

Review



Particulate Matter from the Road Surface Abrasion as a Problem of Non-Exhaust Emission Control

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Abstract: Along with house heating and industry, emissions from road traffic (exhaust and tire, brake, car body or road surface abrasions) are one of the primary sources of particulate matter (PM) in the atmosphere in urban areas. Though numerous regulations and vehicle-control mechanisms have led to a significant decline of PM emissions from vehicle exhaust gases, other sources of PM remain related to road and car abrasion are responsible for non-exhaust emissions. Quantifying these emissions is a hard problem in both laboratory and field conditions. First, we must recognize the physicochemical properties of the PM that is emitted by various non-exhaust sources. In this paper, we underline the problem of information accessibility with regards to the properties and qualities of PM from non-exhaust sources. We also indicate why scarce information is available in order to find the possible solution to this ongoing issue.

Keywords: ambient particulate matter; street dust; exhaust vs. non-exhaust emission; heavy metals; health hazard

1. Introduction

Road traffic emissions caused by both exhaust and non-exhaust sources contribute significantly to the particulate matter (PM) concentration in an urban atmosphere [1–22]. Additionally, very fine particles that are emitted through various road traffic-related processes (e.g., brake wear), can penetrate the human organs [23–25]. A knowledge of physicochemical properties and PM sources is crucial for determining the environmental effects of PM [26–30].

Most of the PM mass, where PM is created by the abrasion of a car body, tires, or a road surface, is composed of particles with diameter ranging between 1 and 10 μ m [9,31–35]. For this reason, coarse PM (fraction of atmospheric particles with an aerodynamic diameter in the 2.5–10 μ m range; coarse PM) usually makes up most of the PM mass close to crossroads. In Switzerland, the PM₁₀ in an urban street canyon may result from brake-disc abrasion (21%), resuspension (38%), or exhaust emissions (41%), while the PM₁₀ found along a freeway consists mainly of resuspension dust (56%) and particles emitted by exhaust emissions (41%) [36]. Both the PM_{2.5} (fraction of atmospheric particles with an aerodynamic diameter not exceeding 2.5 μ m; fine PM) mass share of PM and the ambient concentration of PM_{2.5} in the vicinities of roads are strongly affected by exhaust emissions than by non-exhaust emissions [11,12,20,37]. When compared to PM_{2.5} from other areas, the effect of car fume emissions can mainly be seen in an increase in soot and some organic compounds in the PM_{2.5} mass [3,4,6,11–13,20,21].

Most of the PM mass from non-exhaust emissions, regardless of the type of traffic site (road, crossroad, or highway), is made of resuspension dust. This is a mixture of particles derived from

car body abrasions, brake pads, brake discs, tires, and road surface; and, on the other hand, from soil and particle elements that have settled on the surface of the road (originating from sources other than road traffic) [9,11,12,33,34]. These particles are usually enriched with various organic compounds [15,20–22,38]. In order to reduce the impact of resuspension dust on air quality in urban areas, appropriate methods are employed. For instance, for the prevention of PM resuspension, hardening, unpaved roads may be exposed to chemical-binder agents, as well as water-spraying in dry periods [39–41]. Frequently cleaning the surface via washing and sweeping is a practice used for paved roads. The reduction of PM emissions can also be achieved by limiting both vehicle speed and mass or by using a noise barrier on the roads (e.g., a green-wall sound barrier). The effectiveness of applied methods varies depending the frequency at which particles are removed and local meteorological conditions. The reduction of resuspension dust can reach an efficiency of up to 90% [39–41].

In this work, we demonstrate that the isolation of particles that only results from road surface abrasion from resuspension dust particles is particularly difficult. However, this step is necessary for the proper quantification of non-exhaust emissions in a particular area.

2. Non-Exhaust PM Emissions and Their Relation to Air Pollution in the Vicinity of Roads and Crossroads

Non-exhaust sources of PM include:

- Tires: PM from tire abrasions comprises among others, metals: Cd, Cu, Pb, Zn [33,42–44] and organic compounds such as natural rubber copolymer, organotin compounds, and soot [9].
- Brakes: PM from brake-pad and brake-disc abrasion consists of metals: Zn, Cu, Ti, Fe, Cu, Pb [33,45–51] and other specific compounds such as sulfate silicate, barium sulfate, carbon fibers, and graphite [9,52,53].
- The car body: some particles from the vehicle's consumable parts are released into the air; they may contain small amounts of metal like Zn and Fe [35].
- The road surface: PM from road-surface erosion containing characteristic compounds such as bitumen, cement, and resins [33].
- Paints: the composition of the paint intended for road-surface painting suggests that PM from this source may contain Pb and Ti [54].

