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Tire Particle Emissions: Demand on Reliable Characterization

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ABSTRACT: Tire emissions are recently becoming an important topic in environmental science and among visionary automotive engineers. Deep understanding of the mechanisms involved in tire wear and emissions and their reliable physical/chemical characterization should be assumed as an urgent task. With the aim to contribute to a trustworthy characterization of tire wear and emissions, this paper presents both (i) analysis of state-of-the-art in this field and (ii) a first experimental approach to the collection of particles from a rotating tire in a new developed test box.

KEY WORDS: tire particle emission, tire wear, tire particle characterization, dynamometer test, review

Introduction

Tires are not a negligible component of the car when talking about emissions and environmental impact. Either during its operation or when discarded, the amount of material abraded from tires is clearly a main factor of pollution. Furthermore, the contribution to PM_{10} (particulate matter [PM] such as PM_{10}, $PM_{2.5}$, PM_1, and $PM_{0.1}$ is defined as the fraction of particles with an aerodynamic diameter smaller than 10, 2.5, 1, and 0.1 μm, respectively) clearly exists (even when the literature is full of discrepancies) and will become a hot topic in the next years. The importance of individual sources in terms of their contribution and possible exposure to nanoparticles may become apparent in the future as vehicle exhaust emissions decline due to the use of renewable fuels and low sulfur fuels along with significant improvements in engine technology and exhaust after treatment triggered by the stringent exhaust emission standards [*1*]. Only particles smaller than about 10 μm will reach the alveoli. However, larger particles are deposited higher up in the respiratory system and

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removed on the mucocilliary escalator, but may then be swallowed and subsequently absorbed through the gastro-intestinal tract.

Manufacturers are making significant progress prolonging the life of the tires, mainly by improving the raw materials in the rubber compounds, but also by improving the tire cavity geometry. However, the contact with the road and the increase of temperature are of course unavoidable during the tire operation. Therefore, understanding the intrinsic nature of the generation, life, and destiny of particles originating from tire wear is a challenging and maybe impossible goal, but absolutely necessary if component improvements, control techniques, and future legislation are to be achieved. The last effort in comprehending the topic started in 2006 with the Tire Industry Project (TIP) [*2*] in the context of the Particle Measurement Programme (PMP), which focuses on vehicle emissions since 2001 [*3*].

Tire debris is produced by tire interaction with the road surface. It may be that both the chemical composition and characteristics of the particles generated change compared with the original tire tread due to heat and friction as well as incrustations from the road surface [*4*]. It is well known that no laboratory-based method exists that accurately predicts tire tread wear. Assessment of worn surfaces from laboratory and on-the-road tests, using light microscopy and transmission electron microscopy, showed key differences in the microstructure of tread compounds that may indicate how particles of rubber compound become detached from the tread surface and may indicate that there are different mechanisms involved in the laboratory-based abrasion test and the in-service test [*5*]. Hence, laboratory (precise but artificial) versus field (realistic but not controlled) measurements must be taken together as complementary techniques.

The present work gives a global review of the particle collection methods, measurement devices, physical characterization, and toxicological analysis found in the literature in the last 30 years as well as the first experimental attempt of the authors to assess a reliable characterization.

Literature Review

Methods for Particle Collection and Measurement Techniques

The widely used collection method in the literature is ambient sampling. However, some authors focused on the acquisition of tire/road debris from rotating tires using advanced particle measurement devices. Indoor simulations of tire wear were mainly performed using flat track test machines and road wheels [*6*,*7*], road simulators in closed chambers [*8–10*], a drum testing system [*11*,*12*] with different abraders [*13*], and debris from abraded tires using rasps [*14*]. Among all laboratories, VTI (an interdisciplinary research institute in Sweden) is probably the one that most carefully studied the topic and summarized it in Sjödin et al. [*15*]. Its road simulator consists of four wheels that run up to 70 km h^{-1} due to a direct current motor along a circular track of diameter of 5.3 m and changeable pavement. The devices employed were a stationary aerosol sampler (SAM) for ambient aerosol, a tapered element oscillating microbalance (TEOM) for mass concentration, a DustTrak for comparison of $PM_{2.5}$ and PM_{10}, a scanning mobility particle sizer to measure particle distributions distribution below 1 μm, an aerodynamic particle sizer able to measure up to 20 μm, a small deposit cascade impactor and a wide variety of chemical analyzers, among others [*8*]. Samples were collected with two virtual impactors [*9*] ($PM_{2.5-10}$ and $PM_{2.5}$), two 12 stage cascade impactors, and two high-volume samplers.

