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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

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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

1 Introduction

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

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

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

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

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

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

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

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

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

However, non-exhaust emissions from traffic still represent a relevant PM_{10} and $PM_{2.5}$ source in urban environments. As shown in Fig. 1, the share of NEE on the total emissions from traffic is comparable or even exceeds the contribution of EE (Amato et al., 2014b; Bukowiecki et al., 2010; Düring et al., 2002). The relative contribution of NEE is expected to steadily grow due to the reduction of exhaust emission factors, the phase-out of most polluting cars, the progressive increase of electric vehicles (EVs) and the lack of limiting standards for NEE (Hooftman et al., 2018).

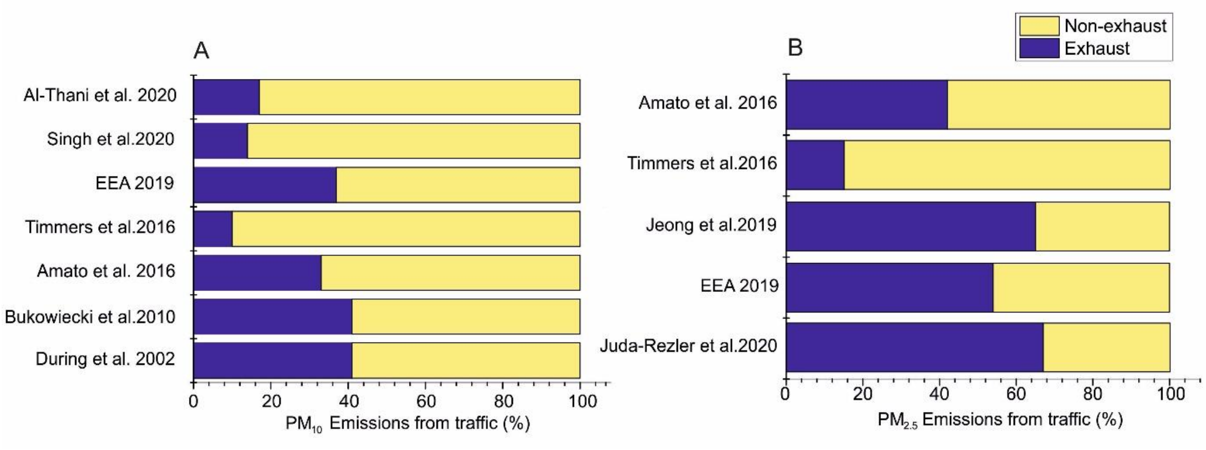

Fig. 1. Relative contribution of exhaust and non-exhaust emissions to total PM_{10} emissions from road traffic, based on the research works conducted by (Al Thani et al., 2020; Amato et al., 2016; Beddows et al., 2015; Bukowiecki et al., 2010; Düring et al., 2002; EEA, 2019b; Jeong et al., 2019; Juda-Rezler et al., 2020; Singh et al., 2020; Timmers and Achten, 2016).

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

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

The literature was reviewed by studying and comparing papers and reports found on search engines and scientific databases. The bibliographic research was performed with a combination of several keywords, among them: exhaust and non-exhaust emissions, PM_{10}, $PM_{2.5}$, road dust, brake, tyre, wear, chemical composition, size distribution and emission factor. The list of references of each source was thoroughly analysed to filter data and to find other relevant sources. Overall, 243 publications were compiled, comprising 199 articles, 12 books or book chapters, 21 technical reports or regulations, 3 emission inventories and 8 patents. The bibliography covers a 46-year time frame (from 1974 to 2020), 84% of the references was published in the last 15 years.

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

2.1 Brake Wear

Brake wear is the result of the frictional process between a brake pad and a rotating disc or drum. It contributes from 16% to 55% by mass of the total non-exhaust traffic-related PM_{10} emissions in urban roads, where braking events are more frequent (Grigoratos and Martini, 2015; Wahid, 2018). To better understand the particle formation process and their release in the environment, it is essential to identify and analyse the brake system components, their materials and properties.

