2. The causes and consequences of non-exhaust emissions from road transport

This chapter describes the sources and processes involved in the generation of non-exhaust emissions, reviews the main vehicle, road, and driving features determining the magnitude of non-exhaust emissions, and reviews the current knowledge on the health impacts of non-exhaust emissions. Non-exhaust emissions are comprised of brake wear, tyre wear, road wear, and road dust resuspension. Considerable uncertainty remains regarding the amount of PM that is emitted by non-exhaust sources in real world driving conditions and how this amount varies with changes in the factors identified above. PM from non-exhaust sources comprise a rising share of PM emissions from road transport. As the global fleet of vehicles becomes newer and the amount of PM from exhaust emissions continues to fall, the vast majority of PM from road transport is expected to come from non-exhaust emissions in future years. Exposure to PM emissions is associated with a variety of adverse health impacts in the short and long term, including an increased risk of cardiovascular, respiratory, and developmental conditions, as well as an increased risk of overall mortality.

2.1. Defining non-exhaust emissions

Emissions of particulate matter (PM) from motor vehicles originate from two main sources: the exhaust from combustion engines and the degradation of vehicle parts and road surfaces. The latter, comprising all airborne particulate emissions generated by vehicle and road wear and the resuspension of road dust, are defined as non-exhaust PM emissions. The proportion of PM emissions from non-exhaust sources has rapidly increased in recent years due to the significant reductions in exhaust emissions over this period, and are now responsible for about 90% of all PM emissions from road traffic (Timmers and Achten, 2016[1]; Rexeis and Hausberger, 2009[2]).

A further distinction can be made between primary and secondary emissions. Whereas primary emissions refer to particles that retain their chemical structure when airborne, secondary emissions refer to particles that, when emitted, react with other chemicals in the atmosphere to form new pollutants. Although most non-exhaust PM emissions are primary, recent findings indicate that they may also contribute to the generation of secondary aerosols due to the degradation of organic compounds found in brake and tyre materials (Kukutschová et al., 2011[3]; Plachá et al., 2017[4]). Particles originating from other sources that have been deposited on road surfaces can be also re-suspended by traffic-induced turbulence.

Following the terminology and hierarchy proposed by Padoan and Amato (2018[5]), non-exhaust PM emissions encompass:

- Direct wear emissions:

  - Brake wear: particles abraded from brake pads and discs that are directly airborne. About half of total brake wear particles are not airborne (Grigoratos and Martini, 2015[6]).
  - Tyre wear: the particles eroded from tyres that are directly airborne.
  - Road wear: particles eroded from road surfaces that are directly airborne.
- Road dust suspension (or resuspension): the particles on (paved) road surfaces that are suspended in the air by vehicle traffic. Road dust can consist of brake, tyre and road wear particles that have been deposited on the road, as well as particles that have migrated to the road from other sources.

The relative proportion of non-exhaust emissions from each source can vary significantly across vehicle types.[1] The classification presented above is based on the emission process, not on the particle type, and is standard practice in emission inventories. The inclusion of PM from resuspension in emission inventorying is the subject of an ongoing debate. Double-counting is a commonly encountered issue in emission inventories and primarily results from using different methodologies to measure the particles emitted from different sources (Pulles and Heslinga, 2007[7]). Some authors claim that, insofar as the particles from direct brake, tyre, and road wear can be deposited on road surfaces and resuspended, the inclusion of these particles as road dust resuspension in emission inventorying implies double-counting.

This report argues that resuspension should be counted in emission inventorying for several reasons. First, the concept of double-counting should not be confounded with the concept of re-emissions. Re-emissions occur at a different time than initial emissions. A brake wear particle is, for example, considered an initial emission when generated and measured at hour t_1, this particle is considered a re-emission when it is deposited on the road surface at hour t_2, and later resuspended and measured again at hour t_3 (where $t_3 > t_2 > t_1$). Although re-emissions may be present in road dust, it also consists of particles that are not re-emissions, that is, that were not originally generated from brake, tyre, or road wear. Examples include particles that have migrated from road shoulders, gutters, or construction sites, and those created by the gradual fragmentation of much larger particles originating from road surfaces and vehicle components.

Double counting becomes problematic in emissions inventorying when the calculation of emission factors for resuspension do not deduct re-emissions. When, for example, an emission inventory combines EMEP/EEA emission factors for direct emissions (i.e. from brake, tyre and road wear) with an emission factor for resuspension that is calculated locally by inverse modelling, i.e. that is calculated without separating out direct wear emissions, this leads to double counting.

Second, recent evidence from PM source apportionment studies demonstrates that resuspension contributes significantly to PM levels even when direct wear emissions are excluded (Amato et al., 2014[8]; Gehrig et al., 2001[9]; Harrison et al., 2012[10]; Lawrence, Sokhi and Ravindra, 2016[11]; Wåhlin, Berkowicz and Palmgren, 2006[12]). Studies based on dispersion modelling demonstrate that including road dust resuspension in emission inventories significantly improves both mass closure and the correlation between simulated and observed PM data (Amato et al., 2016[13]; Denier van der Gon et al., 2010[14]; Schaap et al., 2009[15]).

Another critique of including resuspension emissions as a component of non-exhaust traffic emissions is the fact that ambient PM from resuspension exists even in the absence of vehicle traffic, as wind naturally suspends material on the roads. A variety of experimental evidence suggests, however, that although the wind does lift deposited dust,

it is not responsible for all of the PM emissions from road dust that are measured on roadways. First, HDVs are associated with greater amounts of road dust than LDVs, indicating that vehicle characteristics (and thus vehicles) have some impact on the resuspension of road dust. Second, ambient road dust concentrations are significantly lower on the weekends (Amato et al., 2009[16]; Amato, 2010[17]). Another point of criticism is that resuspension does not depend on traffic itself but on pre-existing dust present on road surfaces (i.e. assuming that with no pollution, there would be no resuspension even with traffic). However, even in the absence of dust on road surfaces, vehicle traffic provokes the fragmentation of very large particles that had not previously been airborne, into smaller particles that can then be suspended.

Given these considerations, including resuspension as source of non-exhaust emissions in emission inventorying is therefore recommended, even if these emissions cannot be easily controlled via regulations regarding vehicle characteristics or via public policies regarding vehicle-kilometres travelled. Urban air quality plans can nevertheless address road dust resuspension through local management measures (see Section 4.1.1).

Emission standards for exhaust particles from motor vehicles are becoming more and more strict worldwide (e.g. EUROx and TIERx) and primary PM emission reduction technologies (e.g. diesel particle filters, continuously regenerating traps) have enabled these new standards to be met. In contrast, non-exhaust PM emissions go largely unregulated. Taken together, these two trends imply that the relative contribution of non-exhaust emissions to PM mass has considerably increased over the past two decades, not only in terms of primary emissions but also in terms of source contributions when secondary PM from vehicle exhaust are considered (Amato et al., 2014[18]; Denier van der Gon et al., 2013[19]; Denier van der Gon et al., 2018[20]).

Non-exhaust emissions have a direct impact on air quality and health not only through their contribution to PM mass, but also via a number of other pollutants, such as ultrafine particles (<0.1 μm in diameter) and heavy metals and metalloids, which are also widely unregulated (Box 2.1). The UN GRPE-PMP (Particle Measurement Program) and the Horizon 2020 EU-funded LOWBRASYS project[2] seek a better understanding of particle number emissions from brake wear, given that combustion-like processes resulting in the formation of ultrafine particles are very different from smaller nanometer-sized wear particles. The European Commission's DG Research unit has also launched a Horizon 2020 project that aims to pave the way for an accurate, reliable, and reproducible test methodology for measuring tyre abrasion rates and the correlation of abrasion rates with PM emissions.

Box 2.1. PM metrics: Mass and number

Atmospheric particles (PM) can be measured in terms of either mass (µg/m3) or number (number of particles/cm^3). Ambient air PM standards are mass-based: PM10 and PM2.5 are defined as the mass concentration of particles with diameters below 10 µm and 2.5 µm, respectively. PM mass concentration is dominated by primary coarse particles (with diameters larger than 1 µm). Particle number concentration is not regulated. It is dominated by particles smaller than 1 µm, and more specifically by ultrafine particles (with diameters smaller than 0.1 µm, UFP).

In contrast to the mass concentration, which is predominantly conservative, particles undergo several processes that modify their number and size, such as new particle formation (nucleation), evaporation, condensation, deposition, and coagulation. Epidemiological studies suggest that negative health effects may be exacerbated with decreasing particle size. Due to their small size, when inhaled, UFP can enter the alveoli and penetrate biological membranes, enabling them to pass into the circulatory system. Once in the circulatory system, these particles can enter all organ systems including the brain and nervous system, as well as traverse the placental barrier. Another recently discovered pathway allows nanoparticles to reach the brain directly from the nose through the olfactory nerve. Toxicological studies suggest that UFP have greater toxicity per mass unit than larger particles and may contribute to the development and progression of various diseases.

Epidemiological evidence for the health effects of UFP is still relatively scarce, mainly due to lack of data. In vitro studies, however, increasingly target these particles and the findings of these studies indicate that smaller particle size is associated with higher toxicity. The hypothesised health effects of UFP include cardiovascular and respiratory morbidity and mortality, local pulmonary and systemic inflammation and oxidative stress, carcinogenesis in several organs, and adverse impacts on the brain and on metabolic processes. A recent meta-analysis suggests short-term associations with inflammatory and cardiovascular changes, which may be at least partly independent of other pollutants. An increasing number of publications however point to hitherto unconsidered specific effects on carcinogenesis (liver, brain, mouth etc), and the onset of diabetes. For the other studied health outcomes, evidence on the independent health effects of UFP remains inconclusive or insufficient.

Source: (Harrison et al., 2018[21]; HEI Review Panel on Ultrafine Particles, 2013[22]; Kettunen et al., 2007[23]; Lanzinger et al., 2016[24]; Meng et al., 2013[25]; Ohlwein et al., 2019[26]; Samoli et al., 2016[27]; Sioutas, Delfino and Singh, 2005[28]; Stafoggia et al., 2017[29]; Tobías et al., 2018[30])

2.1.1. Brake wear

Brake wear PM are generated during the friction between brake pads (linings) and discs (shoes). Not all brake wear debris becomes airborne. It is estimated that approximately 50% of generated brake wear particles become airborne (Garg et al., 2000[31]; Sanders et al., 2003[32]; Barlow et al., 2007[33]; Kukutschová et al., 2011[3]).[3] Particles worn from brake pads and discs contribute almost equally to total PM from braking systems (Hulskotte, Roskam and Denier van der Gon, 2014[34]). While disc brakes typically consist of a rotating disc made of a pearlitic grey cast iron (in some cases made of steel, carbon-carbon, ceramic, or aluminium matrix composites), the composition of brake pads is very heterogeneous. A detailed presentation of the various compositions of brake pads is provided in Box 2.2.

The mass size distribution of brake wear particles (the distribution of particle mass over different categories of particle size) heavily depends on vehicle speed, deceleration and inertia, as well as on the composition of brake materials. Since mechanical wear is the main PM generation process, the mode of the distribution is generally observed to be above 1

µm (Iijima et al., 2007[35]; Iijima et al., 2008[36]; Kukutschová et al., 2011[3]; Sanders et al., 2003[37]). However, due to the high temperatures at the brake/rotor interface and consequent decomposition of brake lining materials, some studies find a mode in the submicrometric region: for example, Garg et al. (2000[38]) found that 33% of the mass of brake wear particles are smaller than 0.1 µm in diameter, varying the average mass median diameters from 0.62 to 2.49 µm depending on the brake pad and temperature. However, it is important to understand whether these conditions are met under typical real-world driving conditions. This is one of the major research topics addressed by the UNECE informal working group on the Particle Measurement Programme (PMP) (UNECE, 2019[39]). Recent PMP findings indicate that 60% of brake wear particles are PM10 and somewhat less than half are PM2.5.

2.1.2. Tyre wear

Tyre wear emissions are generated as a result of the tread abrasion caused by road surfaces. Consequently, it is difficult to measure tyre wear particles separately from road wear particles. As shown by scanning electron microscope (SEM) images and time-of-flight aerosol mass spectrometer studies, the particles formed from the interaction of tyres and pavement appear elongated, with a "sausage-like" shape, and consist of a complex mixture of tread rubber and encrusted mineral particles from pavement (Adachi and Tainosho, 2004[40]; Dall'Osto et al., 2014[41]; Kreider et al., 2010[42]). Tyre wear particles are composed of plasticisers and oils, polymers, carbon blacks and minerals (Kreider et al., 2010[42]), but special attention has been paid to their elemental content (mainly zinc and sulphur) and the abundance of polycyclic aromatic hydrocarbons (PAHs) due to their burden on public health and ecosystems. In Europe, PAHs have been eliminated from tyres since 2009 by the REACH regulation (Chapter 5).

Box 2.2. Chemical composition of brake pads

Brake pads are composed of several materials, including abrasives, binders, fillers, lubricants and reinforcing constituents. A number of chemical compounds are in turn used to produce each of these materials, which makes it difficult to characterise a generic chemical profile of brake pads. According to the combination of ingredients used, brake pads are generally classified as: non-asbestos organic (NAO), mainly composed of organic compounds, mineral fibres and graphite; low metallic (LM), composed of a mixture of metallic components and organic compounds; and semi-metallic (SM), with a metallic (mainly steel and iron) content typically between 30 and 60% by weight.

The chemical composition of pad-worn particles may be different from the bulk composition of the pad due to the physico-chemical transformations occurring during the friction process (e.g. size fractionation, oxidation and volatilisation). However, several components have been commonly identified as possible tracers in literature, mainly iron (Fe), barium (Ba), antimony (Sb), tin (Sn), and copper (Cu), although these elements can also be emitted from other sources, mostly from industrial processes.

The composition of brake pads has varied over time, but in recent years there has been a noticeable reduction in copper and antimony content motivated by the forthcoming implementation of regulations in a number of US states (California, New York, Rhode Island and Washington). There is also an increasing concern about the amount of phenolic resins used in brake pads, which may lead to similar regulations on the use of these constituents in the near future.

Source: (Amato et al., 2009[43]; Gietl et al., 2010[44]; Hulskotte, Roskam and Denier van der Gon, 2014[34]; Kukutschová et al., 2011[3]; Thorpe and Harrison, 2008[45]; Von Uexküll et al., 2005[46])

Concerning tyre wear particle size, less than 1% by volume (which is proportional to the mass) of the particles are less than 10 µm (Kreider et al., 2010[42]). Within PM10, studies conducted on a laboratory road simulator[4] show that more than 60% of tyre wear particles (by mass) are generally between 2.5 and 10 µm (McAtee et al., 2009[47]). However, a bimodal distribution is observed with peaks around 1 µm and between 5 and 8 µm. Several studies demonstrate that the use of studded tyres increases the emissions of tyre-road wear particles by orders of magnitude, and produces particles in the nanometre range from the interaction between the studs and the pavement (Gustafsson et al., 2009[48]).

2.1.3. Road wear

Road wear occurs due to the fragmentation and breakdown of road pavement surfaces as a result of vehicle traffic. Pavements are normally ballast bonded using bitumen, resulting in wear particles composed of minerals dominated by crustal elements but with a significant content of organic carbon (OC). In addition, furnace slag and mixed tyre rubber can be incorporated in road pavements in order to improve their durability and other technical properties.

Road wear is often merged with road dust resuspension in source apportionment studies. The reason for this is that the atmospheric deposition of other mineral particles originating from soil resuspension, traction sanding, building activities and desert dust hampers efforts to measure road wear and road dust separately.

A unique feature of road wear particles is their bitumen content.[5] Bitumen constitutes about 5% of roadside total suspended particles (TSP) mass and bitumen particles have a mean aerodynamic size of about 1 µm (Fauser et al., 2002[49]). Interest in road wear dust particles has been highest in countries where studded tyres are used during winter, as these tyres significantly increase road wear. Traction sand also increases road wear due to the "sandpaper effect" (Kupiainen, Tervahattu and Räisänen, 2003[50]), which results in a higher abrasion of road surfaces due to the interaction with quartz sand particles. In countries where studded tyres are not used, addressing road wear is less of a priority, which means that weaker rocks and less wear-resistant constructions are used. As a result, road wear can nevertheless be an important source of non-exhaust particle pollution in these countries (Gustafsson, 2018[51]).

Road wear emissions are mainly calculated and characterised by means of road simulators, where the contribution of emissions from road dust resuspension can be controlled for and measured separately (see Box 2.3). Gehrig et al. (2010[52]) found that road wear particles had a mass size distribution with a maximum of 6-7 µm in diameter and no particles below 0.5 µm. A number of laboratory tests have also shown that the maximum mass concentration is normally at 5-8 µm, and the total mass of particles below 1 µm is small.

