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Non-exhaust traffic emissions: sources, characterization, and mitigation measures

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Abstract

Non-exhaust emissions (NEE) of particulate matter (PM) from brake, tyre, road pavement and railway wear, as well as resuspension of already deposited road dust, account for up to 90% by mass of total traffic-related PM emitted. This review aims at analysing the current knowledge on road traffic NEE regarding sources, particle generation processes, chemical and physical characterisation, and mitigation strategies. The literature on this matter often presents highly variable and hardly comparable results due to the heterogeneity of NEE sources and the absence of standardized sampling and measurement protocols. As evidence, emission factors (EFs) were found to range from 1 mg km⁻¹ veh⁻¹ to 18.5 mg km⁻¹ veh⁻¹ for brake wear, and from 0.3 mg km⁻¹ veh⁻¹ to 7.4 mg km⁻¹ veh⁻¹ for tyre wear. Resuspended dust, which varies in even wider ranges (from 5.4 mg km⁻¹ veh⁻¹ to 330 mg km⁻¹ veh⁻¹ for cars), is considered the prevailing NEE source. The lack of standardized monitoring approaches resulted in the impossibility of setting international regulations to limit NEE. Therefore, up until now the abatement of NEE has only been achieved by mitigation and prevention strategies. However, the effectiveness of these measures still needs to be improved and further investigated. As an example, mitigation strategies, such as street washing or sweeping, proved effective in reducing PM levels, but only in the short term. The replacement of internal combustion engines vehicles with electric ones was instead proposed as a prevention strategy, but there are still concerns regarding the increase of NEE deriving from the extra weight of the batteries. The data reported in this review highlighted the need for future studies to broaden their research area, and to focus not only on the standardization of methods and the introduction of regulations, but also on improving already existing technologies and mitigating strategies.

Highlights

1. NEE are a relevant PM source in urban areas, exceeding EE contribution.
2. Emission standards and clean air policies must focus on NEE.
3. Standardized NEE sampling and measurement methods are needed to set regulations.
4. Among NEE sources, resuspended dust exceeds brake, tyre, and road wear.
5. EVs have no tailpipe emissions, but still produce significant amounts of NEE.

Keywords: particulate matter, NEE, electric vehicles, air pollution, urban environment

List of abbreviations

Acronym	Meaning
AP-42	Compilation of Air Pollutant Emissions Factors (by USEPA)
APS	Aerodynamic Particle Sizer
BR	Polybutadiene Rubber

Acronym	Meaning
CASQUA	CALifornia stormwater QUality Association
CEN	European Committee for Standardization (original acronym in French)
DLPA	Double Layer Porous Asphalt
DNA	Deoxyribonucleic Acid
EE	Exhaust Emissions
EEA	European Environment Agency
EEC	European Economic Community
EF	Emission Factor
ELPI	Electrical Low-Pressure Impactor
EMEP	European Monitoring and Evaluation Programme
EU	European Union
EV	Electrical Vehicle
HDV	Heavy Duty Vehicle
ICE	Internal Combustion Engine
ICEV	Internal Combustion Engine Vehicle
LDV	Light Duty Vehicle
LRT	Light Rail Trains
LM	Low Metallic
MOUDI	Micro Orifice Uniform Deposit Impactor
NAEI	National Atmospheric Emissions Inventory
NAO	Non-Asbestos Organic
NEE	Non-Exhaust Emissions
NR	Neoprene isoprene Rubber
NURP	Nationwide Urban Runoff Program
PAH	Polycyclic Aromatic Hydrocarbons
PM	Particulate Matter
PMF	Positive Matrix Factorization
RBS	Regenerative Braking System
REACH	Registration, Evaluation, Authorisation and restriction of CHemicals
RP	On-Road generated Particles
SBR	Styrene-Butadiene Rubber
SM	Semi Metallic
SOA	Secondary Organic Aerosol
SUV	Sport Utility Vehicle
TNO	Netherlands Organisation for Applied Scientific Research (original acronym in Dutch)
TP	Tread Particles
TSP	Total Suspended Particles
UITP	International Association of Public Transport (original acronym in French)
UK	United Kingdom
USEPA	United States Environmental Protection Agency
VTI	Swedish National Road and Transport Research Institute (original acronym in Swedish)
WHO	World Health Organization
WP	Tyre Wear Particles
XRD	X-Ray Diffraction

39 1 Introduction

40 Particulate matter (PM) is recognized as one of the main risk factors for adverse health effects and
41 premature deaths worldwide (Brauer et al., 2012; WHO, 2020). Human exposure to PM (especially the
42 fine fraction) is correlated to the outbreak of allergy aggravation, respiratory, cardiovascular and even
43 cerebrovascular diseases (Anderson et al., 2012; Costa et al., 2019; Decesari et al., 2017). Particulate
44 matter can be directly emitted in the environment as primary aerosols or formed in the atmosphere
45 by gas-to-particle conversion processes, i.e. secondary aerosol (Joutsensaari et al., 2018; Sandrini et
46 al., 2016). Sources of particulate matter can be natural or anthropogenic: windblown dust, pollens,
47 plant fragments, sea salt, combustion generated particles, road dust and material produced in the
48 atmosphere by photochemical processes (Fuzzi et al., 2015). The size of airborne particles is very
49 heterogeneous (diameters range from few nanometres to around 100 μm) and depends on the way
50 they originate and on physical and chemical processes occurring in the air (condensation, coagulation,
51 chemical reaction etc.). The need for standardized monitoring systems and regulations led to the
52 definition of particle size classes based on the maximum particle diameter: coarse particles (PM_{10} , i.e.
53 with diameter below 10 μm), fine particles ($\text{PM}_{2.5}$ and PM_1 , i.e. with diameter up to 2.5 μm and 1 μm ,
54 respectively), and ultrafine particles ($\text{PM}_{0.1}$, i.e. with diameter up to 0.1 μm) (Seinfeld and Pandis,
55 2006; Thorpe and Harrison, 2008).

56 The different PM fractions also have different health effects: PM_{10} is the inhalable fraction and
57 includes particles that enter the body through the nose and mouth when breathing; $\text{PM}_{2.5}$ is the
58 thoracic fraction and is composed of particles that penetrate into the lungs under the larynx (Anenberg
59 et al., 2014; Kelly and Fussell, 2012); the respirable fraction (PM_1) encompasses the particles that can
60 reach the lung alveoli (CEN, 1993); and lastly, ultrafine particles ($\text{PM}_{0.1}$) can cross the air-blood barrier
61 in the alveoli and reach internal organs, the brain included (Kelly and Fussell, 2012; Schraufnagel,
62 2020). The fine fraction does not largely contribute to PM mass, but it adds to most of the overall
63 particle number, which is associated to health impact indicators such as the occurrence of hospital
64 admissions due to respiratory and cardiovascular diseases (Atkinson et al., 2010; Stölzel et al., 2007;
65 Wichmann et al., 2000).

66 Vehicle traffic is one of the main contributors to air pollution in the urban environment, and hence
67 to PM (Pant and Harrison, 2013). Traffic-related PM is classified into exhaust emissions (EE), which are
68 the results of fuel oxidation and lubricant volatilization during the combustion process, and non-
69 exhaust emissions (NEE), which are related to brake, tyre, clutch and road surface wear and
70 resuspension of road dust. Another relevant source of non-exhaust PM emissions in urban
71 environments is represented by rail transport (trains, trams and subways) (Abbasi et al., 2013). Besides
72 the diverse generation process, these emissions also differ in size distribution and chemical
73 composition of the released particles (Pernigotti et al., 2016).

74 Exhaust emissions mainly contribute to the fine ($\text{PM}_{2.5}$ and PM_1) and ultrafine ($\text{PM}_{0.1}$) fractions of
75 particulate matter. More specifically, particles formed in the engine and emitted through the tailpipe
76 (primary particles) range from 0.03 μm to 0.5 μm , whereas the secondary particles originating from
77 condensation phenomena in the exhaust plume are below 0.03 μm (Conte and Contini, 2019; Kam et
78 al., 2012; Timmers and Achten, 2016). On the other hand, non-exhaust emissions mainly contribute
79 to PM_{10} and, to a lesser extent, to $\text{PM}_{2.5}$ (Pant and Harrison, 2013; Timmers and Achten, 2016).

80 From a chemical composition point of view, primary EE contain a variety of hydrocarbons and
81 combustion by-products, whereas NEE are mainly composed of heavy metals, such as zinc (Zn), copper
82 (Cu), iron (Fe) and lead (Pb) (Almeida et al., 2020; Thorpe and Harrison, 2008; Timmers and Achten,
83 2016). As it can be expected, a similar chemical composition is also found in the secondary aerosol
84 they generate. The volatile organic compounds in EE react with sunlight and form organic secondary
85 aerosols. On the other hand, NEE mainly produce inorganic secondary aerosols. However, in the case

86 of NEE, it is particularly difficult to discriminate between primary and secondary PM, and therefore to
87 apportion them accordingly (Timmers and Achten, 2016).

88 The potential adverse effects of PM on human health mostly depend on the chemical composition
89 and on the size distribution of particles. From a chemical point of view, transition metals (iron, copper,
90 chromium, cobalt, vanadium, cadmium, arsenic, nickel, etc...) participate in reactions involving the
91 formation of oxygen free radicals, which at high concentration can damage cell structures (Godri et
92 al., 2011; Valko et al., 2006). As a consequence, metal-rich particles potentially cause various
93 modifications in DNA bases and enhance the risk of vascular and neurological diseases and cancer
94 (Berg et al., 2001; Crans et al., 2004; Galán et al., 2001; Halliwell and Gutteridge, 1990; Siah et al.,
95 2005; Valko et al., 2005). As regards the size distribution of PM, small particles are more toxic than
96 large ones since they have a significantly higher surface area available for the adsorption of toxic
97 chemicals (Bogunia-Kubik and Sugisaka, 2002).

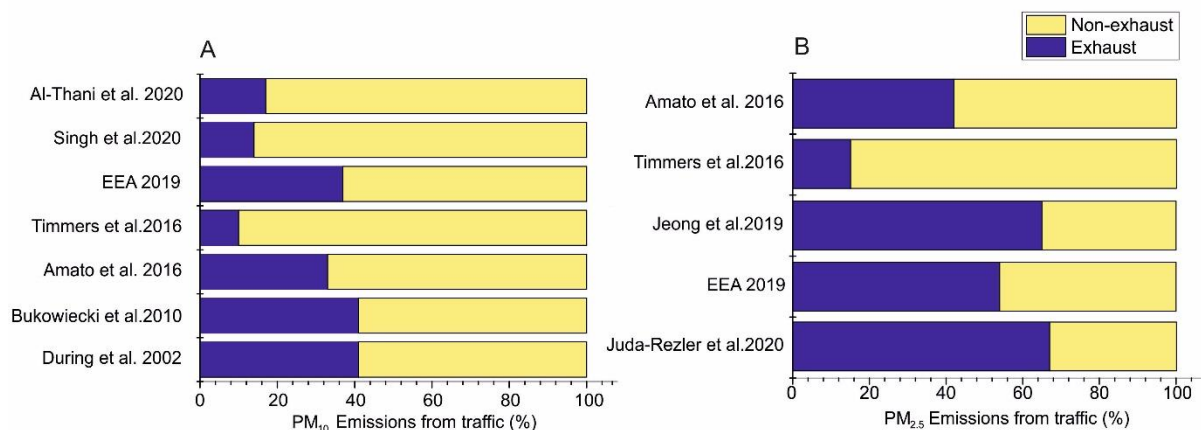
98 To quantify the release of PM into the environment, emission factors (EFs) are used as a tool to
99 estimate the contribution of individual vehicles or vehicle fleet mixtures (Bukowiecki et al., 2009;
100 Bukowiecki et al., 2010). They are typically derived for each category of vehicles (passenger cars, heavy
101 duty vehicle, etc...) and depend on various parameters (brake type, materials, driving style, vehicle
102 weight, etc...). EFs from non-exhaust emissions can be estimated with three different methods,
103 namely 1) direct measurements, 2) receptor modelling, and 3) emission inventories (Timmers and
104 Achten, 2016). Direct measurements involve the characterization of particles sampled during
105 laboratory experiments or field monitoring campaigns. Receptor modelling exploits elemental
106 composition and mass balance analysis to correlate particle emissions to their most probable sources.
107 However, this approach requires a detailed characterization of the source composition and assumes
108 that concentrations measured at the receptor are not altered by other sources (Hopke, 1991).

109 Emission inventories are the most reliable and comprehensive data sources since they derive from
110 the compilation and analysis of a wide range of studies. Authoritative environmental agencies have
111 developed guidelines to standardize the compilation of such inventories, e.g. the “EMEP/EEA Air
112 Pollutant Emissions Inventory Guidebook” (EEA, 2019c) from the European Environment Agency and
113 the “Procedures for Emission Inventory Preparation” from the United States Environmental Protection
114 Agency (USEPA, 1992). Therefore, they are the fundamental tool for air quality management and for
115 the correlation of human activities with the corresponding emissions of pollutants.