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

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

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

2.1.1 Generation

During a braking event, the mechanical friction between the disc and the pad combined with the frictional heat induce the wear of linings and rotors and lead to the release of micronized particles. Mechanical wear is predominant below 300°C and involves a combination of adhesive, abrasive, fatigue and oxidative wear; at higher temperatures (over 300 °C), the less thermally stable components interact with the free gases and oxygen from ambient air (Kukutschová et al., 2009) or undergo a pyrolysis (Plachá et al., 2015). The generation process becomes even more complex when organic brake pads are coupled with cast iron discs. In this case, the braking system tends to deposit a layer of wear particles on the disk boundary. The presence of this layer – that has significantly different physical and chemical properties compared to both pads and disc – creates secondary disk-pad contact plateaus, which are in turn subject to continuous growth and destruction phenomena (Bode and Ostermeyer, 2014; Laguna-Camacho et al., 2015; Merlo et al., 2012; Oesterle and Dmitriev, 2014; Österle and Urban, 2006; Polajnar et al., 2017; Zhang et al., 2019). The synergic effect of all these mechanisms therefore complicates the prediction of the chemical composition, structure and size of the wear debris produced during a braking event (Kukutschová and Filip, 2018).

2.1.2 Characterization

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

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

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

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

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

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

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 (>0.5µm)	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 (>0.5µm)	Unimodal (2 µm)	Unimodal (0.8-1 µm)
Uexküll et al. (2005)	Brake dynamometer	Optical particle counter (>0.3 µm)	Unimodal (2-3 µm)	Unimodal (0.5 -1 µm)
Wahlström et al. (2010)	Brake dynamometer	GRIMM (>0.25µm)		Bimodal (0.28 and 0.35 µm)

2.1.3 Emission factors

Brake wear EFs can be experimentally estimated through direct measurements in laboratory or field tests, namely: 1) brake dynamometer tests, 2) pin-on-disc tribometer, 3) identification of brake wear tracers in ambient air, 4) direct on-road measurement of the source by means of mobile units. The first two procedures involve the controlled generation, collection and characterization of the particles emitted during the test, whereas the latter approaches are based on the direct sampling in the urban environment. Unfortunately, it is difficult to compare results due to the variety of experimental methods and measurement techniques performed. Moreover, each study presents some disadvantages related to the estimation of the EFs. First, for both dynamometer tests and pin-on-disc tribometer, the results are based on a small number of vehicles and the particle generation conditions are considerably far from reality. At the same time, the on-road sampling is inevitably altered by the other traffic-related emissions, making it difficult to perform a correct characterization of the particle matter exclusively emitted by brake wear.

To overcome the disadvantages deriving from experimental methods, the EEA unified the procedure for estimating EFs. The EMEP/EEA Emission Inventory Guidebook (EEA, 2019c) combines PM emission factors in milligrams emitted per kilometre (mg/km) with vehicle kilometres travelled per year. The categories of vehicles considered are passenger cars, light duty vehicles (LDV), heavy duty vehicles (HDV and buses) and two-wheelers. The following equation was reported in the inventory to estimate the PM_{10} EFs (mg km^{-1} veh^{-1}) for brake wear:

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

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

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

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

The variability in the methodology adopted for the estimation of the EFs is reflected in the values found in the literature, which are summarized in Table 3. Brake dynamometer studies reveal brake wear PM_{10} 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 al., 2008; Sanders et al., 2003), with an outlier (Hagino et al., 2015) reporting a much lower value. A wider range of EFs emerged in research works where receptor modelling is applied, with PM_{10} emission factors ranging from 2.2 mg km^{-1} veh^{-1} to 15 mg km^{-1} veh^{-1}. Finally, EFs presented in the emission inventories fall in a range from 7 mg km^{-1} veh^{-1} to 18.5 mg km^{-1} veh^{-1} for PM_{10} and from 2.3 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 mentioned before. The median PM_{10} brake wear emission factor derived from these data is approximately 7.4 mg km^{-1} veh^{-1}, which is slightly higher than the current European emission standard (EURO 6) for gasoline and diesel vehicle (5 mg km^{-1} veh^{-1}) exhaust emissions.