Box 2.3. Road simulators

Three main road simulators have been used for air quality studies. The Model Mobile Load Simulator (MMLS) is a relatively small device simulating approximately one third of the load of the wheels of a light duty vehicle (LDV) at a low speed of 9 km/h.

The Mobile Load Simulator (MLS) is bigger than MMLS and simulates the abrasion processes of the wheels of a full-size heavy-duty vehicle (HDV) at a speed of 25 km/h.

The Swedish Road and Transport Research Institute (VTI) laboratory road simulator consists of an indoor circular track measuring 0.5 metres wide and 16 metres in diameter that can be surfaced with different types of pavement. The machine rotates around a central vertical axis on which six-wheel axles are mounted. The axels can accommodate different types of tyres and simulated speeds can reach up to 70 km/h.

Source: Gehrig et al. (2010[52])

2.1.4. Road dust resuspension

Road dust is a generic description for any form of solid particle deposited on the road surface that can be suspended in the air through traffic-induced turbulence. Road dust emissions contribute significantly to ambient PM10 and PM2.5 levels. Larger particles can also be of concern if they are crushed to generate smaller particles, but since no information is available on the crushing potential, frequency and duration of the fragmentation of larger particles, the first priority of research on road dust resuspension should be to better understand the thoracic fraction, or the fraction of particles the size of PM10 or less.

Road dust can come from a variety of sources, including traffic (exhaust and wear PM deposited on road surfaces), construction and other "dusty" sites, migration from neighbouring environments, deposition from the atmosphere, and the application of road salt and sand. The relative contribution of different sources and the overall chemical composition of road dust likely varies from one site to another, so individual findings in

one environment may not be applicable to others (Amato et al., 2011[53]; Denby, Kupiainen and Gustafsson, 2018[54]).

Road dust emissions strongly depend also on the redistribution of particles, dry and wet losses, and processes affecting the distribution of particle sizes (Denby et al., 2013[55]). The physico-chemical properties of road dust have been studied using different approaches, and targeting different size ranges, yielding a wide variation in documented chemical compositions and size distributions. The chemical profile of road dust for the fraction below 10 µm heavily depends on the sources of these particles, but is typically comprised of minerals (silicon, aluminium, calcium, titanium, strontium among others) with enrichments in antimony, tin, barium, iron, copper and manganese (brake wear), zinc and carbon (elemental and organic), and polycyclic aromatic hydrocarbons, when compared to a general soil dust factor.[6]

In order to avoid double-counting, emission factors used for resuspension should not include the contribution of direct brake, tyre or road wear. However, road dust resuspension should definitively be included in emission inventories to enable a better understanding of the origins of real-world ambient PM concentrations currently observed in large cities and their streets (Denier van der Gon et al., 2018[20]).

The main characteristics of the chemical composition of each category of non-exhaust emissions, as well as the most common approaches used for their measurement are presented in Table 2.1.

2.2. Approaches to measuring non-exhaust emissions

The approaches used to estimate the magnitude of non-exhaust emissions can be grouped in two main categories: i) approaches based on estimates of emissions (e.g. mg/vehicle-km or tons/year); and ii) source apportionment studies based on exposure to air pollutant concentrations (µg/m^3). The most important difference between the two approaches is that source apportionment studies estimate the population exposure at a receptor site, whereas emission estimates analyse the amount of PM emitted at the source, such that the transport and transformation of pollutants are neglected.

This section briefly describes the different methods used in each category in order to provide insight into the reliability of estimates and measurements provided by different approaches, as well as into their limitations.

Table 2.1. Chemical composition of non-exhaust PM and emission measurement

Non-exhaust emissions category	PM10 (% of TSP)	PM2.5 (% of TSP)	Main component (> 1% in mass).*	Common methods/approaches for the estimation of emissions (mg/vehicle-km)
Brake wear	63-98%[1-4]	39-63%[1-4]	**Iron, Copper, Barium, Antimony,** Zinc Aluminium, Chromium, Potassium, Titanium, and Magnesium[9]	Brake dynamometer Pin on disc
Tyre wear	60%[4] 1% of total tread wear[5, 6]	42%[4] 0.4% of total tread wear[5, 6]	**Zinc**, Silicon, Sulfur[10]	Road simulator
Road wear	50%[4]	27%[4]	**Silicon, Calcium**, Potassium and Iron.[11]	Road simulator

Road dust resuspension	2-42%[7,8] of PM<250 µm	1-11%[7,8] of PM<250 µm	Silicon, Calcium, Aluminium, Iron, Potassium, Magnesium, Titanium, Copper, Zinc and Barium[12]	Road dust sampling Kerbside ambient air PM or deposition monitoring

* Most common tracers are in bold.
Note: Total suspended particles (TSP) are PM<50-100 µm.
Data sources: [1]Garg et al. (2000[38]); [2]Sanders et al. (2003[37]); [3]Iijima et al. (2007[35]); [4]Ntziachristos and Boulter (2016[56]); [5]Kreider et al. (2010[42])2; [6] McAtee et al. (2009[47]); [7]Ramirez et al. (2019[57]); [8]Escrig et al. (2011[58]); [9]Grigoratos and Martini (2015[6]); [10]Panko, Kreider and Unice (2018[59]); [11]Gustafsson (2018[51]); [12]Amato et al. (2009[43]).

2.2.1. Emission estimates

This family of approaches focuses on the direct measurement of vehicle emissions. The main advantage of this approach is that it provides highly accurate estimates of primary emissions. The main limitation of these approaches, however, is that they do not address population exposure. A second important limitation of emission estimates is that they do not consider secondary aerosols, which are generated in the atmosphere from gaseous precursors such as nitrogen and sulphur oxides (NOx and SOx), ammonia (NH_3) and volatile organic compounds (VOCs) (see Box 2.4). Although secondary aerosols might not be so relevant for non-exhaust processes, they are relevant for exhaust emissions. As such, they should be considered when non-exhaust emissions are evaluated *relative to* total traffic emissions or emissions from all sources. Secondary aerosols can represent an important share of PM. For example, Amato et al. (2016[60]) estimated that 37-82% of PM2.5 and 40-71% of PM10 were secondary particles in five European cities.

Within this category, there are two main types of studies, those focusing on the measurement of emission factors and those concentrating on the compilation pf emission inventories.

Box 2.4. Primary and secondary aerosols

Understanding pollution from motor vehicles requires a knowledge of both primary PM and gas emissions and of how these pollutants interact and age in the atmosphere. While primary non-exhaust PM is emitted directly in solid or liquid form, secondary PM are formed in the atmosphere by the physico-chemical reaction of gaseous precursors, emitted from vehicles and other sources. The main precursors are SOx, NOx, NH3 and volatile organic compounds. The gas-to-particle conversion is not linear: it depends on a complex set of interactions between gas concentrations, free radicals, air temperature and humidity.

Secondary PM from road traffic are mainly due to exhaust emissions, namely NOx, VOCs and SO2 in countries with high sulphur content in diesel fuel. NOx and SOx interact mainly with ammonia (from agriculture) – forming ammonium nitrate and sulphate – as well as with sea salt and mineral dust cations (mainly sodium and calcium) depending on the local conditions. Secondary nitrate and sulphate salts are named secondary inorganic aerosols (SIA), while secondary organic aerosols (SOA) denote particles formed from volatile organic compounds.

Source: Platt et al., (2017[61])

Emission factors

Emission factors (EFs) measure the mass of PM emitted per unit of activity, e.g. per vehicle-km, from different traffic sources. Studies in this category estimate emission factors for non-exhaust sources in different settings. The main limitation of this group of studies is the representativeness of the retrieved EFs for other environments. Two main approaches are followed within this group: inverse modelling and simulation methods.

Inverse modelling

Studies based on real-world PM measurements, such as experimental measurements next to roads (Amato et al., 2010[62]; Amato et al., 2012[63]; Amato et al., 2016[13]; Bukowiecki et al., 2009[64]; Bukowiecki et al., 2010[65]; Escrig et al., 2011[58]; Gillies et al., 2005[66]; Lawrence, Sokhi and Ravindra, 2016[67]), should be preferred for two reasons. First, they provide a robust average of the PM emitted by circulating traffic, which implies higher representativeness than simulator studies. Second, EFs from different traffic sources, i.e. exhaust, and each non-exhaust source, can be sometimes obtained simultaneously, thereby allowing the evaluation of the relative importance of each source. Measurements can be carried out for PM (together with NOx) at a given height, or for deposited dust across a vertical profile (several heights). Studies focusing on road dust EFs should take care to avoid double counting direct wear emissions.

Road dust emissions are often calculated based on the AP-42 model (U.S. Environmental Protection Agency, 2011[68]).[7] The mathematical formula proposed for the model entails a large variation (up to two-orders of magnitude) of possible observed EFs for a given predicted value. For example, when the predicted EF is 0.5 g/vehicle-km, the observed EF varies from 0.1 to 10 g/vehicle-km (Venkatram, 2000[69]). This variation has led to some criticism for the broader applicability of the model (Kantamaneni et al., 1996[70]; Venkatram, 2000[69]).

For a local emission inventory, it is generally recommendable to experimentally derive EFs by carrying out specific local studies, such as those based on inverse modelling.

Simulator studies

Measurements in these studies are based on a single vehicle or simulator, such as a dynamometer, pin-on-disc or road simulator. While such measurements are useful to investigate the impact of factors influencing the magnitude of emissions, including vehicle mass, speed, and type of tyre and brake, they lack generalisability and are not suitable for direct emission comparisons between different traffic sources.

Emission inventories

Emission inventories compile total emissions in a given geographical domain (e.g. country) using a bottom-up approach. To this end, total activity underpinning each emission source is multiplied by an emission factor (California Air Resources Board, 2019[71]; Centre on Emission Inventories and Projections, 2019[72]; Pachón et al., 2018[73]). Emission factors are generally extracted from the literature and their applicability to a given area of study is not always guaranteed. Emission inventories have the additional limitation that estimates of PM shares depend on the sources included in the inventory. In this context, it is important that emission inventories include resuspension. However, care should be taken to ensure that the EFs used for resuspension do not include direct wear emissions in order to avoid double counting.

Information from emission inventories is not necessarily indicative of population exposure, as e.g. power plants and industries are usually located far from most populated areas.

2.2.2. Source apportionment

This category is the most frequently used, as it reflects concentrations and takes into account secondary aerosols. The latter enables better estimates of relative contribution to PM10 and PM2.5. Within this category, three main measurement approaches can be distinguished:

Standard

The ISO/TS 20593:2017 standard specifies a method for the determination of the airborne concentration (µg/m3), mass concentration (µg/g) and mass fraction (%) of ambient PM from tyre and road wear. ISO/TS 20593:2017 establishes standard practices for the collection of air samples, the generation of pyrolysis fragments from the sample, and the measurement of the generated polymer fragments. The quantified polymer mass is used to calculate the fraction of tyre tread in PM and the concentration of tyre tread in the air. These quantities are expressed on a tyre and road wear particle (TRWP) basis, which encompasses the mass of tyre tread and mass of road wear encrustations, and can also be expressed on a tyre rubber polymer or tyre tread basis.

Receptor models

These models apportion the measured mass of PM at a given site to its emission sources by solving a mass balance equation between the observed PM species concentrations and the predicted ones, expressed as sum of contributions from different sources. Receptor models require a large number of observations (more than 100) of PM component concentrations (PM speciation data), including concentrations of major and trace components.

These models have the advantage of providing information derived from real-world measurements, including the estimation of uncertainty surrounding the model's output. However, it is difficult to separate the contribution of different non-exhaust sources using these models due to a lack of unique tracers. They are widely used for source apportionment at local and regional scales all over the world. In the past decade, the number of scientific publications and technical reports using this method has steadily increased and tools with improved capabilities are in constant development (Belis et al., 2014[74]).

Within receptor models, the most common tool is Positive Matrix Factorization (PMF). PMF offers the advantage of obtaining the chemical profiles of the sources as output, as opposed to Chemical Mass Balance (CMB) that requires them as input information, additional to the chemical composition of ambient air PM.

Dispersion or Chemistry Transport models (CTM)

Dispersion models combine emission inventories, information about weather conditions, and the characteristics of dispersion processes and chemical reactions to simulate PM concentrations at a given receptor site or within a given area. Although dispersion models have been used extensively for PM simulation, their application to source apportionment studies has been limited due to their high computational demand. As a result, only a few such studies are available.

The advantage of receptor models is that they allow apportioning the total (measured) PM concentrations, while dispersion models can only apportion the modelled fraction. This fraction can be significantly lower – likely by 50% in cities – due to the lack of specific sources in the emission inventory and/or physico-chemical reactions in the atmosphere. On the other hand, receptor models are more limited in the number of identifiable sources than dispersion models: the latter can quantify the contribution of all sources included in the inventory. Therefore, attention should be paid when labelling sources: for example, a "road dust" source should always be combined with a "direct wear emission" one; otherwise a generic "non-exhaust" label should be created. Another disadvantage of receptor models is that most secondary inorganic aerosols are identified as a single source, i.e. not apportioned to their precursors' sources.

Table 2.2. Main methods to estimate the magnitude of non-exhaust emissions

	Emission estimates			**Source apportionment**		
Model type (measurement unit)	Emission factors (mg/vehicle-km)		Emission inventories (tonnes/year)	Receptor models (µg/m^3)		Dispersion models (µg/m^3)
	Inverse modelling	Simulator studies		PMF	CMB	
Principle	EFs are estimated by fitting simplified models to observations (least squares fit).	EFs are estimated under specific conditions (e.g. single vehicle, brake/tyre system).	Total emissions are calculated by multiplying EF by activity data (e.g. traffic volume)	A mass balance equation is solved between the observed concentration of PM components at a given receptor and the sum of contributions from different sources.		A "labelling" technique is applied to estimated emissions, allowing to estimate contributions on a given receptor.
Usage	Medium	Medium	High	High	Medium	Low
Type of data used	PM and NOx data measured at kerbside; Vertical deposition profiles in case of resuspension.	Vehicle and driving characteristics (e.g. weight, speed)	EFs and activity data for a given geographical domain	PM speciation data with major and trace components. For PMF a large number of samples is needed (>100)		Emission inventory, meteo fields and a CTM model. "Labelling" tool; High computing demand.

Advantages	**Provides robust average of real-world conditions**; Allows for direct comparison among different traffic sources.	Allows for an investigation of the impact of vehicle/driving characteristics on different emission sources.	**Allows for the evaluation of a large number of sources**; Allows for projections.	**Apportions total measured PM mass**		**Secondary aerosols are apportioned Contributions of all inventoried sources can be quantified;** Allows for projections
				Reflects population exposure; Includes secondary particles		
Limitations	**Does not reflect population exposure; Neglects secondary particles**			**Reduced number of sources identified (collinearity), e.g. it merges road wear and road dust resuspension.**		**Only the modelled fraction of PM is apportioned**
	EFs for road dust should be calculated minimising double-counting of direct wear emissions.		**Estimated share of PM can be biased in the absence of important sources (e.g. resuspension)**	Importance of factor interpretation, in order to avoid double-counting in meta-analyses.	**Requires representative chemical source profiles**	
	Applicability to other environments may be limited	**Limited representativeness of real-world conditions**	Relies heavily on the quality and representativeness of EFs	Very limited apportionment of secondary aerosols		
		Do not allow simultaneous comparison with other non-exhaust sources.	Allows for time series analysis			

Note: Main advantages and limitations are highlighted in bold.

The following section provides a brief overview of the two most common types of studies described in Table 2.2, namely "Emission Inventories" and "Source Apportionment" with the aim of evaluating the importance of non-exhaust emissions, mostly when compared to exhaust emissions on a global perspective.

2.3. The growing importance of non-exhaust emissions for air pollution

2.3.1. Emission inventories

Emission inventories provide important evidence of the growing importance of non-exhaust sources for air pollution. However, this evidence should be considered in light of the limitations associated with emissions inventories discussed in the previous section. Specifically, emission inventories: i) do not reflect population exposure well, as inventories are summed over large areas and include sources located far away from populated areas; ii) do not consider secondary aerosols, which can be significantly increased by exhaust emissions; iii) often discard road dust resuspension, which is the dominant part of non-exhaust emissions; and iv) rely on the quality and representativeness of the emission factors used. The last limitation is also crucial when analysing time trends, since a change in the estimation method of an emission factor may create a break in the series and affect intertemporal analyses.

Keeping these considerations in mind, emission inventories at different administrative levels were explored, prioritising those that enabled a direct comparison of exhaust with non-exhaust emissions from road transport. Inventories providing future projections of

different emission categories were of particular interest for the study. Only a few emission inventories met these criteria.

