
707 rubber production of ~27 million tonnes in 2015 was ~9% the total plastic production (Kole et al.,
708 2017). However, estimating the fluxes of these plastic and rubber materials in the form of
709 microparticles into the environment is a challenge. A proportion of tyre wear will not be released to
710 air but remain on the road surface or enter the environment in run-off. A further consideration in
711 terming tyre wear as plastic arises from the way in which tyre wear particles may be substantially
712 (50%) internally mixed with road-wear fragments (Kreider et al., 2010; Panko et al., 2013; Panko et
713 al., 2019).

714

715 **4.3 Road Surface Wear Particles**

716 It is not possible from field measurements to distinguish unambiguously between direct emissions
717 of road surface wear particles and resuspension of such particles already on the road surface since
718 the chemical composition is often very similar (Bukowiecki et al., 2010). Using mobile load
719 simulators to compare the two contributions, Gehrig et al. (2010) concluded that road abrasion
720 fluxes are normally lower than re-suspension fluxes, and that the latter increase with the dust
721 loading on the road, as expected. However, road-surface wear emissions are substantially enhanced

722 in locations with sub-zero temperature winters where studded tyres are routinely used for additional
723 traction (Denby et al., 2018; Gustafsson, 2018).

724

725 Road surfaces are broadly composed of concrete or asphalt/tarmac. The former is a mixture of
726 mineral aggregate, sand, and cement. The latter comprises ~95% mineral aggregates and ~5%
727 bituminous binder. Since the dominant component in both concrete and asphalt road surfaces is the
728 aggregate, road-wear particles mainly consists of small mineral fragments whose composition
729 varies with the vast array of geological sources of the aggregate. The dominant crustal elements in
730 these minerals are Si, Ca, K, Fe, and Al. Whilst a grouping of these elements is useful to quantify
731 road dust contribution to airborne PM (see next section), it is nearly impossible to separate primary
732 road wear from other mineral dust deposited on the road (Denier van der Gon et al., 2013).

733

734 The bituminous binder in asphalt/tarmac road surfaces comprise many thousands of high molecular
735 weight aliphatic and aromatic hydrocarbons, which may also contain heteroatom functional groups
736 with O, N and S. Smaller amounts of polymers, epoxy resins and metal complexes are also present
737 to enhance binder-aggregate bonding (NIOSH, 2000; Thorpe and Harrison, 2008). There is a
738 paucity of detailed chemical information on road surface composition, but it is unlikely that there is
739 a definitive molecular (or elemental) marker for road surface wear (Boulter, 2005; Thorpe and
740 Harrison, 2008). Many studies have reported elevated concentrations (0.08–149 $\mu\text{g/g}$) of PAHs in
741 urban road dusts (Hwang et al., 2019), and the much greater prevalence of PAHs in road dust from
742 coal-tar-based compared with asphalt-based road surfaces indicates their predominant source is
743 road-surface wear (Metre et al., 2009) (coal-based bitumen containing substantially greater PAH
744 content than oil-based bitumen). However, coal-based tars are now infrequently used.

745

746

747

748 **4.4 Resuspension Particles**

749 The traffic-related resuspension source refers to the uplift into the air by vehicle movement of
750 particles already present on the road surface, mediated for example via tyre shear stress or vehicle
751 wake turbulence. Resuspended particles therefore comprise particles from all three of the sources
752 already described – brake wear, tyre wear, road surface wear – as well as particles from other
753 sources that have also deposited onto the road surface, for example exhaust emission particles,
754 particles from de-icing and gritting, wind-blown dust and biogenic particles. Consequently, the
755 composition of resuspension particles comprise all the chemical components already described, and
756 many more. Chemical analysis of road dust has received particular attention because of its
757 contribution to potential toxicity to near-roadside ecosystems and to aquatic systems via run-off as
758 well as to inhaled airborne PM. The very wide range of elemental and organic components in road
759 dust is highlighted in a number of recent reviews (Haynes et al., 2020; Hwang et al., 2016; Hwang
760 et al., 2019; Khan and Strand, 2018; Lanzerstorfer, 2020; Maeaba et al., 2020). Individual studies
761 identify mutual correlation and/or enrichment in road dust particles of the ‘crustal’ elements
762 associated with road surface mineral aggregates and wind-blown dust, for example Si, Al, Ca, Fe,
763 Ti and Mg (Alves et al., 2020; Amato et al., 2009a; Amato et al., 2011; Amato et al., 2012a;
764 Dall'Osto et al., 2013; Fabretti et al., 2009; Ondráček et al., 2011; Padoan et al., 2017).

765

766 Road dust is also highly enriched and contaminated by elements from tyre and brake wear that have
767 deposited onto the surface, e.g. Sb, Cu, Zn, Sn, Cr (Adamiec et al., 2016; Alves et al., 2020; Cowan
768 et al., 2021; De Silva et al., 2021; Lanzerstorfer, 2018; Padoan et al., 2017; Pipalatkar et al., 2014).

769 A class of elements in road dust particles that has received particular attention due to their toxicity
770 (and potential for recovery) are the platinum group elements Pt, Pd, Rh derived from wear of
771 exhaust after-treatment catalysts (De Silva et al., 2021; Ladonin, 2018; Wang and Li, 2012; Zereini
772 et al., 2012). Whilst the ultimate source of these elements is exhaust emissions, when particles

773 containing these elements deposit on the road surface and are resuspended they contribute to the
774 chemical composition of traffic-derived non-exhaust particles.

775

776 **5. MEASUREMENTS OF NEE IN THE ATMOSPHERE**

777 **5.1 Principles of Measurements**

778 NEE particles are, by their nature, refractory and insoluble in water. This means that many of the
779 common tools for analysing particulate composition (both offline and in situ) such as ion
780 chromatography, thermal desorption gas chromatography and thermal aerosol mass spectrometry
781 such as the aerosol chemical speciation monitor (ACMS), are not well suited for analysis. Instead,
782 offline analysis methods usually focus on the analysis of the metallic fraction. A common method is
783 the digestion of a sample in acid (usually nitric acid) followed by analysis using Inductively
784 Coupled Plasma (ICP) analysis with Optical Emission Spectroscopy (OES, also known as Atomic
785 Emission Spectroscopy or AES) or Mass Spectrometry (MS) (Chow and Watson, 2013; Chow,
786 1995). It is also common to analyse metals in place on a substrate using x-ray spectroscopic
787 methods such as X-Ray Fluorescence (XRF). To increase sensitivity, this may use a synchrotron
788 source (Bukowiecki et al., 2005). The particles can also be analysed using electron microscopy and
789 classified according to morphology and composition (Weinbruch et al., 2014).

790

791 The selective analysis of the carbonaceous component of tyre wear that makes up the majority of its
792 composition is analytically challenging because it is (by design) not volatile or soluble, so cannot
793 generally be analysed using thermal desorption or liquid chromatography (Noziere et al., 2015).
794 However, it is possible to detect certain organic tracers and toxins such as PAHs and plasticisers
795 (Avagyan et al., 2014; Alves et al., 2018). Another method to study the organic fraction of tyre
796 material is pyrolysis, followed by Gas Chromatography (py-GC), a technique that has been
797 successfully used to detect the presence of tyre wear particles in groundwater and soils (Unice et al.,
798 2012). It is also possible to use FTIR to study the polymers present (Mengistu et al., 2019).

