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REVIEW ARTICLE



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What is known and unknown concerning microplastics from tyre wear?

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ABSTRACT

Pollution of the environment by microplastics is a problem that is increasingly visible and worrisome, with tyre wear particles (TWP) being considered, after several studies, as one of the major sources of microplastics. The aim of this paper is to present a review of the current knowledge on microplastics generated by road traffic, more specifically in TWPs. Generation and pathways depend on different factors like characteristics of the tyre and road or physical properties of the particles, among others. Currently, there are some studies on TWPs, both, carried out at the laboratory and under real scale conditions. So far, it has been possible to identify the presence of TWPs in samples, but their quantification involves time-consuming and expensive methods. The big knowledge gap is to find a standardised and practical method for sampling, detection and quantification, so more research on TWPs and their dispersion in the environment is needed.

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Microplastics; tyre wear; road; rubber

1. Introduction

In the mid-nineteenth century, the first plastic material was invented and since then plastics have offered innovative solutions to society's permanent evolving needs and challenges. They improve the quality of life for millions of people across the globe by making our lives easier, safer and more enjoyable. The family of plastics is made up of a wide variety of materials, which are versatile, durable and incredibly adaptable. In 2020 global plastic production was 367 million tons, which meets the needs of end-use markets such as packaging and construction ("Plastics - the Facts, 2021" 2021).

The twenty-first century has seen substantial growth in the production and consumption of plastics, but has not effectively addressed measures to reuse, recycle and control plastic waste. Pollution of the environment with plastics is a serious global threat because it adversely impacts human health, aquatic organisms and the economy (Chae & An, 2017; Näkki et al., 2017; UNEP, 2016; Wagner et al., 2014). The quantity of plastic litter and microplastics observed floating in mid-ocean gyres appear to represent a small fraction of the total amount present in the ocean (UNEP, 2016).

In recent years, much attention has been focused on microplastics as an increasing problem of environmental pollution. The term microplastics comprises plastic fragments smaller than 5 mm and larger than 1 μm (Verschoor, 2015) that are generated by different ways and from different sources, such as synthetic textiles or tyre wear, among others. Microplastics are usually divided into primary and secondary, with different considerations for their definition, depending on whether this division is based on how these microplastics are produced (A), or when they become microplastic (B):

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- (a) Some researchers consider that the primary microplastics are the small particles released directly into the environment, which means that they are directly manufactured as microscopic particles used for commercial applications, while the secondary microplastics originate from the degradation of large plastic objects during usage or after disposal (Essel et al., 2015; Hale et al., 2020). So, the secondary microplastics need a wearing process to become microplastics. Besides, some authors divide these secondary microplastics into two categories that correspond to microplastics that wear away while being used and microplastics that break down once discarded in the environment (Vogelsang et al., 2019).
- (b) An alternative definition used by other researchers describes primary microplastics as plastics directly released into the environment in the form of small particulates, and also those originated from the abrasion of large plastic objects during manufacturing, use or maintenance. In this case, the secondary microplastics emerge when plastic items which are already in the environment are degraded into smaller pieces. Therefore, the primary microplastics reach the environment as small pieces of plastic below 5 mm, while the secondary are pieces of plastic above this size, but they are degraded along time by the environmental conditions until they become microplastic (Boucher & Friot, 2017; Sundt et al., 2014).

In short, there are different alternatives for classifying microplastics (Table 1) so a specific type of microparticles is sometimes classified as primary and other as secondary, depending on the definition used.

This review will focus on microplastics generated on the road due to transportation, in particular on tyre wear particles (TWP), leaving aside road wear particles and microplastics generated during braking or car accidents. Roads can be divided into different types, such as dense asphalt mixture and porous asphalt mixture, among others, and are used depending on the requirements of each particular area. The tyres of the vehicles circulating on them produce wear on the road and the tyre itself that generates particles. From these particles, the highest fraction corresponds to minerals coming from road abrasion, followed by tire and bitumen wear particles (TBWP) (Järlskog et al., 2022). TWP accounts for approximately 50% of TBWP (Järlskog et al., 2020) and the highest contributor to TWP is rubber (Sommer et al., 2018). In relation to this, there is a discrepancy observed in the consideration of rubber as plastic or not. Despite the fact that according to the International Organization for Standardization (ISO) rubber would not be included within the definition for plastic (ISO, 2013), some researchers consider rubber as a kind of plastic (Wagner et al., 2018). There are also discrepancies in TWP classification, being considered secondary microplastics in several studies (Essel et al., 2015; Hale et al., 2020; Vogelsang et al., 2019; Xu et al., 2020), while being framed in the primary fraction by the European Community (Boucher & Friot, 2017; Europeo, 2018; Sundt et al., 2014), stating that they represent up to 28% of primary microplastics (Europeo, 2018). But on one point all authors agree, which is that TWPs are one of the main contributors to microplastic pollution in the environment (Essel et al., 2015; Foss et al., 2015; Jan Kole et al., 2017; Leads & Weinstein, 2019; Sundt et al., 2014; Unice et al., 2019; Wagner et al., 2018), with studies suggesting contributions of 5–18% to the global load of plastics in the oceans, and of 3–7% to the particle fractions smaller than 2.5 µm, known as PM2.5, in urban air [18].