On-road tests were also performed where a vehicle was equipped with five stainless steel sampling tubes in different positions inside the wheel housing in order to distinguish the particle contribution from every possible source [*16*]. Each tube was connected to an engine exhaust particle sizer (EEPS), which provides particle size distribution between 5.6 and 562.3 nm. An aspiration system attached to the tire hub, which included a slot type capture hood connected to a cyclonic cup, which retained particles in the size fraction of 0.3 to 100 μm, was used for on-road collection using two representative driving circuits and three typical tires [*11*]. A vacuum pickup for tire debris and a sticky catch-plate collection were also used [*17*].

A wide range of aerosol, soil, and water sampling methods are available, as well as several measurement devices using different mechanisms for collecting representative samples. High volume samplers and Gelman-A glass filters were used for aerosol and soil sampling [*18*,*19*], and water-containing jars were used for dustfall measurements. For measuring and analyzing, a condensation nuclei counter, a nine-stage impactor, an electrical aerosol analyzer, extraction-infrared technique, thermogravimetric method, and pyrolysis gas chromatography were used. The last method was widely used in the literature [*4*,*11*,*15*,*20*–*26*]. Vinylcyclohexene is a pyrolysis product of both the styrene-butadiene and poly(butadiene) rubbers and can be used as a valid measure of rubber in dust samples [*19*]. Simple plastic bags [*7*,*13*] and vacuum sweeper truck [*27*] were also used for collecting particles along the roads, storm drains, and/or adjacent areas. Quartz, Teflon, and polytetrafluoroethylene filters for collection and real-time dust analyzers were also used [*4*,*9*,*20*,*21*,*27*,*28*]. An aerosol time-of-flight mass spectrometer for ambient aerosol was used in addition [*7*]. The widely used technique for chemical and optical analysis was energy-dispersive X-ray spectroscopy by means of scanning electron microscope (SEM) [*10*,*11*,*13*,*29*,*30*]. Light microscopy, transmission electron microscopy [*5*,*8*], transmission optical microscopy, the laser diffraction method [*11*], and simple microscopes for debris morphology were used in the oldest works [*17*,*31*].

Although several authors [*13*,*32*,*33*] have used Zn as a tracer, this element may have other sources related to traffic emissions in the atmosphere [*28*]. Styrene-butadiene rubber and natural rubber are the main polymer components in the tire tread and naturally the most used as tracers [*4*,*21*,*22*,*24*,*31*]. Other elements, such as benzothiazole, dipentene [*19*], 2-(4-morpholino) benzothiazole [*34*], *N*-cyclohexylbenzothiazole-2-sulfenamide [*35*], and polyaromatic hydrocarbons (PAHs) [*18*], among other commonly used tire chemicals, also have been used as tracers, but they are no longer in use or not unique or representative for tires. However, because the majority of tire dust particles are mixed with minerals coming from the road, the use of chemical tracers may underestimate the particles coming from tire–road interaction and overestimate the resuspension components [*7*].

Characterization

As might be supposed, pavement texture plays a major role in determining the extent and severity of the wear mechanism, in addition to driving habits, climatic conditions, and operational factors [*36*]. Those, and over and above the huge variety of tire compounds found in the market, might be the main reasons why there is a very wide characterization in the literature. In addition to that, the number of sampling methods, sampling sites, and used markers are also augmenting factors to the great variation of tire wear contribution to PM_{10} [*37*]. Abrasion of tire tread has been acknowledged as a predominantly mechanical process, and it has often been assumed that emitted tire wear particles consist exclusively of particles larger than 2.5 μm diameter, while ultrafine particles have been more recently detected and are known to originate due to thermal processes [*38*]. It is evident from the available literature that there is considerable variation in the contribution that tire wear makes to fine and coarse particle emissions. There is also evidence of the emission of a significant proportion of particles larger than PM_{10} [*39*]. In addition, pure tire wear particles are not often found in real world road dust samples and ambient aerosol, but in contrast most of them are internally mixed with minerals and other substances [*7*].