116 To regulate emissions from different sources and prescribe limits to be respected for vehicles
117 produced after a certain year, international authorities set thresholds of emission factor (European
118 Community, 2008; WHO, 2006). The European Union started defining the EURO standards for exhaust
119 vehicle emissions in 1992 with EURO 1 (140 mg km⁻¹ veh⁻¹ for diesel cars) (Williams and Minjares,
120 2016). Since then, the threshold was progressively and rapidly decreased, e.g. EURO 3 (year 2000)
121 brought the emission limit to 50 mg km⁻¹ veh⁻¹ for diesel cars. The current European emission threshold
122 for exhaust PM emissions is 5 mg km⁻¹ veh⁻¹ for both diesel and gasoline cars, imposed with EURO 5
123 (since September 2009) and confirmed by EURO 6 (since September 2014). Similarly, the USA have
124 been setting the TIER standards since 1991 (USEPA, 2020). These stringent regulations fostered the
125 technological upgrade of combustion control and tailpipe emission treatment systems, thus leading
126 to a dramatic decrease in the contribution of vehicle exhaust emissions to PM levels (Fuzzi et al., 2015;
127 Harrison and Beddows, 2017; Millo et al., 2015; Pant and Harrison, 2013; Thorpe and Harrison, 2008).
128 A further improvement is expected with the increasing diffusion of EVs, which have no exhaust
129 emission at all.

130 However, non-exhaust emissions from traffic still represent a relevant PM₁₀ and PM_{2.5} source in
131 urban environments. As shown in Fig. 1, the share of NEE on the total emissions from traffic is
132 comparable or even exceeds the contribution of EE (Amato et al., 2014b; Bukowiecki et al., 2010;
133 Düring et al., 2002). The relative contribution of NEE is expected to steadily grow due to the reduction

134 of exhaust emission factors, the phase-out of most polluting cars, the progressive increase of electric
 135 vehicles (EVs) and the lack of limiting standards for NEE (Hooftman et al., 2018).



136
 137 *Fig. 1. Relative contribution of exhaust and non-exhaust emissions to total PM₁₀ emissions from road traffic, based on*
 138 *the research works conducted by (Al Thani et al., 2020; Amato et al., 2016; Beddows et al., 2015; Bukowiecki et al., 2010;*
 139 *Düring et al., 2002; EEA, 2019b; Jeong et al., 2019; Juda-Rezler et al., 2020; Singh et al., 2020; Timmers and Achten, 2016).*

140 Over the last years, several reviews focused on specific topics regarding non-exhaust emissions,
 141 such as brake wear particle emissions (Grigoratos and Martini, 2015), ecotoxicology of micro-sized
 142 rubber (Halle et al., 2020; Wik and Dave, 2009), tyre wear particles in the environment (Baensch-
 143 Baltruschat et al., 2020; Kole et al., 2017; Wagner et al., 2018), nanoparticle emissions from non-
 144 exhaust sources (Kumar et al., 2013) and PM control methods (Amato et al., 2010; Gulia et al., 2019).
 145 However, comprehensive studies on NEE sources and on measures to reduce these emissions are still
 146 missing.

147 This review aims at filling this gap by providing and comparing information on the generation
 148 mechanisms, on key physical and chemical characteristics (chemical composition, morphology, size
 149 distribution), and on typical emission factors of traffic-related NEE, i.e. brake wear, tyre wear, road
 150 surface wear, rail transport and resuspended dust. Finally, an insight is given on technological and
 151 non-technological measures applicable to abate non-exhaust emissions.

152 The literature was reviewed by studying and comparing papers and reports found on search
 153 engines and scientific databases. The bibliographic research was performed with a combination of
 154 several keywords, among them: exhaust and non-exhaust emissions, PM₁₀, PM_{2.5}, road dust, brake,
 155 tyre, wear, chemical composition, size distribution and emission factor. The list of references of each
 156 source was thoroughly analysed to filter data and to find other relevant sources. Overall, 243
 157 publications were compiled, comprising 199 articles, 12 books or book chapters, 21 technical reports
 158 or regulations, 3 emission inventories and 8 patents. The bibliography covers a 46-year time frame
 159 (from 1974 to 2020), 84% of the references was published in the last 15 years.

160

161 2 Non-exhaust emissions: generation, composition, and morphology

162 2.1 Brake Wear

163 Brake wear is the result of the frictional process between a brake pad and a rotating disc or drum.
 164 It contributes from 16% to 55% by mass of the total non-exhaust traffic-related PM₁₀ emissions in
 165 urban roads, where braking events are more frequent (Grigoratos and Martini, 2015; Wahid, 2018).
 166 To better understand the particle formation process and their release in the environment, it is
 167 essential to identify and analyse the brake system components, their materials and properties.

168 Passenger vehicles are usually equipped either with a disk brake or a drum brake system that mainly
 169 differ in the rotating part (a metal disk or a cylinder). In terms of composition, the rotor or rotating
 170 disc is generally made of grey cast iron, but it can also be made of steel, carbon-carbon, ceramic, or
 171 aluminium matrix composites. State-of-the-art callipers are instead composed of aluminium
 172 (Grigoratos and Martini, 2014; Kukutschová and Filip, 2018). Moreover, to guarantee optimal thermal
 173 and mechanical properties and high resistance to wear, brake linings usually have a more engineered
 174 composition than the rotating parts, which comprise the five components reported in Table 1.

175 *Table 1. Brake lining components and materials. Information summarized from Grigoratos and Martini (2015).*

Brake lining component	Mass share (%)	Materials
Binders	20-40	Phenolic resins or modified phenol-formaldehyde resins
Reinforcing fibres	6-35	Copper, steel, brass, potassium titanate, glass, organic material, Kevlar
Fillers	15-70	Inorganic compounds (barium and antimony sulphate, magnesium and chromium oxides), silicates, ground slag, stone, metal powders
Lubricants	5-29	Graphite, ground rubber, metallic particles, carbon black, cashew nut dust, antimony trisulphide
Abrasives	Up to 10	Aluminium oxide, iron oxide, quartz, zircon

176
 177 In the past, asbestos was used as friction material in brake linings due to its fire resistance and
 178 wear properties, but it was phased out as a result of the asbestos-free European Economic Directive
 179 83/477/EEC (European Community, 1983) because of its carcinogenic effect. Currently used brake
 180 materials are classified in three categories: non-asbestos organic (NAO), semi metallic (SM) and low
 181 metallic (LM). The NAO pads are the softest and produce low brake noise, but they have a lower
 182 friction performance than other types of pad and are sensitive to elevated temperatures. This type of
 183 pads is suitable for day-to-day road driving for light and compact vehicles. The SM pads instead have
 184 the highest metallic content (30-60 % of low carbon steel fibre and/or iron powder), a superior
 185 durability and good heat transfer, but they induce an extensive wear of rotor. Thanks to these
 186 characteristics, they are best suited for high performance race cars. Finally, LM pads are made of a
 187 mixture of organic compounds with a small share of metallic components. They present high friction
 188 performance, durability and good braking capacity at higher temperatures (Grigoratos and Martini,
 189 2015; Kukutschová and Filip, 2018). These features make them suitable for medium sized cars, lorries
 190 and trucks.

191 **2.1.1 Generation**

192 During a braking event, the mechanical friction between the disc and the pad combined with the
 193 frictional heat induce the wear of linings and rotors and lead to the release of micronized particles.
 194 Mechanical wear is predominant below 300°C and involves a combination of adhesive, abrasive,
 195 fatigue and oxidative wear; at higher temperatures (over 300 °C), the less thermally stable
 196 components interact with the free gases and oxygen from ambient air (Kukutschová et al., 2009) or
 197 undergo a pyrolysis (Plachá et al., 2015). The generation process becomes even more complex when
 198 organic brake pads are coupled with cast iron discs. In this case, the braking system tends to deposit
 199 a layer of wear particles on the disk boundary. The presence of this layer – that has significantly
 200 different physical and chemical properties compared to both pads and disc – creates secondary disk-
 201 pad contact plateaus, which are in turn subject to continuous growth and destruction phenomena
 202 (Bode and Ostermeyer, 2014; Laguna-Camacho et al., 2015; Merlo et al., 2012; Oesterle and Dmitriev,
 203 2014; Österle and Urban, 2006; Polajnar et al., 2017; Zhang et al., 2019). The synergic effect of all

204 these mechanisms therefore complicates the prediction of the chemical composition, structure and
205 size of the wear debris produced during a braking event (Kukutschová and Filip, 2018).

206 2.1.2 Characterization

207 The generation mechanism, which can be mechanical or thermal, determines the chemical
208 composition and the size of the wear particles. The dimension of the particles ranges from a few
209 nanometres up to 100 μm (Kumar and Ghosh, 2019). In particular, coarse particles are predominantly
210 generated by mechanical processes and mainly contain residues from friction materials (carbonaceous
211 matter, oxidized metals, degradation products of phenolic resins). Conversely, fine and ultrafine
212 particles are generated by thermal and/or chemical processes (Garg et al., 2000; Grigoratos and
213 Martini, 2015; Nosko and Olofsson, 2017) and encompass thermally stable components (i.e. graphite,
214 zircon, barite) (Filip et al., 2002). More specifically, the study by Wahlström et al. (2010) on LM and
215 NAO brakes found that Fe, Cu, Ti, Al, oxygen and carbonaceous species are the main constituents of
216 the fine fraction of wear particles, and Fe predominates in the coarse fraction. Fe in its oxidized forms
217 (i.e. maghemite, magnetite and hematite) was instead found in the fine and ultrafine fraction by
218 Kukutschová et al. (2011); its presence was attributed to the oxidation of Fe-based ingredients of the
219 low-metallic brake pads tested. Similarly, Oesterle and Dmitriev 2014 observed that the
220 nanocrystalline friction layer or third bodies generated at the contact plateaus in organic brake pads
221 are mainly composed of iron oxides. Finally, Liati et al. (2019) performed energy dispersive X-ray
222 analyses of samples collected on a brake test bench for LM and NAO pads, finding that Fe is dominant
223 in every size fractions, especially the coarsest ones. Smaller size particles contain Ca, sometimes in
224 combination with S and/or P. Other elements found in brake wear particles are Al, Cu, Sn, Mg, Si, Cr,
225 Ti, K and W, while traces of Ni, Zn, Zr, Ba, S, C, P, F, Mn are less frequently found and traces of Bi and
226 Sb are rarely found.

227 The abundant presence of metals in the particulate matter resulting from brake abrasion is
228 exploited in source apportionment studies to quantify exhaust/non-exhaust emissions. A good
229 correlation was observed between Fe, Cu, Mn, Ni, Pb and Sb in response to traffic volume, reflecting
230 that metal emissions are vehicle-related (Almeida et al., 2020; Lawrence et al., 2013). Several
231 researchers tried to identify, through PM sampling and characterization in urban environment, the
232 most suitable tracers for brake wear. Copper (Cu) and Antimony (Sb) are the most common ones found
233 in literature concerning brake wear (Almeida et al., 2005; Almeida et al., 2006; Hagino et al., 2016;
234 Megido et al., 2016; Sternbeck et al., 2002). However, in the future both elements will not be suitable
235 as tracers since brake pad manufacturers are introducing Sb- and Cu-free formulations. Iron, which is
236 mostly present in LM pads (Kukutschová et al., 2011), is also not suitable for the assessment of brake
237 wear emissions due to the fact that it can be released by many different sources. Finally, Gietl et al.
238 (2010) found that Barium (Ba) can be used as a good long-term tracer: barium sulphate (BaSO_4)
239 accounts for 1.1% of brake wear PM_{10} , it is not hazardous and there is no plan to replace it in the near
240 future. However, it must be noted that brake wear composition varies over time and according to the
241 country considered. It is therefore necessary to continuously update the literature data (Beddows et
242 al., 2016).

243 Several studies investigated the mass and the number size distribution of airborne brake wear
244 particles. Most studies found unimodal PM_{10} mass distributions with peaks in the range from 1 μm to
245 6 μm (Grigoratos and Martini, 2015). For example, Kukutschová et al. (2011) found a unimodal mass
246 distribution concentrated between 2 and 4 μm for low-metallic pads. Similar results were obtained by
247 Sanders et al. (2003), who tested several types of pads and found that PM_{10} , with a mean mass
248 diameter of 5-6 μm , accounts for 63-85% of the total brake wear mass. These results are in agreement
249 with the study of Garg et al. (2000), who tested seven brake pad formulations (including semi-metallic
250 brakes and brakes functionalized with potassium titanate or with aramid fibres) and showed that $\text{PM}_{2.5}$
251 and PM_{10} are the 63% and 86% by mass of the airborne PM respectively.

252 Analysing the number size distribution, many researchers also found at least one peak at the
 253 ultrafine fraction (diameter < 0.1 μm). For example, Garg et al. (2000) determined the total number
 254 of particles generated in real time testing with both the Dekati electrical low-pressure impactor (ELPI)
 255 and the TSI electrical aerosol analyser (EAA). The results highlighted that the number of particles
 256 emitted increases with brake temperature and suggested that most of the produced particles are
 257 smaller than 0.03 μm . More recently, Nosko and Olofsson (2017) found a strong correlation between
 258 brake temperature and particle size distribution in the results of their tests performed with a pin-on
 259 disc machine on cast iron discs and LM pads. For disc temperatures below 185°C, a peak at 0.165 μm
 260 and a negligible ultrafine fraction were observed; at temperatures of about 350°C, the size distribution
 261 showed instead a prevalence of ultrafine particles with a peak at 0.011–0.029 μm . Finally, Wahlström
 262 et al. (2010) highlighted that the majority of the airborne particles produced are in the fine fraction.
 263 In particular, they observed a bimodal particle number distribution for the LM and NAO brake pads
 264 tested, with peaks at particle sizes of approximately 0.28 μm and 0.35 μm .