Although TWPs have been studied for a long time, since the 1970s (Cadle & Williams, 1978); currently, they are receiving greater attention and progress is being made in their detection, analysis and

Table 1. Scheme of different divisions of microplastics.

| | | | | | |
|----------------------|-----|-----------|--|-----|------------------------------------|
| MICROPLASTICS | (A) | Primary | They manufacture in this size | (1) | They wear away because of use |
| | | Secondary | They degrade from large pieces | | |
| | (B) | Primary | They manufacture in this size and wear away because of use | (2) | They break down in the environment |
| | | Secondary | They break down in the environment | | |

quantification (Baensch-Baltruschat et al., 2020, 2021; Järnskog et al., 2021; Mengistu et al., 2021; Rauert et al., 2021; Rødland et al., 2022b). One of the major handicaps encountered was the interference during the analysis that is generated by road particles embedded on the rubber particles. To address this problem, it is necessary to consider whether the studies carried out so far refer to road dust, tyre wear particles (TWP) or tyre and road wear particles (TRWP). This difference is explained in Kreider et al. (2010), as well as being addressed for a suitable quantification in Rødland et al. (2022b).

The objective of this paper is to review the current state of knowledge on microplastics from tyre wear and tear, generated by road traffic, which is in constant development as it is an issue of growing international interest. This review addresses the characteristics of these particles, their forms of dispersion in the environment known to date, as well as the way and difficulties encountered when sampling and analysing the particles. It should be noted that this review attempts to obtain information only for TWP, focusing on them and leaving aside road wear particles and microplastics produced by other sources (i.e. braking, car accidents, etc.). Moreover, the TWP are being dispersed throughout the environment, both terrestrial and marina. Although there are studies addressing the sampling and analysis of TWPs collected in different environments (i.e. road surface, air, soil, river, sea, etc.), this review focuses on those collected on the road, which is the closest point to their generation and where more emphasis should be placed to find a solution and mitigate the problem. In relation to the sampling and analysis methods, the existing ones were evaluated, highlighting their strengths and weaknesses in order to focus future research in the achievement of a standardised and practical method, and thus proposing solutions to this international problem with more specific and realistic data.

2. Microplastics from road transport

2.1. Sources and generation of microplastics in roads

The main sources of microplastic particles in road dust are believed to be the rubber in tyre treads, that is the largest contributor, the polymers that sometimes are added to strengthen the bitumen used in asphalt pavements, and the thermoplastic elastomers present in road marking paints (Vogelsang et al., 2019).

The microplastics produced when polymer modified bitumen (PMB) is used have their major source in the use of studded tyres. These tyres are typical in northern Europe to improve traction in snow and ice; however, they increase the production of road wear particles which mainly cause air pollution (Järnskog et al., 2021).

The microplastics from tyre wear, named as TWP, are produced by the frictional contact between the road surface and the moving tyre. This contact causes wear of the tyre, that leads to emissions of rubber particles, and heat friction within the tyre, that may cause volatile tyre components to evaporate. Furthermore, it also causes particles to be released from the road, asphalt or concrete, and from road markings (paint) on the road surface. According to some researchers (Grigoratos & Martini, 2014; Kreider et al., 2010; Panko et al., 2013), road wear particles stick to the rubber wear and tear because of the shear forces and the heat produced in the contact. Moreover, these particles may contain embedded metals or other chemicals coming from the traffic or other environmental sources.

Unfortunately, there are limited data on TWP emissions when compared with other non-exhaust vehicle emissions, such as brake wear (Harrison et al., 2021). The characteristics of the tyre, the vehicle and the road surface, among others, determine the amount of tyre wear (Wagner et al., 2018). For example, a research conducted in the vicinity of an urban area under reconstruction revealed that heavy vehicles were a major source of TWP, among other pollutants (Järnskog et al., 2021). This is confirmed by Youn et al. (2021), which compares an industrial zone with a residential zone and shows a higher number of TWP in the industrial zone associated with a greater number of heavy vehicles. Another example is the study carried out by Arizona Department of Transportation where, taking advantage of the resurfacing of a tunnel, the difference in TWPs emissions between the new asphalt

mixture and the existing concrete pavement was studied under real driving conditions. One of the conclusions was that the tyre wear emission rate per kilometre driven is 1.4–2 times higher on concrete surface than on asphalt road surface (Alexandrova et al., 2007). Additionally, the roughness of the surface layer will increase the wear rate by a factor of two-three times (Kennedy et al., 2002). Thus, the road surface influences tyre wear mainly by its microtexture and macrotexture, referring to the surface of the aggregate particles and binder and mainly to the shape and size of the aggregate, respectively. A deeper microtexture generates a higher skid resistance, but also leads to more tyre wear, resulting in contradictory objectives. Therefore, a balance must be achieved between reducing tyre wear and reducing friction to avoid compromising safety. On the other hand, the degree to which macrotexture affects tyre wear is uncertain (Andersson-Sköld et al., 2020), although some authors (Gustafsson et al., 2019) assert that higher macrotexture increases dust loads.

Moreover, climate, speed, driving conditions and nature of the contact (e.g. rolling versus slipping) also have an influence in the amount of the particles emitted (Alexandrova et al., 2007; Beji et al., 2021; Verschoor et al., 2016). On the one hand, in a study carried out in China (Sun et al., 2022), a higher number of TWP was detected in road dust in northern cities compared to southern cities, which could be due to lower relative humidity and less rainfall, resulting in higher frictional resistance of the roads. These results are in accordance with those obtained from Kang et al. (2022), where they state that the concentration of microplastics in road dust increases in dry periods. On the other hand, in a research carried out in Stockholm over several years (2011–2017), a seasonal dependency has been observed in road dust loads, this is mostly due to the different types of tyres used depending on the weather conditions affecting the road. The highest loads (up to 200 g/m²) occur between late winter and early spring coinciding with the use of studded tyres; while, the lowest values (down to about 15 g/m²) occur in late spring and also early autumn where the use of non-studded tyres is predominant (Gustafsson et al., 2019). In line with the seasonal dependence research, a subsequent research focused on TWP, among other components of road dust, and one of the conclusions drawn was that there is a higher generation of microplastic during the summer than during the winter (Järlskog et al., 2021). The roads had not been swept during the summer, which could explain the results. Further research on seasonal dependence in other parts of the globe is needed in order to confirm the results from these Swedish studies.