799 Another feature of NEE particles that is distinct from many other atmospheric aerosols is their size,
800 owing to the fact they are produced mainly from abrasion rather than combustion or chemical
801 processes. This means that in terms of mass, they tend to occupy predominantly the coarse PM_{2.5-10}
802 fraction rather than the PM_{2.5} fraction. This is evident when samples are obtained using size-
803 segregated methods such as cascade impactors (e.g. Kwak et al., 2013). NEE particles can also be
804 associated with modes identified in the data from aerosol size spectrometers that are optimised for
805 micron sized particles, such as the Aerodynamic Particle Sizer (APS) and optical particle counters
806 (Harrison et al., 2012). By way of example, Harrison et al. (2011) reported a volume concentration
807 size mode at 2 µm for brake dust and at >10 µm for resuspended road dust. By contrast, the mode
808 corresponding to exhaust emissions existed at 0.2 µm. Also, Kreider et al. (2010) reported volume
809 modes of around 70 µm for roadway particles and 100 µm for tyre wear particles.

810

811 It is known that NEE can be a source of ultrafine particles, i.e. those smaller than 100 nm (Kwak et
812 al., 2014; Nosko et al., 2017). These contribute little to the PM mass concentrations, but can
813 dominate number concentrations. While it is possible to measure these in the ambient atmosphere
814 using mobility size spectrometers and condensation particle counters (Wiedensohler et al., 2012),
815 exhaust emissions are known to be a major source of these, so disentangling the NEE contribution
816 is challenging.

817

818 Semi-continuous analysis of these particles is possible using in situ XRF analysis, as used in the
819 XACT monitor (Tremper et al., 2018; Furger et al., 2017). This has the advantage over offline
820 analysis in that it produces data in near real time and can produce data on an hourly basis. Real-time
821 analysis is also offered by single particle mass spectrometers such as the ATOFMS and related
822 instruments (Murphy, 2007; Pratt and Prather, 2012). These use pulsed lasers to vaporise and
823 ionise the components of individual particles and analyse the resultant ions using mass
824 spectrometry. This can identify particle types through numerical clustering of the mass

825 spectrometric data (Rebotier and Prather, 2007). This can assess the relative contributions of NEE
826 particles to ambient number concentrations (Beddows et al., 2016; Dall'Osto et al., 2014). Because
827 these instruments are usually operated using optical detection to trigger the desorption later, the data
828 is limited to particles of around 200 nm or greater.

829

830 **5.2 Receptor Modelling Techniques**

831 The influence of individual sources of particulate matter at a receptor site can be qualitatively
832 inferred from the inspection of tracers (Gietl et al., 2010). This works particularly well if a tracer is
833 unique to a particular source, so in the case of NEE, tracers such as Sb and Cu can be taken as
834 indicative of brake wear, but depending on the environment, it is harder associating tracers such as
835 zinc with tyre wear when it is also associated with wood burning. These can be compared with data
836 from other sources to estimate contributions to the overall particulate budget (Harrison et al., 2012).
837 The ISO/TS 20593:2017 standard covers the estimation of tyre and road wear contributions based
838 on analysis of samples using pyrolysis.

839

840 A common method of aerosol receptor modelling for the purposes of quantitative source
841 apportionment as applied to other aerosol types is the Chemical Mass Balance (CMB) technique,
842 which involves estimating a source's contribution to PM through inspection of tracers and
843 application of knowledge of the emissions from a source (Chow and Watson, 2002). However, this
844 does not work particularly well for NEE because the source profiles are known to vary greatly
845 according to formulation in the case of brake wear and the local composition in the case of road
846 dust.

847

848 As an alternative, non-negative factorisation techniques are often applied, in particular PMF
849 (Paatero, 1997; Paatero and Tapper, 1994). This method is able to deconstruct a multivariate dataset
850 into a specified number of 'factors' using error-weighted residual minimisation without any a priori

851 assumptions regarding the nature of the factors. This method works very well in source
852 apportionment when a given contribution's profile is invariant (has a fixed source profile that is not
853 modified in the atmosphere) and can be applied to a composite of observations, such as the time
854 series of multiple metals derived using ICP-MS analysis. The technique is further improved when
855 data is obtained as a function of time of day, thereby capturing variations in traffic density, so this
856 technique is especially powerful when applied to high frequency sampling and semicontinuous
857 analysis. PMF can also be applied to size distribution data, which can also be used to resolve
858 different sources (Harrison et al., 2011; Vu et al., 2015).

859

860 A limitation of ~~XRF-PMF~~ is that it struggles to separate sources that are covariant at a given
861 receptor site, which can frequently be the case with NEE, so sometimes these are lumped together
862 as a single 'traffic related' factor. Moreover, there is a conceptual problem with this data model
863 when it comes to separating road dust from brake and tyre wear based on chemical measurements,
864 because much of the road dust is contaminated with the chemical components of the other sources
865 (Alves et al., 2018). Likewise, tyre wear particles are also known to contain small road dust
866 particles (Kreider et al., 2010). The only receptor measurements theoretically capable of
867 distinguishing these are the single particle methods such as electron microscopy and single particle
868 mass spectrometry, however these are still not able to explicitly tell whether a given particle was
869 emitted directly to the atmosphere, or initially settled to be subsequently resuspended.

870

871 A further variation of PMF is the application of the Multilinear Engine (ME2) algorithm, which
872 through adding additional penalty functions to the weighted residuals, enables the user to invoke a
873 degree of a priori knowledge, by assuming 'target' profiles or 'pulling' time series towards a
874 desired solution (Amato and Hopke, 2012; Paatero, 1999; Amato et al., 2009b). Another expansion
875 of the PMF technique includes bringing in data from multiple sources; if (say) metal data alone
876 were to be used, the derived factors would only concern the contributions to the available metal data

877 in isolation, but by including other chemical analyses, size spectrometer and/or overall particulate
 878 mass data, the contributions to the PM mass budget can be estimated.

879

880 **5.3 Measured Contributions and Size Distributions**

881 A common method of capturing NEE data is by assessing its contribution to the roadside increment,
 882 i.e. comparing roadside concentrations with equivalent background measurements for a given
 883 conurbation. This can be done for mass concentrations of particulate components and size resolved
 884 data. NEE has been seen as a responsible for a general roadside or kerbside increment in PM_{2.5-10}
 885 and this can be linked to metals associated with NEE (Hopke et al., 2018; Lenschow et al., 2001;
 886 Querol et al., 2008; Lough et al., 2005; Gietl et al., 2010). Road dust can also be sampled directly
 887 using suction and aerodynamic separation, to obtain a sample representative of what could
 888 contribute to ambient PM₁₀ if resuspended by traffic (Amato et al., 2016b; Alves et al., 2020). This
 889 has the advantage of providing a larger volume of material for analysis.

890

891 As shown in Table 2, various metals can be associated with different NEE sources based on
 892 ambient measurements. The associations with specific sources can be based on knowledge of
 893 emissions, analysing the roadside increment and tunnel studies. Some elements can be associated
 894 with multiple sources such as iron present in both brake wear and mineral dust. A common method
 895 of inspecting the brake wear contribution is through evaluation of the Cu:Sb ratio, and values
 896 between approximately 1 and 10 have been reported (Pant and Harrison, 2013, and references
 897 therein).

898

899 **Table 2:** Metal profiles of various NEE and mineral dusts based on selected ambient, tunnel and
 900 road dust studies. *Metals detected at a subset of sites in parenthesis.