It is clear that the level of PM air pollution resulting from non-exhaust sources at roads is dependent on traffic, speed as well as the shape and system of the road interchanges.

Released particles are permanently mixed coming from combustion, industry, exhaust emissions and soil [9,55–59]. PM from anthropogenic sources include particles from so-called municipal emissions (mainly soot, organic matter, and inorganic salts resulting from the combustion of coal and biomass in domestic furnaces, local coal-fired boiler plants, or heat plants [20,60–62]) and from industrial emissions (mainly fine particles enriched in heavy metals or persistent organic pollutants [63–68]). In addition, particles of salt, sand (coarse PM), or a mixture of both are released into the urban atmosphere, especially during the winter season [9,25,35,40,69]. In built-up areas, e.g., city centers, compact residential and service buildings close to traffic arteries significantly reduce air-mass exchange; this results in the accumulation of PM in the ground atmosphere layer [11,12,19,55,56]. The non-exhaust-based particles and particles from other sources creates complex chemical PM mixtures near traffic sites [11–13,21,33,35].

PM-bound elements, including some toxic metals, are subjected to similar phenomena. The ambient concentration of PM-bound elements in the areas affected by road-traffic emissions also depends on the traffic intensity, the vehicle-fleet characteristics car type and speed, the type of road surface, the road-cleaning intensity, and the concentration of PM components in the so-called urban background [70–73]. Given that, it is easy to imagine the difficulty in quantitatively dividing PM-bound elements, located in urban sites that are influenced by traffic emissions, into specific source groups.

Meanwhile, it is extremely important to clearly distribute these element mixtures as such a distribution constitutes the first and most important step in programs and scenarios that aim to reduce PM concentrations in areas characterized by high ambient concentrations of PM. The physical removal of elements from road and street surfaces via washing and sweeping does not sufficiently limit human exposure to PM resulting from traffic emission [55]. The complex process of overlapping the so-called urban background onto road traffic pollutants, along with the associated physical changes and chemical reactions, lead to the conclusion that PM close to roads may be much more toxic or may have a higher carcinogenic potential than PM outside such areas [11,12,20,70,74–77].

Using data derived from several European cities, Querol et al. [78] estimated that PM emissions from road traffic was roughly distributed between exhaust and non-exhaust emission sources. Similarly, in Berlin, Lenschow et al. [79] showed that in areas where air quality is determined by road traffic emissions (on urban roads) half of the PM_{10} mass comes from non-exhaust sources. Studies conducted in Poland in the last decade clearly showed that PM emissions at roads and crossroads [11–13,37]. This contribution was apparent even during smog episodes, when emissions from the combustion of fossil fuels in domestic furnaces determined the air quality in the city of Zabrze [19].

In this respect, the contribution of PM emissions from non-exhaust sources is large, certainly much larger than was previously thought. Nevertheless, we still lack detailed information [75,80–83]. Data and rationales are missing for the reliable subdivision of PM emissions into those from exhaust vs. those from non-exhaust sources, for the separation of road-traffic related PM from PM related to other sources [20,62], and, above all, for the segmentation of PM generally derived from non-exhaust sources into specific source groups (tire abrasion, brakes, road surface, etc.).