Finally, as a result of high braking and acceleration in urban driving styles, the tyre wear emission factor is higher than in highway driving (Knight et al., 2020; Vogelsang et al., 2019). Additionally, there is a high variability, in terms of particle abundance, between traffic densities, though. These results agree with those reported in Panko et al. (2019) where little relevance of traffic density in the tyre wear contribution to PM_{2.5} was found, being also reported that the highest concentrations of particulate matter directly correlate with high braking operations.

2.2. Tyre and TWP composition

The tyres of our vehicles are much more than a black ring of rubber on the wheel. Over time the composition of the tyres has varied up to the modern tyre, that combines up to 12 different sorts of rubbers with other chemicals and construction materials providing the tread with the necessary hardness, wear resistance, durability, elasticity and stickiness (Vogelsang et al., 2019). It can contain between 25 and 30 components (Continental, 2020; GoodYear, 2022). For instance, Figure 1 shows the most popular composition of a summer tyre of Continental (Continental, 2020).

Natural and synthetic rubbers are two types of polymers with excellent properties that are widely used in tyre industry. Each rubber type has its own chemical and physical properties. Natural rubber is characterised as a strong, flexible and heat-resistant material. However, some types of synthetic rubbers are cheaper and have better resistance to temperature, aging and abrasion (Encyclopaedia Britannica, 2017, 2023; Fisher, 1942, *Gmtrubber*). But the main difference between them is the way they are produced. On the one hand, natural rubber is produced naturally from the tree *Hevea brasiliensis*. On the other hand, synthetic rubber is an artificially produced polymer, synthesised from petroleum

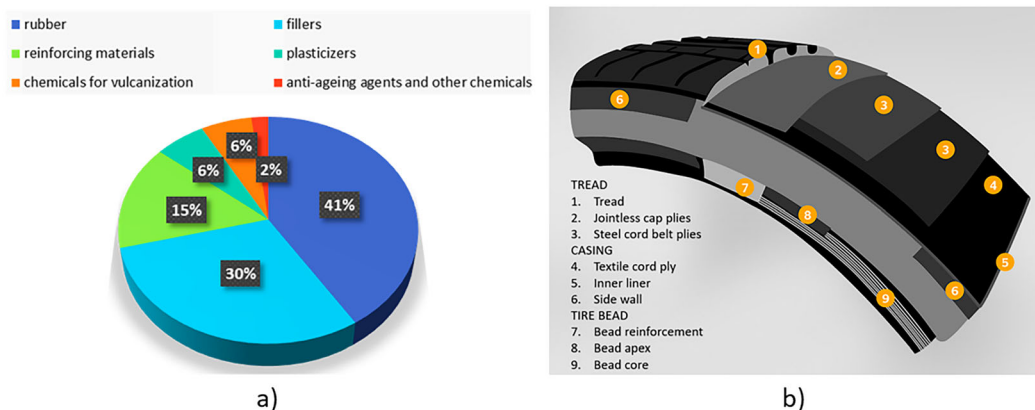


Figure 1. (a) Tyre composition of the most popular summer tyre of Continental. Data collected from Continental (2020); (b) Structure of modern passenger car's tyres. Modified from Continental (2020).

by-products and converted into an elastomer by linking polymer chains using Sulphur bridges. There are more than 20 different classes of synthetic rubbers with their corresponding physical and chemical properties (Essel et al., 2015).

Rubber has different compositions depending on the type of tyre. A mix of styrene–butadiene rubber (SBR) and butadiene rubber (BR) is employed in tyres of passenger cars. However, the type of rubber normally used in heavy duty vehicles is natural rubber (NR), while a higher quantity of polybutadiene rubber (PBR) is applied to non-studded winter tyres in passenger cars to obtain a softer rubber mix which improves the grip (Vogelsang et al., 2019; Wagner et al., 2018). It has been reported (Unice et al., 2012) that the total percentage of rubber in tyre tread is about 50% (w/w), of which 45% is NR in heavy vehicle tyres and 44% is SBR + BR in light vehicle tyres. However, this is an average, as different quantities are used between brands and models. A comparative study of these differences was carried out on several Norwegian and Australian tyres; the results are presented in Rauert et al. (2021). The lack of knowledge about the variability of rubber contained in both car and truck tyres hinders the accurate quantification of TWP in the samples and needs to be addressed.

Furthermore, tyres have other components to provide them with different properties, such as for example, to make them resistant to UV rays. Carbon black is usually added as a filler, although lately it is being partially replaced by silica nanoscale glass balls. The reinforcement of rubber with silica is a complicated process, but the silica can reduce the rolling resistance by 20% and increase the adhesion to the pavement by 12% (Zimmermann et al., 2018). In addition, it is necessary to vulcanise the rubber compounds to give them greater elasticity because natural rubber tends to crystallize spontaneously at low temperatures or when stretched due to its high structural regularity (Wagner et al., 2018). Sulphur or zinc is used as catalysts. Finally, oil is added to make the tyre less stiff and to improve its wet grip performance (Jan Kole et al., 2017). For the purpose of this review the interest is focused on the tyre tread (Figure 1), which contains the components mentioned above, and which are summarised along with their main functions in Table 2.

The tread is the part in contact with the road surface and provides traction and grip, optimal handling, long service life and good mileage. According to Wagner et al. (2018), the particles that come off the original tread of the tyre have their properties and their chemical composition altered due to the interaction of the tyres with the pavement. This is due to the friction and heat generated and to the incorporation of material coming from the road surface (Panko et al., 2013), such as wear from brake pads and tyre studs, exhaust fumes from vehicles, and atmospheric depositions (Wagner et al., 2018). These particulate encrustations were reported to range between 10 and 50% (Klöckner et al., 2021b; Kreider et al., 2010; Sommer et al., 2018; Unice et al., 2019).