901

Reference	Brake wear	Tyre wear	Combined vehicular	Mineral dust
Tanner et al. (2008)	Cu, Cd	Zn		
Fabretti et al. (2009)			Cu, Zn, Sb, Sn	Rb, Sr, Mn, Fe, As
Gietl et al. (2010)			Ba, Fe, Cu, Sb	
Pérez et al. (2010)			Sb, Cu, Ni, Sn	Fe

Amato et al. (2011)	Fe, Cu, Zn, Cr, Sn, Sb	Zn		Al, Ca, Fe, V
Harrison et al. (2012)	Ba, Cu, Fe, Sb	Zn		Si, Al
Visser et al. (2015)	Cu, Zr, Sb, Ba		Fe	Si, Ca
Amato et al. (2016b)	Cu, Sn, Sb, Cr, Ba, Fe, Al, Ca	Ca		Al, Ca, K, Ti, Fe, Mg
Crilley et al. (2017)			Ti, Cr, Mn, Fe, Ni, Cu, Rb, Zr, Mo	Al, Si, Ca, Ti, Mn, Ni, Zn
Alves et al. (2018)			Pb, An, Fe, Cu, Sn, Sb	Al
Jeong et al. (2019)	Cu, Ba			Ca
Alves et al. (2020)			Cu, Zn, Sn, Sb, As, Se, Bi, Cd, Pb	Si, Al, Fe, Ca, K
Daellenbach et al. (2020)			Cu, Fe, Mo, Sb	Al, Se, Ti
Rai et al. (2020)			Fe, Cr, Cu, Mn, Zn, Ba	Ca, Si
Zhang et al. (2020)	Fe, Ba, Zr, Cu, Ti	Si, Zn		Si, Ca
Rai et al. (2021)			Ba, Ni, Mn (Rb, Cr, Fe, Zr, Sr)	Ca, Si, Ti (Sr, Fe, Zr)

902

903 Mineral dust can be associated with Si, Al and Ca, but it is can be harder to unambiguously identify
904 because its composition varies with local mineralogy, nearby sources (e.g. construction) and
905 contamination from exhaust and non-exhaust emissions (Adamiec et al., 2016). In some works, the
906 contribution of traffic to sampled material is expressed as an ‘enrichment’ relative to the local
907 continental crust (using an element such as Al as a reference), thereby capturing all the non-natural
908 components (Alves et al., 2018). Note that this does by no means imply that airborne mineral dust is
909 inherently ‘natural’ as the vehicles are still responsible for its resuspension. In addition, a
910 component of road salt is sometimes reported in the form of Na, Mg and Cl (not shown in table).

911

912 Other components of the brake and tyre particles include elemental and organic carbon as measured
913 using thermal-optical analysis (Amato et al., 2016b), however in isolation, these are difficult to
914 distinguish from exhaust emissions, which are also predominantly carbonaceous in nature. The
915 polymer markers associated with tyre wear, as analysed using py-GC, are relatively unambiguous
916 and can be used to assess its contribution (Panko et al., 2013).

917

918 The application of PMF to urban data has allowed the emissions to be profiled beyond individual
919 elements and complete profiles for the different components of NEE as observed at a receptor site,
920 although as explained above, it is often difficult to separate the brake and tyre wear components.
921 Examples include Fabretti et al. (2009), who were able to divide $PM_{2.5}$ into three categories,
922 specifically resuspension, a combined ‘vehicle abrasion’ factor and exhaust emissions. Another
923 example is the work of Rai et al. (2020), who were able to separate ‘road dust’ and ‘traffic related’
924 factors from background sources and fireworks in XACT data. Further insight can also be gained
925 through the comparison of data collected at multiple sites simultaneously. Through PMF analysis of
926 size-resolved metal data from roadside, urban background and rural measurements in London,
927 Visser et al. (2015) were able to profile and assess the urban and roadside increments due to
928 resuspended dust, brake wear, road salt and other traffic related aerosols. This again found that NEE
929 could be most associated with PM_{10} , but an increment in $PM_{2.5}$ was also detected.

930

931 Some studies have presented a combined factorisation analysis from multiple data sources. For
932 instance, Crilley et al. (2017) were able to use metal data combined with size distributions.
933 Contributions of vehicle wear and road dust to $PM_{2.5-10}$ particles were found to be 13 and 31%
934 respectively at a roadside site, although the contributions to the fine fraction and a background site
935 were not disentangled from general traffic emissions. In a similar vein, Jeong et al. (2019) presented
936 a combined factorisation of $PM_{2.5}$ data obtained in Toronto, using a variety of online techniques
937 including an XACT, ACSM and Aethalometer, and offline analysis. This showed that NEE
938 represented around a third of traffic-related $PM_{2.5}$.

939

940 While many factorisation papers (e.g. Rai et al., 2020) report NEE as a percentage of the total
941 elements measured using a given analysis, these do not strictly represent contributions to the total
942 PM budget because of other components that are not measured during XRF or ICP analysis (e.g.
943 carbon, oxygen and hydrogen). Instead, tracers or a combined factorisation must be used.

944 The papers that do estimate absolute contributions (summarised in Table 3) have presented a
 945 significant contribution of NEE to coarse aerosol in urban environments. Based on tracer-derived
 946 data originally presented by Harrison et al. (2012), Beddows and Harrison (2021) reported the
 947 kerbside PM₁₀ increment at the London Marylebone Road site due to brake wear, tyre wear and
 948 resuspension to be 2.8, 0.5 and 1.5 µg m⁻³ respectively. During a later study at the same site, Crilley
 949 et al. (2017) used combined factorisation to estimate the total contribution of vehicle wear and
 950 resuspension to PM_{2.5-10} to be 1.3 and 3.1 µg m⁻³ respectively. Through the analysis and
 951 classification of samples analysed using electron microscopy, Weinbruch et al. (2014) estimated a
 952 PM₁₀ kerbside increment in Ruhr, Germany due to abrasion and resuspension of 0.8 and 4.2 µg m⁻³
 953 respectively.

954

955 In contrast, estimates of the contribution to PM_{2.5} tend to be more minor. In Toronto, Jeong et al.
 956 (2019) used combined factorisation to estimate the average brake wear and resuspension
 957 contributions to be 0.2 µg m⁻³ each at a downtown site and 0.6 and 0.3 µg m⁻³ respectively at a
 958 highway site. While concentrations were low, it was stressed that these still represented the majority
 959 of some metals such as Ba, Cu and Mn. Using a tracer method applied to samples collected in
 960 London, Tokyo and Los Angeles, Panko et al. (2019) estimated that tyre wear particles represented
 961 only 0.1-0.68% of PM_{2.5} mass. Absolute concentrations varied, but the highest concentration of
 962 0.29 µg m⁻³ was noted at the Blackwall Tunnel approach in London, where significant braking
 963 occurs.

964 Table 3: Summary of quantitative mass apportionment studies of NEE in polluted urban
 965 environments. Notes: (a) Kerbside increment rather than overall concentration reported; (b)
 966 Downtown site; (c) Highway site.
 967

Reference	Brake Wear	Tyre Wear	Combined wear	Resuspension
PM₁₀ (µg m⁻³)				
Beddows and Harrison (2021)	2.8 (a)	0.5 (a)		1.5 (a)
Crilley et al. (2017)			1.3	3.1
Weinbruch et al. (2014)			0.8 (a)	4.2 (a)
PM_{2.5} (µg m⁻³)				
Jeong et al. (2019)	0.2 (b)			0.2 (b)

	0.6 (c)			0.3 (c)
Panko et al. (2019)		≤ 0.29		

968

969 **6. MODELLED CONCENTRATIONS OF NEE**

970 **6.1 Modelling Approaches and Data**

971 The usual purpose of modelling particulate concentrations is to predict annual-mean or daily-mean
972 total mass concentrations in locations where measurements have not been made, or under
973 alternative scenarios. Modelling thus refines estimates of exposure and allows interventions to be
974 tested. Local-scale models provide high output resolution (metres) close to roads (e.g. Hood et al.,
975 2014). Regional models provide coarser output resolution but cover much larger areas (e.g.
976 Simpson et al., 2003). Some models have multi-scale capabilities (e.g. USEPA, 2020b) and
977 different types of models may also be nested (e.g. Vieno et al., 2016). Regardless of the spatial
978 scale, emissions of NEE are usually calculated using the same information which underpins the
979 national inventories, as described in Section 2. These emissions are then aggregated with those
980 from vehicle exhausts for input to the model, meaning that their spatial distribution across road
981 links is closely aligned with that of exhaust emissions. This is not always appropriate, for example
982 the largest braking emissions may occur on road segments where accelerations and hence exhaust
983 emissions are relatively low. Most models of NEE concentrations are mechanistically separated
984 from the emissions calculations. For example, the treatment of local weather conditions for
985 dispersion is independent of any consideration of the impact of rainfall when calculating emissions.
986 Similarly, while traffic-induced turbulence can be varied in some models, this is not used to
987 calculate emissions. Where particle size distributions are defined (e.g. Archer-Nicholls et al., 2014)
988 they do not usually take explicit account of relative source-strengths of the different emission types:
989 exhaust, brake, tyre and road wear or resuspension. The relative contribution to concentrations
990 made by different traffic sources thus mirrors that defined by the emissions inventories.