With the aim of summarizing the results presented in the literature in a way that the reader can understand the full range of the problem, Table 1 shows the values found in the literature for the emission factors, size distribution, percentage contribution to PM, and other possible factors of tire wear particles. This compilation does not detail the techniques, and hence the methods used to obtain the results, and the dates of such works vary significantly, as was mentioned in the previous section. Nevertheless, the interested reader should focus on the details of each reference, since they offer a better understanding of the global phenomena. Moreover, different results are obtained for the same samples tested at the same day using two different techniques [*11*]. Three good reviews of the topic where authors make this observation can be found in Panko et al., Grigoratos and Martini, and Thorpe and Harrison [*4*,*37*,*39*].

Environment and Toxicology

Tire (and brake) wear particles contain transition metals and contribute to near-road PM [*45*]. Tire debris deposited on the road may release a number of chemicals when it interacts with rain and/or runoff water [*46*]. Once deposited on pavement and adjacent soil surfaces, tire wear particles are subjected to physical removal by fluvial and atmospheric processes and to degradation by abiotic and microbial processes [*32*]. Ecotoxicological hazard and risk associated with tire wear particles with special focus on the aquatic environment were exhaustively studied in Wik [*14*], one of the most recent works in this field. All tires tested in Wik and Dave, Goudey and Barton, and Abernethy et al. [*47–49*] were found to be toxic to *Daphnia magna* and other organisms, and all tires showed a higher toxicity after 48 h exposure compared with 24 h exposure. However, not all organisms were sensitive to tire leachates [*50*,*51*], and other artifacts in the experimental setups may affect the results [*52*]. Tire debris organic extract embryotoxicity was studied in Mantecca [*53*], which found that, just at high concentrations, tire debris organic extract was toxic for *Xenopus laevis* embryo development and was linked to malformations. There is a lack of understanding in characterization, since not many studies were carried out, and, as was pointed out in Evans [*54*], they exhibit few comparable aspects with regard to experimental conditions, test organisms, chemical analyses, and physicochemical variables of the water. The last review of Evans [*54*] is an extended work that clearly summarizes the biological effects (toxicity, mutagenicity, and carcinogenicity) of tire exposure to different organisms (plants, mammals, fishes, etc.).

Zinc and sulfur have been widely studied, since they are main components in rubber-compounding and can be used as tracers. As a clear example of the problem, the 1994 study of Ahlbom and Duus [*55*] concluded that approximately 10% of the total concentration of Zn salts found in Swedish cities came from tire wear, but the situation could be even more worrying: estimated release values of Zn from tire tread (using the wear-rate value of 0.05 g_{tread}/km) exceeded all other U.S. atmospheric emissions sources [*32*] except nonferrous metal production. The 0.05 g_{tread}/km wear-rate estimate equals roughly 20% of the total U.S. atmospheric emissions of Zn for 1995. This is comparable to emissions from waste incineration and about seven times that from fossil fuel combustion. While no evidence was found in a 5 year study in Maine [*56*] (which has National Secondary Drinking Water Regulations, a set of non-mandatory water quality standards for 15 contaminants established by the U.S. Environmental Protection Agency) that tire shreds increased the concentration of aluminum, chloride, sulfate, and zinc, it was also found [*29*] that the pH increase in soils containing abraded tire rubber particles proves to be an important factor for the risk assessment of Zn released by rubber tires. It is striking that roadside transect studies have generally identified a higher pH at the contaminated road border compared with a reference site further away from