The simplest way of subdividing road-traffic related PM (though this does not apply to PM-bound elements) into specific sources is to assign a particular PM size, or rather particles belonging to a corresponding size range (i.e., size fractions), to specific sources; these compartments are characterized by so-called lower and upper limit cut-off diameters [84–86]. Many research papers are devoted to investigating the fractional composition of particles related to specific road-traffic sources. In general we can say that non-exhaust emissions mainly include coarse PM, while PM from exhaust emissions consists of fine particles belonging to $PM_{2.5}$ [34,87–89]. If we consider size when examining the distribution of PM particles in non-exhaust emission sources, particles of worn road surfaces usually belong to the coarse PM, while particles from the tires and brake discs are both coarse and fine [90]. In northern European countries, where various methods are used during winter to prevent icing on roads, and winter tires and snow chains are commonly used, $PM_{2.5-10}$ accounts for up to 90% of PM in atmospheric air [73,91]. However, brake discs were selected to be examined, as their usage is the most detailed, in terms of the amounts of emitted particles (emission factors) and their properties [9,33,55,80,87,92,93]. The identification and quantification of PM from other non-exhaust sources are much more difficult. The list of published papers describing the PM emission factors and properties of PM emitted by these sources is therefore much shorter. For example, it was shown that particles with a diameter ranging from several hundred nanometers to several tens of micrometers are produced during the brake-lining and disc-abrasion processes [33]. On the other hand, there are scientific reports showing that non-exhaust emissions (including brake wear) also produce nanoparticles [23–25]. The size of emitted particles depends primarily on the physical properties, shape and structure, and the chemical composition of the abrasive/erosion material, on the nature, value, and complexity of the forces acting on the material, and on the temporal and spatial variability of these parameters [94]. It should be clearly stated that the above-mentioned assignment of specific PM emission sources to appropriate size ranges is contractual and far from sufficient for separating PM into specific emission sources.

Rather than using particle-size distribution, a far more reliable way of achieving this separation involves examining the chemical composition (relatively elemental composition) of PM particles and comparing it with the chemical compositions of particles directly emitted by each of the individual

emission sources [52,95–97]. These were widely described as so-called elemental PM profiles from both exhaust and non-exhaust emissions. Prior to the introduction of a law prohibiting the use of leaded petrol, Pb was a PM marker related to the combustion of gasoline in engines [98]. PM markers from working catalytic converters are metals from the platinum group such as Rh and Pt [99–101]. Zn, Cu, and Ti in urban air are often related to brake-pad erosion [33,47]; particles from brake-disc wear are typically characterized by Fe, Cu, Pb, and Zn [33,45,46,48–51]. To identify PM from tire abrasion, Cd, Cu, Pb, and Zn may be monitored, among others [33,42]. Occasionally, metals such as Fe and Zn are also linked to the corrosion of car-body parts [35].

The most difficult data to find involves the elemental composition of PM arising from the abrasion of road surfaces. The root of the problem lies in the nomenclature given to the material emitted directly by the worn road surface and to the material laid on road and then resuspended. Often, the same particles are attributed to several sources [20,33,62,102].

Fauser (1999) identified asphaltenes and maltenes as potential tracers for road-surface wear [103]. Kupiainen et al. (2003) used the presence of hornblende as a tracer for PM derived from road-surface wear [44]. However, this was in a controlled laboratory test, where the road-surface material constituted the only possible source of this mineral. Polycyclic aromatic hydrocarbons (PAHs) have been identified in bitumen samples, although the concentrations of individual PAHs are very low due to their removal during the distillation process [104]. The lack of a PAH compound unique to bitumen makes such compounds unusable as marker species for road-surface wear [105]. A number of metals have been detected in road bitumen samples including V, Ni, Fe, Mg, and Ca [48,106]. However, a comparison of concentrations in road bitumen with raw bitumen samples revealed much higher concentrations of these metals in the former, indicating that the road-surface material had incorporated these elements from other sources [48]. In conclusion, it may prove very difficult to identify suitable tracer species for road-surface wear [33].

It seems that we lack reliable information on the PM or dust emitted by different road surfaces' erosion processes, and this constitutes a significant gap in the current state of knowledge of non-exhaust sources of road traffic emissions. Such information should include: (i) the fractional composition data (ii) the chemical or elemental composition of the PM /dust particles emitted during the erosion of different road surfaces.

3. Types of Road Surfaces and Their Importance in the Non-Exhaust Emissions of PM

Road pavements can be classified by their construction, deformability, load-bearing capacity, and material types used for the driving layer. Due to the surfaces' deformability, they can be divided as follows:

- susceptible: surfaces with a structure that deforms plastically under the influence of loads (sett, gravel, and bitumen surfaces located on susceptible substrates).
- semi-rigid: asphalt surfaces with a foundation made of concrete, lean concrete, aggregates, or stabilized soils.
- rigid: surfaces with a structure that deforms elastically under a load (cement concrete surfaces) [107–109].

The surface's wearing course can be made of the following mineral-asphalt mixtures:

• asphalt concrete (AC), stone mastic asphalt (SMA), mastic asphalt (MA), very thin-layered asphalt concrete (BBTM), and porous asphalt (PA) [110].