991

992 Concentration modelling presents an opportunity to verify against ambient measurements and thus
993 test the effective performance of NEE emissions factors. Such comparisons are necessarily
994 confounded by uncertainties in the model to which the calculated emissions are input, and made
995 more difficult because of the relatively large contribution of non-traffic sources to roadside
996 concentrations when compared with NO_x. A common approach is to compare modelled and
997 measured concentrations while varying the basis of emissions calculation (e.g. de la Paz et al.,
998 2015).

999

1000 Denby et al. (2013) used concentration modelling to test the performance of NORTRIP emissions
1001 calculations, which take specific account of winter salting and sanding, as well as road wear from
1002 studded tyres used in northern Europe. The model successfully predicted the effect on
1003 concentrations of a reduction in studded tyre use in Stockholm, but performed less well in
1004 Copenhagen, where studded tyres are not used at all. Inclusion of road salt within the emissions
1005 calculations led to a 6-16% increase in mean total concentrations and improved overall model
1006 performance.

1007

1008 Pay et al. (2011) used the CALIOPE model for Spain to compare EEA emissions factors with a
1009 bespoke routine for resuspension, which they derived from ambient measurements made elsewhere
1010 in Europe, adjusted for local rainfall. The bespoke resuspension calculations reduced, but did not
1011 remove, negative bias in the modelled concentrations. Resuspension was predicted to contribute up
1012 to 7 µg/m³ to annual mean PM₁₀ in urban areas. Amato et al. (2016c) predicted concentrations in
1013 Barcelona using a Gaussian line-source model, exhaust and wear emissions from TREMOVE 2.7,
1014 and an experimentally-derived fleet-average resuspension emission rate for Barcelona of 82 mg
1015 vkm⁻¹ which was reduced for periods during and after rainfall using a response curve determined
1016 from local measurements (Amato et al., 2012b). Applying this curvilinear relationship to rainfall
1017 improved model performance compared with turning resuspension on/off or assuming a constant

1018 emission. The bespoke resuspension emission routine removed most negative bias in predicted
1019 concentrations, but the spatially-uniform resuspension emission factor could not account for large
1020 inter-site variations. At traffic sites, resuspension contributed 23-44% of PM₁₀, with other traffic-
1021 related emissions contributing 8-14%. The importance of resuspension was highest in the most
1022 densely-populated parts of the city. Most modelling studies do not include rain frequency as part of
1023 the emissions calculations.

1024

1025 Hood et al. (2018) predicted annual mean PM₁₀ and PM_{2.5} concentrations across London using
1026 ADMS-Urban coupled with EMEP4UK. EEA emissions factors were adjusted based on
1027 measurements made in London (Harrison et al., 2012). AQEG (2019) used these results to show
1028 that NEE contributed more than exhaust emissions to annual mean PM₁₀ and PM_{2.5} and that NEE
1029 thus strongly impacted the predicted spatial variation in annual mean PM_{2.5} across central London.

1030

1031 Daellenbach et al. (2020) used a regional chemical transport model to calculate exposure across
1032 Europe to both PM mass and to its oxidative potential (a measure of the ability to deplete anti-
1033 oxidant defences in the body). NEE emissions were not explicitly represented, but were included as
1034 a function of NO_x emissions, with size distributions subsequently parameterised from ambient
1035 measurements. The model results compared well with measurements and were used to inform a
1036 wider discussion of the oxidative potential of PM.

1037

1038 Machine learning methods may also be used to predict total PM mass concentrations at individual
1039 monitoring sites under alternative traffic management scenarios (e.g. Suleiman et. al, 2019). These
1040 approaches have similarities to source receptor modelling and, while not widely used in this
1041 context, have the potential to determine the site-specific dependence of total concentrations on
1042 individual causal factors, such as rainfall or traffic volume (Carslaw and Taylor, 2009); potentially

1043 allowing alternative scenarios to be tested without resolving the NEE contribution and without
1044 reliance on mechanistic emissions factors.

1045

1046 **7. MITIGATION MEASURES**

1047 **7.1 Engineering Control and Material Reformulation**

1048 There has been consistent effort by automotive manufacturers and aftermarket parts manufacturers
1049 to develop brake systems which have good wear characteristics and good characteristics with
1050 regards fouling of nearby surfaces. The drivers have historically related to cleanliness during
1051 operation (e.g., Gaylard et al., 2010) and controlling operating costs, however, the compromise with
1052 braking performance (including noise, friction characteristics and thermal behaviour) has meant that
1053 brake particulates have not had the focus they are now receiving.

1054

1055 Attention was drawn around 2006 to the contribution of brake pads on the aquatic ecosystem in the
1056 San Francisco Bay area, US as a result of emissions of copper containing particles (e.g., Rosselot,
1057 2006). This attention brought into focus the need for reduction of copper emissions from brake
1058 systems and led to a marked increase in commercial interest in reducing particulate emissions from
1059 brakes evident in the proliferation of related patents at this time. A memorandum of understanding
1060 between USEPA and the automotive industry was signed in 2015 with the aim of phasing out the
1061 use of copper in brake pads (to less than 0.5% by weight) by 2025 (USEPA, 2015), which may
1062 itself resolve the copper issue, but contributed to the awareness of brake systems as sources of
1063 vehicular particulate emissions. The studies investigating technological approaches to emission
1064 reduction or control therefore remain of interest.

1065

1066 There are two technological approaches evident in the literature for reducing airborne particulate
1067 emissions from brakes, namely changing the formulation of the brake friction materials and adding

1068 technology to trap the particles before they are released into the surroundings. It can be inferred also
1069 that vehicle design decisions beyond the friction brake system will influence particle emissions.

1070

1071 **7.2 Friction Material Formulation**

1072 New materials have been evaluated thoroughly in respect of friction characteristics, noise and wear
1073 rates, but only recently has significant attention focused on particle emissions. Low copper
1074 formulations developed in light of the MoU have often focused on wear rather than emissions
1075 limiting the amount of data on the effects of replacing copper fibres in brake pads with alternative
1076 metallic materials (commonly steels), and non-metallic materials (such as natural fibres and
1077 ceramics). Despite the expectation of mass balance, wear rates typically show a weak correlation
1078 with particle number emissions at lower brake material temperatures (Tarasiuk et al., 2020) and are
1079 therefore not a reliable indicator.

1080

1081 There are a myriad of options available to manufacturers when developing new friction materials
1082 such as the materials used to create the primary plateaus and the binders and other materials which
1083 contribute to the secondary plateau formation. Despite only a fraction of this design space having
1084 been explored in the literature, it is clear that brake material formulation has a significant effect on
1085 particle emissions both in laboratory environments (e.g. Lyu et al., 2020) and on road (Farwick zum
1086 Hagen et al., 2019a).