TABLE 1 — *Literature review of the characterization of tire wear particles.*

Ref.	Emission factors	Size distribution	Contribution to PM	Others
[*18*]	0.12–0.48 μg m^{-3}	0.4 μm (volume) 0.09 μm (number) 80% $d > 11$ μm	1.3–2.6%	Hydrocarbon/sulfur emissions: 0.6/0.2 mg kg^{-1}
[*4*]	0.05–0.7 μg m^{-3} (PM$_{10}$) Max: 2.8 μg m^{-3}		0.84% (to PM$_{10}$)	
[*19*]			4.3% (respirable size) 2.5% rubber (max) 4.9% (suspended)	
[*21*]	0.5–10.5 μg m^{-3}		SBR 0.4%–NR 6.3% (total)	Zn concentrat. 0–5.1 pg m^{-3} (no mutagenic activity)
[*22*]	9.4 μg m^{-3}		SBR 3.3%–NR 1.8% (to PM$_{10}$)	
[*11*]		(Roadway/tire particles) 50 μm/75 μm 50 μm/100 μm		
[*31*]	0.2 μg m^{-3} (mostly non-resuspendable) 1 μg m^{-3} (urban/airborne)	0.13 μg m^{-2} $<$ 1.1 μm 0.07 μg m^{-2} $>$ 7 μm	20% airborne PM 2–7% airborne PM 0.02–0.1% (corners)	
[*24*]	7 μg m^{-3} (to TSP) 2.2 μg m^{-3} (to PM$_{10}$) 0.2 μg m^{-3} (to PM$_{3.3}$)	Mass: 2.5 μm		
[*16*]	1×10^{11} No. km^{-1} 1×10^{7} No. cm^{-3}	Mean d: 30–60 nm Mean d: 70–90 nm		No CO$_2$ generation
[*33*]		Mean d: 0.35 μm 7.8% $d < 1$ μm 92.2% $d > 10$ μm		
[*8*]	3.7×10^{11} to 3.1×10^{12} No. vehicle^{-1} km^{-1}	Mean d: 27 nm Mean d: 15–50 nm		
[*15*]	461 ng m^{-3} (to PM$_{10}$) 2.2 mg vehicle^{-1} km^{-1}	8 μm (studded)		16.1 ng m^{-3} of Zn (big)
[*40*]			0.68% (to PM$_{10}$) in roadside samples	

TABLE 1 — Continued.

Ref.	Emission factors	Size distribution	Contribution to PM	Others
[*38*]		0.087 and 1.4 μm 93% $d < 1$ μm 7% $d > 10$ μm	6.6% (to TSP)	Cl, S, Si, Na peaks 20% of total Zn
[*13*]		>100 μm		
[*41*]	0.31–0.5 μg tire^{-1} km^{-1} (to $PM_{2.5}$) 0.54–0.95 μg tire^{-1} km^{-1} (to PM_{10}) 3.5–6.4 mg m^{-3} (larger)	Mean d: 1.7 μm Mass bimodal: 0.3 and 4–5 μm 32.4% $d < 1$ μm 67.6% $d > 1$ μm		
[*29*]		Median d: 74 μm		
[*32*]		Mean d: 10–20 μm		
[*6*]	2.4 g km^{-1} (cornering) 0.03 g km^{-1} (highway)			
[*17*]		Mean d: 25 μm		
[*42*]			No contribution	5–10% becomes airborne
[*43*]	5 mg vehicle^{-1} km^{-1}			70% of PM_{10} is $PM_{2.5}$, 10% is PM_1, and 8% is $PM_{0.1}$
[*44*]	24 μg m^{-3} (to $PM_{4.7}$)			
[*7*]		1–3 μm (field) <100 nm (lab)		

the road. There was also some evidence that tire shreds increase the levels of iron and manganese and exceed the secondary drinking water standard under some conditions. Finally, Nelson et al. [*57*] found that even if toxicity caused by zinc was observed in laboratory tests, it is unlikely that the zinc concentrations leached from the tires used in artificial reefs would ever cause acute or even chronic toxicity. Tires from a floating tire breakwater (structure used to protect an area from wave attack) installed for 10 years did not release chemicals that were toxic to any species tested [*50*]. Two alarming studies of tire particle exposure in rats revealed pulmonary toxicity possibly due to the presence of water soluble Zn and Cu and that at high concentrations these metals may induce cardiac oxidative stress [*45*]. Lung toxicity induced by TP_{10} was primarily due to macrophage-mediated inflammatory events, while toxicity induced by $TP_{2.5}$ appeared to be related more closely to cytotoxicity [*53*]. It was also attributed to the latex allergens or latex cross-reactive material present in tire debris and may cause latex allergy and asthma symptoms associated with air pollution particles [*58*].

Other important components are polyaromatic hydrocarbons (PAHs), which used to amount to about 10% of the rubber material. Considering the carcinogenic properties and the slow biodegradability of the PAHs, tire debris must be taken into account when dealing with particulate air pollution [*33*]. In Fauser et al., the author evaluated the contribution of tire-tread and bitumen abrasion to particulate air pollution. A method for toxicity testing with *Daphnia magna* was presented [*47*] that could be used as a basis for environmental labeling of car tires. In 2005, the European Commission adopted Directive 2005/69/EC, restricting the marketing and use of certain PAHs in extender oils used in tire production. More generally speaking, butadiene and isoprene effects in animals and in population studies have been extensively summarized [*59*]. Both butadiene and isoprene have been reviewed by the International Agency for Research on Cancer, whose overall evaluation is that they are probably carcinogenic to humans (Group 2A and 2B).