A topcoat made of cement concrete can be made in the form of roofed, doweled and anchored, or reinforced plates [111].

Even without any measurements and tests, it is generally recognized that vehicles moving on unpaved roads will generate a significantly greater amount of PM than paved or hardened roads. In particular, the generation of high levels of dust is linked to ground and gravel made of crushed debris and slag roads. In comparison to paved roads (stone-paved, concrete, or asphalt roads), the top layers of unpaved roads (especially particles with a small diameter) move, as a result of the friction of the wheels on the surface. Loose surface grains move under the mechanical stress of the wheels. The free material is lifted under the effect of the vacuum that is generated by the tire surface breaking off from the road surface. Equally, dust is raised from the road surface onto the wheel tread [35,112,113]. However, concerning the impact on air quality unpaved roads are less interesting than paved roads. First of all, the chemical composition of the PM or dust particles emitted by unpaved roads is well known, and it is equal to the composition of soil in the case of forests for example, or to gravel and sand in the case of gravel roads for example [114]. Consequently, in terms of chemical properties, the PM or dust emitted during the erosion of such roads is almost identical, in a given area, with soil particles or the top soil layer [33]. The content in such particles may depend only on the degree of contamination of the soil or sand. Therefore, it basically depends on the road localization [115]. Road dust and roadside soil often contain metals, including Pb, Cu, Cd, and Zn, indicative of contamination by road traffic emissions. Through the calculation of crustal enrichment factors (CEFs), the presence of contaminants in road dust and roadside soil provides a means by which particles arising from anthropogenic sources can be separated from natural or crustal sources [11,12]. However, comparing the contribution of road-surface wear to that of resuspended road dust will require the development of an alternative approach [33]. Additionally, unpaved roads are usually located outside urban centers, in less-populated areas such as villages, small municipalities, and around small housing estates (access roads). They are therefore of little importance when compared to the state of air quality in city centers, where PM concentrations matter to authorities and sanitary services. Furthermore, it seems that the impact of unpaved-road emissions on the quality of atmospheric air is difficult and, almost impossible to estimate due to the high variability in time [116].

This is not the case for paved roads. First of all, they are built from mixes that are produced through strictly-defined processes. Their composition must therefore differ from the composition of natural components such as soil or sand. In addition, they are made of road surfaces where vehicles are able to move at high speeds, and varied with types ranging from compact passenger cars to several-ton trucks on highways. It seems, therefore, that the size of PM or dust particles generated during erosion may fit within a wide range Their composition determined by the structure of building material's original mixture may also vary depending on the place (road age, renovations, degree of wear, etc.) [116,117].

Most paved road surfaces have the same base: a mixture of aggregates such as bitumen or cement with different grain sizes, and modifiers such as fillers and binders [35]. The choice and proportions of the ingredients dictate the exact differences in chemical composition of mixtures made for the production of road surfaces [118]. Road surfaces can be widely classified as being made of either concrete or asphalt. Asphalt consists mainly (~95% of the mass) of mineral aggregates and various geological materials [93]. Its remaining content is mostly bituminous binder, modified by the addition of fillers and adhesives [106]. The bitumen contains many thousands of high-molecular-weight organic compounds (about 500–50,000 u), most of which are aliphatic and aromatic hydrocarbons [33].

As a result of increasing requirements of road surfaces' strength parameters, the continuous growth in their heavy traffic load, and the fact that extreme weather conditions occur at various latitudes, diverse types of modifiers are introduced into the bitumen-bonding compound or asphalt mix. This aims to improve both the properties of the road surface and the bond between the binder and mineral components. Adhesives are joined to aggregates using fillers and reinforcing fibers such as glass, fly ash, and shredded used tires [33,119]. This solution is increasingly prevalent due to problems related to the utilization of tires. Polymers, epoxy resins, and low-carbon steel are used as modifiers, while sulfur is used to increase the rigidity of the binder [33].

Concrete pavements are made of mineral aggregates, sand, and cement [35]. The literature provides little information on the chemical composition of concretes that are used to produce roads and on the dust emitted by such surfaces. This is related to the fact that there are many ways of choosing the composition and proportion of materials used to make a concrete mix. As a consequence there is

no universal molecular formula for the concrete mix and therefore for particles emitted during the erosion of such road surfaces [33].