Table 2. Examples of tyre tread components and their main function (Andersson-Sköld et al., 2020).

| Component | Function |
|---|---|
| Natural rubber | Main components providing elastic properties, which provide comfort, traction, and friction. |
| Synthetic rubber polymers | |
| Fillers | Strengthen the rubber, i.e. improve tensile strength and prevent tear and abrasion. |
| Vulcanisation agents | Transform the rubber into a solid product through vulcanisation (i.e. curing of the rubber) by crosslinking the polymer chains. |
| Activators | Activate the curing during vulcanisation. |
| Accelerators | Speed up the vulcanisation. |
| Inhibitors (pre-vulcanisation inhibitors) | Inhibit premature vulcanisation. |
| Antioxidants | Prevent the rubber from degrading due to temperature or oxygen. |
| Antiozonants | Prevent degradation caused by ozone exposure. |
| Peptizes | Accelerate the mastication process (i.e. when the viscosity is reduced to aid mixing). |
| Plasticizers | Function as softening agents and processing aids by reducing the viscosity during processing. |
| Other processing aids | Aid mixing and processing (e.g. dispersing agents). |

2.3. Physical properties of tyre wear

While there is a clear understanding of tyre tread composition, the characteristics of the particles generated during the use of the tyre are not well known. One of the major uncertainties on the subject is whether the physical properties depend on where the sampling is performed (i.e. airborne, soil, road surface, water, etc.). Several research studies (Kreider et al., 2010) have been carried out concerning the physical properties of TWPs. These properties, including shape, size and density are considered to be important factors in the dispersion of microplastics from tyre wear in the environment, so a better understanding of its dynamics is essential to decrease pollution (Klößner et al., 2020).

2.3.1. Shape

According to Gunawardana et al. (2012), Klößner et al. (2021b), Kreider et al. (2010), tread wear particles are generally elongated in a sausage-like shape with rough surface. Theoretically, these fragments implant less deep into the asphalt mixtures than spherical particles due to the entanglement of the angular particles in the pores (Waldschläger et al., 2020). Some researchers also analysed the morphology of the collected road dust and particularly TWPs, qualitatively using scanning electron microscopy (SEM), as shown in Figure 2, where mineral incrustations are evident in the photo of greater magnification, and quantitatively using transmission optical microscopy. These pictures show TWPs from a road simulator. However, according to Kreider et al. (2010), when comparing the particles generated at the laboratory under simulated driving conditions and on-road collected samples, similar features were found on the particles, though generally on-road samples were smaller on average.

2.3.2. Size

The available studies on size distributions of TWPs show heterogeneous results. The size distribution depends on factors such as speed, temperature, aging, type of pavement, tyre's composition (Jan Kole et al., 2017) and driving behaviour (Andersson-Sköld et al., 2020). It is important to point out that the particle size reported by a particular experiment also depends on the experimental setup (Jan Kole et al., 2017), because different methods are used to generate samples and analyse the particles, which can be collected either at real-scale test conditions or at the laboratory test using road simulators (Andersson-Sköld et al., 2020; Jan Kole et al., 2017).

Some authors (Jan Kole et al., 2017) provide an overview of the size ranges covered and detected in four key studies (Aatmeeyata et al., 2009; Dahl et al., 2006; Kreider et al., 2010; Mathissen et al., 2011) focusing on the size distribution of tyre wear. Interpretation of the experimental results is complicated due to test set up, analytical difficulties to separate tyre from road particles, and the enormous variety in experimental conditions and analytical experiment. More studies are needed to determine the

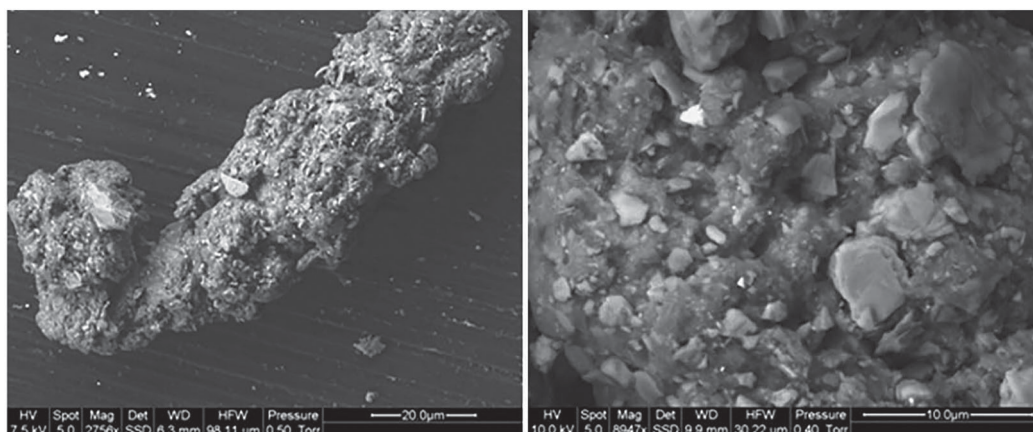


Figure 2. Scanning electron microscope images of particles collected on a simulated laboratory driving course. Images obtained from Kreider et al. (2010).

size of the particles generated under realistic driving conditions. However, in Jan Kole et al. (2017), the findings from four separate studies on TWP were combined. According to it, TWPs fell within a size range of approximately 10 nm to several hundred microns; these data are in accordance with the results obtained in a recent research (Parker-Jurd et al., 2021).