Europe

Under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) and the EU National Emission Ceilings Directive (2016/2284/EU), European Union Member States, EFTA countries and Turkey report their national emissions according to the following categories: passenger cars, light duty vehicles (LDVs), heavy duty vehicles (HDVs) and busses, mopeds and motorcycles, gasoline evaporation, automobile tyre and brake wear, automobile road abrasion (i.e. road wear).

Traffic-induced resuspension of deposited road dust is not included in the European emission inventory, at least not as a mandatory category. Ntziachristos and Boulter (2016, pp. 3,10[56]) justify the exclusion of resuspension from the European emission inventory guidebook as follows: "The focus is on (…) particles emitted directly as a result of the wear of surfaces —and not those resulting from the resuspension of previously deposited matter. (….). Due to the open discussion with regard to the definition of resuspension as a primary source, and the uncertainty in the methods used for the estimation of its effect, no methodology to estimate PM concentrations from resuspension is provided". However, some countries already consider resuspension in their national inventories, such as the United Kingdom, which includes it in a "natural sources" category.8

The growing importance of wear sources relative to exhaust emissions in the EU-28 is illustrated in Figure 2.1, which draws on PM10 data for the period 2000-2014. While a clear downward trend is observed for exhaust emissions due to the implementation of EUROx directives, the sum of reported wear emissions is quite stable (see the dark grey and dark blue bars at the bottom of the columns). The same pattern is observed for PM2.5, although the percentage of wear emissions in total road transport is considerably lower than that for PM10.

Kousoulidou et al. (2008[75]) projected the evolution of non-exhaust PM emissions in European urban environments, finding that the share of non-exhaust emissions to total PM2.5 is on the order of 77% for gasoline PCs, 12% for HDVs and 8% for diesel PCs. The high share of non-exhaust PM2.5 for gasoline PCs in particular is an important finding, as these emissions are not always estimated in studies dealing with air-quality targets, as exhaust emissions have the primary focus.

To put it into perspective, gasoline PC non-exhaust PM2.5 emissions are roughly twice as high as the exhaust PM2.5 of diesel PC at the Euro 5 level. The increasing share of non-exhaust PM with respect to total PM has significant implications for the assessment of the effectiveness of emission control measures in the attainment of air-quality standards. Specific measures for PM control introduce the widespread application of diesel particle filters (DPFs) for Euro 5 and Euro 6 diesel PCs and Euro VI HDVs. According to the findings of Kousoulidou et al. there is clear evidence that non-exhaust sources become increasingly important and that vehicle categories previously not considered as key sources of PM emissions, such as gasoline passenger cars, now need to be taken into account in the total emissions.

The contribution of exhaust emissions may be somewhat underestimated in the inventory, as the emission factors underlying the estimation of exhaust emissions are based on measurements according to type-approval tests. Under real-world driving conditions, PM emissions from vehicle exhaust have been shown to be higher. As a result, the share of wear emissions as a part of total emissions reported is likely to be an upper bound.

However, keeping in mind these limitations, and also the fact that resuspension is not included in the analysis, Figure 2.2 shows a break point for PM10 in 2014, when PM from wear emissions were already larger (51%) than PM from primary exhaust emissions (49%) at the EU-28 level. For PM2.5 this point has not been reached yet; wear represented 34% of total emissions from road transport in EU-28 in 2014.

In terms of percentages to total *primary* emissions (secondary PM is not considered) from all sources, wear emissions showed a clear increase since 2000, reaching a 5% share of total PM10 and 4% of total PM2.5 in 2014 (Figure 2.2). Exhaust emissions represented a similar share of primary PM10, and a share of primary PM2.5 almost double that of wear (8%) in that year.

In the UK, vehicle exhaust emissions are already estimated to be a smaller source than non-exhaust PM and expected to be less than 10% of total road transport PM by 2030. As other emission sources of PM are addressed, it is estimated that the non-exhaust component will increase in importance, growing from less than 8% of national emissions in 2017 to 10% in 2030.

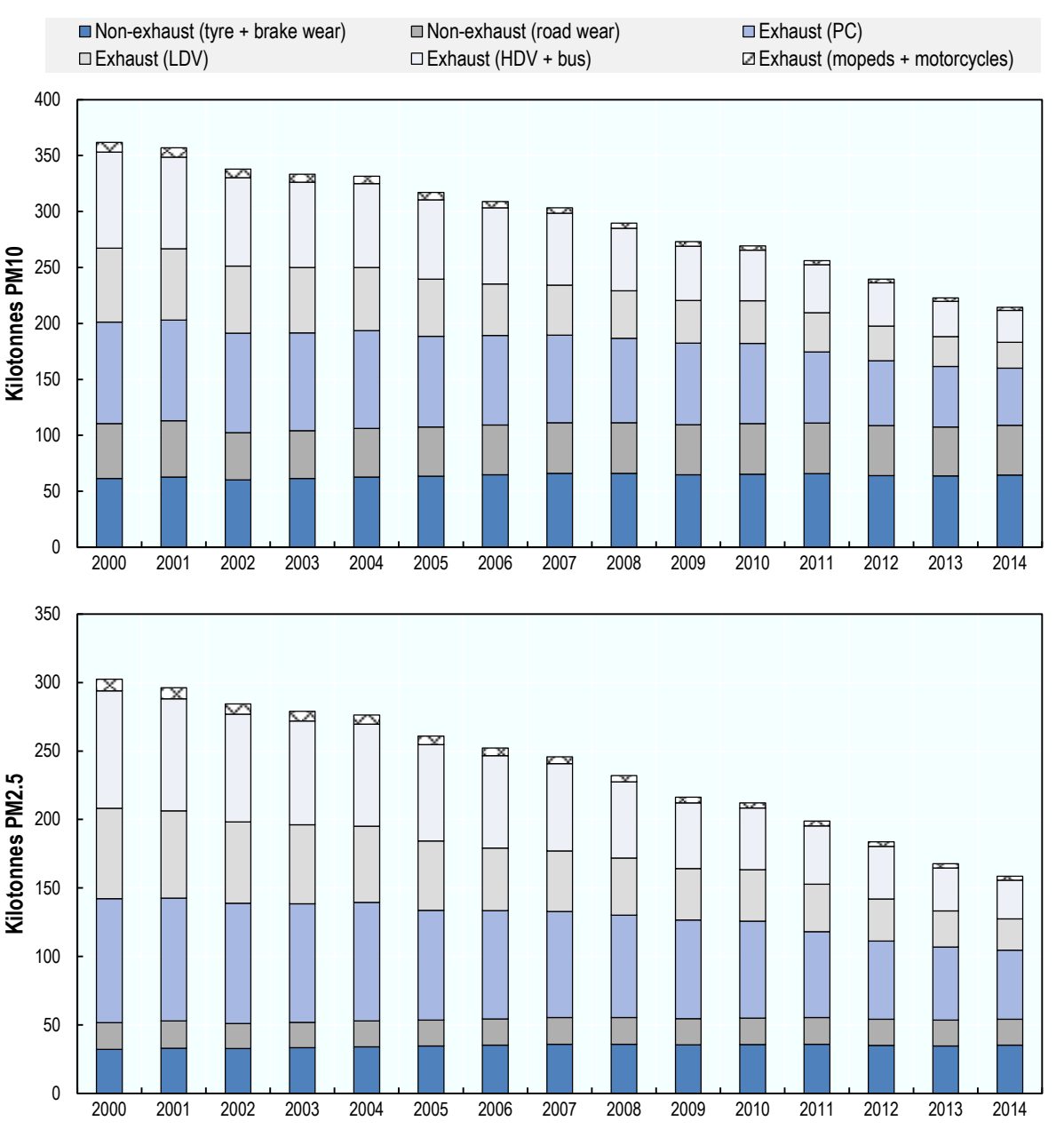
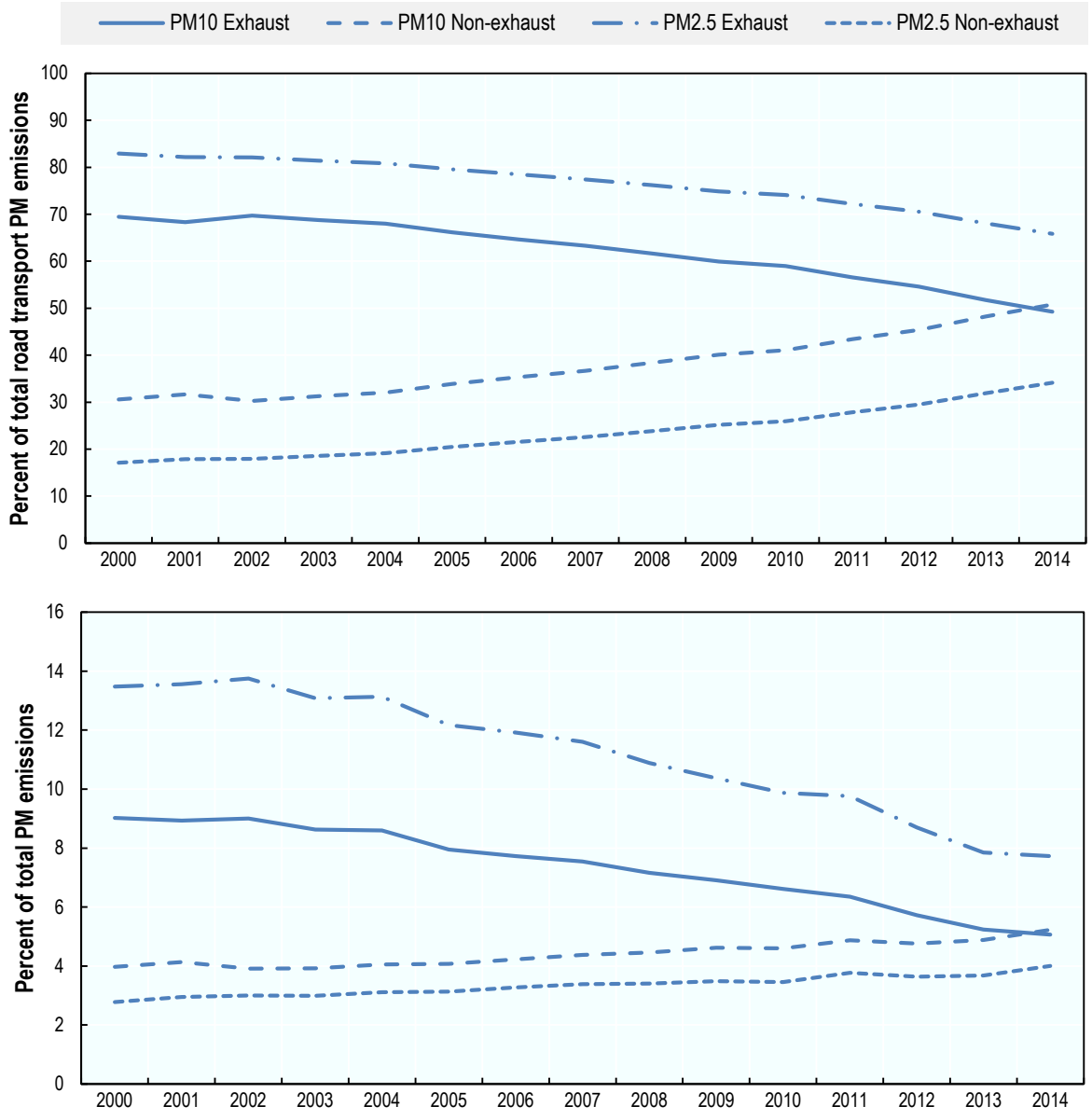

Non-exhaust (tyre + brake wear)
Non-exhaust (road wear)
Exhaust (PC)
Exhaust (LDV)
Exhaust (HDV + bus)
Exhaust (mopeds + motorcycles)
Kilotonnes PM10
400
350
300
250
200
150
100
50
0
2000
2001
2002
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
Kilotonnes PM2.5
350
300
250
200
150
100
50
0
2000
2001
2002
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014

Figure 2.2. Exhaust vs. non-exhaust wear PM emissions

Percentage of total PM emissions from road transport (top) and total PM emissions (bottom)



Source: Centre on Emissions Inventories and Projections (2019)

United States

In the United States, the National Emission Inventory (NEI) provides aggregate emission data for 3 years (2008, 2011 and 2014) (U.S. Environmental Protection Agency, 2019[76]).[9] While road dust emissions are clearly identified as "Paved road dust", brake wear and tyre wear emissions are merged in the "on-road mobile" category. Road dust emissions are calculated based on the AP-42 model (U.S. Environmental Protection Agency, 2011[68]), which, as mentioned before, has certain limitations. Taking into consideration these limitations, emissions from road dust represented at least 74-76% and 51-58% of PM10

and PM2.5 emissions from road traffic respectively in the period 2008-2014 (Table 2.3). In absolute levels, emissions show a slight decrease of around 10% in 6 years. With respect to total sources, road dust emissions represented at least 5% of PM10 and 4% of PM2.5, while mobile emissions (including brake and tyre wear) less than 2% and 3-4% respectively (Table 2.3).

The California Air Resources Board (CARB) website provides emission data on a 5-year basis from 2000 and includes projections until 2035 (California Air Resources Board, 2019[71]). A CARB project is ongoing to update the emission factors, as they are considered dated. However, with the information available today, brake wear and tyre wear are also included in the "on-road mobile" category, but they are specifically labelled and can be extracted; road dust resuspension is labelled as "paved roads". The sum of non-exhaust emissions can thus be calculated by subtracting brake and tyre wear from the total "on-road mobile" category and adding the resulting amount to that of the "paved road" category. It is important to note that road wear is not identified as a separate category in the inventory.

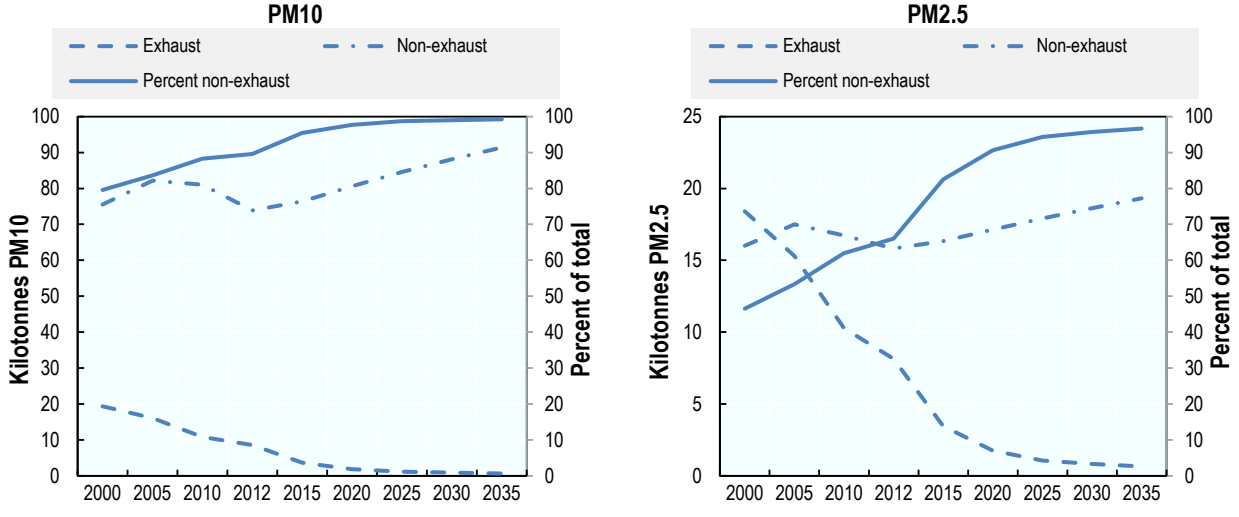
Table 2.3. US-wide estimates for road dust vs. on-road mobile emissions

		Kilotonnes		% of road traffic		% of total	
PM10		Road dust	Mobile[1]	Road dust	Mobile[1]	Road dust	Mobile[1]
	2008	1 037.320	331.899	76	24	5	2
	2011	1 047.690	370.825	74	26	5	2
	2014	944.948	304.269	76	24	5	2
PM2.5							
	2008	259.330	252.603	51	49	4	4
	2011	261.922	197.527	57	43	4	3
	2014	229.466	163.092	58	42	4	3

Note: [1] Includes brake and tyre wear emissions.
Data source: (U.S. Environmental Protection Agency, 2019[76]).

Figure 2.3. Exhaust vs. non-exhaust emissions of PM10 and PM2.5, California, 2000-2035

Non-exhaust PM10 and PM2.5 emissions from road transport in kilotonnes/year and percentage of non-exhaust PM emissions from road transport



Source: California Air Resources Board (2019[71]).