1087

1088 Ceramic fibre and steel fibre based friction materials have been shown to produce higher particle
1089 number emissions than copper based materials at lower operating temperatures (<100°C) (Wei et
1090 al., 2020) raising concerns about the risk of particle emissions increasing as a result of reducing
1091 copper content. It is well recognised that a transition temperature exists at which the resins in the
1092 formulation start to decompose creating a large increase in fine particle emissions (Perricone et al.,
1093 2018). This transition temperature is affected by resin choice, with higher resin molecular weight

1094 (Joo et al., 2020) and higher heat resistance of binders (Joo et al., 2019) correlating well with low
1095 particle emission rates, particularly under higher temperature testing.

1096

1097 The location at which the emissions are formed is that of the friction contact. The dissipation of
1098 energy at this contact results in locally high temperatures, particularly on the pads (which explains
1099 the impact of fibre choice on particle emissions).

1100

1101 The temperature achieved, which relates to the particulate emissions is influenced by the thermal
1102 behaviour of both the brake pad and disc. Thermal conductivity of the brake disc has consequently
1103 been shown to influence the particle emissions, with particle emissions increasing as disc thermal
1104 conductivity reduces, even when the lower thermal conductivity is only at the surface through a thin
1105 oxynitride layer (Seo et al., 2021). Hardening of the disc has been demonstrated to reduce particle
1106 emissions as a result of the harder material behaviour as part of the friction surface (Perricone et al.,
1107 2018).

1108

1109 **7.3 Vehicle Design Decisions**

1110 The importance of brake material temperatures highlights the influence of vehicle design decisions
1111 on brake particle emissions. Frictional power has been shown to be a significant metric determining
1112 the particle emission rates (Alemani et al., 2018), disproportionately increasing emissions when
1113 considered on a per unit energy dissipated basis likely as a combined effect of contact temperatures
1114 and contact area. This suggests that vehicle mass and regenerative braking systems, both of which
1115 reduce the frictional power demand for a given deceleration event, will significantly influence
1116 particle emission rates. Although regenerative braking will reduce frictional power demand, there is
1117 a trend of increasing vehicle weight (USEPA, 2020c) which will somewhat counter this. Vehicle
1118 aerodynamic design, specifically with respect to brake cooling and airflows will also affect brake
1119 contact temperatures and therefore emissions.

1120 Studies which have compared enclosed and open brake systems (i.e. drum brakes versus disc
1121 brakes) have shown that the emitted airborne particulates can be ~10 times lower when the brake
1122 system is enclosed (Hagino et al., 2016). Drum brakes seem to have fallen out of favour in many
1123 markets, however, the drop in particle emissions is somewhat attributed to the enclosing of the
1124 friction surfaces which may give a technological opportunity to reduce particle emissions not
1125 dissimilar in some ways to a passive particle filter system.

1126

1127 **7.4 Brake Particle Filters/Traps**

1128 Filtration systems have been developed for many industrial applications and are now common in
1129 automotive internal combustion engine exhaust systems. This technology is translating across to
1130 brake systems with examples of forced induction filtration systems (developed by Tallano) (Hascoet
1131 and Adamczak, 2020; Gramstat et al., 2020) and passive filtration systems (developed by Mann
1132 Hummel) (Bock et al., 2019).

1133

1134 The Tallano system uses a grooved pad with an extraction system drawing air out of the groove
1135 through a filter using an electric motor powered from the vehicle electrical system. The system has
1136 been tested on brake dynamometer rigs and on vehicle. Mass based measurement was primarily
1137 used to determine the trapping effectiveness by comparing the weight loss of the friction materials
1138 to the weight gain of the filter, supported by particle measurements. The results indicate that
1139 filtration efficiencies between 40 and 90% are achievable depending on the braking behaviour,
1140 particle size range and filter condition (Hascoet and Adamczak, 2020). The filtration efficiency
1141 appeared to degrade to the lower levels when brake temperatures exceeded the decomposition
1142 temperatures – suggesting potential difficulties in this particular design in trapping finer particles.
1143 Comments on filter capacity suggest that limited life of the filter (consumable) may still need to be
1144 resolved as well as understanding its behaviour as a function of weather (Gramstat et al., 2020).

1145

1146 The Mann-Hummel system consists of a passive filter element located downstream of the brake
1147 pads and caliper with the intention of using the induced flows to transport the airborne particles
1148 through a filter layer. This has the advantage of being retrofittable due to the minimal changes to the
1149 existing braking system (Bock et al., 2019). It is reported to have been demonstrated on many trial
1150 vehicles under many conditions but limited supporting data is currently published. They claim filter
1151 life comparable with pad life and filtration efficiencies as high as 80%. The flow rate through the
1152 filter will depend on the filter permeability which will change over time, thereby changing the
1153 bypass ratios and filtration efficiencies. Conceptually similar filters have reported unevidenced
1154 filtration efficiencies of up to 92% (Fieldhouse and Gelb, 2016). Partial flow filters used in exhaust
1155 systems exhibit similar behaviour with filter loading and flow condition, achieving filtration
1156 efficiencies between 30 and 70% (Schrewe et al., 2012). This comparison should clearly be taken
1157 with caution, but does indicate the degree of variation that is typical of partial flow systems.

1158

1159 **7.5 Road Surface Cleaning**

1160 Road maintenance has been implemented for many generations to reduce the debris and loose
1161 matter on the streets. Technologies to achieve this include sweeping systems, vacuum systems
1162 (often combined with sweeping) and street washing.

1163

1164 There have been a number of studies attempting to elucidate the effect of street sweeping,
1165 vacuuming and washing on airborne particulate number, the majority of which have struggled to
1166 identify a statistically significant effect due to the influence of, for example meteorological effects.
1167 Daily averages were highlighted to be a problematic metric for identifying effects of road
1168 maintenance. The majority of the older studies have already been reviewed well (Amato et al.,
1169 2010b) to which the reader is directed. The difficulties in measuring PM change amongst other
1170 external effects meant that many studies were inconclusive. The minority identified improvements
1171 in PM₁₀ of the order of 2-3 $\mu\text{g}\cdot\text{m}^{-3}$ and 6-30% reduction. The most significant differences arose in

1172 the hours immediately after the road maintenance, with almost twice the reduction in coarse
1173 particles compared to fine particles (Amato et al., 2010a).

1174

1175 Since that review (Amato et al., 2010b), there have been few focused studies on the effects of street
1176 maintenance on airborne PM. A longer duration study in Nevada US, overcame many of the issues
1177 with short term studies through analysis of a year's worth of data alongside street maintenance data
1178 (Zhu et al., 2012). They concluded that winter road treatments have an extended impact on
1179 emissions throughout subsequent months through the transport and degradation of traction additives
1180 (e.g. sand, gravel and salt), with the recommendation to perform street maintenance (sweeping and
1181 vacuuming) after winter storms to remove the remaining traction materials having recognised
1182 statistically significant improvements in roads with similar maintenance schedules. Other analysis
1183 focused studies indicate benefits of $\sim 2 \mu\text{g}\cdot\text{m}^{-3}$ and 2-15% reduction in PM_{10} (Karanasiou et al.,
1184 2011).

1185

1186 The importance of transport of emission material from secondary and tertiary roads was highlighted
1187 by Zhu et al. (2012) with similar results in a more recent study reported by Monks et al. (2019). In
1188 this latter 2017 study a road which was heavily loaded with suspendable matter due to industrial use
1189 was subject to street maintenance. Combined sweeping and washing contributed to a measurable
1190 benefit in PM_{10} whereas sweeping alone was reported not to, supporting earlier conclusions from
1191 Amato et al. (2010b).