265 The heterogeneity of the results proved that it is difficult to perform a precise characterization of
 266 the brake wear emissions and to compare the results of different tests due to a lack of standardized
 267 sampling procedures and measurement techniques (Grigoratos and Martini, 2015). Additionally, the
 268 amount and composition of the brake wear particles produced depend on various parameters, both
 269 intrinsic and external: bulk frictional material (Kukutschová et al., 2011); driving behaviour, frequency
 270 and intensity of braking (Kwak et al., 2013); speed, state and care history of the vehicle; and
 271 environmental conditions, such as ambient temperature and chemicals present in the environment
 272 (Grigoratos and Martini, 2015; Kukutschová et al., 2011).

273 A summary of the results reported in the literature is presented in Table 2.

274 *Table 2. Summary of brake wear studies on size distribution.*

Reference	Generation method	Detection method	Mass size distribution	Number size distribution
Sanders et al. (2003)	Brake dynamometer	MOUDI -ELPI	Unimodal (4-5 μm)	Unimodal (1 μm)
Iijima et al. (2007)	Brake dynamometer	APS (>0.5 μm)	Unimodal (3-6 μm)	Unimodal (1 - 2 μm)
Kukutschová and Filip (2018)	Brake dynamometer	APS – SMPS - BLPI	Unimodal (2-4 μm)	Bimodal (100 and 300 μm)
Garg et al. (2000)	Brake dynamometer	MOUDI (>0.1 μm)	Unimodal (0.1-1 μm)	
Iijima et al. (2008)	Brake dynamometer	APS (>0.5 μm)	Unimodal (μm)	(2 Unimodal (0.8-1 μm))
Uexküll et al. (2005)	Brake dynamometer	Optical particle counter (>0.3 μm)	Unimodal (μm)	(2-3 Unimodal (0.5 -1 μm))
Wahlström et al. (2010)	Brake dynamometer	GRIMM (>0.25 μm)		Bimodal (0.28 and 0.35 μm)

275
 276 **2.1.3 Emission factors**

277 Brake wear EFs can be experimentally estimated through direct measurements in laboratory or
 278 field tests, namely: 1) brake dynamometer tests, 2) pin-on-disc tribometer, 3) identification of brake
 279 wear tracers in ambient air, 4) direct on-road measurement of the source by means of mobile units.
 280 The first two procedures involve the controlled generation, collection and characterization of the
 281 particles emitted during the test, whereas the latter approaches are based on the direct sampling in

282 the urban environment. Unfortunately, it is difficult to compare results due to the variety of
 283 experimental methods and measurement techniques performed. Moreover, each study presents
 284 some disadvantages related to the estimation of the EFs. First, for both dynamometer tests and pin-
 285 on-disc tribometer, the results are based on a small number of vehicles and the particle generation
 286 conditions are considerably far from reality. At the same time, the on-road sampling is inevitably
 287 altered by the other traffic-related emissions, making it difficult to perform a correct characterization
 288 of the particle matter exclusively emitted by brake wear.

289 To overcome the disadvantages deriving from experimental methods, the EEA unified the
 290 procedure for estimating EFs. The EMEP/EEA Emission Inventory Guidebook (EEA, 2019c) combines
 291 PM emission factors in milligrams emitted per kilometre (mg/km) with vehicle kilometres travelled
 292 per year. The categories of vehicles considered are passenger cars, light duty vehicles (LDV), heavy
 293 duty vehicles (HDV and buses) and two-wheelers. The following equation was reported in the
 294 inventory to estimate the PM₁₀ EFs (mg km⁻¹ veh⁻¹) for brake wear:

$$EF_{PM10} = 0.98 \cdot EF_{TSP} \quad \text{Eq. 1}$$

295 where the coefficient 0.98 was proposed by USEPA (USEPA, 1995) and TNO (Berdowski et al., 1997),
 296 while EF_{TSP} (mg km⁻¹ veh⁻¹) is the emission factor of the total suspended particles (TSP) generated
 297 from brake wear. This emission factor can be derived from:

$$EF_{TSP} = f \cdot WR \quad \text{Eq. 2}$$

298 where WR is the wear rate (mg km⁻¹ veh⁻¹) that must be estimated or calculated from surveys,
 299 whereas f is the share of the brake pad mass loss that becomes airborne PM (TSP). This coefficient
 300 varies between 30% (Garg et al., 2000) and 70% (Sanders et al., 2003).

301 The variability in the methodology adopted for the estimation of the EFs is reflected in the values
 302 found in the literature, which are summarized in Table 3. Brake dynamometer studies reveal brake
 303 wear PM₁₀ EFs in the ranges from 2.9 mg km⁻¹ veh⁻¹ to 8.1 mg km⁻¹ veh⁻¹ (Garg et al., 2000; Iijima et
 304 al., 2008; Sanders et al., 2003), with an outlier (Hagino et al., 2015) reporting a much lower value. A
 305 wider range of EFs emerged in research works where receptor modelling is applied, with PM₁₀
 306 emission factors ranging from 2.2 mg km⁻¹ veh⁻¹ to 15 mg km⁻¹ veh⁻¹. Finally, EFs presented in the
 307 emission inventories fall in a range from 7 mg km⁻¹ veh⁻¹ to 18.5 mg km⁻¹ veh⁻¹ for PM₁₀ and from 2.3
 308 mg km⁻¹ veh⁻¹ to 3 mg km⁻¹ veh⁻¹ for PM_{2.5}, in agreement with the results of the other types of studies
 309 mentioned before. The median PM₁₀ brake wear emission factor derived from these data is
 310 approximately 7.4 mg km⁻¹ veh⁻¹, which is slightly higher than the current European emission standard
 311 (EURO 6) for gasoline and diesel vehicle (5 mg km⁻¹ veh⁻¹) exhaust emissions.

312 *Table 3. Brake wear PM₁₀ and PM_{2.5} emission factors found in the literature. For median calculation, the same weight*
 313 *was attributed to each reference. When ranges were provided, the mean value was considered.*

Reference	Data source	Brake PM ₁₀ EF (mg km ⁻¹ veh ⁻¹)	Brake PM _{2.5} EF (mg km ⁻¹ veh ⁻¹)
Garg et al (2000)	Brake dynamometer study	2.9-7.5	1.8-2.8
Sanders et al. (2003)	Brake dynamometer study	8.1	
Iijima et al. (2008)	Brake dynamometer study	5.8	
Hagino et al. (2015)	Brake dynamometer study	0.04-1.4	0.04-1.2
Luhana et al. (2004)	Receptor modelling	8.8	
Bukowiecki et al. (2009)	Receptor modelling	8	
Bukowiecki et al. (2010)	Receptor modelling	15	
Sjodin et al. (2010)	Receptor modelling	2.2	

Dahl et al. (2006)	Receptor modelling	7.4	
EEA (2019b)	Emission inventory	7.4	2.9
USEPA (2014b)	Emission inventory	18.5	2.3
UK NAEI (2018)	Emission inventory	7	3
	Median	7.4	2.3

314

315 2.2 Tyre Wear

316 Tyre manufacturing requires the use of a wide variety of chemicals, fillers and polymers, reported
 317 in Table 4. In detail, the primary components of tyre tread are carbon black, elastomer compounds,
 318 steel cords, fibres and other organic and inorganic compounds (Kreider et al., 2010; Landi et al., 2016).

319 *Table 4. Average composition of tyres (Wagner et al., 2018).*

Compounds	Content (%)	Ingredients
Rubber/Elastomer	40-60%	Polybutadiene (BR), styrene-butadiene (SBR), neoprene isoprene (NR), polysulphide
Fillers	20-35%	Carbon black, silica, silanes
Process oils	12-15%	Mineral oils
Vulcanization agents	1-2%	ZnO, S, Se, Te, thiazoles, organic peroxides, nitro compounds
Additives	5-10%	Preservatives, antioxidants, desiccants, processing aids
Textile and metal reinforcement	5-10%	Steel, polyester, nylon, rayon, aramid fibre

320

321 The core of tyre tread is composed of thermoset polymers (natural or synthetic rubber), which are
 322 liquid or soft solid polymers irreversibly hardened by curing (Halle et al., 2020; Hirata et al., 2014; Kole
 323 et al., 2017). Fillers are used as reinforcing agents to improve the hardness, wear and UV resistance of
 324 tyres. Oils, pine tar, resins and waxes are instead softeners and extenders used to increase rubber
 325 stickiness and stiffness and to improve wet grip performance. For many years, polycyclic aromatic
 326 hydrocarbons (PAHs) were used in extender oils for tyre production. PAHs, which are formed during
 327 the incomplete burning of coal, oil, gas and other organic matters, are known to be cancerogenic.
 328 Therefore, the REACH directive (Registration, Evaluation, Authorisation and restriction of Chemicals)
 329 prohibited the use of the "PAH-rich" extender oil in tyres produced after January 2010 (European
 330 Community, 2006). Anti-degradants are instead used in tyre manufacturing to protect them against
 331 degradation by ozone, oxygen, heat or other factors. Finally, the vulcanization process requires curing
 332 agents, which are accelerators, activators and retardants (Gent and Walter, 2006; Grigoratos and
 333 Martini, 2014; Wik and Dave, 2009).

334 Some of the chemical components involved in the manufacturing of tyres can be used as markers
 335 for tyre wear particles, such as benzothiazole (Kim et al., 1990; Rogge et al., 1993) and benzothiazole
 336 compounds, such as 2-(4-morpholinyl) benzothiazole (Kumata et al., 1997; Park et al., 2019), styrene
 337 butadiene rubber (SBR) (Mengistu et al., 2019; Pierson and Brachaczek, 1974), and zinc, which is added
 338 as an activator for the vulcanization processes (Adachi and Tainosho, 2004; Fauser et al., 2000).

339 2.2.1 Generation

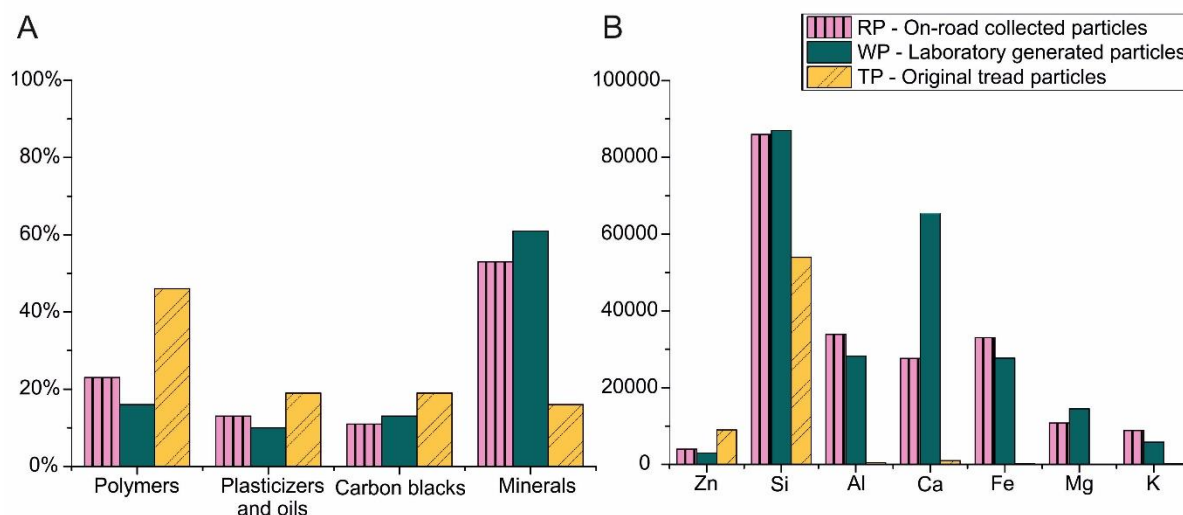
340 Tyre abrasion due to the contact with the road surface leads to the release of large quantities of
 341 small rubber particles (the size range includes PM₁₀), whose chemical composition and characteristics
 342 can differ from the original tyre tread due to heat, friction and incorporation of material from the road
 343 surface (Adachi and Tainosho, 2004; Kreider et al., 2010; Panko et al., 2013). The amount and size of
 344 wear particles released depend on several factors: climate, composition of the tyre, road surface and

345 conditions, speed and driving behaviour. The wear process is dominated by both heat generation and
 346 shear forces between the tread and the road pavement. Similarly to brake wear, mechanical shear is
 347 responsible for the generation of coarse particles (PM₁₀), whereas the high temperature reached in
 348 the tyre surfaces causes the volatilization of small particles in the fine mode (PM₁) (Grigoratos and
 349 Martini, 2014; Kole et al., 2017; Kreider et al., 2010; Mathissen et al., 2011).

350 Experimental tests for the characterization of tyre-related NEE can be either performed on real
 351 particle samples directly collected from the environment or on micronized rubbers specifically
 352 generated in laboratory-controlled conditions. Depending on the particle origin, it is possible to divide
 353 all the micro-rubbers into three main categories: tread particles, tyre wear particles and tyre road
 354 wear particles. Tread particles (TP) include particles manually abraded from never used tyres and
 355 therefore do not contain road wear. Tyre wear particles (WP) are instead generated by tyre wear on
 356 a road or in a laboratory road simulator, thus they also include road wear. On-road generated particles
 357 (RP) are a mixture of rubber tyre particles, road dust and other car emissions; this form of particles is
 358 mostly sampled from the environment. Because of the different generation process, these micro-
 359 rubbers potentially present different characteristics in term of size and/or chemical composition.