2.3.3. Density

The density of the microplastic particles influences how they spread and where they accumulate. The density is affected, among others, by whether the particle is a pure tyre particle or if it is made up of a mixture of tyre, bitumen and minerals from the roadway (Andersson-Sköld et al., 2020). The density of tread tyre is 1.15–1.18 g/cm³, but because the particles also contain a mix of road surface materials, the final density varies from 0.94 to 2.5 g/cm³ (Parker-Jurd et al., 2021; Sommer et al., 2018; Unice et al., 2019; Verschoor et al., 2016; Vogelsang et al., 2019). Using this range of values, a density separation method is recently being investigated to isolate the TWPs from the rest of the dust collected in the sample in order to analyse and quantify them more efficiently (Goßmann et al., 2021; Järlskog et al., 2020, 2021; Klöckner et al., 2019; Oh et al., 2022). Moreover, according to some authors (Klöckner et al., 2020), the density of TWP particles may be affected by the aging of the particles.

The effect of the size and density of TWPs on road runoff was studied by Bondelind et al. (2020) using a three-dimensional hydrodynamic model. One of the conclusions was that the settlement of particles in the river was influenced by their size and density. Microplastics with sizes around 75 µm and 1.9 g/cm³ (larger and heavier particles) together with one third of microplastics with sizes around 20 µm and 1.7 g/cm³ (medium-size particles) settled in the river, while the small microplastics with density close to 1.0 g/cm³ did not.

2.4. Estimation on the amount of road related microplastics

Two different approaches are mainly used to estimate the amount of wear and tear from tyres. The first uses emission factors per vehicle-km multiplied by the total mileage, and the second uses the number of tyres multiplied by the weight loss of these tyres during use (Baensch-Baltruschat et al., 2020; Jan Kole et al., 2017). Both methods have been used with comparable results, which means that the two methods can be reliably used (Jan Kole et al., 2017). In Europe, the manufacturer or importer must collect and process tyres after use (European Parliament and Council, 2000).

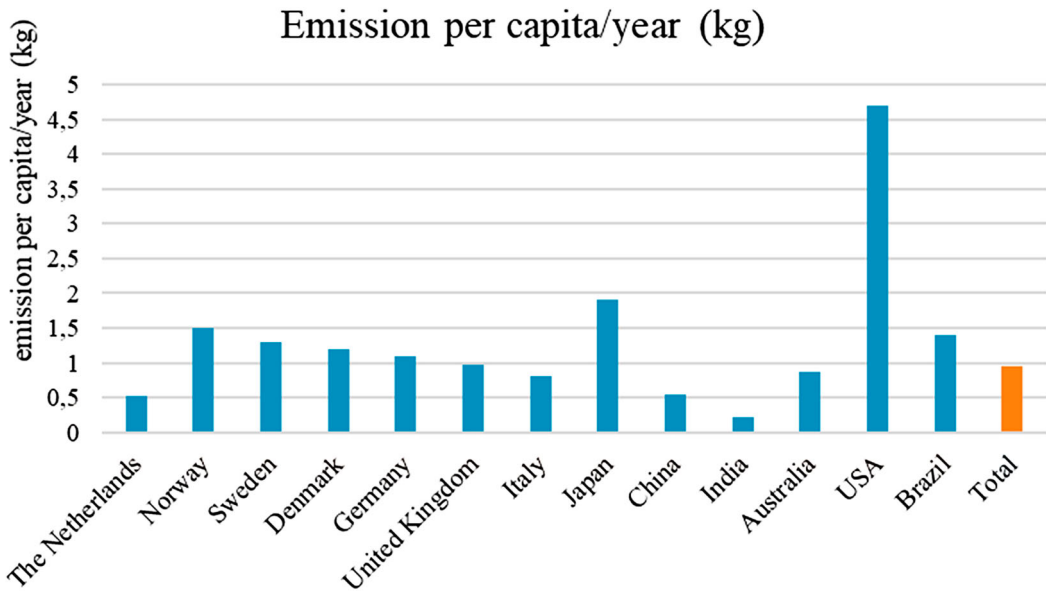


Figure 3. The amount of wear of car tyres per capita per year. Data collected from Jan Kole et al. (2017).

A literature review conducted in Jan Kole et al. (2017) compiled national estimations of the amount of tyre's wear in eight countries. Japan leads the studies of the tyre's wear, followed by Western European countries. Japan, United Kingdom and Italy use the tyre's weight loss method whereas The Netherlands use the emission factor per vehicle-km approach. On the other hand, Germany, Denmark, Sweden and Norway use both emission estimation approaches. This information is not available for all countries, though. Therefore, to obtain a global estimate of the amount of wear emitted to the environment, data on mileage and the number of tyres were collected from countries for which national emission estimates were missing. This search returned results for China, India, Australia, the United States and Brazil. Some researchers used the emission factor method based on data from Japan and several European countries to estimate the national tyre wear emission of these countries. The emission per capita and year in kilograms estimated by this study is shown in Figure 3 (Jan Kole et al., 2017).

According to Figure 3, India is the country with the lowest emissions (0.23 kg/capita-year), in contrast to the USA that presents the highest emissions (4.7 kg/capita-year). The USA is the leader due to the existence of a greater number of cars and to the larger distances that are travelled along the country, especially by trucks. In contrast, the fewer emissions in India and China (0.55 kg/capita-year) are due to the smaller percentage of cars per person in these countries. It should be noted that the low emissions of The Netherlands (0.52 kg/capita-year) are due to the fact that only 5% of the total emitted particles are taken into account, since the other 95% are considered to be captured and retained by porous asphalt roads (van Duijnhoove et al., 2014). For the rest of the countries the estimates are between 0.81 and 1.9 kg/capita-year (Jan Kole et al., 2017).

3. Possible pathways to the environment

TWPs are generated on the road, but their final destination is uncertain. These particles end up in different areas of the environment depending on the different possible pathways that exist for their transportation. In addition, different factors control how the microplastics spread in the environment. These factors include physical properties (shape, size and density of the particles) and precipitation

(Andersson-Sköld et al., 2020). Moreover, maintenance and weather conditions influence the accumulation of tyre wear on the road (Jan Kole et al., 2017; Wagner et al., 2018), for example the build-up of microplastics depends on variables such as precipitation, moisture of the surface road or wind. These parameters rule the microplastics transportation by run-off, air and swirling (Järnskog et al., 2020).