In contrast with the European case, the sum of non-exhaust emissions represented in 2015 already 95% of total primary PM10 from road traffic. The large difference between the contribution of non-exhaust emissions to primary PM10 from traffic in Europe and California can be attributed to two reasons: first, the inclusion of resuspension in the Californian inventory leads to much higher estimates of non-exhaust emissions; second, the amount of exhaust emissions per vehicle kilometre is lower in California due to the very low penetration of diesel light duty vehicles (their share to the total stock of light duty vehicles is around 0.5%.

Besides a decrease in 2012 due to the change of the methodological approach,[10] a clearly increasing trend can be observed for non-exhaust emissions, due to an increase in vehicle kilometres travelled. In contrast, exhaust emissions keep decreasing until 2035. Emission projections show an increase of non-exhaust emissions to up to 98% of total primary PM10 emissions from road traffic and 15% of total primary PM10 emissions already from 2020. The share then remains quite stable until 2035.

With respect to PM2.5, non-exhaust emissions represented already from 2015 more than 80% of road traffic emissions. Their share is projected to increase to 97% in 2030 and then remain relatively stable until 2035. Their share to total PM2.5 emissions is projected to be around 13% in 2035. Exhaust emissions in 2035 are projected to represent about 3% and 1% of road traffic and total PM2.5 emissions respectively.

Latin America

In Latin America, non-exhaust emissions were found to be included only in three local (municipal or metropolitan level) emission inventories: Mexico D.F. (Metropolitan area), Bogotá (city) and Santiago de Chile (metropolitan region).

Mexico

The emission inventory of the metropolitan area of Mexico D.F. is reported every 2 years (Secretaria del Medio Ambiente de la Ciudad de Mexico, 2018[77]). Following the MOVES software, it distinguishes between mobile sources (including exhaust, brake and tyre wear) and road dust resuspension. Road wear is not considered. Similarly to the US National Emission Inventory, it does not distinguish the contribution of brake and tyre wear from the contribution of other mobile sources.

Intertemporal analysis of emissions does not point to a clear trend, probably due to methodological changes which are not always reported. Two periods can be distinguished: from 2002 to 2008, emissions from road dust resuspension were lower than emissions from mobile sources, but they were increasing over time. From 2010 to 2016, emissions from road dust resuspension remain quite stable, while those from mobile sources increase (sharply from 2014). The authors claim that these drastic changes are due to modifications in the methodology used to estimate emissions (Secretaria del Medio Ambiente de la Ciudad de Mexico, 2018[77]), which eventually hampers intertemporal analysis. The most recent (2016) scenario highlights the current importance of road dust emissions, which represent 35% of road traffic emissions of PM10 (21% of PM2.5) and 16% of total emissions of PM10 (9% for PM2.5).

Bogota

For the city of Bogota in Colombia, (Pachón et al., 2018[73]) and (Pérez-Peña et al., 2017[78]) have published PM10 and PM2.5 emission inventories using the AP-42 method

(U.S. Environmental Protection Agency, 2011[68]) for road dust, the COPERT IV model for brake and tyre wear, and the MOVES model for motor exhaust. They found road dust resuspension to be the dominant source of total emissions of PM10 (46%) and PM2.5 (56%), even after correcting emissions for mitigation due to precipitation. Brake and tyre wear represented 1.4% of total PM10 emissions and 0.8% of PM2.5 ones, while motor exhaust 3% and 11%, respectively. The authors concluded that road dust emissions were likely overestimated due to the low applicability of the AP-42 method. Further research is underway to estimate real-world emission factors for road dust and apply an adjustment factor that considers the effects of land use, for example.

Santiago

In Santiago de Chile, the Regional Government published an inventory with projections for 2010, using the MODEM model which separates road dust resuspension from mobile sources. The mobile category is separated between urban roads and highways. While urban mobile sources are further broken down to engine exhaust, brake and tyre wear, this breakdown does not exist for highway sources. Brake and tyre wear are not considered in PM2.5 emissions. Road wear is not considered at all. Similarly to the case of Bogota, the inventory revealed a dominant role of non-exhaust emissions, in the area of 54% of PM10 and 28% of PM2.5 total emissions. Mobile sources (including brake and tyre wear in highways) represented only 6% of total PM10 emissions and 21% of total PM2.5 emissions (DICTUC, 2007[79]).

Summarizing, emission inventories worldwide allow concluding that already in 2014 non-exhaust emissions represented at least 50% of total traffic emissions (primary PM) and 5% of total PM10 emissions, even excluding resuspension. Regarding PM2.5, the corresponding shares decrease to 34% and 4% respectively. In California, where resuspension is taken into account in emission inventories, non-exhaust emissions were found to represent 95% of primary traffic PM10 emissions and 15% of PM10 emissions from all sources (for 2015). Estimates are somewhat lower for PM2.5, but still very significant: non-exhaust emissions account for 82% of primary PM2.5 caused by road traffic and 12% of primary total PM2.5. However, the quality of such estimates is directly linked to the representativeness of emission factors used for resuspension, which may be questionable for the AP-42 model. The same problem applies to the local inventories in Latin America, where road dust contributions may be overestimated due to the lack of reliable emission factors.

2.3.2. Source apportionment studies

When compared to emission inventories, source apportionment studies have the advantage of reflecting population exposure, since they are based on measured (or simulated) concentrations at receptors. Another important advantage is that they consider secondary PM, so that estimates of the share of non-exhaust emissions in traffic PM, or total PM are much more comprehensive. However, caution should be taken when comparing results from different studies: a miss-interpretation of factors can lead to double counting. Padoan and Amato (2018[5]) reviewed about 100 peer-reviewed articles providing more than 250 estimates of non-exhaust sources contributions (µg/m3) worldwide. There is a clearly increasing trend in the number of articles per year from 2000 onwards.

The most common method used in the reviewed studies is receptor modelling (84%), dominated by Positive Matrix Factorization (PMF),[11] which offers multiple advantages but cannot easily separate road wear from road dust resuspension (Table 2.2), while only 10% of studies were performed by means of CTM modelling. The remaining studies used other techniques or measures different from PM mass.

Shares of the relative contribution of each non-exhaust emission source to PM10 and PM2.5 concentrations are listed in Table 2.4, distinguishing between urban background and traffic sites. Road dust was identified much more frequently than wear sources, which can be due to several reasons. If contributions from wear sources are merged under contributions from another source (e.g. road dust or exhaust), summing over cells in a row of Table 2.4 would lead to double-counting. If wear contributions are excluded because they are too small to be quantified by the receptor model, no such risk for double counting exists.

Considering this limitation, Table 2.4 shows that PM10 contribution ranges for road dust resuspension for urban background – places in urban areas where levels are representative of the exposure of the general urban population – are wide due to the important influence of local factors, such as microclimate and road conditions. The contribution of road dust is the highest among all traffic sources (median of 21%), followed by exhaust emissions (16%) and brake and tyre wear (4% each). For urban background PM2.5, exhaust emissions clearly dominate (median of 22%), followed by road dust resuspension (7%), and brake and tyre wear (5%).

Table 2.4. Percent of non-exhaust and exhaust emissions contributions to ambient PM10 and PM2.5: Estimates from source apportionment studies

	Brake wear	Tyre wear	Road dust resuspension /Road wear	Exhaust emissions
		Background sites		
PM10 (observations)	7*	5*	50	59
Range	3-6	2-6	6-59	2-64
Median	4	4	21	16
Mean	4	4	22	20
PM2.5 (observations)	3**	3**	59	60
Range	5-9	3-5	1-31	3-57
Median	5	5	7	22
Mean	7	5	9	24
		Traffic sites		
PM10 (observations)	4	7	11	13
Range	5-20	2-8	11-76	13-36
Median	7	6	23	20
Mean	8	5	29	21
PM2.5 (observations)	-	4***	6	9
Range	-	0-1	5-31	12-62
Median	-	0.1	18	26
Mean	-	0.4	17	30

Note: * A mixed brake/tyre wear component was found in 3 studies; ** A mixed brake/tyre wear component was found in 2 studies, *** A mixed brake/tyre wear component was found in 1 study. Ranges for PM10 and PM2.5 are not derived from the same number of observations. For exhaust emissions, only the studies identifying also non-exhaust emissions are considered.

The number of studies decreases significantly at traffic sites, but the overall picture does not change significantly except for the higher contributions, as expected. Road dust resuspension is still the most important source (median of 23%) followed by exhaust (20%), brake wear (7%) and tyre wear (6%). Regarding PM2.5, street level contributions mainly result from exhaust emissions (26%) and road dust resuspension (18%). Negligible contributions were found for tyre wear (0.1%), while no data on brake wear are available,

possibly due to the specific local driving conditions at these sites. If brakes do not face severe stops, their temperature does not increase above 170 C, thus the emitted PM is mostly coarse (Alemani et al., 2015[80]).

The relatively wide ranges observed in Table 2.4 for the contribution of road dust resuspension suggests an important impact of weather conditions and other local features. In order to explore this hypothesis further, contributions are presented for selected countries with different climate conditions in Table 2.5. Road dust contributions are highest during spring months in Sweden, where the use of studded tyres is the main reason behind a median contribution of 74% of kerbside PM10. On an annual basis, mean contributions go down to 23%, which is also the maximum observed in Europe. The median contribution found by studies in Spain is 17% of PM10, and by studies in central European countries 11%. On an annual basis, emissions from road dust are highest in India and China, where median contributions reach 36% and 31% respectively. For the Unites States, only PM2.5 estimates are available and reach a 6% median contribution, which is lower than China, India and Spain.

Recent analyses of trends in atmospheric concentrations of pollutants reported additional evidence of the increasing importance of non-exhaust emissions. Masri, Kang and Koutrakis (2015[81]) found an increase of the share of coarse particles in PM10, as PM2.5 have decreased at higher rates than PM10. Likewise, PM2.5 have declined more rapidly than coarse PM over the past decade at various European sites (Barmpadimos et al., 2012[82]). A similar pattern has also been found by Font and Fuller (2016[83]) for the city of London. In Southern Spain, road dust contributions to PM10 levels measured at a number of sites did not decrease in the period 2004-2011, whereas vehicle exhaust contributions decreased by 0.4 (0.24-0.57) mg/m3 year (Amato et al., 2014[18]).

Table 2.5. Average share of urban PM explained by road dust and exhaust emissions

	Urban PM10		Urban PM2.5	
	Road dust	Vehicle exhaust	Road dust	Vehicle exhaust
United States, 28 studies	-	-	**2-25% (6%)**	3-40% (22%)
Sweden (kerbside in spring), 3 studies	**66-76% (74%)**	<20%*	-	-
Central EU (CZ and DE), 2 studies	**9-20% (11%)**	11-17% (15%)	-	-
Spain, 10 studies	**8-34% (17%)**	10-31% (16%)	**8-31% (11%)**	10-32% (22%)
China, 15 studies	**7-59% (30%)**	7-62% (13%)	**1-13% (9%)**	6-22% (17%)
India, 11 studies	**18-51% (36%)**	6-26% (31%)	**6-26% (17%)**	31-57% (38%)

Note: * not specified. Road dust contributions may contain also wear emissions in some cases.

2.4. The negative consequences of exposure to non-exhaust emissions

The evidence for adverse health effects of particulate matter has grown dramatically in the past 20 years, and PM2.5 is understood to be associated with particularly harmful health effects. The Global Burden of Disease study ranked exposure to ambient fine particulate matter as the seventh most important risk factor for mortality – causing 4.2 million

premature deaths in 2016 globally (OECD, 2017[84]; Wang et al., 2016[85]).[12] A large fraction of urban populations are exposed to levels of fine particulate matter in excess of limit values set for the protection of human health. In Europe, 8.6 months of YPLL (Years of Potential Life Lost) have been blamed to excessive PM2.5 exposure. Numerous epidemiological studies have also demonstrated correlations between PM exposure and the occurrence of acute respiratory infections, lung cancer, and chronic respiratory and cardiovascular diseases (de Kok et al., 2006[86]; Heinrich and Slama, 2007[87]).

Although they note that no comprehensive studies have directly linked brake wear PM with adverse health outcomes, Grigoratos and Martini (2014[88]) review evidence demonstrating the negative health impacts of PM and of the chemical components of non-exhaust emissions. The mechanisms underlying the health effects of inhaled PM have been well-studied in the laboratory and there is general agreement regarding the key roles played by cellular injury and inflammation. In 2013, the WHO noted the possibility of the negative impacts of non-exhaust emissions, identifying them among the main sources of ambient PM and stating that the toxicological evidence increasingly shows that non-exhaust emissions could be responsible for some of the health effects of traffic-related pollution (WHO Regional Office for Europe, 2013[89]).

Particle mass, number, size and chemistry all affect PM toxicity. Regarding PM chemical composition, evidence continues to accumulate on the adverse effects that oxidative stress, which is often related to transition metals and redox active organics, such as quinone, has on human health (Ayres et al., 2008[90]; Borm et al., 2007[91]; Cassee et al., 2013[92]; Kelly, 2003[93]). Ambient PM derived from vehicles has a high oxidative potential (Kelly, 2003[93]) and it has been found that a clear increment in roadside particulate oxidative potential is associated with metals arising from non-exhaust emissions (Fedotov et al., 2014[94]; Thorpe and Harrison, 2008[45]; Schauer et al., 2006[95]). The roadside increments of particulate oxidative potential are significant and the metal components identified as determinants of this oxidative activity have associated with toxicity in human beings (Atkinson et al., 2009[96]). These results are important, as they highlight the contribution of currently non-regulated non-exhaust pollutants to negative health outcomes.

While it is unlikely that transition metals can explain all of the health effects observed in epidemiological studies at present ambient levels, measures to reduce them will most likely lead to improvements in the health status of the population. It is also unlikely that metals concentrations in ambient air are due exclusively to non-exhaust sources. Metal oxides are substances traditionally considered to be relatively inert chemically, but in very small size ranges, they have been linked with significant oxidative stress-mediated toxicity (Duffin, Mills and Donaldson, 2007[97]).

The presence of aluminium and silicon in particles has been associated with health problems, particularly respiratory ones (Batalha et al., 2002[98]; Rhoden et al., 2004[99]; Wellenius et al., 2003[100]). Other elements, including iron, copper, zinc and sulphur have also shown associations with health impacts, such as cardio-pulmonary oxidative stress, heart-rate variability and tissue damage in vivo (Gurgueira et al., 2002[101]; Kodavanti et al., 2005[102]; Rohr et al., 2011[103]). For example, in tests with laboratory animals, it has been shown that combined exposure to tyre wear dust and zinc and copper at high concentrations can lead to cardiac oxidative stress (Gottipolu et al., 2008[104]).

This part summarizes the work reported by Amato et al. (2019[105]), who performed a systematic review of epidemiological studies on non-exhaust source contributions and 15 possible tracers (Cu, Pb, Zn, Fe, Mn, Ca, Al, Si, Ti, K, Ni, Cr, Mg, Sb, and Ba). That report extracts results for Cu, Zn, Fe, Ca, Si, Sb, and Ba considering these elements are the most reliable tracers according to Table 2.1. Epidemiological studies have assessed the impact of non-exhaust emissions through two main exposure indicators: i) elements, also defined

as tracers (even though there are no unique tracers of non-exhaust sources); and ii) source contributions obtained from modelling activities. The main study designs used by epidemiologists to investigate the short- and long-run health effects of air pollution are presented in Box 2.5.

2.4.1. Elements

Studies based on elements are more common (45 studies). The main advantage of this approach is that elements can be analysed separately and there is no need for a full chemical speciation of PM samples. The main limitation is that elements are less suitable exposure indicators than source contributions, since they are usually emitted by multiple sources. However, most of the variance can be due to a single non-exhaust source. Among elements, studies analysed always a discrete group of elements including one or some non-exhaust tracers (Table 2.1) but often including also other elements/components.

Based on Table 2.1, results are presented for copper, iron, antimony, barium, calcium, silicon and zinc, which are more often identified as suitable tracers, although not unique. Among them, silicon and calcium, are probably those which serve less, since their non-traffic share is generally higher. Although antimony and copper will be significantly reduced in the future manufacturing, they can still offer valid information for time-series and case-crossover studies using historical data. The remaining elements of Table 2.1 have been excluded since they are less suitable tracers: their variance in ambient air is more due to other sources rather than to non-exhaust emissions. It is also noteworthy that barium have rarely been analysed, while it potentially offers very useful information, since it is more unique tracer than others in the sense that fewer other sources of barium exist.

Results from element-based studies can be influenced by the method employed. For example, for some trace elements, X-Ray Fluorescence (XRF) has a much higher detection limit than Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) and this may affect determination of trace elements at low concentrations.