360 2.2.2 Characterization

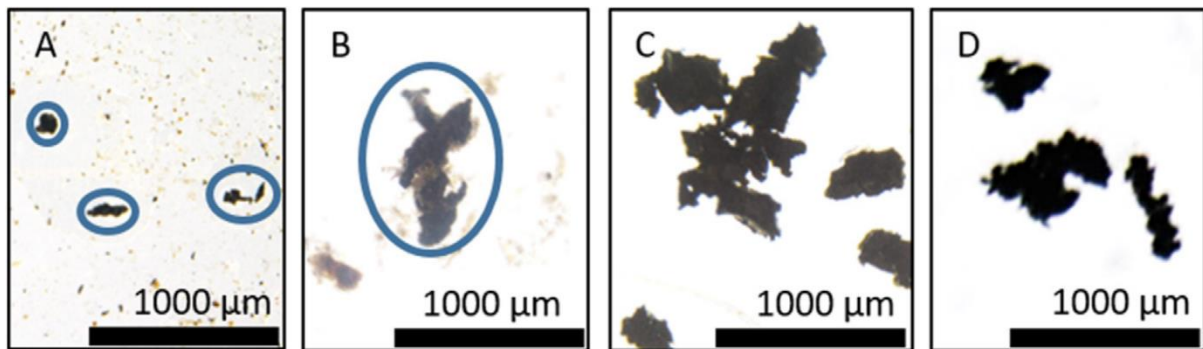
361 Adachi and Tainosho (2004) compared the chemical composition of tyre dust (RP) to the one
 362 belonging to original tyre tread (TP), and found out that tyre dust is enriched of heavy metal particles
 363 (i.e. Ti, Fe, Cu, Zn etc.), which derive from brake linings and asphalt pavement materials (i.e. CrPbO₄).
 364 Kreider et al. 2010 highlighted the chemical composition differences of original tread particles (TP),
 365 laboratory generated particles (WP) and on-road generated particles (RP) measured through
 366 thermogravimetric analysis. As shown in Fig. 2A, both WP and RP present an enrichment in mineral
 367 content and a reduction of polymer amount due to the contribution of other traffic-related emission
 368 sources, whereas more polymers and less mineral content were found for TP. As shown in Fig. 2B, WP
 369 and RP are enriched in Al, Si, Fe, Ca, Mg and K from pavements and other traffic-related sources; on
 370 the other hand, WP and RP are less rich in zinc, a metal used in tyre manufacturing in the form of ZnO,
 371 because of a dilution from the pavement. Consequentially, the authors concluded that tyre can be
 372 considered as the major contributor of zinc in road dust.



373
 374 *Fig. 2. General composition analysis with values expressed in % weight (A) and analysis of metallic content with values*
 375 *expressed in parts per million (B) of particles from Kreider et al. (2010) for on-road collected particles (RP), laboratory*
 376 *generated particles (WP) and original tread particles (TP).*

377 Besides the identification of the chemical composition, many attempts were also made with the
 378 purpose of comparing the morphology and size distribution of different types of particles, such as on-

379 road samples or laboratory generated particles under simulated driving conditions (Kreider et al.,
380 2010; Wagner et al., 2018). The results obtained were very heterogeneous and the measured sizes
381 proved to be dependent not only on the particle type, but also on the experimental setup and
382 procedure and on the analytical techniques used in each study (Kole et al., 2017). The morphologies
383 of tyre wear micro-rubber found were different according to the experimental conditions applied,
384 such as tyre rotation velocity, force and type of surface abrasion. Nevertheless, abraded micro-rubber
385 particles sampled in nature (Fig. 3A-B) and produced in laboratory (Fig. 3C-D) have a similar shape, i.e.
386 particles are elongated, warped, porous, near spherical, jagged and irregular (Halle et al., 2020).



387

388 *Fig. 3. Diversity of rubber from laboratory production to sampling along a road. A: RP (circled in blue) sampled on gel-*
389 *tape from a road surface. B: Water sample of RP (circled in blue) collected from a road surface drain. C: Laboratory generated*
390 *WP by cryogrinding in liquid nitrogen. D: Laboratory generated WP produced by pressing a tyre on a coarse grinding stone*
391 *(Halle et al., 2020).*

392 Size distributions of tyre wear particles available in the literature (Table 6) vary according to the
393 wear processes, i.e. mechanical abrasion forms coarse particles, whereas thermal processes generate
394 fine particles. Kreider et al. (2010) tested summer and friction tyres in a road simulator and, through
395 laser diffraction analysis, identified a wide unimodal particle size distribution ranging from 5 µm to
396 200 µm, with a mode centred at ~ 75 µm for RP, and from 4 µm to 350 µm, with a mode centred at ~
397 100 µm for WP. A limitation of this study is represented by the inability of the measurement device to
398 detect particles below a diameter of 0.3 µm. Similarly, Aatmeeyata et al. (2009) found a bimodal mass
399 distribution of tyre wear particles (0.3 µm and 4/5 µm), but the peak at 0.3 µm was probably altered
400 by the operational limit of the detector.

401 The introduction of more advanced characterization methods – which are able to explore the
402 nanometre range and analyse the number particle size distribution – allowed to extend the
403 investigation also to the finest fraction of tyre wear emissions. The road simulation study conducted
404 by Sjödin et al. (2010) reported a unimodal number size distribution with peaks at 30 nm. A similar
405 mode was described by Dahl et al. (2006) in their road simulation studies performed with studded and
406 friction tyres on different types of asphalts. The authors reported a unimodal number size distribution
407 with a measured peak in the ultrafine mode (from 15 nm to 50 nm), thus proving that the interaction
408 at the road-tyre interface represents a significant source of sub-micrometer particles. Their results
409 agree with the road simulation study from Mathissen et al. (2011), who found a unimodal number size
410 distribution with a peak ranging from 70 nm to 90 nm.

411 In conclusion, literature results on tyre wear were found highly variable depending on the type of
412 study performed (road simulator, pin-on-disc or on-road direct measurement) and on the measuring
413 instrumentation adopted; for these reasons, they are hardly comparable.

Table 5. Summary of tyre wear studies on size distribution.

Reference	Generation method	Detection method	Mass size distribution	Number size distribution
Olofsson et al. (2018)	Pin-on-disc	Spectrometer FMPS (5.6-560 nm) & OPS (0.3-10 µm)	Unimodal (5-10 µm)	0.2, 1 and 2 µm
Kreider et al. (2010)	Road simulator	Suction system collecting particle (>0.3 µm) + Laser diffraction	Unimodal (75 µm)	Bimodal (5 and 25 µm)
Aatmeeyata et al. (2009)	Road simulator	GRIMM analyzer (>0.3 µm)	Bimodal (0.3 and 4/5 µm)	Bimodal (0.33 µm and 1.75 µm)
Sjödin et al. (2010)	Road simulator	APS (> 0.5 µm) & SMPS	Unimodal (2-4 µm)	Unimodal (30 nm)
Panko et al. (2009)	Road simulator	APS (> 0.5 µm) & SMPS	Bimodal (1.0 µm and 5-8 µm)	Unimodal (30-90 nm)
Mathissen et al. (2011)	Road simulator	Spectrometer (5.6-560 nm)	-	Unimodal (70-90 nm)
Kim and Lee (2018)	Road simulator	Spectrometer (5.6-560 nm) & APS (> 0.5 µm)	Unimodal (3-4 µm)	-
Park et al. (2018)	Road simulator	Light-scattering laser photometer & Spectrometer (5.6-560 nm)	Unimodal (2 µm)	Unimodal (< 0.5 µm)
Dahl et al. (2006)	Road simulator	SMPS (14-660 nm)		Unimodal (15-50 nm)
Kwak et al. (2013)	Road simulator	APS (> 0.5 µm)	Unimodal (2-3 µm)	
Kreider et al. (2010)	On-road direct measurement	Suction system collecting particle (>0.3 µm) + Laser diffraction	Unimodal (50 µm)	Unimodal (25 µm)
Alves et al. (2020)	On-road direct measurement	SMPS (14-660 nm) & APS (> 0.5 µm)	Unimodal (> 0.5 µm)	Unimodal (< 0.5 µm)

415

416 2.2.3 Emission factors

417 Most of the studies on brake particulates mentioned above also report an estimation of emission
418 rates for airborne tyre wear particles, for both PM₁₀ and PM_{2.5} fractions. The methods used to estimate
419 tyre wear emission factors include derivation from emission inventories, receptor modelling and direct
420 measurement in laboratory experiments using a road simulator. Alternatively, the emission factors
421 can be derived by experimental measurements in wind tunnels (which reproduce a realistic condition)
422 or through road sampling and mobile on-board measurement.

423 Based on experimental results, several authors proposed models to evaluate the PM₁₀ and PM_{2.5}
424 emission factors, such as the formula of Aatmeeyata et al. (2009):

$$EF_i = \frac{(s \times L + c) \cdot V_{set-up}}{M} \quad \text{Eq. 3}$$

425 where i stands for PM₁₀ or PM_{2.5}, EF_i is the emission factor ($\mu\text{g km}^{-1} \text{tyre}^{-1}$) of i , s ($\mu\text{g m}^{-3} \text{kg}^{-1}$) is the slope of
426 the trend line of the variation of PM _{i} emission with increasing load and c ($\mu\text{g m}^{-3} \text{tyre}^{-1}$) is its intercept, L is the

427 load (kg tyre^{-1}), V_{set-up} is the volume of set up (m^3), and M (km) is the average km run during the test (5-minute-
 428 long). Similarly, Wang et al. (2016) found a linear correlation between vehicle load and tyre wear, whereas
 429 Salminen (2014) proposed an exponential correlation.

430 Table 6 summarizes the results of studies on tyre wear emission factors. Two different road
 431 simulation studies reported a PM_{10} emission factor of $3.8 \text{ mg km}^{-1} \text{ veh}^{-1}$ and $9 \text{ mg km}^{-1} \text{ veh}^{-1}$ for
 432 summer and friction tyres respectively (Kupiainen et al., 2005; Sjödin et al., 2010). An intermediate
 433 value of $7.0 \text{ mg km}^{-1} \text{ veh}^{-1}$ was estimated in the on-road study conducted by Panko et al. (2013). More
 434 recently, a substantially lower value of about $2 \text{ mg km}^{-1} \text{ veh}^{-1}$ for PM_{10} was estimated with a road
 435 simulation study (Alves et al., 2020). These values are close to PM_{10} emission factors reported in both
 436 the USEPA (2014b) emission inventory for light-duty vehicles ($6.1 \text{ mg km}^{-1} \text{ veh}^{-1}$) and the UK National
 437 Atmospheric Emission Inventory (NAEI, 2018) for passenger vehicles ($7.0 \text{ mg km}^{-1} \text{ veh}^{-1}$). Also, the
 438 more updated emission inventory by EEA (EEA, 2019a) confirmed a similar value of $6.4 \text{ mg km}^{-1} \text{ veh}^{-1}$.
 439 As for $\text{PM}_{2.5}$, Panko et al. (2013) estimated an emission factor of $0.3 \text{ mg km}^{-1} \text{ veh}^{-1}$ for light duty
 440 vehicles. Similarly, a $\text{PM}_{2.5}$ EF of $0.9 \text{ mg km}^{-1} \text{ veh}^{-1}$ was stated by USEPA (2014a), whereas higher values
 441 of $4.5 \text{ mg km}^{-1} \text{ veh}^{-1}$ and $5 \text{ mg km}^{-1} \text{ veh}^{-1}$ were reported respectively by EEA (2019a) and NAEI (2018).

442 The literature analysis reveals some variability in the EF values, which can be attributed to the
 443 employment of different sampling and quantification methods, as well as operating conditions
 444 (meteorological conditions, type of tyre, road surface type and vehicles) and driving behaviour (speed
 445 and braking events) (Mathissen et al., 2011; Pant and Harrison, 2013). Overall, PM_{10} EFs fall in the
 446 range from $3.8 \text{ mg km}^{-1} \text{ veh}^{-1}$ to $9 \text{ mg km}^{-1} \text{ veh}^{-1}$, while $\text{PM}_{2.5}$ EFs range from $0.3 \text{ mg km}^{-1} \text{ veh}^{-1}$ to 5 mg
 447 $\text{km}^{-1} \text{ veh}^{-1}$ (Table 6).

448 *Table 6. Summary of PM_{10} and $\text{PM}_{2.5}$ emission factors for tyre wear. For median calculation, the same weight was*
 449 *attributed to each reference.*

Reference	Data source	Tyre PM_{10} EF ($\text{mg km}^{-1} \text{ veh}^{-1}$)	Tyre $\text{PM}_{2.5}$ EF ($\text{mg km}^{-1} \text{ veh}^{-1}$)
Dahl et al. (2006)	Tests with road simulator (70 km/h)	1.9	
Sjödin et al. (2010)	Tests with road simulator	3.8	
Kupiainen et al. (2005)	Tests with road simulator (30 km/h)	9	2
Alves et al. (2020)	Tests with road simulator	2	
Aatmeeyata et al. (2009)	Tests with road simulator	3.7	2
Panko et al. (2013)	Roadside study	7	0.3
Luhana et al. (2004)	Receptor Modelling	7.4	
EEA (2019a)	Emission inventory	6.4	4.5
USEPA (2014b)	Emission inventory	6.1	0.9
UK NAEI (2018)	Emission Inventory	7	5
	Median	6.3	2

450

451 2.3 Road Surface Wear

452 Road surfaces are typically concrete-based or asphalt-based: the former are made of coarse
 453 aggregate, sand and cement (EEA, 2019c), while the latter are mainly composed of mineral aggregate
 454 (95%). The remaining 5% is composed of bitumen, which is a highly viscoelastic black blend of
 455 hydrocarbons and their derivatives obtained from the fractional distillation of crude oil (Lindgren,
 456 1996). The composition of road mixtures can be further enriched and optimized with additives like

457 polymers (e.g. tyre crumbs) and different types of fillers (e.g. waxes, asbestos, clay, silica etc.), which
458 lead to the modification of the pavement properties (Porto et al., 2019).