Table 3. Brake wear PM_{10} and $PM_{2.5}$ emission factors found in the literature. For median calculation, the same weight 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**

2.2 Tyre Wear

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

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

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

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

2.2.1 Generation

Tyre abrasion due to the contact with the road surface leads to the release of large quantities of small rubber particles (the size range includes PM_{10}), whose chemical composition and characteristics can differ from the original tyre tread due to heat, friction and incorporation of material from the road surface (Adachi and Tainosho, 2004; Kreider et al., 2010; Panko et al., 2013). The amount and size of wear particles released depend on several factors: climate, composition of the tyre, road surface and conditions, speed and driving behaviour. The wear process is dominated by both heat generation and shear forces between the tread and the road pavement. Similarly to brake wear, mechanical shear is responsible for the generation of coarse particles (PM_{10}), whereas the high temperature reached in the tyre surfaces causes the volatilization of small particles in the fine mode (PM_1) (Grigoratos and Martini, 2014; Kole et al., 2017; Kreider et al., 2010; Mathissen et al., 2011).

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

2.2.2 Characterization

Adachi and Tainosho (2004) compared the chemical composition of tyre dust (RP) to the one belonging to original tyre tread (TP), and found out that tyre dust is enriched of heavy metal particles (i.e. Ti, Fe, Cu, Zn etc.), which derive from brake linings and asphalt pavement materials (i.e. $CrPbO_4$). Kreider et al. 2010 highlighted the chemical composition differences of original tread particles (TP), laboratory generated particles (WP) and on-road generated particles (RP) measured through thermogravimetric analysis. As shown in Fig. 2A, both WP and RP present an enrichment in mineral content and a reduction of polymer amount due to the contribution of other traffic-related emission sources, whereas more polymers and less mineral content were found for TP. As shown in Fig. 2B, WP and RP are enriched in Al, Si, Fe, Ca, Mg and K from pavements and other traffic-related sources; on the other hand, WP and RP are less rich in zinc, a metal used in tyre manufacturing in the form of ZnO, because of a dilution from the pavement. Consequentially, the authors concluded that tyre can be considered as the major contributor of zinc in road dust.

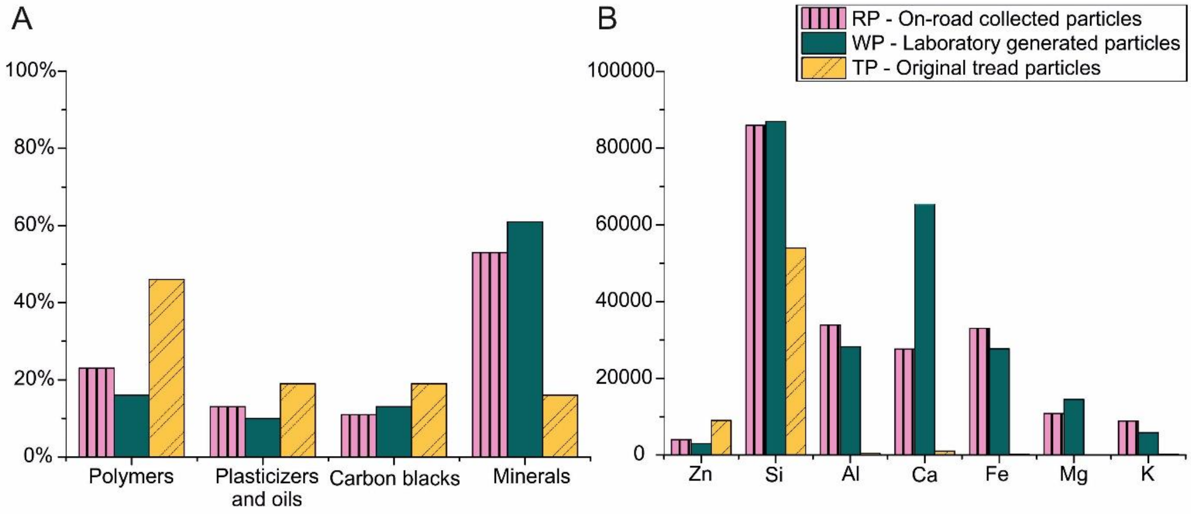

Fig. 2. General composition analysis with values expressed in % weight (A) and analysis of metallic content with values expressed in parts per million (B) of particles from Kreider et al. (2010) for on-road collected particles (RP), laboratory generated particles (WP) and original tread particles (TP).