Despite all the facts presented above, the two most recent works in the context of the Tire Industry Project (TIP) concluded exactly the opposite: tire road wear particles (TRWP) should be considered a low risk to aquatic ecosystems and no-observable-adverse-effect-level of TRWP in rats [*60*,*61*]. Some points of the ongoing discussion of environmental effects may be clarified through new studies.

PMP and TIP

A special section of this paper is dedicated to the Particle Measurement Programme (PMP) and the Tire Industry Project (TIP), since they are the set of prevalent groups working in exhaust and nonexhaust vehicle emissions. PMP [*3*] is an intergovernmental research program that operates under the auspices of the Working Party on Pollution and Energy (GRPE) within the United Nations Economic Commission for Europe (UNECE) aimed at developing new vehicle exhaust particle measurement procedures for regulatory use. The mandate given to the PMP Informal Group by GRPE was to develop new particle measurement techniques to complement or replace the existing particulate mass measurement, with special consideration to measuring particle emissions at very low levels [*62*]. The particulate matter (PM) mass measurement systems and methodologies have been continuously refined in order to improve measurement accuracy. The PMP within the UNECE developed a solid particle number measurement method in order to improve the sensitivity of particulate emission measurement from vehicles. Consequently, particle number (PN) limits were implemented into the regulations in Europe from 2011. Portable emission measurement systems (PEMS) for in-use vehicle emission measurements are also attracting attention currently in North America and Europe, and real-time PM mass and PN instruments are under evaluation [*63*].

Within the context of PMP, the TIP [*2*] was started in 2006 with the support of the 11 most prominent tire producers, whose main goals were to anticipate the potential long-term environmental and health issues related to tire materials, tire road wear particles, end of life tires, and recycling management [*64*]. Last year, the work of a group of researchers from Cardno ChemRisk [*4*,*25*,*26*,*35*,*60*,*61*,*65*] with cooperation from Ford [*16*] and VTI [*8*,*15*], among others, was presented in regular sessions, most of them available in open access libraries.

Experimental Approach

None of the existing measurement devices can satisfactorily register particle number, mass, and size of tire particles. They are all designed and calibrated under specified norms for exhaust emissions. Since no legislation still applies for tire (and brake) emissions, even if companies are now starting to take care of the research and development aspects, no device is still available for that purpose. Having said that, any instrument and measurement technique that can offer a better understanding of the phenomena must be carefully studied and taken into account with all its limitations and benefits.

Likewise, the collection technique is another key method, since two main factors affect the correct estimation of the real number of particles and their size distribution: the tendency of tire debris to become electrically charged and adhere to the walls of the sampling equipment and other surfaces [*27*] and the devices that in the collection process may retain a certain amount of debris or introduce possible errors from aerosol growth and wall losses [*18*].

Particle Collection and Analysis

Considering the expected wide range of particle size and composition, an ample variety of devices should be applied in order to reach a reliable characterization. The Automotive Engineering Group at TU Ilmenau use the following equipment. For particle size, number, and mass measurements: (i) ELPI®+ by Dekati (Kangasala, Finland) enables measurement of real-time particle size distribution and concentration in the size range of 6 nm to 10 μm; (ii) the DMS500 fast particle analyzer by Cambustion (Cambridge, United Kingdom) combines electrical mobility measurements of particles with sensitive electrometer detectors to give outputs of particle size, number, and mass in real time and up to 1 μm; (iii) the Mexa 2100 Solid Particle Counting System by Horiba (Irvine, California) uses laser scattering condensation particle counting to measure the number of solid particles from engine exhaust gas in real time and up to 2.5 μm; (iv) the PN Portable Emissions Measurement System by AVL (Graz, Austria) uses the advanced diffusion charger technology for exhaust particle number measurement on the car in real driving conditions