459 2.3.1 Generation

460 Road surface is a relevant source of both primary and secondary PM in urban areas. The former
461 originates from the fragmentation of the road pavement surface due to the interaction with vehicle
462 tyres: the friction between the tyre and the road surface leads to the abrasion of the road surface and
463 the consequent release of airborne particles in the environment. As for the secondary aerosol, it
464 derives from the transformation of the organic asphalt-based materials that are released in the air
465 during the road wear process (Khare et al., 2020).

466 2.3.2 Characterization

467 The chemical characterization of the wear particles is difficult to perform due to the complex
468 composition of the road mixtures. The presence of abundant stone fractions in the pavement results
469 in a mineral wear that is mainly composed of Si, Ca, K, Fe and Al (Lindgren, 1996). Moreover, dust
470 particles with similar mineralogical compositions are released from other sources, such as
471 construction sites and winter sanding/salting. As dust from these sources deposits onto the road
472 surface and is resuspended by wind or vehicle-induced turbulence, it becomes difficult to identify
473 particulate from road wear (Gehrig et al., 2010). Several studies tried to distinguish road abrasion
474 particles from dust resuspension with different approaches. Some authors suggested to employ
475 bitumen as a tracer for road wear particles (Fauser et al., 2000; Fauser et al., 2002; Thorpe and
476 Harrison, 2008). Fauser et al. (2000) analysed size-segregated aerosol sampling and found that 5.8-
477 10.1 wt.% of roadside total suspended particulate (TSP) is made of asphalt particles with a mean
478 aerodynamic diameter of about 1 μm . Amato et al. (2014a) instead used positive matrix factorization
479 (PMF) modelling, that is a statistical factor analysis tool based on the law of mass conservations and it
480 is used to apportion sources of airborne particulate matter in the atmosphere (Hopke, 2000; Hopke,
481 2016; Paatero and Tapper, 1993; Paatero and Tapper, 1994). On average, the authors estimated a 20%
482 relative contribution of road wear source to the total road dust mass loadings.

483 Laboratory-controlled experiments can be helpful to overcome the problems related to the source
484 apportionment in real-world studies. To this purpose, Gehrig et al. (2010) adopted a mobile load
485 simulator, i.e. a device where wheels are pulled in a closed loop at specific speed and loading
486 conditions. This device is generally used to test the properties and durability of road pavements and
487 it allows the separation of road abrasion particles from resuspended road dust. The resulting mass
488 size distribution of the pavement wear particles presented a peak at 6-7 μm and no particles below
489 0.5 μm were observed. Similarly, a study of the Swedish National Road and Transport Research
490 Institute found a mass particle size distribution with a peak at 5-8 μm and low particle mass below 1
491 μm (Gustafsson and Johansson, 2012). The experiments were conducted testing different pavement
492 types (porous asphalt and stone mastic asphalt) in a road simulator. Contrary to what expected, the
493 authors observed that the PM_{10} mass size distribution did not vary significantly between different
494 pavements, thus suggesting that the particulate formation process is more relevant than the material
495 itself.

496 Gustafsson and Johansson (2012) completed their study analysing the chemical composition of
497 PM_{10} generated from road surfaces wear. Particles in the coarser fractions were characterized by
498 mineral contents, mostly silicon (Si), calcium (Ca), potassium (K) and iron (Fe). Conversely, the fraction
499 of particles below 1 μm was dominated by sulphur (S) and chlorine (Cl) related to tyre rubber or
500 bitumen. Finally, small quantities of zinc, that is commonly associated with tyre wear, was found in
501 PM_{10} . These evidences corroborated the results from Kreider et al. (2010), who identified aluminium
502 and silicon as the elements present in the highest quantities in RP and WP. As a consequence, since
503 these elements are commonly detected in higher quantities in asphalt (Legret et al., 2005; Lindgren,

1996) and just in smaller quantities in TP, it can be inferred that road powder principally originates from the pavement surface wear and not from tyre. Moreover, other metal components of pavements, such as iron (Fe), sodium (Na), calcium (Ca), potassium (K) and magnesium (Mg), were found in higher quantities in RP and WP, rather than in TP.

2.3.3 Emission factors

Since the chemical composition of bitumen makes the estimation methods (e.g., chemical mass balance and receptor modelling) inapplicable, only few studies provided road wear PM₁₀ emission factors and even fewer provided PM_{2.5} EFs. In this framework, the EMEP/EEA Air Pollutant Emissions Inventory Guidebook uses the estimation methodology originally proposed by Klimont et al. 2002, who solved the problem of quantifying particle emissions from road surface wear by subtracting tyre wear, brake wear, and re-suspension from the total non-exhaust emission factors. However, their study presents some limits due to the high uncertainty of the values and the limited information obtained.

Another variable that strongly affects road wear emissions is the use of studded tyre and traction sand, commonly employed in some Nordic European countries. Kupiainen et al. (2005) tested non-studded and studded tyres at 30 km/h, and obtained PM₁₀ emission factors of 9 mg km⁻¹ veh⁻¹ and 40 mg km⁻¹ veh⁻¹, respectively. The use of traction sand further increases PM₁₀ EFs, reaching values of 108 mg km⁻¹ veh⁻¹ and 155 mg km⁻¹ veh⁻¹ for non-studded and studded tyres respectively. Due to the lack of information on the matter, no EFs are included in the EMEP/EEA Emissions Inventory Guidebook for road surface wear associated with the use of studded tyre.

Table 7 summarizes the results reported in literature for LDV and HDV. Although only few studies are available, the positive correlation between vehicle weight and NEE EFs is largely confirmed. As a consequence, the growing preference of customers for sport utility vehicles (SUVs) over compact cars is expected to increase road wear contribution to NEE. Indeed, the average SUV weight is estimated to be 21% higher than average cars (Burnham, 2012).

Table 7. Road wear surfaces PM₁₀ and PM_{2.5} Emission Factors. LDV; light duty vehicles, HDV, Heavy-duty vehicles. For median calculation, the same weight was attributed to each reference.

Reference	Data source	Road PM ₁₀ EF (mg km ⁻¹ veh ⁻¹)	Road PM _{2.5} EF (mg km ⁻¹ veh ⁻¹)
Kupiainen (2005)	Road Simulator (30 km/h)	9 (LDV non-studded tyres) 40 (LDV studded tyres)	2 8
Gehrig (2010)	Road Simulator (asphalt concrete)	3 (LDV) 7 (HDV)	
Luhana (2004)	On road measurement	3.1 (LDV) 29 (HDV)	
EMEP/EEA (2019c)	Emission inventory	7.5 (LDV) 38 (HDV)	4.1 -
UK NAEI (2018)	Emission inventory	8 (LDV) 38 (HDV)	4 21
	Median for LDV	7.75	4.05
	Median for HDV	33.5	-

2.4 Resuspended Road dust

2.4.1 Generation

Most of non-exhaust particles derive from the resuspension of material already deposited on the road surface (between wheel tracks, on curb side or into the pores of the asphalt) due to tyre shear, vehicle-generated turbulence and the action of the wind (Grigoratos and Martini, 2015). Therefore, the terms “road dust”, “road sediments” or “street dust” include any form of solid particle on the road

537 surface that can be suspended in the atmosphere through traffic or windblown action (Denby et al.,
538 2018). Only particles with a size below 70 μm can be airborne, however, all the deposited dust can be
539 resuspended as larger particles may be ground into finer ones by the vehicle weight.

540 There are several sources that contribute to road dust: deposited brake/tyre/road wear particles,
541 deposited particles from exhaust emissions, particles from nearby environments, fugitive loading from
542 constructions, roadsides and/or unpaved roads, dry and wet deposition from atmosphere, application
543 of salt during freezing periods, traction sand, deposition of pollen and plant materials (Amato et al.,
544 2014b). The dominant contributor to road dust largely varies according to the environment: in colder
545 regions the predominant source is the road wear from studded tyres (Norman and Johansson, 2006),
546 in urban environments the relevant sources are tyre and brake wear (Bukowiecki et al., 2010), while
547 in some cases there may be no dominant source. Because of the heterogeneity of dust sources, it is
548 difficult to distinguish between “direct” wear emissions (tyre, brake or road wear) and “resuspended”
549 wear emissions, and therefore to separate their relative contribution to atmospheric PM levels (Denby
550 et al., 2018).

551 2.4.2 Characterization

552 Road dusts have natural or anthropogenic origins, and their composition varies depending on
553 geographical location, climate factors, resuspended soil and anthropogenic sources (Candeias et al.,
554 2020). Many attempts were made to identify the chemical composition, the size distribution, the
555 morphology and the source apportionment of urban dust (Adamiec, 2017; Alves et al., 2018; Amato
556 et al., 2016; Candeias et al., 2020; Cesari et al., 2019).

557 Road dust is sampled, collected and analysed with various approaches that consequentially lead to
558 different results. For example, Amato et al. (2011) collected road dust samples with a transportable
559 resuspension chamber in Zurich (Switzerland), Barcelona and Girona (Spain). The X-ray diffraction
560 (XRD) mineralogical characterization revealed the presence of quartz, alkali feldspars, carbonate and
561 clay minerals in all the examined cities. The abundance of those elements is related to the road
562 pavement, which is made of granite stones. Heavy metals (Cu, Mo, Co, Zr, Ni, Sb, As, Nb, Zn and Cr)
563 were found in larger concentrations in Zurich compared to Barcelona due to a higher traffic
564 contamination. Alves et al. (2018) used the same method to characterize road dust in Oporto and
565 Braga (Portugal), observing an abundance of inorganic materials (Pb, Zn, Fe, Cu, Sn and Sb). XRD was
566 also used by Candeias et al. (2020) to examine road dust samples collected with a vacuum cleaner in
567 Viana do Castelo (Portugal). Chemical elements relative to traffic sources (such as Br, Cl, Cr, Cu, P, Pb,
568 S, Sn, W and Zn) were found mostly in the finest fractions of the road samples ($<0.074 \mu\text{m}$). The most
569 abundant mineral found was quartz, particularly present in the coarser fraction, followed by
570 muscovite, albite, kaolinite, Fe-enstatite and graphite. The grain size distribution of road dusts showed
571 a marked unimodal distribution with a peak in the range from 10 μm to 106 μm .

572 The size distribution observed in road dust samples strongly depends on the accuracy of the
573 sampling method. Dry sampling methods, based on dry brushing and vacuuming of the road surface,
574 might not be efficient for the collection of finer particles, and therefore lead to an underestimation of
575 the silt loads. This issue was addressed by Gustafsson et al. (2019), whose experiments were
576 conducted in 5 central streets of Stockholm using the VTI wet dust sampler. This instrument uses
577 pressurized water to clean a circular portion of the road surface during a specified interval of time.
578 Then compressed air is applied to move the sample from the washing unit to a sampling vial. The
579 authors found a road dust load between 15 g/m^2 and 200 g/m^2 , with a percentage of particles below
580 10 μm in the range from 2% to 30%. However, as stated by the authors themselves, their results are
581 hardly comparable with those of studies performed through dry sampling methods (Amato et al.,
582 2009a). To the authors’ knowledge, no studies have so far addressed the inter-comparison between
583 dry and wet sampling methods, and therefore their relative collection efficiency is still unknown.

584 2.4.3 Emission factors

585 The ambiguity of resuspended road dust measurements also affects the estimations of emission
 586 factors. EFs are estimated through direct measurements (mobile technique or roadside samplings) or
 587 through inverse modelling, with road dust dispersion based on PM monitoring data. The USEPA
 588 compilation of air pollutant emission factors AP-42 (USEPA, 2014a) provides the following formula for
 589 the estimation of resuspended dust from a vehicle ($\text{mg km}^{-1} \text{veh}^{-1}$):

$$EF = k(sL)^{0.91}W^{1.02} \quad \text{Eq. 4}$$

590 where sL (g/m^2) is the silt load on the road, W (t) is the average weight of vehicles travelling the
 591 road expressed in tons, and k ($\text{mg km}^{-1} \text{veh}^{-1}$) is a particle size multiplier that depends on the range
 592 chosen. In particular, $k = 150 \text{ mg km}^{-1} \text{veh}^{-1}$ for $\text{PM}_{2.5}$ and $k = 620 \text{ mg km}^{-1} \text{veh}^{-1}$ for PM_{10} . The
 593 equation is valid for silt loads sL ranging from 0.03 gm^{-2} to 400 gm^{-2} , the vehicle weight W ranging
 594 from 1.8 t to 38 t, and the vehicle speed up to 88 km/h. Reduction factors are also provided to take
 595 into account the rainfall frequency. The silt loading sL is a key input and it is measured through
 596 sampling with a vacuum.

597 The AP-42 method is the most used for the estimation of resuspended dust EFs and several
 598 examples of its application are reported in Table 8. The results reported are highly variable, also due
 599 to the inclusion of motorbikes and HDV in the datasets. The median value of $33 \text{ mg km}^{-1} \text{veh}^{-1}$ is given
 600 as an order of magnitude of resuspended dust EF which, however, strongly depends on the road type.