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

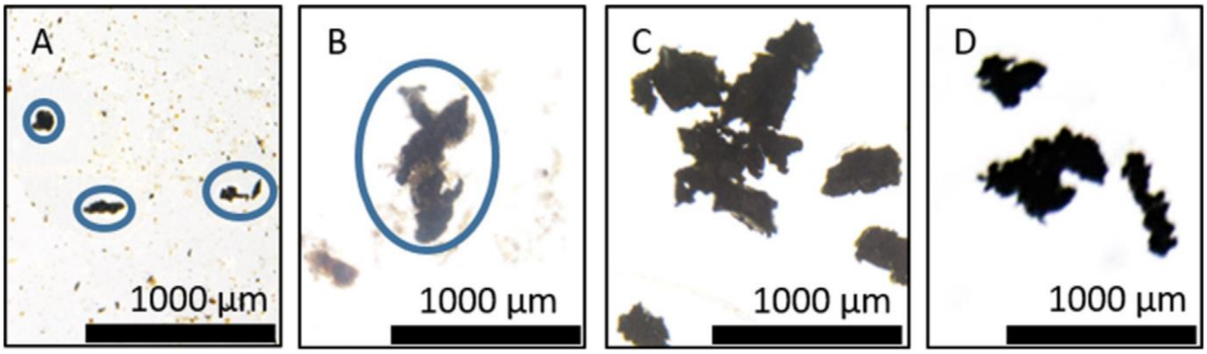

Fig. 3. Diversity of rubber from laboratory production to sampling along a road. A: RP (circled in blue) sampled on gel-tape from a road surface. B: Water sample of RP (circled in blue) collected from a road surface drain. C: Laboratory generated WP by cryogrinding in liquid nitrogen. D: Laboratory generated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

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

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

In conclusion, literature results on tyre wear were found highly variable depending on the type of study performed (road simulator, pin-on-disc or on-road direct measurement) and on the measuring 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)

2.2.3 Emission factors

Most of the studies on brake particulates mentioned above also report an estimation of emission rates for airborne tyre wear particles, for both PM_{10} and $PM_{2.5}$ fractions. The methods used to estimate tyre wear emission factors include derivation from emission inventories, receptor modelling and direct measurement in laboratory experiments using a road simulator. Alternatively, the emission factors can be derived by experimental measurements in wind tunnels (which reproduce a realistic condition) or through road sampling and mobile on-board measurement.

Based on experimental results, several authors proposed models to evaluate the PM_{10} and $PM_{2.5}$ emission factors, such as the formula of Aatmeeyata et al. (2009):

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

where i stands for PM_{10} or $PM_{2.5}$, EF_i is the emission factor (µg km^{-1} $tyre^{-1}$) of i, s (µg m^{-3} kg^{-1}) is the slope of the trend line of the variation of PM_i emission with increasing load and c (µg m^{-3} $tyre^{-1}$) is its intercept, L is the 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-long). Similarly, Wang et al. (2016) found a linear correlation between vehicle load and tyre wear, whereas Salminen (2014) proposed an exponential correlation.

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

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

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

Reference	**Data source**	**Tyre PM_{10} EF (mg km^{-1} veh^{-1})**	**Tyre $PM_{2.5}$ EF (mg km^{-1} 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**

2.3 Road Surface Wear

Road surfaces are typically concrete-based or asphalt-based: the former are made of coarse aggregate, sand and cement (EEA, 2019c), while the latter are mainly composed of mineral aggregate (95%). The remaining 5% is composed of bitumen, which is a highly viscoelastic black blend of hydrocarbons and their derivatives obtained from the fractional distillation of crude oil (Lindgren, 1996). The composition of road mixtures can be further enriched and optimized with additives like polymers (e.g. tyre crumbs) and different types of fillers (e.g. waxes, asbestos, clay, silica etc.), which lead to the modification of the pavement properties (Porto et al., 2019).