When concrete pavements are made, many different additives affect the strength of the material when they are introduced into the mix. Due to the fact that the structures are designed for a maximum period of 30 years, old concrete surfaces are significantly damaged and therefore not always adapted to heavy traffic and vehicle loads. This may result in emissions of PM and dust into the atmosphere, not only from the base-surface components released during the vehicle's movement but also from the processes and materials used for the surfaces' temporary repairs [120].

In order to promote the use of cement and concrete in the transport infrastructure, the non-profit European Concrete Paving Association (EUPAVE) was established in 2007. Their mission is to advocate and enable the wider use of cement and concrete applications in European transport infrastructures. They are involved in engagement with EU, national, and local decision makers, disseminating technical know-how, and presenting the benefits of using this material. According to EUPAVE, the use of cement and concrete in road-building implements a circular economy. Considering the hierarchy of waste management, in accordance with the European Commission's recommendations (prevention, reuse, recycling, recovery, and utilization), it seems obvious that construction using concrete, adheres to these principles. Concrete pavements have always been valued for their durability and low-maintenance costs, which are simple ways of obtaining ecological benefits [120].

Surface type also determines the wear of vehicles that move on it [35] The volume of dust emissions from used car tires in Britain in 1996 was 5.3×10^7 kg, while in Japan it was 2.1×10^8 kg in 2001. Within a year, Germany's emissions range between 55 and 657 kg·km⁻¹, depending on the type of road [121]. The asphalt surface causes less tire abrasion than concrete surfaces. In Arizona (in the United States), PM emissions are 1.4 to 2 times lower for asphalt roads than for concrete roads [9,122]. However, it is important to note what type of tires are used. Numerous studies have shown that winter tires generate much more dust, and this is mainly due to greater friction on the ground compared to their summer equivalents [9,35,123–126]. Furthermore, the use of metal constructions on tires, in the form of pins or chains, influences an increase in the level of road-surface abrasion [9,44,127]. In Sweden, studded tires are approved for use from October to April. The emission factor for PM₁₀ from tire abrasion close to roads and junctions is clearly higher during this period than in other months [127].

Finally, the emission of particulate matter by road-surface abrasion is difficult to isolate in PM and dust tests in the field, and such test, which aim to define its characteristics, are therefore performed under controlled laboratory conditions [36,44,125,128,129]. In many countries, practically no road with a wearing course of cement-based concrete existed until the second half of the 20th century. When this technology was reintroduced in the mid-1990s, there were many new modifications in the production of concrete. For example, some special additives, when introduced into the mix (e.g., silica), affect the surface strength. Because the surfaces are designed to last for about 30 years, there is now a noticeable increase in damaged old concrete surfaces, which are not always adapted to today's increase in traffic and vehicle loads. This can cause significant emissions of PM or dust into the atmosphere from both base-surface elements released during vehicle movement and materials used for the emergency repair of these surfaces [119].

4. Conclusions

Current research shows that direct road-surface abrasion is of minor importance when the road is undamaged and that emissions of PM from direct road-surface abrasion are then significantly lower than emissions from other sources of road dust like abrasion of car body, brakes or tires [36,44,125,128–130]. Nevertheless, data on this subject is clearly lacking. This scarcity concerns both the availability of data on PM/dust emissions from road-surface abrasion in various places, and studies on PM/dust emission factors and related elements from different types of road surfaces as current research were restricted to asphalt surface only. The return to the technology of road construction and renovation with concrete in recent years is more challenging in terms of PM mass, composition and size ranges emitted from

abrasion roads in non-exhaust emission sources than results from current studies. The amounts of PM (emission factors) created during abrasion of concrete surfaces for different types of vehicles are not known, what is more, a chemical composition of particles released during this process is also unknown. The constant concrete upgrades aimed at their mechanical improvement can lead to the presence of dangerous elements in the environment. The increasing production of silica, used in concrete mixtures, could be a good example. Thus, more extensive research to establish emission factors and chemical composition of PM emitted from currently used road pavements are necessary. Such studies should be conducted under laboratory, controlled conditions for defining emission factors and chemical composition of PM (depending on vehicle type, speed, environmental conditions). On the other hand, analyzing the impact of abrasion emission reflecting realistic conditions is also very important. Defining how such emissions quantitatively and qualitatively change the character of PM near roads is crucial as it allows researchers also to assess PM parameters determining the power and scale of its influence in the environment. Gathering such information will allow for the revision of forecasts and scripts of PM impact on the environment and human health.

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