601 A modified version of the AP-42 formula is reported in Boulter et al. (2006):

$$EF = c(sL)^{0.65}W^{1.5} \quad \text{Eq. 5}$$

602 where the constant c (mg/m^2) is set to 260 mg/m^2 for $\text{PM}_{2.5}$ and 560 mg/m^2 for PM_{10} . Hence, the
 603 vehicle-induced turbulence increases with the size and vehicle weight, leading to an increased rate of
 604 resuspensions (Barlow, 2014; Garg et al., 2000; Kupiainen et al., 2005).

605 The key role of dust loading is confirmed by other correlations, such as the one proposed by Amato
 606 et al. (2011) based on measurements conducted in Zurich (Switzerland), Barcelona and Girona (Spain):

$$EF = 12.35 \cdot RD^{0.81} \quad \text{Eq. 6}$$

607 where RD (mg/m^2) is the road dust loading.

608 As pointed out by Amato et al. (2011), the formulae (Eq. 4, Eq. 5, Eq. 6) have the strong limitation
 609 of neglecting important factors such as the vehicle speed. Several studies confirm an increase in
 610 resuspended dust emission factors with higher vehicle speed (Amato et al., 2017; Lee et al., 2013;
 611 Pirjola et al., 2010). For example, Hussein et al. 2008 quantified the dependence of road particle
 612 emission and resuspension on vehicle speed using a mobile measurement system, recording particle
 613 mass concentrations at 100 km/h about 10 times higher than those at 20 km/h. Similar results were
 614 obtained by Gustafsson et al. (2008) with their road simulator, where the effect of speed velocity was
 615 studied on stone mastic asphalt pavement. They found that a higher speed increases particle mass
 616 and number concentration for both studded and non-studded tyres. Conversely, other studies (Amato
 617 et al., 2012; Gillies et al., 2001) pointed out that roads with a high average speed and intense traffic
 618 (e.g. motorways) potentially have lower resuspended dust emission factors compared to slower roads.
 619 The authors explain this behaviour with the fact that both variables (traffic and speed) contribute at
 620 reducing the amount of dust on the road surface available for its resuspension.

621 *Table 8. Road dust PM_{10} emission factors estimated by means of the USEPA AP-42 method reported in the literature.*
 622 *Median values were calculated setting each category within each study as one sample. When ranges were provided, the mean*
 623 *value was considered.*

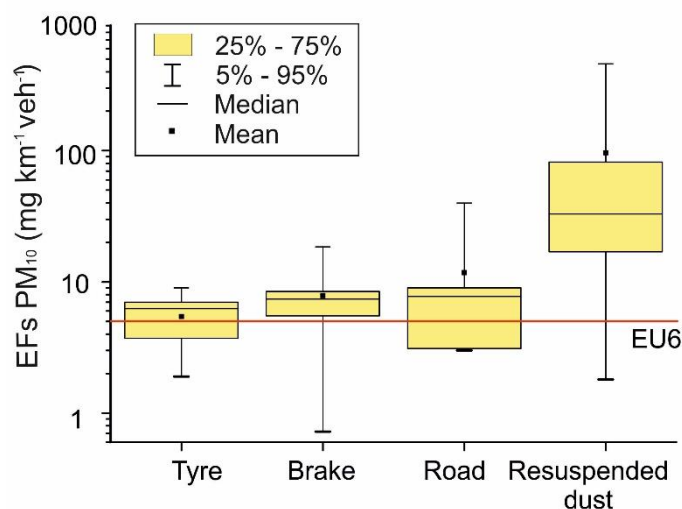
Reference	Location	PM_{10} EF ($\text{mg km}^{-1} \text{veh}^{-1}$)
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Candeias et al. (2020)	Viana do Castelo, Portugal	49 (asphalt road) 330 (cobble stones road)
Amato et al. (2017)	Milan, Italy	13-32
Amato et al. (2016)	Paris, France	5.4-9.0 (inner roads) 17 (ring road)
Alves et al. (2018)	Braga, Portugal	33 (road tunnel)
Amato et al. (2012)	Barcelona, Spain	187-733 (heavy duty vehicles) 33-131 (light duty vehicles) 9.4-36.9 (cars) 0.3-3.3 (motorbikes)
Lamoree and Turner (1999)	St. Louis (USA)	30-40 (highway)
Median		33

624

625 A summary of the results mentioned in the previous chapters is provided in Fig. 4. The graph
626 reports the mean, median, 25/75 percentiles and 5/95 percentiles for PM₁₀ emission factors of NEE
627 sources reviewed in Table 3 (brake wear), Table 6 (tyre wear), Table 7 (road wear), and Table 8
628 (resuspended dust). Resuspended dust appears as the prevailing NEE source, followed by road wear,
629 brake wear and tyre wear. However, resuspended dust composition is extremely broad, both in terms
630 of particle size and chemical composition. The finest metal-enriched fractions (size < 10 µm), which
631 are the most toxic ones, constitute only a small fraction of total resuspended dust mass (between 2%
632 and 30%).

633 Fig. 4 highlights the variability of results obtained in different studies. The most variable results
634 were found for resuspended dust, since environmental and road conditions are significantly
635 influencing the EF estimation. The median value of each NEE sources illustrated in Fig. 4 exceeds the
636 exhaust PM₁₀ emission factor set by EURO 6 (5 mg km⁻¹ veh⁻¹). This confirms the need for the definition
637 and regulation of emission standards for NEE.



638

639 Fig. 4. Mean, median, 25/75 percentiles for box, and 5/95 percentiles for PM₁₀ emission factors of NEE sources, based on
640 the literature review reported in Table 3 (brake wear), Table 6 (tyre wear), Table 7 (LDV road wear), and Table 8
641 (resuspended dust).

642 2.5 Rail transport non-exhaust emissions

643 Railways for light trains, subways and trams represent a vital infrastructure for urban areas. At the
644 end of 2017, there were metros in 182 cities in 56 countries, carrying on average a total of 168 million
645 passengers per day (UITP, 2019). Tram and light rail systems (LRT) are instead used in 389 cities around
646 the world. Melbourne is the city with the longest cable train system, with around 250 km of tramway,

647 followed by Saint Petersburg (246 km), Moscow (208 km), Berlin (193 km), Milan (180 km), Vienna
648 (178 km) and Los Angeles (159 km). In Europe only, the total annual ridership for LRT in 2018 was
649 around 10,422 million, which is comparable to the number of passengers served by metro systems
650 and 10 times higher than those traveling by plane (UITP, 2018).

651 Metros and LRT play a relevant role in the sustainable evolution of modern cities since they
652 positively contribute to the improvement of air quality, due to the reduction of traffic congestion and
653 the absence of tailpipe emissions. However, rail transport is also an appraisable source of non-exhaust
654 particle emissions (Uherek et al., 2010) and can have a negative impact on air quality at local scale.
655 PM produced by railway systems tends to accumulate inside the vehicles themselves or in the
656 immediate vicinity of railways, especially in stagnant or quasi-stagnant areas (e.g. subways, urban
657 canyons) and close to deceleration segments (e.g. railways stations, tram stops), thus representing a
658 potential risk for passengers and drivers.

659 Most research on railway PM emissions focused on subways (Carteni and Cascetta, 2018; Loxham
660 et al., 2013; Loxham and Nieuwenhuijsen, 2019). As an example, several studies (Adams et al., 2001;
661 Pfeifer et al., 1999; Saunders et al., 2019; Seaton et al., 2005; Smith et al., 2020) reported a significantly
662 higher concentration of PM_{2.5} in the London Underground than in other surface means of transport.
663 More specifically, Smith et al. 2020 observed an average PM_{2.5} concentration of 88 µg/m³, with peaks
664 of more than 400 µg/m³. This value was much higher than the average concentration detected in
665 roadside environments in central London (22 µg/m³). A similar result was also reported in Stockholm,
666 where the average PM₁₀ and PM_{2.5} concentrations inside an underground station (470 µg/m³ and 260
667 µg/m³ respectively) were 5-10 times higher than the corresponding values measured in one of the
668 busiest streets of the city centre. Ryswyk et al (2017) instead conducted a study on the exposure of
669 PM_{2.5}, PM₁₀, ultrafine particles and black carbon in the metro system of Toronto, Montreal and
670 Vancouver, Canada. The authors estimated that a typical commute of 70 minutes contributes to 21%
671 of the daily PM_{2.5} exposure in Toronto, 11% in Montreal and 12% in Vancouver. The high concentration
672 of particulate matter in the subway environment was also confirmed by Kim et al. (2008), whose
673 research revealed that the levels of PM₁₀ and PM_{2.5} in platforms of the Seoul Metropolitan Subway
674 exceeded the daily acceptable threshold limits regulated by the USEPA. Finally, Martins et al. (2016)
675 assessed the indoor air quality in the Barcelona subway system, where the mean PM_{2.5} concentrations
676 on the subway platforms were found between 1.4 and 5.4 times higher than the one outdoors.

677 Similar studies were also carried out in tram cabins to assess commuter and driver exposure to PM.
678 Papp et al. 2020 compared PM concentration inside trams with the outside air quality at a nearby site
679 in Debrecen (Hungary). The researchers found that the mass concentrations of PM inside the vehicles
680 were 5–20 times higher than in the outdoor air both for the coarse (aerodynamic diameter larger than
681 2.5 µm) and the fine (aerodynamic diameter smaller than 2.5 µm) fractions. Moreover, since the
682 particle elemental composition was found to be enriched in Cr, Zn, Cu, the study concluded that
683 resuspended dust, rail wear and abrasion of the overhead wire were the main particulate sources.
684 Similar results were obtained also in Vienna (Strasser et al., 2018), Barcelona (Moreno et al., 2015)
685 and Helsinki (Asmi et al., 2009), where number and mass concentrations of fine and ultrafine particles
686 with high metal content were found higher inside tram cabins than the background urban levels.

687 To the authors' knowledge, there are instead only few investigations about the particle release
688 from trams and trains in open urban environments. For example, Kumar et al. (2014) monitored PM
689 concentration at various "hot spots" in Kanpur city (India), while Lorenzo et al. (2006) identified and
690 quantified particles emitted at an increasing distance from a busy railway line at Juchhof (Switzerland).
691 Both studies highlighted that railroad contributes to PM emissions in these "hot spots". However,
692 further research should be conducted on this topic to quantify the actual impact of these PM sources
693 and set specific mitigation measures.

694 2.5.1 Generation

695 Rail transport NEE include brake, wheel and rail wear and the usage of power supply materials:
696 friction and spark erosion of the current collectors (pantographs) and the overhead contact lines
697 (Abbasi et al., 2013; Moreno et al., 2018).

698 2.5.2 Characterization

699 The characteristics of particulate NEE from railways vary according to many factors, such as rail
700 system infrastructures, vehicle technical specifications and operational factors (weight, speed etc.),
701 and power transmission type (Abbasi et al., 2013). The characterization outcome is also affected by
702 the methods adopted for sampling, the meteorological conditions found, and the analytical technique.
703 The majority of PM particles are generated by friction processes and mechanical wear at the rail-
704 wheel-brake interfaces (Moreno et al., 2018), which generate highly ferruginous particles with traces
705 of heavy metals (Mn, Cr, Cu, Sb, Ba and Zn) (Cui et al., 2016; Martins et al., 2016; Perrino et al., 2015;
706 Van Ryswyk et al., 2017). As well as for brake wear emissions, these particles derived from metal-rich
707 sources and transition metals pose a serious threat to human health due to their ability to catalyse
708 the generation of reactive oxygen species.

709 Regarding the particle size distribution of the railway non-exhaust emissions, various studies
710 identified a number peak at approximately 0.35 µm in diameter (Abbasi et al., 2011; Fridell et al., 2011;
711 Gustafsson, 2009). In addition, three other laboratory studies by Sundh et al. (2009) and Abbasi et al.
712 (2012a; 2012b) found peaks at 0.28 µm and 0.6 µm, respectively. Finally, Tokarek and Bernis (2006)
713 discovered that the dominant fraction is composed of even finer particles, with a number peak at
714 diameters below 0.05 µm. As a consequence, the generation of sub-micrometer particles is well stated
715 and the peaks of the number size distribution are comparable to those found in the brake wear studies
716 by Garg et al. (2000), Nosko and Olofsson (2017) and Wahlström et al. (2010).

717 2.5.3 Emission factors

718 To the authors' knowledge, there are no legislations or regulations that control railway NEE, and their
719 quantification is even less studied than the NEE from road vehicles. No concentrations and no
720 recommended emission factors and methodologies are available in international inventories such as
721 the EMEP/EEA Emissions Inventory Guidebook (EEA, 2019c). However, a few countries included
722 emission estimates in their national inventories. For instance, estimates of the PM₁₀ emission factors
723 for abrasion and wear of contact line, braking systems and tyres on rails (reported in Table 9) are
724 provided by the German railroad company Deutsche Bahn AG (Boettcher et al., 2020). Emissions from
725 other wear sources (e.g. the current collector) are not estimated. In the absence of specific
726 information, PM_{2.5} was assumed to be 50% of PM₁₀, whereas TSP was assumed equal to PM₁₀. The
727 report also suggests that non-exhaust emissions from abrasion and wear exceed 90% of total PM
728 emissions, i.e. the contribution of exhaust emissions from diesel trains is very small.