1192

1193 **7.6 Dust Suppressants**

1194 For unpaved roads (particularly those in mining industries), dust suppressants are being widely used
1195 to help bind the surface road matter and reduce the amount of transfer to airborne PM. Water can
1196 have a short-term benefit widely recognised through meteorological effects in other studies and

1197 industrial applications of water sprays, however, dust suppressants aim to have a longer lasting
1198 effect. Typical formulations include salts, polymers and resins.

1199

1200 Results, like those for street sweeping, vary depending on the location, with focused reviews on this
1201 area showing benefits in areas of high surface loading of suspendable materials (e.g. areas with
1202 significant use of winter road traction additives, industrial areas or unpaved roads), with benefits of
1203 typically 25-50% in PM₁₀ (USEPA, 2015). The effect of dust suppressants on industrial roads in
1204 Spain identified an ~8% reduction in kerbside PM₁₀ compared to ~18% for street washing (Amato
1205 et al., 2016d). There is a distinct lack of available recent literature for more lightly loaded road
1206 surfaces.

1207

1208 Research communities, particularly for the mining industries continue to develop dust suppressants
1209 with improved durability and effectiveness through improved mechanical properties and ability to
1210 be robust to wetting events, for example through the careful use of hydrophilic and hydrophobic
1211 components in a polymer dust suppressant to improve water durability (Lo et al., 2020). However,
1212 the application is focused on centimetres of depth of loose material and therefore offers little insight
1213 into the effects on paved roads where the resuspendable layer is substantially thinner.

1214

1215 **8. BATTERY ELECTRIC AND AUTONOMOUS VEHICLES**

1216 Many countries have now set deadline dates for terminating the sale of light duty internal
1217 combustion engine vehicles (ICEVs), after which new passenger car sales are expected to be
1218 restricted to battery electric vehicles. The average battery electric vehicle is heavier than the nearest
1219 equivalent ICEV, and Timmers and Achten (2016) raised the possibility that non-exhaust particle
1220 emissions from such a vehicle would exceed all emissions, including exhaust, from an ICEV. Their
1221 estimates were based upon assumed increases in tyre and road surface wear, and greater
1222 resuspension due to increased vehicle mass, and included much reduced brake wear due to

1223 regenerative braking (a system in which vehicle inertia is used to rotate the electric motor, which
1224 then acts as a generator) on electric vehicles. A scenario in which battery electric vehicles became
1225 mandatory in Ireland (Alam et al., 2018) showed an increase in PM_{2.5} emissions based upon the
1226 emission factors assumed by Timmers and Achten (2016).

1227

1228 Beddows and Harrison (2021) used the vehicle weight dependence of emission factors to estimate
1229 the consequences of a change from an ICEV to a battery electric passenger car. The outcomes
1230 differed for gasoline and diesel vehicles, primarily due to a weight difference, and between rural,
1231 urban and motorway driving due to differing levels of wear emissions according to driving mode.
1232 However, the largest influence was the reduction in brake wear due to regenerative braking. A
1233 review by OECD (2020) found estimates of the reduction in brake wear ranging from 25-95%.
1234 Based upon an estimate of 90% reduction due to regenerative braking, Beddows and Harrison
1235 (2021) report reductions of up to 23% in total emissions of PM_{2.5} (including exhaust for the ICEV)
1236 for a battery electric vehicle during urban driving, with smaller improvements in rural and
1237 motorway driving. OECD (2020) used similar methods of calculation, but assumed identical
1238 emissions from gasoline and diesel vehicles, and a 75% reduction in brake wear due to regenerative
1239 braking. They calculated non-exhaust emission factors for passenger cars, SUV and light
1240 commercial vehicles with ranges of 100 and 300km, finding net reductions in PM_{2.5} emissions of
1241 11.2 – 13.3% for the 100 km range vehicles and an increase of 2.6 – 7.8% for the 300 km range
1242 vehicles. Exhaust emissions were not however included. Both studies therefore show little impact
1243 of electrification of the vehicle fleet upon PM_{2.5} emissions, with broadly similar conclusions for
1244 emissions of PM₁₀.

1245

1246 There are, as yet few hard data regarding autonomous (self-driving) vehicles. Current opinion is
1247 that the additional weight of batteries required for the computerisation of operation would
1248 detrimentally affect emissions compared to a conventional BEV, while the reduced braking due to

1249 smoother operation would be beneficial. The balance between the two has yet to be tested in
1250 practice.

1251

1252 **9. NON-EXHAUST EMISSIONS OF VOLATILE ORGANIC COMPOUNDS**

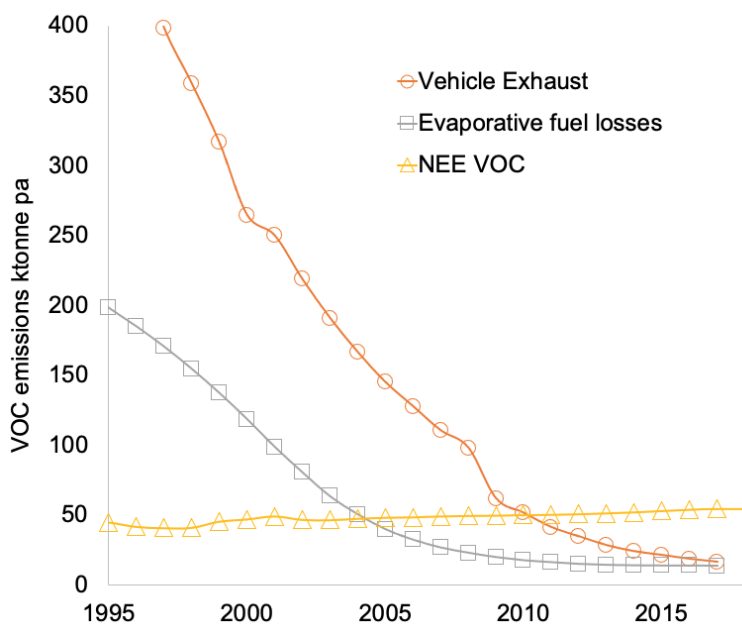
1253 Whilst particulate emissions are the most frequently cited atmospheric emission arising from
1254 vehicle non-exhaust sources, and currently the largest by mass, some vehicles are associated with
1255 non-exhaust emissions of VOCs. VOC emissions from road transport have reduced dramatically
1256 over the last thirty years, both from the tailpipe and from the evaporative loss of fuels. Emissions
1257 from the tailpipe (all vehicle types) in the United Kingdom reduced from 594 kt pa in 1990 to ~16
1258 kt in 2017 (NAEI, 2020)¹, driven by the universal adoption of the highly effective three-way
1259 catalytic convertor. Tailpipe emissions of VOCs typically comprise a mixture of unburned fuel
1260 (most significantly, toluene, xylenes and a range of C4-C10 alkanes), and products from partial
1261 combustion conditions (such as ethene, formaldehyde, acetaldehyde, propene) (NAEI, 2020).

1262

1263 There are non-exhaust emissions of VOCs from ICE vehicles linked to the evaporative loss of fuel
1264 (predominantly gasoline vapour but also some emissions of intermediate volatility VOCs from
1265 diesel fuel). The speciation of those emissions is unsurprisingly linked to fuel formulation, in the
1266 UK the dominant VOCs released from evaporative fuel loss are C6 and C5 alkanes, xylenes,
1267 ethanol, butane, propane and toluene along with smaller amounts of a range of other alkanes and
1268 alkenes (NAEI, 2020). Technical controls on non-exhaust fuel evaporative losses have also been
1269 highly effective, reducing from 261 kt pa in the UK in 1990 to 13 kt in 2017. In combination, the
1270 emission from tailpipe and fuel loss can be considered as emissions that are intrinsic to ICE
1271 vehicles, and that over time are likely to be eliminated as these are replaced by EV/ low carbon
1272 equivalents.