2.4.2. Source contributions

Studies based on source apportionment are much fewer (8 studies), due to two main factors: i) a large number of elements/compounds is needed, increasing considerably the costs; ii) the difficulty of receptor modelling tools to identify clearly non-exhaust contributions. The main advantage is that a source contribution is interpreted as the total contribution from that source, including all contained toxics. The main limitation is that one can hardly separate all non-exhaust sources, implying that if the factor is identified as a mix, e.g. "non-exhaust" or "road dust/brake wear", the estimated health effects cannot be attributed to a specific non-exhaust source; and that if the factor is not correctly labelled, e.g. labelled as "road dust" while also including direct wear emissions, the estimated health effects are erroneously attributed only to a single source.

The review presented below shows only the 8 studies where the contribution of non-exhaust emissions was well separated from other mineral sources (Table 2.6). In fact, most of the source apportionment studies showed a generic "road dust/crustal" and "soil/road dust" or even a "soil" source with the road dust component simply mentioned in the text as a possible contributor of the mixture, and were therefore excluded from the analysis. A study using PM1-10 mass concentration is also included to draw on the health effects of road and tyre wear emissions due to the use of studded tyres.

Box 2.5. Study designs for air pollution epidemiology

Studies of short-term effects

The two main study designs for investigating the short-term effects of air pollution are time-series and case-crossover studies. In time series studies, exposure is measured or estimated for each time unit (e.g. day) homogeneously across the study area. Health outcomes, such as daily health indicators (e.g. mortality or morbidity) are also measured in the same area, so that temporal variability in exposure can be related to temporal variability in the health outcome. An important advantage of time-series studies is that the exposed population is compared with itself, hence time-invariant population characteristics are automatically controlled for. Confounders like population dynamics and weather conditions are controlled for in the regression models. Models also control for time trends and seasonality.

In case-crossover studies, the underlying principle is that when the effect of an exposure is immediate and reversible, the observed cases can be matched with themselves on past periods when the studied outcome had not occurred. Then the exposure on the case day and the exposures on case-free (control) days are contrasted to derive an estimate of the effect of interest. This approach has the advantage of preventing confounding by individual time-fixed characteristics, since comparisons are done within subjects. The "time-stratified" approach (Levy et al., 2001[106]), uses same weekdays of the case period within the same month and year as control periods.

In general, time series analyses are more flexible than case-crossover designs to model time trend, but case-crossover designs are better suited to analyse individual characteristics as effect modifiers, or are the only possible option when the daily exposure of interest is not an average of a domain, but is heterogeneous over space (Stafoggia and Faustini, 2018[107]).

Studies of long-term effects

Long-term effects are investigated through cohort studies and survival analysis. A cohort is a group of people who share a common exposure within a defined period and whose effect on health may occur much later in time. Then, a cohort study is a particular form of longitudinal study enrolling a cohort and following it over time usually until the occurrence of a specified outcome, or the end of the study. Exposure is usually assessed at the residential address of cohort participants by land-use regression, dispersion models or hybrid approaches. Exposure can be "fixed" (when attributed only once at baseline or as a long-term average), or time-dependent (when assessed repeatedly over the follow-up years for each individual). At any point in time, the "risk set" is made of all subjects at risk of incurring the study outcome at that time, while the cases are the events occurring at that time.

In the analysis of long-term effects of air pollution, it is assumed that the risk of mortality/morbidity is higher for increasing levels of long-term exposure at the residence. In order to test this hypothesis, conventional approaches of survival analysis (analysis of time-to-event data) are applied. The most used survival model is the "Cox proportional-hazards model" which assumes that the effect of a unit increase in the exposure is multiplicative with respect to the hazard rate, and it imposes no assumption on the shape of the hazard function. In the analysis of the association between long-term exposure to air pollution and mortality/morbidity outcomes, individual hazard rates are regressed against (baseline or time-dependent) exposure attributed at the residential address of each subject, while controlling for individual-level covariates (e.g. age, gender, lifestyle variables and comorbidities) and area-level indicators of socio-economic status (Stafoggia and Faustini, 2018[107])

Source: Levy et al. (2001[106]); Stafoggia and Faustini (2018[107]).

2.4.3. Impacts of non-exhaust emissions on mortality

Short-term effects

The countries under study were the United States (6 studies), Chile (2), Canada, Korea, China, Spain, UK and a group of 5 Southern European cities (one study each). Thirteen studies used elements as indicators of exposure,[13] while three studies applied source apportionment. However, only one study clearly disentangled a non-exhaust source contribution (Ostro et al., 2011[108]). Results show that:

Seven studies found significant association between exposure to Zinc and natural/all cause causes (2 studies), cardiovascular (5 studies), respiratory (2 studies) and ischaemic heart disease (1 study) mortality; other four studies investigating natural/all cause (4 studies), cardiovascular (3 studies) and respiratory (3 studies) mortality did not find significant associations.

Six studies found significant association between exposure to Iron and natural/all cause (3 studies), cardiovascular (3 studies), respiratory (1 study) and ischaemic heart disease (1 study) mortality; other four studies investigating natural/all cause (3 studies), cardiovascular (3 studies), respiratory (2 studies), COPD (1 study) and cerebrovascular (1 study) mortality did not find significant associations.

Five studies found significant association between exposure to Copper and natural/all cause (3 studies), respiratory (2 studies) and ischaemic heart disease (1 study) mortality; other four studies investigating natural/all cause (2 studies), , cardiovascular (3 studies), respiratory (3 studies), COPD (1 study) and cerebrovascular (1 study) mortality did not find significant associations.

Four studies found significant association between exposure to silicon and natural/all cause (2 studies), cardiovascular (2 studies) and respiratory (1 study) mortality; other four studies investigating natural/all cause (3 studies), cardiovascular (4 studies), respiratory (4 studies), COPD (1 study) and cerebrovascular (1 study) mortality did not find significant associations.

Two studies found significant association between exposure to Calcium and natural/all cause (2 studies), cardiovascular and respiratory mortality (1 study); other four studies investigating natural/all cause (4 studies), cardiovascular (5 studies), respiratory (4 studies), COPD (1 study) and cerebrovascular (1 study) mortality did not find significant associations. Antimony and Barium were both associated with cardiovascular and respiratory mortality (1 study), but not with natural mortality (1 study).

The only study on source apportionment clearly identifying a non-exhaust contribution was carried out in Barcelona (see also Table 2.6) (Ostro et al., 2011[108]) where authors disentangled the road dust contribution from other mineral sources. They applied a constrained Positive Matrix Factorization on PM10 and PM2.5 filters in Barcelona to estimate daily contributions from eight sources from 2003 to 2007, including road dust and motor exhaust. They applied the case-crossover design (see Box 2.5) to estimate the association between daily source contribution and all-cause and cardiovascular mortality. They found a significant association between road dust PM2.5 and all-cause deaths, with mortality increasing by 4.2% per 1.8 μg/m3 increase in a daily road dust load in PM2.5. The estimate was similar to that found for the vehicle exhaust: mortality rises by 3.7% per 5.2 μg/m3 increase. For PM10, only vehicle exhaust emissions had a statistically significant impact on all-cause mortality, amounting to a 3.6% increase per 5.2 μg/m3. For cardiovascular mortality, road dust showed an excess risk of 6.7% per 1.8 μg/m3 increase

in PM2.5 while vehicle exhaust had no significant effect. In contrast, vehicle exhaust emissions were associated with a 6.4% increase in the risk of cardiovascular mortality per 5.2 µg/m3 increase of PM10, while the impact of road dust was statistically insignificant.

In summary, 13 out of 14 studies found at least one statistically significant association between one of our selected indicators for non-exhaust emissions (either elemental or source contribution) and all-cause mortality as well as for specific causes, such as cardiovascular, respiratory and ischaemic heart disease. The only study with no positive associations for the selected indicators did find however significant association for Mg which has also been used as a possible indicator of road dust (Amato et al., 2019[105]).

Long-term effects

Long-term mortality studies have been carried out in the United States (7 studies) and Europe (5 studies). Most studies are based on the elemental tracer approach,[14] while two studies have used source apportioned data (Desikan et al., 2016[109]; Tonne et al., 2016[110]). Four studies found significant association between exposure to Iron and natural/all cause (2 studies),, cardiovascular (2 studies) and ischaemic heart disease (3 studies) mortality; other six studies investigating natural/all cause (3 studies), cardiovascular (2 studies), IHD (2 studies) and respiratory, pulmonary and lung cancer (1 study each) mortality did not find significant associations.

Three studies found significant association between exposure to Copper and natural/all cause (1 study), cardiovascular (1 study) and ischaemic heart disease (2 studies) mortality; other five studies investigating natural/all cause (3 studies), cardiovascular (2 studies), and respiratory, IHD and lung cancer (1 study each) mortality did not find significant associations.

Three studies found significant association between exposure to Zinc and natural/all cause (2 studies), cardiovascular (2 studies), and ischaemic heart disease (3 studies) mortality; other six studies investigating natural/all cause (3 studies), cardiovascular (2 studies), and respiratory, IHD, pulmonary, CHF, MI, COPD, Diabetes and lung cancer (1 study each) mortality did not find significant associations.

Five studies found significant association between exposure to Silicon and natural/all cause (2 studies), respiratory (1 study), cardiovascular (1 study), ischemic heart disease (2 studies), cardio-pulmonary (1 study) and pulmonary (1 study) mortality; other four studies investigating natural/all-cause (3 studies), cardiovascular (2 studies), ischemic heart disease (2 studies) and lung cancer (1 study) mortality did not find significant associations.

Two studies found significant association between exposure to Calcium and respiratory, CHF, MI, COPD, and diabetes mortality; other four studies investigating natural/all-cause (2 studies), ischemic heart disease (2 studies), cardiovascular (1 study) and lung cancer (1 study) mortality did not find significant associations. Barium was not associated with all-cause or cardiovascular mortality, and antimony was not associated with cardiovascular mortality

With respect to source apportionment studies, long term mortality was investigated for all causes and stroke (Table 2.6). Tonne et al. (2016[110]) investigated both long-term all-cause mortality and myocardial infarction morbidity using a modelling approach for exhaust and non-exhaust PM in Greater London. They followed more than 18 000 patients from the myocardial infarction National Audit Project for both death and readmission for myocardial infarction for the years 2003–2010. They found that most air pollutants were positively associated with all-cause mortality alone and in combination with hospital readmission. The largest associations with all-cause mortality per interquartile range (IQR) increase of pollutant were observed for non-exhaust contributions to PM10 (Hazard ratio

= 1.05, IQR = 1.1 μg/m3). For the PM2.5 fraction, the hazard ratio was slightly lower (1.04), for an IQR equal to 0.3 μg/m3. For both PM fractions, exhaust emissions were not significantly associated with morbidity or mortality.

Desikan et al. (2016[109]) combined a high-resolution urban air quality dispersion model for South London with a population-based stroke register to explore associations between long-term exposure to modelled non-exhaust and exhaust PM and mortality risk in post-stroke patients (within 5 years). The authors acknowledge that one limitation of their study is that they did not quantify individual pollution exposure, but instead used pollution levels at residential postcode addresses as a proxy for individual exposure to pollutants. Neither non-exhaust emissions of PM2.5 nor of PM10 were associated with increased mortality in post-stroke patients.

Of 12 studies, 11 found a statistically significant association between some non-exhaust indicator and long-term mortality. Besides all-cause and natural mortality, non-exhaust PM were also significantly associated with ischemic heart disease, cardiovascular/cardiopulmonary, respiratory, CHF, MI, COPD, and diabetes mortality.

2.4.4. Impact of non-exhaust emissions on morbidity and cognitive development

Short-term effects

Thirteen studies in this review investigate the impact of several constituent elements of non-exhaust emissions on short-term morbidity.[15] Zanobetti et al. (2009[111]) (Suh et al., 2011[112]; Tiittanen et al., 1999[113]; Sun et al., 2016[114]; Samoli et al., 2016[115]; Zanobetti et al., 2009[116]) also used source contributions as exposure indicators. A number of studies were conducted in the United States, and one study was conducted each in Chile, China, the UK, and a group of five Southern European cities.

Five studies found a significant association between exposure to Copper and cardiovascular and respiratory hospitalisations (4 studies and 2 studies, respectively). Three other studies investigating cardiovascular, respiratory, and stroke morbidity (1 study each) did not find significant associations between these elements and short term morbidity. Four studies found a significant association between exposure to Zinc and natural/all cause, cardiovascular and respiratory hospitalisations (1 study, 3 studies, and 2 studies, respectively). Another five studies investigating cardiovascular and respiratory conditions (3 studies each), and myocardial infarction, congestive heart failure, diabetes, stroke and heart rate morbidity (1 study each) did not find significant associations.

Four studies found a significant association between exposure to Iron and natural/all cause, cardiovascular and respiratory hospitalisations (1 study, 3 studies, and 1 study, respectively). Another six studies investigating cardiovascular and respiratory morbidity (2 studies each), and diabetes, stroke, heart rate and cough morbidity (1 study each) did not find significant associations with exposure to Iron. Three studies found a significant association between exposure to Calcium and natural/all cause, cardiovascular and respiratory hospitalisations (1 study, 1 study, and 2 studies, respectively). Six other studies investigating cardiovascular and respiratory morbidity (3 studies each) and diabetes and stroke morbidity (1 study each) did not find significant associations.

Three studies found a significant association between exposure to Silicon and natural/all cause, cardiovascular and respiratory hospitalisations (1 study, 1 study, and 3 studies, respectively). Five studies investigating cardiovascular and respiratory morbidity (4 studies and 3 studies, respectively) and diabetes, stroke, congestive heart failure and myocardial infarction morbidity (1 study each) did not find significant associations.

Antimony and Barium have been associated with natural/all cause and respiratory hospitalisations (1 study).

Results from studies analysing biomarkers and clinically specific outcomes should be considered with some caution, since they generally report wide confidence intervals. This also extends to studies of long-term morbidity effects, presented in the next subsection.

The only study using source apportionment that separates a non-exhaust contribution on morbidity was conducted by Kioumourtzoglou et al. (2014[117]) (Table 2.6). In this study, the authors examined the effects of PM2.5 sources on emergency cardiovascular hospital admissions among Medicare enrolees in Boston, MA between 2003 and 2010. They also studied the effect of uncertainty in source contributions using a block bootstrap procedure. While inconsistent associations across different source apportionment methods were observed for exposure to road dust, exposure to exhaust PM2.5 was associated with increased admissions.

In summary, six of the 13 studies reviewed found at least one statistically significant association between an indicator of non-exhaust PM (either elemental, ionic or source contribution) and short-term morbidity. Respiratory and cardiovascular morbidity were the principle types of morbidity associated with exposure to indicators of non-exhaust PM. Of the seven remaining studies, five found a significant association between other elements (e.g. Ni, Al and Mg) that have been also used as possible indicators of non-exhaust PM (Amato et al., 2019[105]) and short term morbidities.

Long-term effects

Fifteen studies included in the review investigated the association between long-term exposure to non-exhaust PM and hospitalisations or other biomarkers. Thirteen studies were carried out in Europe, mostly as part of the ESCAPE and TRANSPHORM projects[16] and two studies were conducted in the United States (Basu et al., 2014[118]; Vedal et al., 2013[119]). Eleven studies examined exposure to specific elements and four used source apportionment. Results show that:

Six studies found a significant association between exposure to Iron and cardiovascular and lung cancer hospitalisations, impaired lung function, high blood pressure, low birthweight and markers of inflammatory (1 study each). Four studies investigated cardiovascular morbidity, impaired lung function, pneumonia and low birthweight (1 study each) and did not find significant associations.

Six studies found significant association between exposure to Zinc and pneumonia (1 study) and lung cancer (1 study) hospitalisations, lung function (1 study), low birthweight size (2 studies) and inflammatory marker (1 study); other four studies investigating cardiovascular (2 studies), lung function, and blood pressure (1 study each) morbidity did not find significant associations

Five studies found significant association between exposure to Copper and a marker of cardiovascular disease, lung cancer hospitalisations, low birthweight and inflammatory markers (1 study, 1 study, 2 studies, and 1 study, respectively). Another five studies investigated cardiovascular morbidity (2 studies) and lung function, pneumonia and blood pressure morbidity (1 study each) did not find significant associations with exposure to Copper.

Three studies found a significant association between exposure to Silicon and cardiovascular hospitalisations, blood pressure and low birthweight (1 study each). Six studies investigating indicators of cardiovascular disease, lung cancer, impaired lung

function, pneumonia, low birthweight and inflammatory markers (1 study each) did not find significant associations with exposure to Silicon.

Calcium was investigated for low birthweight (1 study) and carotid intima-media thickness and coronary artery calcium (1 study), finding no significant associations. Antimony and Barium were not associate with coronary artery calcium nor with carotid intima-media thickness.