2.3.1 Generation

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

2.3.2 Characterization

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

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

Gustafsson and Johansson (2012) completed their study analysing the chemical composition of PM_{10} generated from road surfaces wear. Particles in the coarser fractions were characterized by mineral contents, mostly silicon (Si), calcium (Ca), potassium (K) and iron (Fe). Conversely, the fraction of particles below 1 µm was dominated by sulphur (S) and chlorine (Cl) related to tyre rubber or bitumen. Finally, small quantities of zinc, that is commonly associated with tyre wear, was found in PM_{10}. These evidences corroborated the results from Kreider et al. (2010), who identified aluminium and silicon as the elements present in the highest quantities in RP and WP. As a consequence, since 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 PM_{10} 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 at al. (2005) tested non-studded and studded tyres at 30 km/h, and obtained PM_{10} emission factors of 9 mg km^{-1} veh^{-1} and 40 mg km^{-1} veh^{-1}, respectively. The use of traction sand further increases PM_{10} 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 PM_{10} 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 surface that can be suspended in the atmosphere through traffic or windblown action (Denby et al., 2018). Only particles with a size below 70 µm can be airborne, however, all the deposited dust can be resuspended as larger particles may be ground into finer ones by the vehicle weight.

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

2.4.2 Characterization

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

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

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

2.4.3 Emission factors

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

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

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

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

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

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

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

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

$$EF = 12.35 \cdot RD^{0.81} \qquad \textit{Eq. 6}$$

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

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

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

Reference	**Location**	**PM$_{10}$ EF (mg km^{-1} veh^{-1})**

Candeias et al. (2020)	Viana do Castelo, Portugal	49 (asphalt road) 330 (cobbled 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**

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

Fig. 4 highlights the variability of results obtained in different studies. The most variable results were found for resuspended dust, since environmental and road conditions are significantly influencing the EF estimation. The median value of each NEE sources illustrated in Fig. 4 exceeds the exhaust PM_{10} emission factor set by EURO 6 (5 mg km^{-1} veh^{-1}). This confirms the need for the definition and regulation of emission standards for NEE.

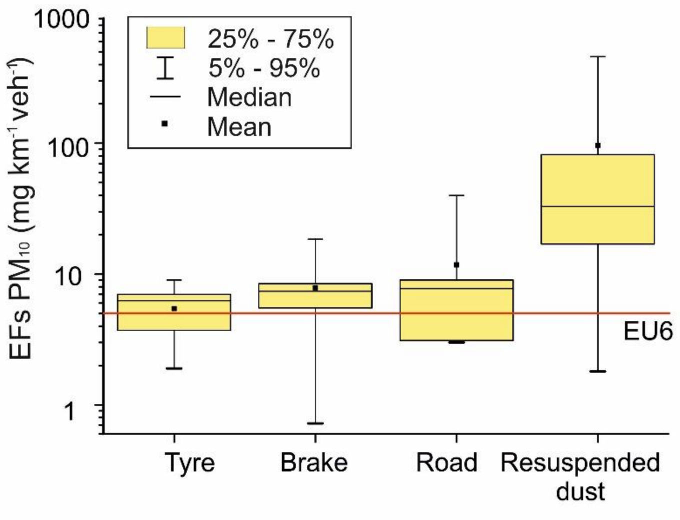

Fig. 4. Mean, median, 25/75 percentiles for box, and 5/95 percentiles for PM_{10} emission factors of NEE sources, based on the literature review reported in Table 3 (brake wear), Table 6 (tyre wear), Table 7 (LDV road wear), and Table 8 (resuspended dust).

2.5 Rail transport non-exhaust emissions

Railways for light trains, subways and trams represent a vital infrastructure for urban areas. At the end of 2017, there were metros in 182 cities in 56 countries, carrying on average a total of 168 million passengers per day (UITP, 2019). Tram and light rail systems (LRT) are instead used in 389 cities around the world. Melbourne is the city with the longest cable train system, with around 250 km of tramway, followed by Saint Petersburg (246 km), Moscow (208 km), Berlin (193 km), Milan (180 km), Vienna (178 km) and Los Angeles (159 km). In Europe only, the total annual ridership for LRT in 2018 was around 10,422 million, which is comparable to the number of passengers served by metro systems and 10 times higher than those traveling by plane (UITP, 2018).