As said before, there are different pathways to disperse the TWP. Generally, small particles are liberated into the air and dispersed, while large particles will settle on the road surface, where some of the particles will be trapped and other transported in different ways (Jan Kole et al., 2017). Non-airborne emissions make up from 90 to 99.9% by mass of the total emissions (Baensch-Baltruschat et al., 2021; Grigoratos & Martini, 2014; Panko et al., 2013) and can be found deposited on the road or on the side of the road (Panko et al., 2013).

TWP concentrations decrease with increasing distance from the source, i.e. as it moves away from roads (Baensch-Baltruschat et al., 2020). This indicates that the deposition of suspended particles is a key pathway of TWPs to the aquatic environment, which together with two point sources: treated wastewater effluent and surface runoff via storm water drainage are considered the three principal routes of TWPs to the marine environment (Parker-Jurd et al., 2021). According to Parker-Jurd et al. (2021), Sundt et al. (2014), the highest emissions of TWP to aquatic environments come from stormwater runoff, which are four times higher than those coming from the atmosphere.

Roads play an important role in the spread of microplastics as they are considered an intermediate step between the generation of rubber microparticles and the environment, with the exception, according to some studies, of porous asphalt surface layers which may retain a larger number of particles than conventional roads due to their void content (Jan Kole et al., 2017). In this sense, according to van Duijnhove et al. (2014), porous asphalt roads in The Netherlands are believed to collect 95% of the microplastics coming from tyre wear. This peculiarity of porous asphalt mixtures deserves special attention, because if this theory is confirmed, it would importantly reduce the diffusion of TWP into the environment. However, measurements to confirm this hypothesis have not been found. Depending on the proportion of voids, the depth of the macrotexture, the meteorological conditions and the speed and composition of traffic, among others, these particles may stick permanently to the voids or remain for some time before being transported (Andersson-Sköld et al., 2020; Jan Kole et al., 2017).

Several ways in which microplastics generated by tyre wear can be dispersed into the environment are described in the next sections.

3.1. Wind and the passing of vehicles

In dry conditions, the wind and the passing of vehicles can suspend or re-suspend the particles deposited on the road, as shown in Figure 4. Turbulence below and around the vehicle, which depends on the speed, number of tyres and vehicle size, causes road dust to swirl up. On the other hand, in wet conditions, passing of vehicles can also swirl up particles due to the splash and spray that occurs when vehicles travel on a wet surface (Vogelsang et al., 2019).

Both the above-mentioned particles and those emitted directly into the air are deposited in different land and water environments via dry or wet deposition. Dry deposition means that the particles are deposited on surrounding surfaces, whereas wet deposition means that the particles are washed out from the air and deposited via precipitation. Dry and wet deposition can occur in a lot of surfaces, such as, for example, the road itself, the areas around the road, built environments, or on land and water areas further away. Another possibility is that the particles are inhaled by humans and animals that are exposed to polluted air. This usually occurs in the vicinity of high traffic roads and urban landscapes where concentrations are comparatively high, which increases the risk (Andersson-Sköld et al., 2020).

3.2. Road runoff

During precipitation and snowmelt, an important dispersal pathway for TWPs is the runoff from roads and traffic areas (Andersson-Sköld et al., 2020; Jan Kole et al., 2017; Parker-Jurd et al., 2021). The amount

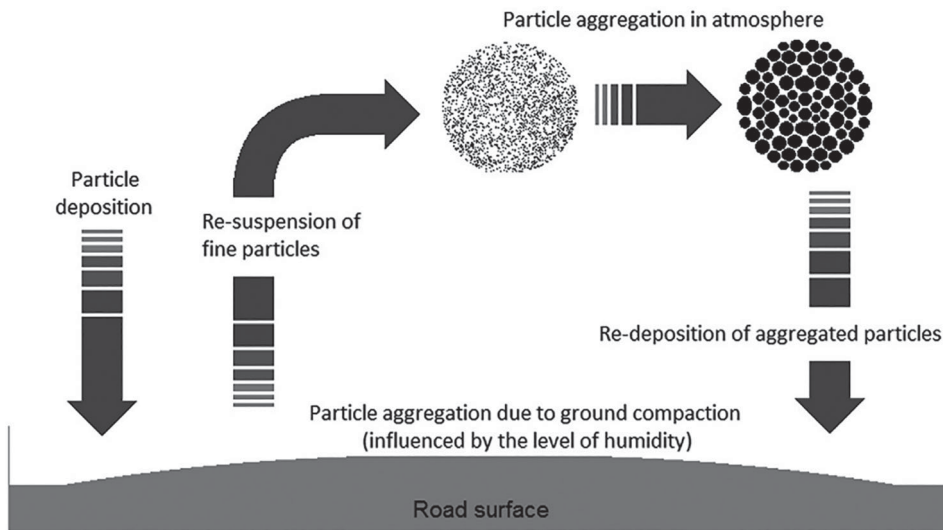


Figure 4. Redistribution of road dust caused by wind and vehicle-induced turbulence during dry weather period. Picture obtained from Wijesiri et al. (2016).

of TWP released to the aquatic environment depends on the infrastructure for collection and treatment of road runoff (Sieber et al., 2020). In areas with stormwater systems, which is commonly the case in built-up areas, several tyre wear compounds and transformation products have been detected by some researchers (Johannessen et al., 2021) at concentrations of mg/L in urban waters receiving stormwater runoff; the dispersal of which will depend on the design of these stormwater systems (Andersson-Sköld et al., 2020). In contrast, in areas without runoff management systems, TWPs will drain off with the rain onto land adjacent to the road and into any open ditches, from where they can be transported to other nearby waterways (Wagner et al., 2018).