Long-term effects on morbidity were investigated by four source apportionment studies (Table 2.6) (including the one by Tonne et al. (2016[110]), while another study investigated the effect of traffic sources on cognitive development in children. Willers et al. (2013[120]) investigated the effects of exposure to particulate matter fractions (PM1 and PM1-10) on respiratory health in the Swedish adult population, using an integrated assessment of sources at different geographical scales. They assumed PM1-10 to be a proxy of road-tyre wear particles, which is a reasonable assumption in the case of Sweden due to the use of studded tyres, implying that the dominant source of coarse PM is wear and road dust resuspension. The study was based on a nationwide environmental health survey performed in 2007, including 25 851 adults aged 18–80 years. Individual exposure to PM at residential addresses was estimated by dispersion modelling of regional, urban and local sources. Associations between different size fractions or source categories and respiratory outcomes were analysed using multiple logistic regression, controlling for individual and contextual factors. Exposure to locally generated wear particles showed associations for blocked nose or hay fever, chest tightness or cough, and restricted activity days with odds ratios of 1.5–2 per 10 µg/m3 increase.

Crichton et al. (2016[121]) combined a high resolution urban air quality model for South London with a population-based stroke register to explore associations between long-term exposure to PM sources and stroke incidence (2005–2012). No associations were observed between non-exhaust sources and overall ischemic or haemorrhagic incidence, while a 20% increase of total anterior circulation infarct for an interquartile range (0.78–0.96 µg/m3) of exhaust PM was found.

Dadvand et al. (2014[122]) investigated a hospital cohort of pregnant women (N=3182) residing in Barcelona, Spain, during 2003–2005. Positive Matrix Factorization source apportionment (PMF) was used to identify sources of PM10 and PM2.5 samples obtained by an urban background monitor, resulting in the detection of eight sources. They separated brake wear and vehicle exhaust and generated a comprehensive indicator of combined traffic sources, including also a fraction of secondary nitrate/organics. For the exposure during the entire pregnancy, they found a 44% increase in the risk of preeclampsia associated with one IQR increase in exposure to PM10 brake dust (4.7 µg/m3), and an 80% increase from one IQR rise in exposure to PM10 from all traffic-related sources (15.7 µg/m3) combined. For vehicle exhaust alone, they did not find a significant association with preeclampsia.

Finally, Basagaña et al. (2016[123]) investigated associations between traffic-related air pollution exposure at schools and cognitive development. Using a cohort of 2 618 schoolchildren (average age of 8.5 years) belonging to 39 schools in Barcelona, they found that an interquartile range (3.8 µg/m3) increase in motor exhaust PM2.5 was associated with reductions in cognitive growth equivalent to 22% of the annual change in working memory, 30% of the annual change in superior working memory, and 11% of the annual change in the inattentiveness scale. Non-exhaust PM2.5 sources were not associated with adverse effects on cognitive development.

In summary, 12 of the 15 studies found at least one statistically significant association between an indicator of non-exhaust PM (either elemental or source contribution) and

long-term morbidity or increase in biomarkers. Non-exhaust PM were associated with several causes of morbidity, mainly cardiovascular, lung cancer, lung function, pneumonia, myocardial infarction, respiratory markers and preeclampsia. Significant associations were also found with other endpoints, such as carotid intima-media thickness (CIMT), inflammatory markers, fibrinogen, birthweight and blood pressure in children.

Table 2.6 summarizes results from source apportionment studies, allowing a comparison between vehicle exhaust and non-exhaust emissions. Since epidemiological studies imply a linear dose-response function, the increased risk can be normalised by the IQR, so that estimates per µg/m3 are obtained. Following this approach, road emissions in Barcelona resulted in a higher risk (2.3% per µg/m3) than exhaust emissions (0.7%) of short-term all-cause and cardiovascular mortality (Ostro et al., 2011[108]), but with some overlap of their confidence intervals (0.8-3.9 and 0.1-1.3 respectively). Since the study did not separate any direct wear emission factor, the "road dust" source should be probably interpreted as a general "non-exhaust" source. Kioumourtzoglou et al. (2014[117]) found that cardiovascular emergency visits in Boston were instead significantly associated only with exhaust emissions, but not with road dust.

For long-term effects, Tonne et al. (2016[110]) found that all-cause long-term mortality in London was positively associated with non-exhaust emissions, for both PM2.5 and PM10. Other long-term studies concluded that respiratory symptoms and preeclampsia were only associated with non-exhaust PM, rather than with exhaust ones, while the opposite was found for stroke incidence and cognitive development in children.

Table 2.7 summarizes the qualitative associations between exposure to non-exhaust emission indicators and increased risk of short-term and long-term mortality and morbidity.

Table 2.6. Overview of findings of epidemiological studies using source apportionment analysis

Study	Country	Period	Health outcome	Non-exhaust source	Increased risk (95% confidence interval) per increase of non-exhaust PM	Increased risk (95% confidence interval) per increase of exhaust PM
Ostro et al. (2011[108])	Spain	2003-2007	Short-term total mortality	Road dust (PM2.5)	4.2% (1.5-7.0%) per 1.8 µg/m3	3.7% (0.7-6.7%) per 5.2 µg/m3
				Road dust (PM10)	No effect	3.6% (0.1-7.2%) per 5.2 µg/m3
			Short-term cardiovascular mortality	Road dust (PM2.5)	6.7% (2.4-11.3%) per 1.8 µg/m3	No effect
				Road dust (PM10)	No effect	6.4% (1.5-11.6%) per 5.2 µg/m3
Kioumourtzoglou et al. (2014[117])	MA, USA	2003-2010	Short-term cardiovascular emergency visits	Road dust (PM2.5)	No effect	1.44% (0.02- 3.11%) per 1.1 µg/m3
Tonne et al. (2016[110])	UK	2003-2010	Long-term all-cause mortality	Non-exhaust contribution (PM10)	5% (0-10%) per 1.1 µg/m3	No effect
				Non-exhaust contribution (PM2.5)	4% (0-9%) per 0.3 µg/m3	No effect

Desikan et al. (2016[109])	UK	2005-2013	Long-term all-cause mortality	Non-exhaust contribution (PM10 and PM2.5)	No effect	No effect
Willers et al. (2013[120])	Sweden	2007	Long-term respiratory symptoms	Road and tyre wear (PM1-10)	1.5–2 odds ratio* per 10 µg/m3	No effect
Dadvand et al. (2014[122])	Spain	2003-2005	Preeclampsia in pregnant women	Brake wear (PM10)	1.44 (1.07-1.94) odds ratio per 0.5 µg/m3	No effect
Crichton et al. (2016[121])	UK	2005-2012	Long-term stroke incidence	Non-exhaust contribution (PM10 and PM2.5)	No effect	20% (1-41%) per 0.78–0.96 µg/m3
Basagaña et al. (2016[123])	Spain	2012-2013	Cognitive development	Road dust (PM2.5)	No effect	22% (2-42%) per 3.8 µg/m3

Note: Only studies clearly identifying a non-exhaust source are presented in this table. Studies analysing biomarkers and/or clinical specific outcomes should be considered with caution, since they reported very wide confidence intervals. *Range of observed symptoms, confidence intervals reported in Willers et al. (2013).

Table 2.7. Health effects of non-exhaust tracers concentrations and source contributions

Exposure indicator	Mortality		Morbidity		Other symptoms/biomarkers
	Short-term	Long-term	Short-term	Long-term	Long-term
Copper	All-cause, respiratory and ischemic heart disease	All-cause, cardiovascular and ischemic heart disease	Cardiovascular and respiratory	Lung cancer, carotid intima-media thickness	Low birthweight and inflammatory marker
Zinc	All-cause, cardiovascular, respiratory and ischemic heart disease	All-cause, cardiovascular and ischemic heart disease	Non-accidental, cardiovascular and respiratory	Pneumonia and lung cancer	Lung function, low birthweight and inflammatory marker
Iron	All-cause, cardiovascular, respiratory and ischemic heart disease	All-cause, cardiovascular and ischemic heart disease	Cardiovascular and respiratory	Cardiovascular and lung cancer	Lung function, blood pressure, low birthweight and inflammatory marker
Silicon	All-cause, cardiovascular and respiratory	All-cause, cardiovascular, respiratory, pulmonary and ischemic heart disease		Cardiovascular	Blood pressure and low birthweight
Calcium	All-cause, cardiovascular and respiratory	Respiratory	Non-accidental, cardiovascular and respiratory		
Barium	Cardiovascular and respiratory		Non-accidental and respiratory		
Antimony	Cardiovascular and respiratory		Non-accidental and respiratory		
Source contributions	All-cause and cardiovascular	All-cause		Myocardial infarction and Preeclampsia	Respiratory

2.5. The causes of non-exhaust emissions

This section is aimed at describing the mechanisms, vehicle and road features underpinning the generation and dispersion of non-exhaust emissions and analysis of their role in

determining the magnitude of emissions. The different methodological approaches used to quantify the impact of each of these drivers are also reviewed.

2.5.1. Brake wear

When drivers press the brake pedal, kinetic energy is transformed mostly into frictional heat between the brake lining (pad) and a rotor (disc or drum) and wear particles are generated. A friction process is always accompanied by the formation of friction products. Friction products include an external layer on the pad surface made of newly formed materials, brake wear PM emissions and emissions of gaseous pollutants.

The air quality burden depends mostly on the generation of airborne particles with a diameter below 10 µm. During the friction process, not all worn particles become airborne, Sanders et al. (2003[37]) report that only 50% of brake wear was emitted as airborne and the PM10 fraction accounted for 63%-85% of the airborne material. The wear of pads and rotors generate particles of various sizes and morphology, and each combination of speed, pressure, and temperature leads to a different amount of wear (Kukutschová et al., 2009[124]). In addition, the amount (measured as mass or number) of emitted particles likely depend also on braking frequency, vehicle weight, and the composition and age of pad and disc. In sum, the size and number of emitted particles depends on driving, vehicle and brake characteristics.

Quantifying the role of each factor is a very difficult task since the operation of brake systems is quite complex and often stochastic in nature, so it is not possible to simulate all braking scenarios. This hampers quantitative comparisons among different studies, since testing devices, testing procedures, sampling conditions and brake materials are different. This is mostly because no recommended approach for the generation, measurement, and expression of brake wear emissions exists yet (Panko, Kreider and Unice, 2018[59]). For example, braking tests can be performed with simplified laboratory tests (e.g., pin-on-disc), or by subscale and full-scale brake dynamometers and real field tests.[17] Nevertheless, these tests can hardly predict the wear behaviour of real brake systems (Grigoratos and Martini, 2015[6]; Kukutschová and Filip, 2018[125]; Lee and Filip, 2013[126]). Depending on the set-up, laboratory studies can differ in size of the tested pad samples, adopted sliding velocity and deceleration, particle generation rate, airflow regime and driving regime from real-world conditions. These differences further result in differences in energy generated per area and mass and per unit time to heat up the tested materials, which can also influence parameters of wear particles (Kukutschová and Filip, 2018[125]).

Rotor temperature

Rotor temperature is the most studied parameter. Typically, wear increases with increasing temperature (Filip, 2013[127]; Filip, Weiss and Rafaja, 2002[128]). Depending on the temperature reached, we can distinguish between “mechanical” and “oxidative” wear. At high temperatures (probably reached under extreme braking) wear of polymer matrix pads is accompanied with oxidative processes associated with considerable mass loss due to polymer degradation and formation of volatiles. This is characteristic for high-temperature braking scenarios, when numerous thermally less stable components (e.g., phenolic resin, rubber, graphite, coke) interact with available gases and oxygen from the ambient air (Kukutschová et al., 2009[124]) or undergo a pyrolysis (Plachá et al., 2017[4]). This degradation of organic components is associated with emissions of very fine amorphous carbon particles with negligible contribution to PM mass and volatile organic compounds (Kukutschová et al., 2011[3]; Plachá et al., 2017[4]). At lower temperatures, “mechanical” wear dominates. In general, the adhesive wear mechanism is combined with abrasive wear, fatigue wear mechanisms, and oxidative wear as well. Thus, the produced brake wear

debris is a complex mixture containing particles with sizes ranging from several nanometres to millimetres and the chemistry of wear debris is significantly different compared with the original pad material constituents, but typically comparable with the chemistry of a friction layer (Kukutschová et al., 2009[124]; Roubicek et al., 2008[129]).

Oxidative wear can generate very fine (submicron-sized), typically round-shaped particles. These submicrometric particles are formed by condensation of volatile gaseous compounds generated by thermal degradation of organic binder in brake pads. Iron oxide particles, produced not only by oxidative mild wear of the cast iron disc but also from oxidation of iron-based ingredients of metallic pads, are typically present in friction layer and together with elemental carbon from resin and other organic pad constituents represent one of the main components of airborne wear debris.

Mechanical wear (abrasive and fatigue wear) typically leads to the release of larger particles, belonging mainly to PM10 or PM2.5 fractions. These particles usually have sharper edges and irregular morphology (Kukutschová et al., 2011[3]). Alemani et al. (2016[130]) found that with temperature increasing from 100 to 300ºC, the ultrafine particle emissions intensifies, while the coarse particle emission decreases. Similarly, Kukutschová et al. (2011[3]) found that testing conditions of a relatively cold rotor (below and around 200ºC) led to negligible emissions of submicron particles. After the rotor temperature reached 300ºC, a gradual increase of the finest fractions, including the nanosized particles (<100 nm) reaching concentration up to 10^6 per cm^3, was detected. From the shape of the particle size distributions and their variation with time, it could be assumed that submicron particles are formed by the evaporation/ condensation process with a subsequent aggregation of the primary nanosized particles. In contrast, the detected concentrations of larger microsized particles were not so strongly affected by increasing rotor temperature.

Sliding speed, deceleration and contact pressure

Mosleh et al. (2004[131]) generated wear particles from commercial metallic truck brake pads but focused on settled particles only (not airborne). The study addressed effects of contact pressure, sliding speed, and continuity of sliding contact on particle size distribution and the chemistry of collected wear particles. The generated particles had a bimodal distribution, with a first peak at approximately 350 nm (composition corresponding to the cast iron disc), and a second peak between 2 and 15 µm, depending on the pressure and sliding speed. Importantly, when the motion was discontinuous at a repeated brake action, smaller wear particles were generated. Wahlstrom et al (2017[132]) found that the specific wear rate of the disc decreases with increasing contact pressure and sliding velocity. The particle mass and number rates seem to decrease with increasing contact pressure and sliding velocity until the disc temperature is about 200 °C; thereafter they increase.

Few studies have evaluated the impact of acceleration or deceleration rate on non-exhaust PM generation. In general, acceleration results in slightly increased brake emissions due to the drag effect (zum Hagen et al., 2019[133]). Hagino, Oyama and Sasaki (2015[134]) focused on the quantification of PM10 and PM2.5 emissions by mass and the evaluation of resuspended particles. Airborne wear particles were not only detected at deceleration as a direct brake wear but also occurred at acceleration (non-braking event), suggesting resuspension of wear particles. These particles had increasing concentration during acceleration with an increasing initial speed. Based on their findings, the resuspended particles should be included in emission measurements.

Riediker et al. (2008) tested pad materials of six different passenger cars under controlled environmental conditions and found a bimodal PN distribution with peaks at 80 nm (depending on the tested car and braking behaviour) and at 200-400 nm (0.2-0.4 µm). They

found that complete stops result in higher nanoparticle production compared with normal deceleration. Sanders et al. (2003[37]) found that emission factors were increasing by a factor of 4 when comparing a deceleration rate of 0.6-1.6 m/s^2 (typical urban driving pattern) to an aggressive deceleration rate of 1.8 m/s^2. Lee et al. (2013[135]) found that when a vehicle decelerates rapidly (2.6 m/s^2), considerable brake wear particles and particles from tyre/road interface are generated. The maximum value of the mass size distribution was located at 1-5 µm, and, unlike constant speed driving conditions, the ratio of nano-particles measuring 50 nm or less was very high.

Vehicle weight

Garg et al. (2000[38]) claim that the inertia weight being stopped is one of the factors contributing to brake wear rate, but they did not perform any tests with varying weights to confirm this. Several individual studies have found higher non-exhaust emission factors for heavier vehicle categories, indicating a correlation with weight. Despite varying definitions for the weight of vehicle categories, the consensus is that light duty trucks (including vans, pick-up trucks and SUVs) emit more PM than passenger cars (Luekewille et al., 2001[136]). Garg et al. (2000[38]) found that the brakes of large cars emit 55% more total suspended particles (TSP), PM10 and PM2.5, than small cars. Large pick-up trucks were found to emit more than double the quantity of particulates compared with small cars. The EMEP/EEA inventory guidebook provides PM10 and PM2.5 emission factors for brake wear for light duty trucks which is 55% higher than passenger cars, and a linear increase with the percentage of HDV load.[18] The recent UK government Survey "Call for Evidence on non-exhaust emissions" acknowledged that emissions vary hugely as a function of weight.