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

Most research on railway PM emissions focused on subways (Cartenì and Cascetta, 2018; Loxham et al., 2013; Loxham and Nieuwenhuijsen, 2019). As an example, several studies (Adams et al., 2001; Pfeifer et al., 1999; Saunders et al., 2019; Seaton et al., 2005; Smith et al., 2020) reported a significantly higher concentration of $PM_{2.5}$ in the London Underground than in other surface means of transport. More specifically, Smith et al. 2020 observed an average $PM_{2.5}$ concentration of 88 $\mu g/m^3$, with peaks of more than 400 $\mu g/m^3$. This value was much higher than the average concentration detected in roadside environments in central London (22 $\mu g/m^3$). A similar result was also reported in Stockholm, where the average PM_{10} and $PM_{2.5}$ concentrations inside an underground station (470 $\mu g/m^3$ and 260 $\mu g/m^3$ respectively) were 5-10 times higher than the corresponding values measured in one of the busiest streets of the city centre. Ryswyk et al (2017) instead conducted a study on the exposure of $PM_{2.5}$, PM_{10}, ultrafine particles and black carbon in the metro system of Toronto, Montreal and Vancouver, Canada. The authors estimated that a typical commute of 70 minutes contributes to 21% of the daily $PM_{2.5}$ exposure in Toronto, 11% in Montreal and 12% in Vancouver. The high concentration of particulate matter in the subway environment was also confirmed by Kim et al. (2008), whose research revealed that the levels of PM_{10} and $PM_{2.5}$ in platforms of the Seoul Metropolitan Subway exceeded the daily acceptable threshold limits regulated by the USEPA. Finally, Martins et al. (2016) assessed the indoor air quality in the Barcelona subway system, where the mean $PM_{2.5}$ concentrations on the subway platforms were found between 1.4 and 5.4 times higher than the one outdoors.

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

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

2.5.1 Generation

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

2.5.2 Characterization

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

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

2.5.3 Emission factors

To the authors' knowledge, there are no legislations or regulations that control railway NEE, and their quantification is even less studied than the NEE from road vehicles. No concentrations and no recommended emission factors and methodologies are available in international inventories such as the EMEP/EEA Emissions Inventory Guidebook (EEA, 2019c). However, a few countries included emission estimates in their national inventories. For instance, estimates of the PM_{10} emission factors for abrasion and wear of contact line, braking systems and tyres on rails (reported in Table 9) are provided by the German railroad company Deutsche Bahn AG (Boettcher et al., 2020). Emissions from other wear sources (e.g. the current collector) are not estimated. In the absence of specific information, $PM_{2.5}$ was assumed to be 50% of PM_{10}, whereas TSP was assumed equal to PM_{10}. The report also suggests that non-exhaust emissions from abrasion and wear exceed 90% of total PM emissions, i.e. the contribution of exhaust emissions from diesel trains is very small.

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

	$PM_{2.5}$ (mg km^{-1})	**PM_{10} (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

3 Mitigating strategies for PM

The pervasive presence of air pollution in urban areas and its associated adverse effects on human health led to an increasing introduction of measures to abate PM concentration levels. Since road traffic was identified as a main contributor to urban air pollution (Pant and Harrison, 2013), current regulations are focusing on reducing not only exhaust emissions, but also non-exhaust emissions from brake, tyre, road surface wear and resuspension of road dust. The different mitigating strategies adopted can be grouped into technological and non-technological measures: the former include technologies applied to improve the emission performance of existing vehicles or to substitute them (e.g. introduction of electric vehicles), the latter involve management policies or remediation measures. At the same time, it is possible to differentiate between prevention or mitigation strategies adopted to reduce PM levels: the former aim at avoiding particle emissions (paving the access to unpaved lots or imposing road traffic restrictions), the latter at removing the already deposited dust (sweeping, street washing, etc…) (Amato et al., 2010). Fig. 5 shows the classification of the existing abatement methods described above.