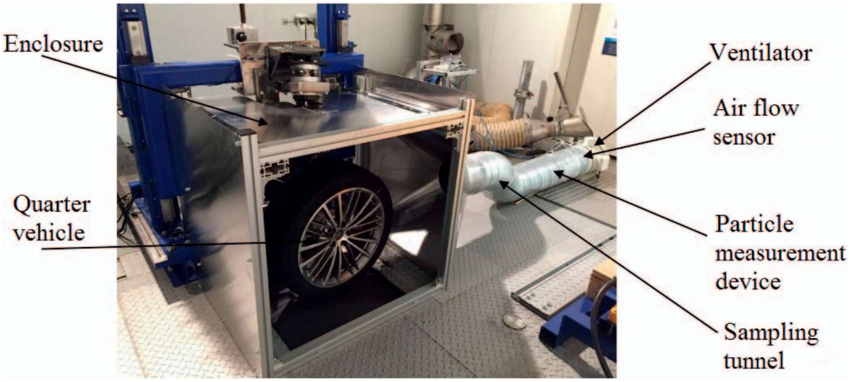

FIG. 1 — *New developed box built in the quarter car system over the chassis dynamometer.*

(particle size up to 10 μm); (v) the ultrafine condensation particle counter model 3776 by TSI (Shoreview, Minnesota) has a lower particle detection size limit of 2.5 nm and single particle counting up to 300,000 particles cm^{-3}. Finally, for chemistry composition and analysis, a JSM-6610 Series SEM by Jeol (Peabody, Massachusetts) is available in the laboratory.

Designed Collection Box

Based on the large experience of the group in brake emissions, a tire sampling box was designed and built in order to collect and measure the particles coming from a tire mounted on a quarter car system and rotating over a 1.9 m diameter drum (Fig. 1). The corner module test rig was developed by the Automotive Engineering Group at Technische Universität Ilmenau. The test rig was designed to analyze suspension systems and tire characteristics on a roller of a four chassis roller dynamometer. Camber angle, toe angle, and wheel load can be adjusted continuously. The maximum speed reachable is 250 km/h, while the maximum load is 1250 kg. In addition, it is possible to control the temperature of the test environment between –20 and +45° C and humidity conditions [*66*].

The enclosure is a sectional aluminum structure of size 2.4 m × 2.0 m × 3.2 m with totally detachable steel walls that can be adapted for any tire dimension. The bell shaped section allows the air flow entering from the space between the drum and the floor and carrying the particles generated in the tire–drum contact to come easily to the sampling tunnel thanks to a Solar & Palau (Jacksonville, Florida) tube fan located at its end. With the fan, it is possible to adjust the airflow through the sampling tunnel. The total length is 2.7 m, and the measurement devices are located by means of a probe introduced into the tube. The measurement system evacuates with a constant volume flow from the probe. An air flow sensor and an air filter are located in front of the ventilator.

Since our focus is on the physical and chemical characterization, no environmental or health studies will be performed from our side. Now, the gaps that appear in the literature are mainly due to a lack of defined test procedures and the lack of proper devices for measuring tire emissions. We intend to fulfill these two aspects. The designed collection box follows the previous experience of the department in the collection of brake particles, but an overall understanding for tire particles is required. Our attention is on the comprehension of the generation of particles to attempt its best measurement protocol. This means a deep physical understanding that could lead to a global acceptance of what we want to measure and for what purpose (with future legislation in mind).

Conclusions

Despite the effort of all the authors mentioned in this literature review, there is a lack of knowledge in tire particle emission and its risk for the environment and human health. Most of the characterization works were carried out several years ago, and hence the information about concentration, size distribution, chemical characterization, and so on may be not accurate anymore. The recent works were mostly focused on the environmental impact by means of collected samples without a standardized procedure, and no clear conclusions about concerns emerged. The resulting discrepancies and deficits were clearly presented in Table 1.

The two principal factors conditioning the amount and characteristics of the tire emissions are the tread and pavement composition and geometry. Tire raw materials, blends, tire inner cavity geometry (shape), and surface macro-roughness are essential players in tire particle generation. Braking, accelerating, and cornering are the three more cited driving events that influence tire emissions, essentially due to the high tire–road contact temperatures generated, which could be the dominant cause for ultrafine particle formations. Tire load and pressure, air temperature and humidity, velocity, camber angle, and the suspension system are also important, even if their influence has not been studied properly yet. Understanding those external factors that influence tire wear is half of the problem and should not be neglected. The absence of (i) standardized sampling methods, (ii) understanding of mechanisms, (iii) physical models, and (iv) relevant data turn this topic into a puzzle that must be addressed simultaneously from several angles.

This work intends to introduce the emergent problem by summarizing the literature results and to begin the attempt to characterize the intrinsic nature of the tire particles. Works are currently taking place in a newly developed laboratory environment and will lead to a deeper understanding, not focusing on the discussion of the dimension of the problem but taking it as an existing one and generating the necessary experience and data concerning future regulations.

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