Assuming a linear relationship between weight and brake wear emission factor, Simons (2016[137]) estimated that brake wear PM10 emissions per vehicle-km increase by 0.004 per kg of vehicle weight, using the ecoinvent v2 database. A slightly higher value of 0.0053 can be obtained by dividing the EMEP/EEA PM10 emission factor by 1400, which is the average weight of EU passenger cars (ICCT, 2018[138]; Ntziachristos and Boulter, 2016[56]). For PM2.5 emissions per vehicle-km, the estimates of Simons (2016[137]) and EMEP/EEA factors would be 0.0017 and 0.0021 mg per kg of vehicle weight, respectively.

Brake pad type

Brake lining materials underwent a distinct development in the past three decades. This is related to an effort to develop materials responding to the demand for higher transportation safety, fuel economy, comfort, and performance, as well as to increasing environmental concerns. As a rule, the brake lining material should have: (i) appropriate mechanical and thermal properties, (ii) adequate and stable coefficient of friction in a wide range of operating conditions (temperature, pressure, environment, e.g., dust, water, de-icing agents), and (iii) high resistance to wear and good compatibility with the rubbing counterpart. Ideally, they should operate reliably under hot, dry, wet, or cold conditions, and without pollution and noise, and should be easily manufactured at low costs.

Despite the replacement of asbestos and a gradual elimination of copper in brake pad materials, they have environmental consequences. Wahlström et al. (2010[139]) indicated that the low metallic (LM) pads showed higher friction performance and caused more wear to the rotor than the non-asbestos organic (NAO) pads, resulting in higher mass losses and more concentrations of airborne wear particles. Although there were variations in the measured particle concentrations, similar size distributions of airborne wear particles were obtained regardless of the pad material. Perricone et al. (2016[140]) produced a ranking of pad-rotor combinations. This ranking revealed that NAO pads have the lowest emission

factors with respect to mass, but the highest emission factors with respect to particle numbers. LM pads had higher emission factors with respect to mass, and lower ones with respect to particle number.

Sanders et al. (2003[37]), estimated that 60% of the wear debris comes from the rotor and 40% from the pads, as confirmed also by (Hulskotte, Roskam and Denier van der Gon, 2014[34]). The EU-funded LOWBRASYS project aims at demonstrating a novel and low environmental impact brake system that will reduce micro and nanoparticles emissions by at least 50%. Both, particulate emissions in the micrometer range (important for PM mass reduction), as well as ultrafine particles (important for Particulate Number, PN reduction) are addressed.

2.5.2. Tyre and road wear

Vehicle weight

Few studies link tyre wear to vehicle weight, mostly using tyre wear simulator and computational methods (Chen and Prathaban, 2013[141]; Li et al., 2012[142]; Salminen, 2014[143]; Simons, 2016[137]; Wang et al., 2017[144]). Studies agree that tyre wear increases with vehicle weight, but they do not reach a consensus on the shape of this relationship. Wang et al. (2017[144]) and Li et al. (2012[142]) found that the tyre wear (mass loss) increased linearly with increasing vertical load (or sprung mass). Furthermore, the vertical load had a marked influence on the contact pressure distribution. Despite the longer contact length, the vertical contact pressure becomes higher with increasing vertical load, which leads to higher slip forces, and then to more severe wear. Plotting the average wear rates of vehicles with different ranges of maximum admissible vehicle weight, Pohrt (2019[145]) found that the central value in each vehicle category correlates almost linearly with the emission rate. Salminen (2014[143]) simulated numerically an exponential increase but the experimental data used for validation indicated a rather linear relationship between vehicle weight and tyre wear.

Assuming a linear relationship between vehicle weight and tyre wear emissions and using information from the ecoinvent v2 (emission factors and vehicle weight) database, Simons (2016[137]) calculated that tyre wear PM10 emissions per vehicle-km increase at a rate of 0.0041 mg per kg of vehicle weight. A similar value (0.0046) can be obtained for tyre wear by dividing the EMEP/EEA emission factor for passenger car by 1400 Kg which is the mean curb weight in the EU market (ICCT, 2018[138]).[19] For PM2.5 emissions per vehicle-km, the estimates of Simons (2016[137]) and EMEP/EEA factors would be 0.0029 and 0.0032 mg/kg respectively.

The relationship between road wear and vehicle weight has hardly been studies in the literature. While Simons (2016[137]) calculated a value of 0.0049 mg PM10 emissions per kg of vehicle weight per km – assuming a linear relationship – the EMEP/EEA provides the same emission factor for cars and vans, implying that road wear is assumed to be insensitive to weight changes. For PM2.5 emissions per vehicle-km, the estimate of Simons (2016[137]) is 0.0026 mg/kg.

For road wear induced by heavy duty vehicles, (Žnidarič, 2015[146]) estimates a power law of 4 between increased axle load and road wear, but the applicability of this relationship to light duty vehicles is questionable.

Speed and acceleration/deceleration

According got the recent response on UK Survey on non-exhaust emissions, In 30% of tyre wear could be attribute to driving style, so the promotion of better, smoother, more

efficient driving styles by incorporating 'eco' driving into standard driver training and custom courses could be a good means of reducing non-exhaust and other emissions.

Speed determines the amount of mechanical stress in the tyre material and thus the wear and temperature of the tyre. Chen and Prathaban (2013[141]) estimated that truck speed has an influence on tyre wear which can vary exponentially from 0.0242 to 0.0244 mm/100km, when vehicle speed increases from 20 km/h to 120 km/h. Wang et al. (2017[144]) found that the normalized amount of tyre wear increases from 0.812 to 1.148 as the rolling velocity increases from -40% to 40% compared with its initial value. Li et al. (2012[142]) found a linear relationship between tyre wear and vehicle speed between 10 and 40 m/s. Foitzik et al. (2018[147]) found similar results for particle number emissions.

Gustafsson et al. (2008[148]) investigated the wear of road surface on a road simulator at different speed levels (30, 50 and 70 km/h) concluding that road wear increases linearly with speed (on a stone mastic asphalt pavement). Speed increased particle mass concentration in the simulator for both studded and friction tyres, but the magnitude of the increase is much higher for studded tyres. Decreased speed of the road simulator results also in a lower particle number concentration, but with the same distribution.

Salminen (2014[143]) found that the wear increases exponentially with speed. He also found that the wear rate increases significantly with longitudinal slip, which occurs when braking or accelerating[20]: tyre wear can vary up to a factor of 2 within a longitudinal slip range of [-0.3,0.3]. Foitzik et al. (2018[147]) found similar results for particle number emissions.

Tyre type and properties

Three types of tyres are available in the market: summer tyres, winter tyres and studded tyres. Winter tyres have a higher natural rubber content that keeps them supple in the cold. They also have a deep tread pattern. Summer tyres provide better all-round performance in the warmer months (temperatures above 7ºC). They are composed of a relatively hard compound that softens in milder temperatures to be able to adapt to dry as well as wet roads. Studded tyres contain metal studs embedded within the tread that are designed to dig into ice, providing added traction. When the driving surface is not covered in ice, however, studded tyres can damage road suraces. Many countries limit their use during non-winter months and some states have outlawed them completely. Early tests have shown a marked difference in PM10 generation by different pavement constructions with different rock aggregates when compared to friction tyres resulting from studded tyres (Gustafsson et al., 2009[48]).

Chen and Prathaban (2013[141]) use mathematical models to simulate the effect of varying inflation pressure from 55 to 165 psi on tyre wear concluding that wear decreases exponentially with higher inflation pressure (which is related to the contact patch area) from 0.0265 to 0.0240 mm/100km. Similar findings were found by Li et al. (2012[142]), Salminen (2014[143]) and Wang et al. (2017[144]). Tyre wear is also inversely proportional to tyre diameter and width, whilst it is invariant to tyre groove depth (Chen and Prathaban, 2013[141]; Le Maître, Süssner and Zarak, 1998[149]).[21]

Low rolling resistance tyres are designed to reduce the energy loss as a tyre rolls, decreasing the required rolling effort and improving vehicle fuel efficiency. As more research is being done, wear rates will likely improve, but, at present, no sources suggest that low rolling resistance tyres will have significantly lower wear rates.

Different tyre brand/model can have up to fourfold different wear rate (Grigoratos et al., 2018[150]). Tyre age was found to be an influencing parameter on tyre wear. New tyre can have 10% higher wear rate than used ones (Sakai, 1996[151]).

Road pavement properties

In Finland and Sweden, research has been conducted in similar laboratories to assess the influence of pavement properties on particle emissions, with a strong focus on PM10 and wear resistant pavement constructions, such as stone mastic asphalt (SMA). The Los Angeles abrasion test, measuring the fragmentation capacity of the road pavement material, has been proposed as a proxy measure of PM10 emission potential, since road simulator studies have found a correlation between different Los Angeles test value pavements and emission rates (Gustafsson and Johansson, 2012[152]).

In Finland, Räisänen, Kupiainen and Tervahattu (2005[153]) tested different road pavement using traction sand to increase wear, and concluded that a pavement made with a granitic aggregate composition with higher resistance to abrasive wear resulted in lower PM10 emissions than a pavement made with a mafic volcanic rock. However, it is not self-evident that a material of high abrasion resistance also generates lower PM10. Döse and Åkesson (2011[154]) (cited in Gustafsson and Johansson (2012[152])) studied the amount of PM10 produced in the Nordic ball mill test (with studded tyres) and showed that some materials have higher ball mill values (less resistant), but still do not produce higher PM10 amounts than far more resistant rock aggregates. For rocks with Nordic ball mill values below 10, the authors suggested that for each unit lower value, each ton of rock will produce 4 kg less PM10. However, the results shown are based on a single experimental design and on the specific studded tyre behaviour, so it is not relevant for soft materials like those used in normal tyres.

It is also believed that the diameter of aggregates in the pavements influences the total wear in as much as coarser material results in lesser wear (Jacobson and Wågberg, 2007[155]). Gustafsson and Johansson (2012[152]), China and James (2012[156]) and Amato et al. (2013[157]) found a -1.5 power relationship between mean size of pavement aggregates and road dust loading (R^2=0.52). Padoan et al. (2018[158]) found a -0.2 power relationship with the corrected aggregate mode (correcting the aggregate size mode by the textural depth). Gustafsson and Johansson (2012[152]) concluded that the lower the maximum size of coarse aggregate and the lower the Nordic abrasion value of the aggregate material, the lower the particle formation.

Most of European and American road surfaces consist of open asphalt. According to manufacturers, it is around 90% share of the EU market. Open asphalt has 15-25% hollow space which can retain wear particles (Kole et al., 2017[159]), but it has been found that lower-density asphalt had a higher wear rate (Do et al., 2003[160]; Gothie and Do, 2003[161]). Asphalt roads are also found to have higher rolling resistance (Ejsmont et al., 2014[162]) and higher wear rate than concrete roads. Because asphalt is an aggregate of particles with a bitumen binder, a distinction between the macro texture (size, distribution and geometrical configuration of particles) and the micro texture (of the individual particles) can be made (Pohrt, 2019[145]). With the wearing away of the bitumen binder and the resulting increase of surface voids, the macro texture tends to increase over time (Pohrt, 2019[145]). In contrast, the micro texture tends to diminish due to polishing (Veirh, 1992[163]).

The state of the pavement is also a relevant parameter. Gehrig et al (2010[52]) found that damaged asphalt concrete was emitting 10 times more road wear particles than the same pavement in good conditions, by means of a road simulator for HDV. Furthermore, alternative designs and materials, such as rubber mixed asphalt, furnace slag asphalt, porous asphalt, and cement concrete, have been tested for their PM10 emissions. Alternative materials for use in asphalt pavements as well as alternative constructions are often considered not only to improve pavement duration and properties but also to find ways to use or reuse residual or waste materials. Rubber asphalt with a gap grading was shown to slightly reduce wear PM10 production, whereas open-graded rubber asphalt did

not differ from a reference SMA pavement. Furnace slag pavements were tested in (Viman and Gustafsson, 2015[164]). PM10 production from SMA8 and SMA11 slag asphalts were at similar levels to most asphalt wearing courses made from natural aggregates.

In cement concrete, cement replaces bitumen as a binder, while aggregate may be the same as the asphalt pavement Cement concrete pavements are more durable and wear resistant, but also more expensive to build than asphalt pavements (Wiman et al., 2009[165]). As they have advantages from a fire perspective, they are often considered for use in road tunnels (Bonati et al., 2012[166]). Cement concrete has been tested for PM10 emissions, and the results show that they emit more PM10 than the reference asphalt with the same rock used for the conglomerate stones (aggregates), even if the total wear is lower (Gustafsson et al., 2015[167]).

2.5.3. Road dust resuspension

The three main factors determining road dust emissions are vehicle speed and size, and road dust loading. The importance of each parameter on the magnitude of emission potential has been the object of research since several decades and empirical models have been developed to infer emission factors.

Speed and acceleration

Few studies have been undertaken on the effect of vehicle speed on road dust suspension. Lee et al. (2013[135]) used a mobile laboratory to track emissions in order to evaluate the concentration of roadway particles at different speeds of the vehicle. They found an increase in the mean concentrations only from 80 to 110 km/h, but with high standard deviations. Pirjola et al. (2009[168]; 2010[169]) found a linear increase in dust concentrations, measured behind the rear tyre of two testing vehicles, exploring the range 50-80 km/h, but no emission factors were calculated. Hussein et al. (2008[170]) found an important dependence of road dust emissions on vehicle speed when studded tyres are used: the particle mass concentrations behind the tyre at 100 km/h were about 10 times higher than that at 20 km/h. Although these studies were not able to attribute the speed dependence to direct wear and/or resuspension emissions, it is likely that results of Pirjola et al. (2009[168]; 2010[169]) and Hussein et al. (2008[170]) are to a large extent due to road/wear and resuspended dust, given the importance of these sources in Scandinavian countries. However, the speed dependence could also be attributed to the use of studded tyres. Interestingly, they eliminated from their studies the high PM concentrations observed during braking and hard acceleration (>0.5 m/s^2), which suggests that acceleration may also play also a role in road dust emissions.

Etyemezian et al. (2003[171]) and Zhu et al. (2009[172]) found that on the same road (i.e. same dust loading) emissions increase with vehicle speed to the power of approximately 3. More recently, Amato et al. (2017[173]) investigated the impact of traffic speed on road dust emissions in Milan. They used vertical deposition fluxes and calculated the emission factors at three sites along the same road with different instantaneous traffic speeds. Emission factor values were 24.6, 40.9 and 48.4 mg/VKT for instantaneous traffic speeds of 36, 47 and 57 km/h, respectively, which suggests that road dust resuspension increased with a power of 1.5 of vehicle speed. The 1.5 exponent is lower than that reported by Sehmel (1973[174]), who found that resuspension increased with the square of the car speed using fluorescent silica as tracer, but higher than the estimates of Nicholson and Branson (1990[175]), who suggested that only around 20% increase of emissions from 36 km/h to 57 km/h, using zinc sulphide as tracer. The difference between these estimates can be due to several factors, such as the amount, type and age of road dust, type of road dust (tracers were used by the aforementioned studies), road pavement and type of vehicles.[22]

Vehicle size/type

The fact that larger vehicles provoke higher road dust resuspension belongs to our everyday experience: on an unpaved road it can be readily observed that more dust is uplifted by a truck than a car.[23] Assuming higher resuspension for larger vehicles also on paved roads seems logical. Emission factors for HDVs are about 10 times higher than for LDVs. A similar estimate, namely a ratio of HDV over LDV of 9, was adopted by Schaap et al. (2009[176]) for modelling road dust emissions over Europe.

Whether such difference is only due to the larger size of the vehicle or also to the heavier load, is an open question, which needs further research. From an aerodynamic perspective only size matters, but heavier loads may enhance the tyre-induced lifting forces on deposited dust, which would enable more particles to be re-entrained in the air. In fact, the U.S. Environmental Protection Agency (2011[68]) AP-42 model for estimating road dust emissions, uses vehicle weight as predictor variable, adopting a nearly linear relationship with the emission factor (power of 1.02). Düring et al. (2002[177]), used instead a 2.14 power law relationship. A linear approach was followed by Timmers and Achten (2016[178]) to estimate the increase of road dust emissions due to the extra weight of electric vehicles.