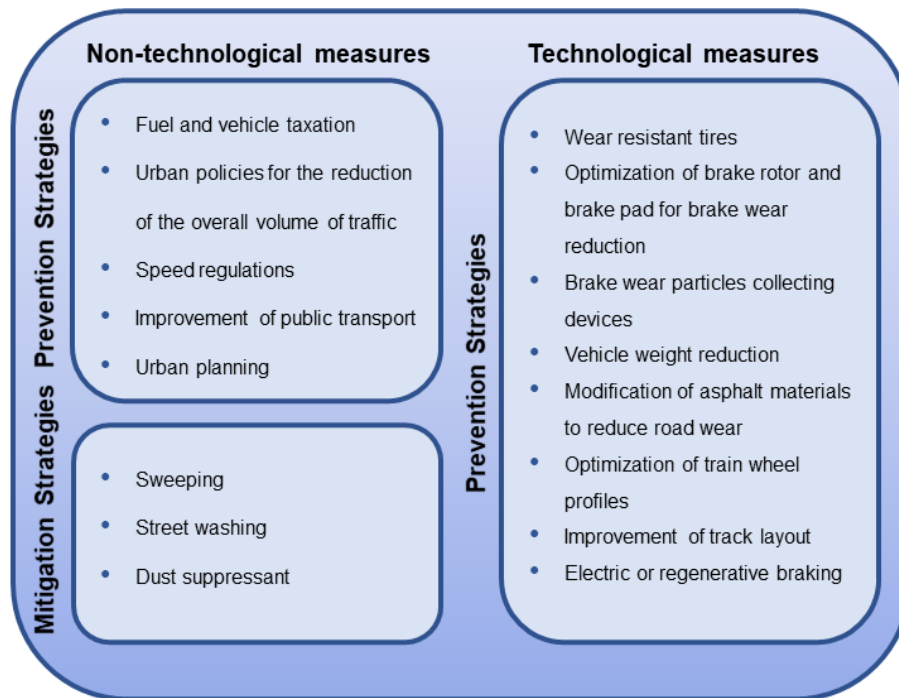
729 *Table 9. Emission factors for railway abrasive emission in Germany (Boettcher et al., 2020).*

	PM_{2.5} (mg km⁻¹)	PM₁₀ (mg km⁻¹)	Cr (mg km⁻¹)	Cu (mg km⁻¹)	Ni (mg km⁻¹)
Contact line	0.16	0.32	-	0.33	-
Tyres on rails	9	18	-	-	-
Braking system	4	8	0.08	-	0.16

730 3 Mitigating strategies for PM

731 The pervasive presence of air pollution in urban areas and its associated adverse effects on human
732 health led to an increasing introduction of measures to abate PM concentration levels. Since road
733 traffic was identified as a main contributor to urban air pollution (Pant and Harrison, 2013), current

734 regulations are focusing on reducing not only exhaust emissions, but also non-exhaust emissions from
 735 brake, tyre, road surface wear and resuspension of road dust. The different mitigating strategies
 736 adopted can be grouped into technological and non-technological measures: the former include
 737 technologies applied to improve the emission performance of existing vehicles or to substitute them
 738 (e.g. introduction of electric vehicles), the latter involve management policies or remediation
 739 measures. At the same time, it is possible to differentiate between prevention or mitigation strategies
 740 adopted to reduce PM levels: the former aim at avoiding particle emissions (paving the access to
 741 unpaved lots or imposing road traffic restrictions), the latter at removing the already deposited dust
 742 (sweeping, street washing, etc...) (Amato et al., 2010). Fig. 5 shows the classification of the existing
 743 abatement methods described above.



744
745

746 *Fig. 5. Classification of strategies to abate PM pollution from non-exhaust emissions.*

747 **3.1 Non-technological measures**

748 **3.1.1 Prevention strategies**

749 Non-technological prevention strategies entail a wide variety of solutions for the reduction of the
 750 overall traffic volume (fuel and vehicle taxation, road traffic management policies etc.), the renewal
 751 and transformation of urban vehicle fleets, and the improvement of public transport and urban
 752 planning (pedestrians and green areas, cycle lanes, low emission zones). For example, cleaning
 753 procedures can be imposed for vehicles exiting construction sites, waste-management plants, and
 754 similar places which, otherwise, would bring large quantities of dust on the road.

755 As already discussed in Chapter 2.4.3, the imposition of speed limits and the promotion of a smooth
 756 driving style reduce braking events and dust resuspension, thus improving passenger safety and air
 757 quality (Gustafsson et al., 2008; Querol et al., 2018). For instance, Kwak et al. (2013) observed a
 758 significant increase in PM concentrations of both road wear and tyre wear particles during
 759 deceleration events. Since both the amount and size of the generated particles strongly depend on
 760 the intensity and duration of the braking events, brake wear emissions were found to be influenced
 761 also by the driving style (Chapter 2.1.1). Stronger and longer braking events produce higher disk

762 temperatures which, in turn, induce the generation of a large amount of fine and ultrafine particles
763 (Garg et al., 2000; Nosko and Olofsson, 2017; Wahlström et al., 2012).

764 3.1.2 Mitigation strategies

765 Mitigation measures aimed at reducing resuspendable dust are sweeping, street washing,
766 sediment removal and dust suppressant. Street sweeping reduces the quantity of dust on the streets
767 and, hence, the fugitive dust that can be re-entrained into the atmosphere by car traffic. The most
768 common types of sweeping vehicles are mechanical broom sweepers, regenerative-air sweepers and
769 vacuum sweepers (Amato et al., 2010; Kang et al., 2009). More specifically, mechanical broom
770 sweepers collect debris into a hopper through a pick-up broom. Regenerative air-sweepers are instead
771 equipped with a gutter, which directs the materials toward a pick-up head, while air is blown onto the
772 pavement to dislodge particles entrained within cracks. As regards vacuum sweepers, they are
773 equipped with gutter brooms and strong vacuum heads for collecting both large and small debris
774 (Calvillo et al., 2015).

775 Street sweeping is a traditional method adopted in most cities for centuries, but investigations on
776 their impact have been carried out only over the last 40 years. Some researcher analysed both
777 standard mechanical street brooms and air-regenerative sweepers for a wide range of cleaning
778 frequencies and street textures, with the aim of understanding the impact of the practices on surface
779 water quality. The evaluation of the street sweeping efficiency was obtained through stormwater
780 quality assessment pre- and post-sweeping. The first studies on the matter were conducted as part
781 of the Nationwide Urban Runoff Program (NURP), which concluded that street sweeping was largely
782 ineffective at reducing the mean concentration of pollutant in urban runoff during a rain event (Pitt,
783 1979; USEPA, 1983). Subsequent studies questioned the NURP conclusions due to the development
784 of sweepers with the ability to retain finer particles (PM_{10}) with specific filtering mechanisms (Amato
785 et al., 2010). Indeed, more recent research projects reported quantitative evidence that street
786 sweeping directly improves runoff water quality (Curtis, 2002; Martinelli et al., 2002). As an example,
787 Selbig (2016) demonstrated a significant reduction in mean total suspended solids concentrations in
788 samples collected from a gutter of a street swept by mechanical sweeper (74% of reduction) and
789 vacuum sweeper (85%).

790 However, there is still considerable uncertainty with regard to pollutant reduction efficiencies,
791 which are extremely variable depending on the frequency and timing of sweeping between storms
792 and the high variability of stormwater quality loads (Hixon and Dymond, 2018; Kang et al., 2009;
793 Sutherland and Jelen, 1997). Regardless, all the different street sweeping techniques proved to be
794 more efficient for the removal of non-resuspendable coarser particles. Although this may seem
795 negligible from the point of view of air quality, it should be remembered that fine particles also
796 originate from the fragmentation of the coarser ones. For this reason, the utility of this traditional
797 cleaning technique should not be underestimated (Amato et al., 2010).

798 Another mitigation measure is water flushing, that consists in the employment of water jet –
799 generally applied in combination with street sweeping – to remove residues from street surfaces.
800 Since the water jet alone cannot easily move the dust towards the sewage system unless an intense
801 water flow is applied, no significant and long-lasting differences were found in PM_{10} concentrations
802 after the application of this technique (Norman and Johansson, 2006). Other studies analysed the
803 combined effect on urban air quality of street sweeping and water flushing. Chang et al. (2005) tested
804 a combination of modified regenerative-air vacuum sweeper and washer, observing a short-term (3-
805 4 h) direct impact on ambient PM emissions. Amato et al. (2009b) evaluated the performances of a
806 vacuum-assisted sweeper followed by manual washer that resulted in a reduction of daily PM_{10} levels
807 between 7% and 10%. More recently, Kryłów and Generowicz (2019) observed a 17.3% reduction of
808 PM_{10} and a 15.4% reduction of $PM_{2.5}$, which lasted up to three days after sweeping and street washing
809 in Cracow. However, they observed a short-term negative effect due to an increase of PM

810 concentration during the street cleaning, as a result of traffic jams and dust resuspension.
811 Nevertheless, the overall effect of street sweeping and washing proved positive.

812 Dust suppressants can be instead classified, according to their chemical composition, as
813 surfactants, salts, polymers, resins and bitumen (Gromaire et al., 2000). These chemicals are water
814 soluble and are spread on the road in a water mixture. They form a film on the particles, which induces
815 the reduction of water evaporation rate and the absorption of moisture from the air, thus resulting in
816 the dust attachment to the road (Gulia et al., 2019). For example, Amato et al. (2014c) evaluated the
817 effectiveness of calcium acetate and $MgCl_2$ in reducing road dust emissions in a Mediterranean city.
818 The authors observed episodic reductions of PM after the application of calcium acetate, but the
819 results were not statistically significant or systematic. Similarly, the application of $MgCl_2$ slightly
820 reduced mineral and brake wear tracers. Better results were obtained in Sweden, with a reduction of
821 the daily mean PM_{10} concentration up to 35% (Norman and Johansson, 2006). The studies carried out
822 so far therefore highlighted that the effectiveness of dust suppressant is strongly influenced by local
823 conditions, such as the amount of dust and climate.

824 3.2 Technological measures

825 3.2.1 Brake wear

826 The reduction of brake wear particle emissions is pursued by changing the chemical composition
827 of the brake pads and/or the rotor and through the introduction of brake dust collection systems.
828 Various approaches that proposed to improve the composition of the friction components of the
829 brake, resulted in the reduction of brake pad particle emitted. Grey cast iron is currently the most
830 used material in the manufacturing of brake rotors. Several studies were performed to improve the
831 wear resistance of cast iron through the addition of titanium (0.1-0.25% weight), which also improves
832 the hardness (Chen, 2014), or the addition of niobium (0.1-0.3% weight), which improves the thermal
833 fatigue resistance and the overall performance at high temperatures (Ying et al., 2014).

834 Copper is one of the most important ingredients in brake pads, since it improves their thermal and
835 mechanical properties (Merlo et al., 2012; Zhang et al., 2020), but it must be phased out due to its
836 toxicity (Lyu et al., 2020). As a consequence, a California law enacted in 2010 (California Senate Bill
837 (SB) 346 (Kehoe)) set a program to eliminate copper use in brake manufacturing, requiring its content
838 in brake pads to be lower than 0.5% by 2025 (CASQUA, 2019). This deadline forced most
839 manufacturers to develop “copper-free” brake pads (Antonyraj and Vijay, 2019; Singaravelu et al.,
840 2019; Vijay et al., 2020).

841 Other experiments were performed on the employment and performances of the coating layers
842 composed of ceramic and metal powder to reduce wear (Lampke and Özer, 2011; Lembach and
843 Mayer, 2012). Research on brake pads was further conducted to improve the hardness and mechanical
844 strength of the linings. Santamaria Razo et al. (2015) developed a new generation of reinforced
845 mineral fibres with enhanced friction material surface. Sun (2014) instead proposed an inorganic nano
846 modified phenolic resin brake pad with improved thermal stability, heat resistance, frictional stability,
847 wear resistance, and a simple preparation.

848 In the last years, several research activities addressed the collection of the particles generated by
849 the brake wear. Rocca Serra (2014) invented an autonomous suction device for drawing off brake wear
850 particles, that consisted in an intake opening (installed close to the pad and the rotor), a collection
851 chamber and an impeller (placed onto the rotor), which drives particles into the intake opening.
852 Fieldhouse and Gelb (2016) developed a brake pad waste collection system composed of a shell that
853 encapsulate the rotor and the calliper. A vented disc connected to the shell generates a positive air
854 flow that leads debris into a filtering system. Combined with magnets, the filters collect particles up
855 to 40 μm and, based on the tests performed, over 92% of brake wear dust was collected and removed.
856 Hascoët and Adamczak 2020 developed TAMIC, an aspiration system composed of a turbine and a

857 high efficiency filter. The device proved a PM₁₀ removal efficiency higher than 85% for both particle
858 mass and number.

859 Despite the significant amount of research in the field of brake wear particle collection, the related
860 market is still poorly developed due to the absence of any specific legislation for the reduction of brake
861 wear emissions.