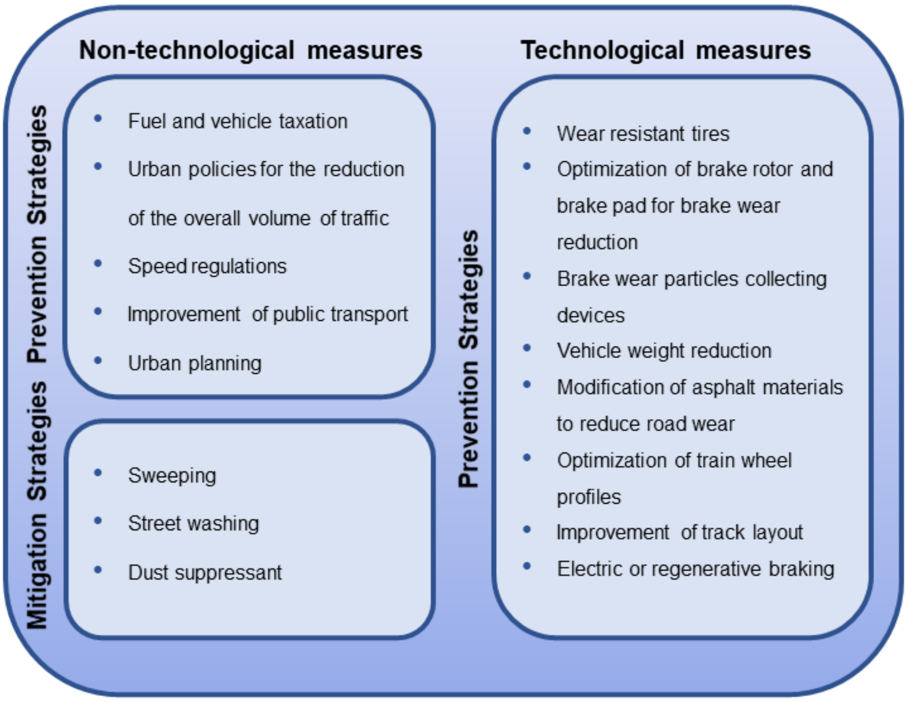

Fig. 5. Classification of strategies to abate PM pollution from non-exhaust emissions.

3.1 Non-technological measures

3.1.1 Prevention strategies

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

As already discussed in Chapter 2.4.3, the imposition of speed limits and the promotion of a smooth driving style reduce braking events and dust resuspension, thus improving passenger safety and air quality (Gustafsson et al., 2008; Querol et al., 2018). For instance, Kwak et al. (2013) observed a significant increase in PM concentrations of both road wear and tyre wear particles during deceleration events. Since both the amount and size of the generated particles strongly depend on the intensity and duration of the braking events, brake wear emissions were found to be influenced also by the driving style (Chapter 2.1.1). Stronger and longer braking events produce higher disk temperatures which, in turn, induce the generation of a large amount of fine and ultrafine particles (Garg et al., 2000; Nosko and Olofsson, 2017; Wahlström et al., 2012).

3.1.2 Mitigation strategies

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

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

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

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

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

3.2 Technological measures

3.2.1 Brake wear

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

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

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

In the last years, several research activities addressed the collection of the particles generated by the brake wear. Rocca Serra (2014) invented an autonomous suction device for drawing off brake wear particles, that consisted in an intake opening (installed close to the pad and the rotor), a collection chamber and an impeller (placed onto the rotor), which drives particles into the intake opening. Fieldhouse and Gelb (2016) developed a brake pad waste collection system composed of a shell that encapsulate the rotor and the calliper. A vented disc connected to the shell generates a positive air flow that leads debris into a filtering system. Combined with magnets, the filters collect particles up to 40 µm and, based on the tests performed, over 92% of brake wear dust was collected and removed. Hascoët and Adamczak 2020 developed TAMIC, an aspiration system composed of a turbine and a high efficiency filter. The device proved a PM_{10} removal efficiency higher than 85% for both particle mass and number.