Road dust loading

Road dust emissions also depend on the amount of material deposited on the road. Several empirical formulas were obtained relating emission factors with the amount of dust as silt loading (<75 μm) or thoracic dust loading (<10 μm) following generally a power law relationship (Amato et al., 2011[53]; Cowherd and Englehart, 1984[179]; Düring et al., 2002[177]; U.S. Environmental Protection Agency, 2011[68]) alone or in combination with vehicle weight. The variance of emission factors explained solely by the road dust loading is generally low, indicating that other factors, such as vehicle weight and speed are influencing emission rates. The power law relationships found generally have exponents lower than 1, indicating that changes in road dust loading are less influential than equivalent changes in vehicle weight (linear relationship) and speed (exponent varying between 1.5 and 2).

Road dust loading is a road property which in general corresponds to a steady-state condition between a complex combination of production, loss and redistribution processes. Production processes are mainly due to traffic intensity (i.e. wear and exhaust particle generation), atmospheric deposition, dust sources on the roadside, road shoulder, sanding/salting in countries with a lot of snowfall, the presence of additional fugitive sources (building and maintenance activities), and the deposition of pollen and other organic materials. Loss processes are mainly the resuspension itself – traffic or wind-induced – drainage, and road cleaning. Redistribution processes are particle crushing, aggregating and migration. Most of the aforementioned processes are heavily affected by road surface conditions, such as age, state, composition, texture, porosity and moisture.

Given the wide range of influencing factors, a wide spatial variation of road dust loadings has been observed at various scales. Important differences were found comparing Southern and Central European cities pointing at the presence of uncontrolled fugitive sources, lower vegetative cover and lower moisture as main responsible factors. Important variations were also found at the urban scale, with lower loadings generally found in high-speed roads, average values at urban roads and higher loadings at sites affected by construction activities, or next to unpaved areas (Amato et al., 2009[43]; 2011[53]; 2016[13]; 2017[173]).

Amato et al. (2011[53]; 2012[180]; 2014[18]; 2016[13]) investigated the source apportionment of the thoracic fraction (less than 10 μm) of road dust in several European cities, separating from 3 to 4 sources depending on the city. In the 2011-2012 studies (Northern Spain,

Netherlands and Switzerland), the main sources were related to tyre wear (16-38%), brake wear (27-44%), motor exhaust (5-20%), and mineral dust (13-37%) which likely involved road wear, soil and building dust, but could not be disentangled due to the chemical affinity of these sources. In Southern Spain, a carbonaceous factor (associated with tyre wear, motor exhaust and worn bitumen from asphalt) was found to dominate the mass (50% as average), while lower contributions were found for worn mineral particles from asphalt (20%) and brake wear (12%) on average (Amato et al., 2014[18]). In Paris, the carbonaceous factor was responsible for only one third of road dust mass, and the rest was equally apportioned between brake wear and road wear (Amato et al., 2016[13]).

Conversely, very little is known in terms of the time variability and seasonality of effects. Kantamaneni et al. (1996[70]) found that the addition of traction sand material on the road increased the PM10 emission factor from 1.04 to 1.45 g per vehicle-kilometre. Moreover, when roads were sanded, the correlation found between emission factors and relative humidity was not observed. On the other hand, Amato et al. (2013[181]) found that the 1-month variability of PM10 fraction of road dust was determined solely by rain, confirming that road dust loading can be seen as a constant feature of the road, which deviates from the equilibrium value only when there is precipitation or extraordinary dust intrusion (construction or desert dust).[24]

Porous pavements

Porous pavements are mainly used to reduce tyre-pavement noise and to increase water drainage from the surface. These pavements have been reported to decrease resuspension of road dust (Costabile et al., 2017[182]; Gehrig et al., 2010[52]) through reducing the amount of dust on the road surface. Direct wear emissions do not seem to be affected by porous pavements. Stationary and mobile measurements in Stockholm showed that the difference in PM10 emissions between the pavements (porous and reference), with studded tyres in use, was around 15% (Gustafsson and Johansson, 2012[152]). In a more recent study, no effect on PM coarse emission factors was observed after two years of implementation of porous asphalt on a Swedish highway (Norman et al., 2016[183]). One of the possible reasons was the higher wear rate of porous asphalt under studded tyre conditions, which makes it difficult to extrapolate their result to other regions.

Vehicle underside

It has been argued that a flatter undercarriage should improve the aerodynamics of the car and reduce resuspension. The only reference found on this matter is relative to emissions from unpaved roads (Gillies et al., 2005[66]), where it was concluded that the vehicle undercarriage area and the number of wheels have weak and no discernible relationships with emission factors. However, today' cars, and especially electric ones, have likely flatter undercarriage, thus more research is needed in order to evaluate the impact of this on the emission factor.

Weather conditions

Ambient air humidity and wind speed have the potential to influence emission factors from resuspension. Ambient air humidity directly affects road moisture, which is probably the most important determinant of the temporal variability of emissions (at least in countries where no studded tyres are used and no road sanding/salting is deployed). On urban paved roads, a negative correlation between road humidity and dust emission factors has been found. Kantamaneni et al. (1996[70]) estimated that a change of relative humidity from 10 to 70% was associated with a reduction of the road dust emission factors from 2.5 to 0.5 g/km, with relative humidity explaining 41% of the PM10 emission factors variance.

Wind speed increases emissions, but it has been difficult to isolate its impact. Amato et al. (2012[180]) identified a negative correlation between wind speed and road dust loadings by means of Principal Component Analysis (PCA). Thorpe et al. (2007[184]) investigated the effect of wind speed on road dust emissions at two sites in London. At the Bloomsbury background site, they found evidence of an increase in resuspension emission factors with wind speed, with the gradient appearing to diminish at higher wind speeds (above 5 m/s). The Bexley site suggested instead a different dependency, with emission factors appearing to decrease initially with increasing wind speed (below 3 m/s) before increasing during periods of high wind speed. Charron and Harrison (2005[185]) also suggested that PM2.5–10 concentrations were elevated as wind speed increased, based on the data collected at Marylebone Road, suggesting that road dust entrainment due to wind is apparent when a speed threshold (around 7 m/s) is reached.

Once airborne, the dispersion of non-exhaust particles in the atmosphere will depend mostly on atmospheric conditions (wind speed, direction and turbulence). The main removal mechanisms are dry and mostly wet deposition. Both removal mechanisms depend on the physico-chemical properties of the particles including size, shape, hygroscopicity and chemical composition.

Table 2.8 summarizes qualitatively the current knowledge on the impact of different vehicle, road and weather features, as well as driver choices, on each component of non-exhaust emissions. A preliminary attempt of estimating the share of emissions due to road or vehicle/driver features is presented at the bottom of the table based on the weight that each factor likely has in the literature.

Table 2.8. Overview of the influence of vehicle, driving and road features and weather conditions on non-exhaust emissions

	Brake wear	Tyre wear	Road wear	Resuspension
Vehicle features				
Rotor temperature	↑			
Vehicle size	↑	↑	↑	↑
Vehicle weight	↑	↑	?	?
Metal content in brake pads	↑			
Studded tyres		↑	↑	↑
Tyre diameter		↓		
Tyre width		↓		
Tyre tread depth		–		
Vehicle undercarriage				?
Tyre rolling resistance		?	?	
Tyre tread wear rating		–		
Mileage	?	↓		
Road features				
Allowed max speed	↑	↑	↑	↑
Allowed max weight	↑	↑	↑	↑
Pavement age/state		?	↑	↑
Resistant ballast rocks		↑	↓	?
Size of stones for road pavement conglomerate			↓	↓
Asphalt porosity		?	?	↓
Rubber asphalt		?	?	?

Cement concrete pavement		?	↑	?
Road dust loading			↑	↑
Road moisture		?	↓	↓
Side-slip angle	?	↑	?	?
Driving features				
Aggressive driving style	↑	↑	↑	↑
Speed	↑	↑	↑	↑
Acceleration/Deceleration	↑	↑	?	?
Tyre pressure		↓		
Wheels imbalance		↑		
Weather conditions				
Temperature	?	↑	?	↑
Humidity	?	?	?	↓
Precipitation	?	↓	?	↓
Wind speed				↑
	Brake wear	Tyre wear	Road wear	Resuspension
Estimated share due to road features	10%	30%	60%	70%
Estimated share due to vehicle/driving features	90%	70%	40%	30%

Note: ↑ indicates a positive effect (increases emissions); ↓ indicates a negative effect (decreases emissions); − indicates an insignificant effect; ? indicates that while there is likely an effect, it is unknown due to lack of evidence or mixed findings; empty cells indicate a hypothesis of no effect.

2.6. Implications for policy

More than 100 source apportionment studies reveal that, on a global level, non-exhaust emissions contribute similarly to exhaust emissions to ambient air PM2.5 and more than exhaust emissions to ambient air PM10. Road dust resuspension is the most polluting source of PM10 from road traffic, and the second most polluting source of PM2.5 (behind exhaust emissions). Moreover, emission trends and projections reveal that non-exhaust emissions may have already surpassed primary exhaust emissions of both PM2.5 and PM10, and their relative contribution to total emissions from road traffic is likely to continue increasing.

The impact of non-exhaust emissions is particularly severe in countries where studded tyres (and traction sand) are used to improve friction under snow/ice conditions. Beside these extreme conditions, maximum contributions to PM10 are found in India and China, while lower, but still significant, concentrations are recorded in European cities, with Mediterranean countries seemingly more affected than Central Europe. In the United States, non-exhaust sources' contributions to PM2.5 have been found to be lower than China, India and Spain.

More than 50 epidemiological studies have investigated the health outcomes associated with non-exhaust PM exposure indicators. Most of the studies used elemental tracers while a few applied source apportionment methods. Results indicate that exposure to PM emissions, and PM2.5 in particular, is associated with a variety of short- and long-term health effects. These impacts come in the form of increased risk of cardiovascular, respiratory, and developmental conditions, as well as overall mortality (Amato et al., 2019[105]).

Such evidence implies a need for immediate policy action to mitigate non-exhaust emissions and prevent their consequences for air quality and public health. In order to identify the most important drivers of non-exhaust emissions, a comprehensive literature review on the impact of different road, vehicle, driver and weather features has been carried out. Among the first three categories, vehicle weight, speed and acceleration, brake, tyre and pavement type, road moisture and road dust loading have been identified as the most important determinants of non-exhaust emissions.

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[1] For a review of the literature, see Table 2.1, and for the estimations reported in this analysis see Tables 3.4 and 3.5.

[2] The LOWBRASYS has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 636592. The project is managed by the Innovations and Networks Executive Agency (INEA) of the European Commission.

[3] According to a JARI study published at the 48th PMP Meeting, the share of PM10 in brake wear emissions may be lower than the values mentioned above.

[4] A laboratory road simulator is a piece of equipment, which can be used to generate and study abrasion particles that are formed by the interaction between the tyre and pavement.

[5] Fauser et al. (2000[189]) suggested and used a method to identify bitumen in aerosols. They found that asphalt is the only source contributing to organic molecules with a molecular weight more than 2000 g/mol.

[6] The size distribution of road dust particles is also affected by existing sources, but is generally characterised by a coarse size distribution with size mode well above 10 µm, typical of any crustal material (Bi, Liang and Li, 2013[195]; Escrig et al., 2011[58]; Fedotov et al., 2014[196]; Janhäll et al., 2016[197]; Padoan, Romè and Ajmone-Marsan, 2017[198]; Ramírez et al., 2019[199]).

[7] The AP-42 model for estimating PM10 emissions from paved roads was developed by Midwest Research Institute under contract with the U.S. Environmental Protection Agency (Cowherd and Englehart, 1984[179]; U.S. Environmental Protection Agency, 2011[68]). The latest version of the model expresses the emission factor (EF), from a paved road in terms of the silt loading (sL), mean vehicle weight (W), days of precipitation (P) and total days (N) as follows:

$$EF = k\,(sL)^{0.91} \times (W)^{1.02} \times (1 - P/4N)$$

where k is a particle size multiplier for particle size range and units of interest. This formula was obtained by a least squares regression between observed and predicted EFs using a dataset consisting of about 60 observations for a variety of roads ranging from public paved ones to unregulated industrial ones.

[8] Finland likely also includes resuspension within the "road wear" category in their inventory, as the emission factor used for road wear in the Finnish inventory is significantly higher than that in neighbouring countries that do not include resuspension.

[9] See also the U.S. Environmental Protection Agency website: https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei. Additional years before 2014 are available but need to be calculated with the Motor Vehicle Emission Simulator (MOVES) model. While road dust emissions are clearly labelled as "Paved road dust", brake wear and tyre wear emissions are included in the "on-road mobile" categories and can be retrieved only by running the MOVES software, which uses emission factors in combination with activity (mileage) data.

[10] Since 2012, the road dust emission factor is corrected for precipitation, and brake and tyre wear have been subtracted from road dust to avoid double counting with mobile emissions (California Air Resources Board, 2019[71]).

[11] PMF offers multiple advantages, but cannot easily separate road wear from road dust resuspension.

[12] See also http://www.who.int/gho/phe/outdoor_air_pollution/burden/en.

[13] (Atkinson et al., 2016[191]; Basagaña et al., 2015[192]; Burnett et al., 2000[193]; Cahill et al., 2011[194]; Cakmak et al., 2009[200]; Franklin, Koutrakis and Schwartz, 2008[201]; Li et al., 2014[202]; Lippmann et al., 2013[203]; Ostro et al., 2007[204]; Ostro et al., 2008[205]; Son et al., 2012[206]; Valdés et al., 2012[207]; Zhou et al., 2011[208])

[14] Badaloni et al., 2017[209]; Beelen et al., 2015[210]; Lipfert et al., 2006[211]; Lippmann et al., 2013[203]; Ostro et al., 2010[212]; Ostro et al., 2015[213]; Thurston et al., 2016[214]; Vedal et al., 2013[215]; Wang et al., 2014[216]; Wang et al., 2017[217].

[15] Suh et al., 2011[113]; Tiittanen et al., 1999[114]; Sun et al., 2016[115]; Samoli et al., 2016[116]; Zanobetti et al., 2009[117]

[16] Basagaña et al., 2016[218]; Bilenko et al., 2015[219]; Crichton et al., 2016[220]; Dadvand et al., 2014[221]; Eeftens et al., 2014[222]; Fuertes et al., 2014[223]; Hampel et al., 2015[224]; Lagorio et al., 2006[225]; Pedersen et al., 2016[226]; Raaschou-Nielsen et al., 2016[227]; Tonne et al., 2016[228]; Willers et al., 2013[229]; Wolf et al., 2015[230].

[17] The advantage of small-size testers is typically related not only to cost but also to a considerably higher accuracy of detected physical variables compared to large-scale and field tests and often allows for a better understanding of wear mechanisms.

[18] See https://www.eng.auth.gr/mech0/lat/PM10.

[19] For tyre wear, the EMEP/EEA inventory guidebook attributes an approximately 60% higher emission factor to light duty trucks than passenger cars, and a linear increase of tyre wear emissions with the percentage of HDV load.

[20] In the extreme braking condition, locked wheels yet still moving forward, the value of the longitudinal slip reaches $\lambda = -1$. In the extreme acceleration condition, standing still while the tyre is spinning, the value of the longitudinal slip reaches $\lambda = 1$. In normal conditions, only the range $-0.3 \leq \lambda \leq 0.3$ is of interest.

[21] It has been argued that the tread wear rating (TWR) provided on the sidewall of the tyre and marking the expected durability of the tyre, could be a measure of tyre wear potential. Grigoratos et al. (2018[150]) discovered that, in general, the tyre tread mass loss shows no obvious statistical relation to PM10, PM2.5 or particle number concentration. A higher tread mass loss does not imply higher PM or PN emissions, since the size distribution of tread wear is very coarse and PM10 fraction is only 1%. Particle number is poorly correlated to mass, as it depends mostly from ultrafine particles which have negligible mass. Tires of the same tread wear rate but of different brands showed different behaviour in terms of material loss, PM, and PN emissions under the selected testing conditions. This means that it is not feasible to categorise tyres of different brands in terms of their emissions based on their TWR.

[22] Denby et al. (2013[55]) specified a quadratic dependence on speed for spray (water, dust and salt) emissions in the NORTRIP emission model but did not specify it for road dust suspension, stating that the model remains uncertain and requires further refinement based on experimental studies.

[23] Gillies et al. (2005[66]) estimated a linear relationship of 3 mg per kg of weight (using it as a proxy of size) and vehicle-km on an unpaved road, using vertical profile measurements of mass concentration at three instrumented towers. This is several orders of magnitude higher than Simons' (2016[137]) estimates for wear emissions. The reason for this is that Gillies et al.'s (2005[66]) study draws on measurements on unpaved roads, where the road dust reservoir is the road itself.

[24] For example, Amato et al. (2012[180]) calculated that a Saharan dust intrusion event in Barcelona, increasing ambient air PM10 concentrations by 6 µg/m3 on the daily average, provoked a 35% increase of dry deposition flux and a 30% increase in the instantaneous mobile road dust load.

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