In relation to the fate of runoff water, one study (Ziajahromi et al., 2020) investigated the concentration of microplastics in a wetland, whose main findings were that higher concentrations of microplastics were observed in the sediment compared to the aquatic phase, and that around 15–38% of the microplastics found in the sediment were rubber carbon filled particles, which are suspected to come mainly from TWPs that are transported by road runoff.

Locally, amounts released will vary depending on kilometres driven, climate and topography. Moreover, suspended particles in the water can sediment and, on the contrary, water flow can be strong enough to re-suspend sediments and take the particles elsewhere (Andersson-Sköld et al., 2020). An estimate for OSPAR Commission, essentially the European countries bordering the North-East Atlantic Ocean, suggested that the amount of microplastics transported to local marine environments from tyre wear was comparable to land-based litter (Hale et al., 2020). For example, in Sweden 8700 tons of tyre and road wear particles are theoretically released into the environment every year (Järlskog et al., 2020). In Norway, 3942 tons of TWPs are expected to end up in the ocean every year, which amounts to 31.9% of all plastics dumped into oceans (Sundt et al., 2014). In contrast, in The Netherlands it is estimated that only 0.9% (261 tons per year) of the total amount of plastics in the ocean corresponds to TWPs (Jan Kole et al., 2017).

3.3. Snow removal and de-icing salt

In winter, the greatest contribution to the spread of microplastics occurs when snow must be removed from built-up areas so it is transported elsewhere. The TWPs are then moved to where the snow is placed. Conversely, outside built-up areas the snow is cleared by moving it by the side of the road. A

recent study (Rødland et al., 2022a) stated that snow on roadsides in peri-urban and urban environments is highly contaminated and therefore meltwater runoff should be treated before it is released into the environment because otherwise, the TWP spread according to the location specific dispersal pathways related to the road runoff (Andersson-Sköld et al., 2020). Furthermore, according to the conclusions obtained from Rødland et al. (2020), the concentration of microplastics in rock and sea salt used to de-icing winter roads is very low, being considered a negligible source of microplastics compared to others with a greater impact.

3.4. Street cleaning

Depending on the type of the road sweeper and the technology used, street cleaning can generate dust and airborne particles. One solution to reduce the generation of dust is using water while sweeping (Andersson-Sköld et al., 2020) so that this water is collected in stormwater or sewage systems while road dust and sludge fractions are sent to landfills (Vogelsang et al., 2019). Road cleaning has been shown to be an effective way of removing the larger microplastics ($> 100\text{--}125\text{ }\mu\text{m}$) (Amato et al., 2010) and microplastics over $20\text{ }\mu\text{m}$ (Järlskog et al., 2021); so TWP might be prevented from reaching receiving stormwater through weekly sweeping (Järlskog et al., 2020). Meanwhile the smaller ones will more easily accumulate within the pavement microstructure (Vogelsang et al., 2019).

3.5. Vehicle washing

TWPs can stick to the surface of vehicles travelling along the road, when vehicles are washed, these particles may follow the same dispersal pathways as road runoff (Andersson-Sköld et al., 2020). This applies to the smallest size particles in the air, so the total volume of particles that adhere is considered to be relatively limited (Vogelsang et al., 2019). In addition, washing the car in the street is prohibited in some countries because it is illegal to let the vehicle wash water run into a storm drain. Vehicle washing facilities are equipped with a water treatment management system, connected to a stormwater or wastewater sewer system to prevent environmental contamination from car wash water.

4. Artificial generation, sampling and analysis

The sampling, detection and quantification of TWPs is one of the main challenges in terms of knowing the real presence of these particles in the environment and, therefore, the damage they can cause both to the environmental and to human health. As mentioned above, microplastics can be found in both terrestrial and marine environments. However, the focus of this section is on sampling and analysing microplastics on the road since it is the area closest to the emission source and therefore a particularly relevant area to study. The different methods available for sampling and further analysis in the laboratory are described and discussed. In addition, a first section focused on the artificial generation of TWPs has been developed to see the different options currently used in the laboratory.

4.1. Artificial generation of TWPs

Laboratory scaled methods can be used to obtain information on the chemical composition but not on the physical characteristics, since, as evaluated in article (Kreider et al., 2010), particles obtained by laboratory techniques differ in size and morphology from those collected in real samples. The laboratory methods differ in their complexity, starting from very basic sampling and test procedures passing through more complex and evolved ones (Figure 5) to end in large-scale test facilities (Figure 6). Thus, a simple method is, for example, to cut different pieces from the tyre using a knife (Camatini et al., 2001). Another commonly used simple method is to use a file or metal grater to abrade a piece of tyre, which gives coarse tyre filings rather than whole pieces (Redondo-Hasselerharm et al., 2018). Sometimes, samples are frozen using liquid nitrogen, after which they are ground into finer materials to be

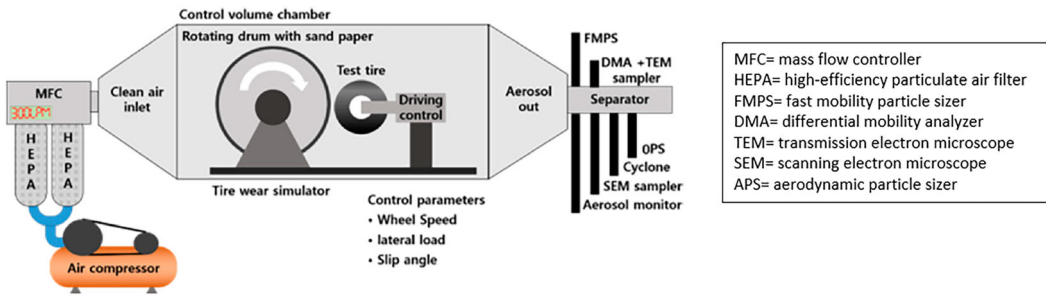


Figure 5. Schematic illustration of a simulator for tyre wear and associated equipment for emission analysis (Park et al., 2018).