862 3.2.2 Tyre and road wear

863 Research for the abatement of tyre and road wear is focused on the improvement of materials that
864 generate the greatest quantity of wear particles, i.e. rubber compounds and asphalts. Over the last
865 years, the increase in traffic speed and vehicle weight load shortened the life of asphalt pavements.
866 Thus, various modifiers and additives – such as polymers, chemical modifiers, extenders, oxidants and
867 antioxidants, hydrocarbons and anti-stripping additives – were used to enhance road performance
868 properties (Porto et al., 2019; Yue et al., 2019). Vieira et al. (2019) suggested the application of a
869 Double Layered Porous Asphalt (DLPA) to abate PM₁₀ emissions. DLPAs are composed of a top layer
870 containing fine aggregates and a second layer beneath, that includes coarser aggregates with a high
871 air void ratio (Liu et al., 2016). Thanks to its layered composition, this asphalt is expected to reduce
872 both traffic noise and road wearing. The air quality measurements performed close to a DLPA road
873 highlighted a 52% reduction of PM₁₀ concentrations, compared to measurements with non-porous
874 stone mastic asphalt. Other studies proposed the addition of rubber crumbs – derived from tyre
875 grinding or other secondary rubbers sources – to the asphalt mixture to reduce traffic noise, road and
876 tyre wear (Bressi et al., 2019; Frolova et al., 2016). This technological solution not only increased the
877 road durability (which may reduce particle emission due to asphalt wear), but also allowed to reduce
878 tyre wear emissions by 30%-50% compared to conventional concrete pavements.

879 As regards tyre wear, molybdenum disulphide was proposed as an additive to improve the abrasion
880 resistance (Park, 2006) and, more recently, carbon nanotubes were proposed as reinforcing materials
881 (Huh, 2005). The research activity on tyre wear also focused on the reduction of tyre surface
882 temperature. Park et al. 2019 achieved a reduction of 2-3°C of the average surface temperature of
883 front tyres by introducing cooling air ducts, which resulted in a 4.6% reduction of tyre wear particle
884 emissions.

885 3.2.3 Electric vehicles

886 Electric vehicles were proposed as a potential technological solution to urban air pollution. The
887 absence of tailpipe emissions results in a reduction of nitrogen oxides (NOx) and other exhaust
888 pollutant emissions in urban areas. However, the impact of pollutants emitted during the production
889 of the electricity needed to fuel the battery must also be considered. As for NOx in particular, the
890 overall emission factor for EVs is expected to be far lower than combustion engine cars, since EVs can
891 exploit the excess of electricity produced by photovoltaic and wind power plants (Richardson, 2013).

892 However, EVs contribute to air pollution through non-exhaust emissions (NEE) of particulate
893 matter, and hence they cannot be defined 'zero emission vehicles', but rather 'zero exhaust emission
894 vehicles' (AQEG, 2019). As well as emissions from conventional fossil fuel powered cars, NEE from
895 electric vehicles can be mainly ascribed to tyre and road wear, and dust resuspension. Brake wear
896 emissions for hybrid and full-electric vehicles are instead expected to be lower than combustion
897 engine cars thanks to the possibility to install regenerative braking systems (RBS) in addition to
898 conventional frictional brakes. Regenerative braking is an energy recovery system that slows down a
899 moving vehicle and converts its kinetic energy into another form, most commonly electric energy. This
900 energy can be either directly used for the vehicle propulsion or stored until needed. Thanks to the
901 regenerative system, conventional frictional brakes can be used less frequently during the driving
902 experience (Hall, 2017), thus reducing the particle emitted from brake wear.

903 Various rates of abatement of brake wear emissions thanks to the RBS have been estimated so far.
904 Based on a visual examination of brake pads of passenger cars, Barlow (2014) concluded that EV brake
905 wear emissions are virtually null and braking is mostly based on RBS. A more conservative estimate
906 was provided by Nopmongcol et al. (2017), who predicted a 25% reduction of brake wear to assess
907 the air quality impact of electric vehicles in the USA. Althaus and Gauch (2010) considered a 90%
908 reduction of brake wear emissions based on behaviour analyses of EV drivers. Antanaitis (2010) found
909 that the implementation of a RBS reduces the average operating disc temperatures from 200°C to less
910 than 100°C, with an evident positive effect on the abatement of PM, especially in the finest fraction.
911 On the other hand, tyre and road wear and resuspended dust emission are expected to be higher for
912 EVs because of their increased weight. As reported in the previous paragraphs, there is a positive
913 relationship between vehicle weight and non-exhaust emissions, especially for PM deriving from dust
914 resuspension (Garg et al., 2000; Simons, 2016; Timmers and Achten, 2016). As highlighted by several
915 authors, EVs are heavier than their counterparts with an internal combustion engine (ICE). For
916 example, Moawad, et al. (2013) estimated that EVs are between 43% and 56% heavier than ICE
917 vehicles, whereas Bauer et al. (2015) and Timmers and Achten (2016) found the increase of the weight
918 to be 24%. Moreover, Burnham (2012) estimated that the weight of electric cars and SUVs is 43% and
919 52% higher than their ICE counterparts, respectively. The higher weight is expected to compensate
920 the absence of tailpipe emissions, thus leading to comparable primary PM emission factors for EVs
921 and ICE vehicles. As an example, Timmers and Achten 2016 estimated that the additional weight due
922 to vehicle electrification leads to an overall increase of NEE equal to $12.1 \text{ mg km}^{-1} \text{ veh}^{-1}$ for PM_{10}
923 (corresponding to an increase of $1.1 \text{ mg km}^{-1} \text{ veh}^{-1}$ for tyre wear, $1.4 \text{ mg km}^{-1} \text{ veh}^{-1}$ for road wear and
924 $9.6 \text{ mg km}^{-1} \text{ veh}^{-1}$ for resuspended dust) and $4.4 \text{ mg km}^{-1} \text{ veh}^{-1}$ for $\text{PM}_{2.5}$ (corresponding to an increase
925 of $0.8 \text{ mg km}^{-1} \text{ veh}^{-1}$ for tyre wear, $0.7 \text{ mg km}^{-1} \text{ veh}^{-1}$ for road wear and $2.9 \text{ mg km}^{-1} \text{ veh}^{-1}$ for
926 resuspended dust). This increase substantially compensates the particulate emission saving induced
927 by the diesel/gasoline-to-electric transition, which is estimated to be equal to $12.4 \text{ mg km}^{-1} \text{ veh}^{-1}$ for
928 PM_{10} ($3.1 \text{ mg km}^{-1} \text{ veh}^{-1}$ for tailpipe and $9.3 \text{ mg km}^{-1} \text{ veh}^{-1}$ for brake wear) and to $5.2 \text{ mg km}^{-1} \text{ veh}^{-1}$ for
929 $\text{PM}_{2.5}$ ($3 \text{ mg km}^{-1} \text{ veh}^{-1}$ for tailpipe and $2.2 \text{ mg km}^{-1} \text{ veh}^{-1}$ for brake wear).

930 Secondary organic aerosols (SOA) – which originate from precursors emitted from tailpipes –
931 should also be considered when evaluating the EV impact on air quality (Gentner et al., 2017). EVs do
932 not contribute to SOA emission, and hence, their increasing introduction in the circulating fleet is
933 expected to reduce the formation of secondary particulate due to traffic. However, it is still very
934 difficult to quantify SOA and, therefore, the contribution of EVs in their abatement.

935 3.2.4 Rail transport

936 The management solutions to abate PM emission from railways include better track layouts,
937 optimising train wheel profiles and applying friction modifiers. As regards track layouts, it was proved
938 that the use of a variable slope of the rail line longitudinal profile allows to reduce both energy
939 consumption and mechanical braking usage, thus also reducing PM emissions (Carteni and Cascetta,
940 2018). Similarly to the case of electric cars, brake use and wear can be reduced thanks to RBS, which
941 allow to recover the kinetic energy of electric trams and trains and convert it into electrical energy.
942 Another possible approach to reduce PM emissions is the transition to rubber-tyred metro systems,
943 which eliminates the steel-on-steel interaction that is the main responsible for particulate generation.
944 On the other hand, this leads to a higher rolling resistance, and hence to a higher energy consumption
945 and higher costs for tyre replacements (Carteni and Cascetta, 2018). Finally, radial grooves in brake
946 discs were found to reduce brake wear debris (Mosleh and Khemet, 2006). As regards the passenger
947 and driver exposure to PM inside tram, metro and train cabins, the introduction of more efficient air
948 conditioning and filtration systems and a minor employment of external ventilation through open
949 windows can be exploited to reduce PM levels.

950 4 Conclusions and future needs

951 Particulate matter from traffic is recognized as one of the main risk factors for adverse health effects
952 and premature deaths worldwide. In the last decades, different measures were implemented to
953 reduce PM levels, such as the adoption of exhaust emission standards, the development of after-
954 treatment systems for tailpipe emissions and the promotion of electric vehicles. However, PM still
955 represent an air quality issue worldwide, especially in urban areas. This is due to the high incidence of
956 NEE, which are currently estimated to contribute up to 90% of the overall vehicle PM₁₀ emissions and
957 up to 85% of PM_{2.5} emissions from road traffic. However, non-exhaust emissions of PM are scarcely
958 addressed by regulation and technology development.

959 Despite numerous studies in the field of non-exhaust PM, the definition of NEE limits is still a
960 complex issue. The results of NEE characterization studies are strongly influenced by the type of
961 research performed (field or laboratory), the sampling and detection method, as well as the local road,
962 traffic and climatic conditions. The resulting size distributions and the EFs for resuspended dust, brake,
963 tyre, road and railway wear span over wide ranges. It is therefore difficult to derive generalizable
964 values and set universal emission limits.

965 From this review emerged the following considerations:

- 966 • Resuspended dust is the prevailing NEE source, with PM₁₀ emission factors ranging from
967 5.4 mg km⁻¹ veh⁻¹ to 330 mg km⁻¹ veh⁻¹ for cars. Its emission factors are mostly influenced
968 by road type and vehicle weight, whereas vehicle speed was found to contribute to a lesser
969 extent.
- 970 • Brake wear PM₁₀ emission factors range from 1 mg km⁻¹ veh⁻¹ to 18.5 mg km⁻¹ veh⁻¹ and
971 most of the available studies agree on a unimodal mass size distribution centred between
972 2 µm and 6 µm.
- 973 • Tyre wear PM₁₀ emission factors vary in a narrower range compared to brake wear, i.e.
974 from 2 mg km⁻¹ veh⁻¹ to 9 mg km⁻¹ veh⁻¹. However, studies on size distributions showed
975 contradictory results, which vary from unimodal mass distributions with a peak at 75 µm
976 to bimodal mass distributions with peaks at 0.3 µm and 4 µm.
- 977 • Road surface PM₁₀ emission factors range from 3 mg km⁻¹ veh⁻¹ to 40 mg km⁻¹ veh⁻¹,
978 presenting the highest values for HDVs and vehicles with studded tyres. The mass size
979 distributions reveal a peak in the range from 5 µm to 8 µm.
- 980 • Railway wear PM₁₀ emission factors (from trams, metros and urban trains) are often
981 neglected in emission inventories. However, the wheel-railway friction, the brake wear
982 and, to a lesser extent, the friction of the contact lines are known to release very fine
983 ferruginous particles (unimodal size distribution peaking between 0.28 µm and 0.6 µm),
984 which pose a serious threat to human health at the local scale (i.e. inside vehicle cabins or
985 in underground stations). The quantification of railway emissions and the development of
986 solutions to reduce their impact on urban air quality should therefore be included in NEE
987 studies.
- 988 • The orders of magnitude of the EFs reported in this review largely exceed the most recent
989 exhaust PM₁₀ emissions standards (i.e., 5 mg km⁻¹ veh⁻¹ set by EURO 6), and consequently
990 emphasize the need for future research on particulate emissions from traffic to give more
991 prominence to NEE, rather than EE.
- 992 • Among all the PM fractions, the finest ones have the highest impact on human health since
993 they are more likely to penetrate the human body and are enriched in redox-active
994 transition metals. However, the contribution of these fractions to the overall PM mass
995 emission is negligible, whereas their presence is extremely relevant in terms of particle
996 number concentration. Consequently, the use of mass-based emission factors is not

997 sufficient to fully describe the NEE problem. Future policies should therefore also regulate
998 particle number emissions, and not only mass emissions, to be effective in reducing PM
999 effects on human health.

1000 Current actions to reduce non-exhaust emissions include non-technological prevention (such as
1001 fuel taxation, speed limits in urban areas) and mitigation measures (sweeping, street washing, dust
1002 suppressants), as well as technological measures, such as the modification of the chemical
1003 composition of brake rotors, brake pads, road pavements, tyres, and the introduction of brake dust
1004 collection devices. The extensive use of EVs (i.e. cars, trams, metros and other railway means of
1005 transport) is also considered a technological measure to abate PM emissions in urban environment
1006 thanks to the elimination of tailpipe emissions and the introduction of regenerative braking systems
1007 (RBS) to reduce brake wear. However, in the case of electric cars, the extra weight of the batteries
1008 generates a substantial increase of resuspended dust, tyre and road wear emissions, which together
1009 compensate the lack of tailpipe and brake wear emissions. It is therefore improper to consider the
1010 current state EVs as “zero impact vehicles”, and hence, as the definitive solution to urban PM
1011 pollution. NEE from electric vehicles still contribute to PM pollution and need to be regulated and
1012 reduced to effectively improve urban air quality. A great margin of improvement in both technological
1013 and normative measures to abate NEE particulate still needs to be achieved.

1014 Considering the vast impact of all the different non-exhaust contributors to pollution, future
1015 studies should expand their research field, focusing not only on the standardization of characterization
1016 methods and the introduction of regulations, but also on improving already existing technologies (e.g.
1017 reducing the weight of EV battery pack, or applying RBS to different means of transport), and
1018 developing less polluting materials and systems. Mitigating strategies instead should be more
1019 comprehensive and focus on preventing not only the generation and deposition of primary
1020 particulate, but also the formation of precursors, and therefore the generation of SOA. From this
1021 review emerged the need for future studies on NEE to be as wide and various, as their generation
1022 processes are.

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