¹The UK reports emissions from road transport separately on a 'fuel sold' and on a 'fuel used' basis. For tracking compliance with the UK's emissions ceilings, the 'fuel used' figures are used derived from kilometres driven. The UK figures shown in this report are on a fuel used basis.

1273 An often-overlooked component of non-exhaust VOC emissions are those associated with solvents
1274 used in ancillary systems and for car care. Most notable are screen-wash and windscreen de-icer.
1275 These emissions are agnostic to the propulsion system and will likely persist even with low carbon
1276 vehicle fleets. The non-exhaust emissions from this class fall into two types, non-aerosol and
1277 aerosol (referred to in EMEP reporting for inventories as: Non-aerosol product – automotive
1278 products NFR Code 2D3a 258 and Aerosols - car care NFR Code 2D3a 274). In combination, these
1279 non-exhaust VOC emissions from vehicles are estimated in the UK at ~55 kt pa, around 3.4 times
1280 the amount of VOC emitted from vehicle tailpipes in that year. Considered in the broader context,
1281 non-exhaust emissions of VOCs (and excluding fuel evaporation) represents around 6.8% of total
1282 UK anthropogenic VOC emissions. Non-aerosol non-exhaust emissions from screen-wash are
1283 dominated by VOCs such as ethanol, ethylene glycol, methanol and 2-methoxy ethanol. Methanol
1284 is particularly notable because of its direct toxicity and EU regulations introduced using the
1285 REACH Directive in 2018 limited its use in screen-wash to 0.6% v/v, now replaced most
1286 commonly by ethanol. Use of aerosol care car products (predominantly for de-icer, but other
1287 sources such as cleaners and polishes contribute also) releases *n*- and *iso*-butane and propane (as the
1288 propellant) and ethanol / 2 propanol as the active ingredient for de-icing. The trends since 1995 in
1289 the UK exhaust and fuel evaporative and NEE VOCs associated with all road transport for the UK
1290 are show in Figure 3.
1291



1292

1293

1294 **Figure 3:** UK emissions of NMVOCs from all road transport by sources according to the United
 1295 Kingdom National Atmospheric Emissions Inventory, summary data available at
 1296 <https://naei.beis.gov.uk>. Methodology for the construction of the VOC element of the NAEI is
 1297 described in Passant et al., 2002.

1298

1299 Whilst the total mass of emissions of VOC from non-exhaust sources is now dominated by ancillary
 1300 emissions from care car products, the differences in speciation between the various non-exhaust
 1301 sources are worth noting. Considering the Photochemical Ozone Creation Potentials (POCP) of the
 1302 two different NEE VOC sources, evaporative fuels are a more significant ozone source on a tonne-
 1303 for-tonne emission basis, since these contain some highly OH-reactive species such as
 1304 monoaromatics and alkenes. The alcohols which make up a significant component of VOCs from
 1305 aerosols and de-icers, whilst still capable of forming ozone, are somewhat lower reactivity. The
 1306 exact ozone formation potentials from different NEE VOC mixtures will depend on the wider
 1307 photochemical environment and is beyond this review.

1308

1309 In summary, whilst the total mass of VOCs emitted from vehicles has reduced dramatically over the
 1310 last three decades, there remains a significant non-exhaust source of emissions, through the release
 1311 of solvents used as consumable products in screen-wash and windscreen de-icing. NEE solvent

1312 emissions from vehicles substantially exceed the total currently being emitted from the tailpipe and
1313 from evaporative fuel losses and would likely persist with future decarbonised fleets.

1314

1315 **10. FUTURE TRENDS**

1316 With traffic levels predicted to grow in many countries, non-exhaust emissions of PM are expected
1317 to further increase if no actions are taken to control them. The UK's emissions inventory predicts
1318 emissions from tyre and brake wear and road abrasion will increase by 13% by 2030, relative to
1319 2018 levels. With exhaust emissions, and emissions from other sources, expected to decline over
1320 this time period as new vehicles meeting tighter exhaust emission standards enter the fleet, non-
1321 exhaust emissions are predicted to be responsible for 90% of all road transport emissions and 10%
1322 of all UK primary emissions of PM_{2.5} by 2030. Emission projections for all countries reported in
1323 this section are taken from official inventory reporting to EMEP Centre on Emission Inventories
1324 and Projections (EMEP, 2021)

1325

1326 The future trend is expected to be similar elsewhere in the world where traffic is predicted to
1327 increase. Emission inventory projections reported by countries in Europe for PM_{2.5}, all excluding
1328 resuspension, are not always complete or comparable making it difficult to compare predicted
1329 trends in non-exhaust emissions between countries. Many countries do predict a growth in non-
1330 exhaust emissions or for the trend to remain roughly flat, although some countries do consider a
1331 reduction in emissions will occur without any clear explanation why. According to the latest
1332 available inventory data, France and Germany each predict a 2% and 4% increase in non-exhaust
1333 emissions of PM_{2.5} from 2018 to 2030, respectively, with the share in these emissions to overall
1334 road transport emissions increasing from 42% to 74% in France and from 62% to 86% in Germany,
1335 a trend similar to the UK situation (EMEP, 2021). Based on inventories for all those countries in
1336 Europe which have made national inventory predictions of non-exhaust emissions, the contribution
1337 made by these sources to total road transport emissions of PM_{2.5} increases, on average, from 50% in

1338 2018 to 71% by 2030. Therefore, the overall picture in Europe is clearly one in which non-exhaust
1339 emissions are expected to become the dominant source of overall traffic emissions of PM by 2030.

1340

1341 Data on future trends in emissions are sparse for other countries and regions of the world. The
1342 California Air Resources Board (CARB) has a comprehensive set of inventory and projections data
1343 showing a similar trend in non-exhaust emissions as in Europe (CARB, 2016; 2017). As described
1344 earlier, the California inventory includes emissions from tyre and brake wear and resuspension and
1345 these are predicted to increase by 11% and 12%, respectively, from 2018 to 2030. To compare with
1346 the scope of PM_{2.5} emissions shown in Table 1 for 2018, tyre and brake wear emissions combined
1347 as a share of total road transport emissions in California (i.e. excluding resuspension) are expected
1348 to increase from 71% in 2018 to 89% in 2030. Including resuspension further increases the
1349 contribution of non-exhaust emission in future years.

1350

1351 Future trends in non-exhaust emissions according to emission inventories are largely based on
1352 predictions in traffic growth, changes in the vehicle fleet and emission factors, but the general
1353 assumption is made that emission factors for non-exhaust sources on a g/km basis remain
1354 unchanged. A major uncertainty in these predictions is whether this is the case and whether PM
1355 factors will change with changes in vehicle design, weight and tyre and brake wear materials and
1356 technologies. A particular area of uncertainty is the effect the future fleet penetration of hybrid and
1357 battery electric vehicles will have on non-exhaust emissions. As discussed in previous chapters,
1358 emissions may be impacted in either direction through the effect of increased vehicle mass which
1359 may increase emissions and the effect of regenerative braking which may reduce emissions. There
1360 is an urgent need for measurements to confirm emission factors for these vehicle types as there are
1361 currently no confirmed or widely accepted emission factors for use in inventories that reflect the
1362 effect these technologies have on tyre and brake wear emissions under real-world conditions.