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

3.2.2 Tyre and road wear

Research for the abatement of tyre and road wear is focused on the improvement of materials that generate the greatest quantity of wear particles, i.e. rubber compounds and asphalts. Over the last years, the increase in traffic speed and vehicle weight load shortened the life of asphalt pavements. Thus, various modifiers and additives – such as polymers, chemical modifiers, extenders, oxidants and antioxidants, hydrocarbons and anti-stripping additives – were used to enhance road performance properties (Porto et al., 2019; Yue et al., 2019). Vieira et al. (2019) suggested the application of a Double Layered Porous Asphalt (DLPA) to abate PM_{10} emissions. DLPAs are composed of a top layer containing fine aggregates and a second layer beneath, that includes coarser aggregates with a high air void ratio (Liu et al., 2016). Thanks to its layered composition, this asphalt is expected to reduce both traffic noise and road wearing. The air quality measurements performed close to a DLPA road highlighted a 52% reduction of PM_{10} concentrations, compared to measurements with non-porous stone mastic asphalt. Other studies proposed the addition of rubber crumbs – derived from tyre grinding or other secondary rubbers sources – to the asphalt mixture to reduce traffic noise, road and tyre wear (Bressi et al., 2019; Frolova et al., 2016). This technological solution not only increased the road durability (which may reduce particle emission due to asphalt wear), but also allowed to reduce tyre wear emissions by 30%-50% compared to conventional concrete pavements.

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

3.2.3 Electric vehicles

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

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

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

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

3.2.4 Rail transport

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

4 Conclusions and future needs

Particulate matter from traffic is recognized as one of the main risk factors for adverse health effects and premature deaths worldwide. In the last decades, different measures were implemented to reduce PM levels, such as the adoption of exhaust emission standards, the development of after-treatment systems for tailpipe emissions and the promotion of electric vehicles. However, PM still represent an air quality issue worldwide, especially in urban areas. This is due to the high incidence of NEE, which are currently estimated to contribute up to 90% of the overall vehicle PM_{10} emissions and up to 85% of $PM_{2.5}$ emissions from road traffic. However, non-exhaust emissions of PM are scarcely addressed by regulation and technology development.

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

From this review emerged the following considerations:

- Resuspended dust is the prevailing NEE source, with PM_{10} emission factors ranging from 5.4 mg km^{-1} veh^{-1} to 330 mg km^{-1} veh^{-1} for cars. Its emission factors are mostly influenced by road type and vehicle weight, whereas vehicle speed was found to contribute to a lesser extent.
- Brake wear PM_{10} emission factors range from 1 mg km^{-1} veh^{-1} to 18.5 mg km^{-1} veh^{-1} and most of the available studies agree on a unimodal mass size distribution centred between 2 µm and 6 µm.
- Tyre wear PM_{10} emission factors vary in a narrower range compared to brake wear, i.e. from 2 mg km^{-1} veh^{-1} to 9 mg km^{-1} veh^{-1}. However, studies on size distributions showed contradictory results, which vary from unimodal mass distributions with a peak at 75 µm to bimodal mass distributions with peaks at 0.3 µm and 4 µm.
- Road surface PM_{10} emission factors range from 3 mg km^{-1} veh^{-1} to 40 mg km^{-1} veh^{-1}, presenting the highest values for HDVs and vehicles with studded tyres. The mass size distributions reveal a peak in the range from 5 µm to 8 µm.
- Railway wear PM_{10} emission factors (from trams, metros and urban trains) are often neglected in emission inventories. However, the wheel-railway friction, the brake wear and, to a lesser extent, the friction of the contact lines are known to release very fine ferruginous particles (unimodal size distribution peaking between 0.28 µm and 0.6 µm), which pose a serious threat to human health at the local scale (i.e. inside vehicle cabins or in underground stations). The quantification of railway emissions and the development of solutions to reduce their impact on urban air quality should therefore be included in NEE studies.
- The orders of magnitude of the EFs reported in this review largely exceed the most recent exhaust PM_{10} emissions standards (i.e., 5 mg km^{-1} veh^{-1} set by EURO 6), and consequently emphasize the need for future research on particulate emissions from traffic to give more prominence to NEE, rather than EE.
- Among all the PM fractions, the finest ones have the highest impact on human health since they are more likely to penetrate the human body and are enriched in redox-active transition metals. However, the contribution of these fractions to the overall PM mass emission is negligible, whereas their presence is extremely relevant in terms of particle number concentration. Consequently, the use of mass-based emission factors is not

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

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

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

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