1363 The report by the OECD on non-exhaust emissions estimated that there would be a 53.5% increase
1364 in non-exhaust emissions of PM from passenger vehicles worldwide, from 0.85 million tonnes in
1365 2017 to 1.3 million tonnes in 2030 based on the predicted growth in the global vehicle stock
1366 (OECD, 2020). This was for a Business-as-Usual scenario that assumed an uptake of lightweight
1367 electric vehicles (EVs) to 4% of the vehicle stock by 2030 and the emission estimates considered
1368 the effect of vehicle weight and regenerative braking on emissions. The study concluded that a
1369 doubling of the EV numbers to 8% of the vehicle stock would have a minimal effect on the rate of
1370 increase in non-exhaust emissions. However, higher emissions would occur if the uptake were with
1371 heavier, larger range EVs.

1372

1373 Beddows and Harrison (2021) recently considered the dependence of non-exhaust emissions from
1374 battery electric vehicles on vehicle mass. The study considered the issue on a g/km emission factor
1375 basis for tyre and brake wear, road abrasion and resuspension under different driving conditions. It
1376 concluded that overall there would only be a modest change in total PM emissions from a passenger
1377 car built to current Euro 6 standards as gains made by removal of the exhaust emissions are offset
1378 by the likely increase in non-exhaust emissions due to the increased weight of EVs. However, the
1379 outcome is critically dependent upon the extent of regenerative braking which is essential if there is
1380 to be a net benefit of EVs relative to conventional internal combustion engine vehicles. Studies like
1381 this highlight the importance of gathering relevant emission factors for each aspect of non-exhaust
1382 emissions from EVs if accurate predictions are to be made on future trends in PM emissions from
1383 the global vehicle fleet as increasing numbers of EVs enter service.

1384

1385 **11. CONCLUSIONS**

1386 Nearly all non-exhaust vehicle emissions (brake wear, tyre wear, road wear and resuspension
1387 particles, and vehicle-care VOCs) arise regardless of the type of vehicle and its mode of power. No
1388 legislation is currently in place specifically to target non-exhaust emissions, in contrast to

1389 progressively stringent legislation that has been effective in reducing exhaust emissions.
1390 Consequently, in most developed countries national emissions inventories indicate that non-exhaust
1391 particle and VOC emissions now exceed their respective exhaust emissions. Future projections are
1392 for non-exhaust emissions to be an increasingly dominant proportion of road transport-related
1393 particle and VOC emissions. From an air quality perspective it is therefore essential to recognise
1394 that further reductions in PM_{2.5} and PM₁₀ in relation to road sources requires attention to non-
1395 exhaust as well as to exhaust emissions.

1396

1397 Comparison between inventories for non-exhaust particle emissions in different regions of the
1398 world is complicated by the different scope in the sources that are covered, for instance with
1399 resuspension being included in inventories reported in the U.S. and Latin America, but not in
1400 Europe. Nevertheless, there is a consistent trend showing at least 50% of PM_{2.5} emissions from
1401 road transport come from tyre, brake wear and road abrasion in much of Europe and the U.S. while
1402 estimates of resuspension in the U.S. suggest around a further 50% occur from this source meaning
1403 a total dominance in all non-exhaust source emissions.

1404

1405 Non-exhaust emission factors used in inventories are sparse, have not been updated since the 1990s
1406 and span a wide range. Therefore, the exact contribution of non-exhaust emissions to air quality
1407 locally and nationally is subject to considerable uncertainty. It is strongly recommended that efforts
1408 continue on development of consistent approaches internationally for measurement of non-exhaust
1409 emission factors. Methods must strive to capture real-world variation in non-exhaust emissions
1410 arising from different materials used, type of road, surface wetness, different cycles of braking,
1411 cornering, etc.

1412

1413 Measurement evidence is clear that non-exhaust emissions increase concentrations of PM₁₀ and
1414 PM_{2.5} and some metals at roadside. Studies suggest contributions (including resuspension) of

1415 several (4-5) $\mu\text{g m}^{-3}$ of PM_{10} at busy roadsides, mostly in $\text{PM}_{2.5-10}$, and in the region 1-2 $\mu\text{g m}^{-3}$ for
1416 the urban background in central London.

1417

1418 Emission inventories for non-exhaust PM sources are largely based on old measurements and do not
1419 account for contemporary vehicle designs, technologies, tyre and brake systems and materials.

1420 Emission factors are held constant at current values and assumed not to change over time. The
1421 effect of electrification of the vehicle fleet on non-exhaust PM emissions is highly uncertain.

1422 Important future determinants of non-exhaust particle emissions are the extent of regenerative
1423 braking in the vehicle fleet and vehicle mass. The former eliminates direct brake-wear emissions
1424 but where batteries, sensors and computer hardware increases vehicle mass, the result may be
1425 greater tyre, road and resuspension emissions. A limited number of recent studies show little impact
1426 of electrification of the vehicle fleet upon $\text{PM}_{2.5}$ emissions, with broadly similar conclusions for
1427 emissions of PM_{10} .

1428

1429 Other non-exhaust emission mitigation approaches (in addition to driving less) include: managing
1430 driving patterns towards lower speeds and less braking; on-vehicle brake-wear capture;
1431 development of low-wear tyres and road surfaces; and road sweeping/washing and application of
1432 dust suppressants to road surfaces, although there is little evidence at present that the latter
1433 measures are effective at mitigating non-exhaust particle emissions in the long term.

1434

1435 Technological innovation on reducing non-exhaust emissions is encouraged, including via
1436 regulation, but the net effect of all on-vehicle changes (e.g. regenerative braking, particle capture,
1437 autonomous driving, vehicle mass) must be evaluated holistically across all sources of non-exhaust
1438 emissions, including any trade-offs between particle mass and particle number emissions.

1439

1440

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1443 the inventory data for NEE.

1444

1445 **CONFLICT OF INTERESTS**

1446 The authors declare no competing financial interest.

1447

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2271 **TABLE LEGENDS:**

2272

2273 **Table 1:** Contribution of non-exhaust sources to road transport and all primary sources of PM
2274 emissions in Europe and the U.S. These are according to current estimates from
2275 emission inventories for the respective regions, with comparisons done on a like-for-
2276 like basis, as far as is possible. Data are derived for the UK from NAEI (2020), for
2277 EU28 from EEA (2021), for California from CARB (2016) and CARB (2017) and
2278 for the USA from USEPA (2020a).

2279

2280 **Table 2:** Metal profiles of various NEE and mineral dusts based on selected ambient, tunnel
2281 and road dust studies. *Metals detected at a subset of sites in parenthesis.

2282 **Table 3:** Summary of quantitative mass apportionment studies of NEE in polluted urban
2283 environments. Notes: (a) Kerbside increment rather than overall concentration
2284 reported; (b) Downtown site; (c) Highway site.

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2287 **FIGURE LEGENDS:**

2288

2289 **Figure 1:** Total exhaust and non-exhaust road transport emissions of PM_{2.5} from EU28
2290 countries from emission inventories reported by Parties under the Convention on
2291 Long-range Transboundary Air Pollution in 2020, covering years up to 2018 (EEA,
2292 2021).

2293

2294 **Figure 2:** Percentage share of total non-exhaust emissions of PM_{2.5} and PM₁₀ for EU28
2295 countries combined, as a percentage share of total road transport and of all national
2296 primary emissions (shown in the figure marked 'Total'). Based on data from EEA
2297 (2021).

2298

2299 **Figure 3:** UK emissions of NMVOCs from all road transport by sources according to the
2300 United Kingdom National Atmospheric Emissions Inventory, summary data
2301 available at <https://naei.beis.gov.uk>. Methodology for the construction of the VOC
2302 element of the NAEI is described in Passant et al., 2002.

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