Figure 6. Left: vertical drums from BAST; right, VTI's road simulator (Andersson-Sköld et al., 2020).



Figure 7. Sampling in a city street with WDS. Picture: Roosa Ritola (SYKE and SKYE, 2020).

analysed for chemical composition. On the other hand, to obtain more realistic samples, TWPs can be generated at the laboratory simulating the wearing process occurring at the road. For example, in the Republic of Korea, the Korea Institute of Machinery and Materials has a test rig where a tyre is worn against a drum covered in sandpaper, which is enclosed in a chamber through which purified air flows (the installation diagram can be seen in Figure 7). Air samples are collected downstream of the contact point between the tyre and the drum (Park et al., 2018).

Finally, there are at least two types of large-scale test facilities, where real surfacing materials are used. The first type is a vehicle and road test bench (PFF) build at BAST; while the second is a road simulator, located in Sweden. In Figure 6 both facilities are shown.

The former consists of a standing drum with a road surface on the inside, equipped with an internal wheel platform. It is mainly used to test the skid resistance of various road pavements, including wet road surfaces (Kreider et al., 2010). 'Bundesanstalt für Straßenwesen' BAST and 'Karlsruhe Institute of Technology' KIT have this system. In Kreider et al. (2010), the characteristics of the particles generated by cryogenic breaking of tread rubber, on-road collected samples and particles generated at the PFF facility at BAST were compared. In this facility, the tyre's wear is collected by cyclonic vacuum cleaners installed behind the wheels. The most relevant conclusions of the study are that the particles collected in real conditions and those generated in the laboratory at the PFF simulator have similar morphology, although the former are smaller on average. In addition, it is shown that all particles have a significant amount of incorporated material.

The other large-scale simulator is the circular road simulator (CRS) built at Swedish National Road and Transport Research Institute (VTI). Under controlled conditions, the CRS implements accelerated tests that simulate the effect of traffic on roads materials. The VTI's CRS consists of a circular track where any type of road surface material can be tested (Institute, Kuttah, and Kalman 2020). CRS is used for pavement wear testing (Gustafsson et al., 2009) and also for the generation and sampling of TWPs, focusing on the inhalable fraction (PM10). Some researchers used the CRS to look at the wear that occurs on different tyres at a constant speed and analysed the particle mass size distribution of the emitted particles (Grigoratos et al., 2018).

4.2. Methods for sampling tyre wear microplastics

Sample collection can be carried out in several ways, mainly by analysing the amount of material per square metre or by setting and collecting a specific sample volume. To ensure that the samples collected are as relevant and representative as possible, different aspects must be considered, including among others where particles are accumulated, the wind direction, the recent precipitation and the time of the year when the sample is collected. In addition, samples of microplastics from road traffic should be taken from different locations and at different distances from the source to increase the knowledge of where, when and how these microplastics are transported and deposited. This is important to carry out risk assessments, take appropriate measures and assess the spreading (Andersson-Sköld et al., 2020).

Different sampling methods and devices have been used to collect TWPs. The simplest method is to use a broom and dustpan (Abbasi et al., 2019; Chae et al., 2021; Kang et al., 2022) or a basic vacuum cleaner (Roychand & Pramanik, 2020; Youn et al., 2021). A combination of these could also be used, as in Vaze and Chiew (2002), where the dust was first vacuumed without any brushing and then the same surface is lightly scrubbed using a fibre brush so that most of the fine pollutants attached to the surface are released to be vacuumed again. These simpler methods are valid for dust road, but if you need to research microplastics, collecting them can be difficult due to their tiny size. One step ahead is the VTI which uses a proprietary sampling machine called the Wet Dust Sampler (WDS). This is a more complex instrument consisting of a device that uses high pressurised water to clean a small circular road surface area during a specified amount of time, and compressed air to move the material from the washing unit to a sample bottle (Figure 7) (Lundberg et al., 2019). These device is used in some research studies, such as in Gustafsson et al. (2019).

Another method that appears quite often in the literature is the one developed by Amato et al. (2009). This method arose because of the difficulties associated with collecting the fine particles and the electrostatic adhesion of the particles to the hairs of the tweezers/brushes and the sieve meshes. In order to reduce the losses during the sampling procedure, based on a vacuum pump, a resuspension chamber was introduced in addition to a PM10 separator and a filter (Figure 8).

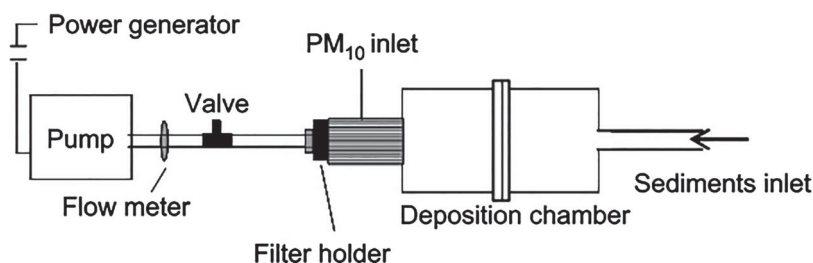


Figure 8. Amato's sampler. Illustration from Amato et al. (2009).

Despite the development of these specific devices, there are still uncertainties about their accuracy in terms of the particle size they can collect, as well as their performance in different asphalt mixtures since these devices had generally been used for dense mixtures, leaving aside porous asphalt mixtures, which normally have more than 20% of voids. In this case, the greatest challenge is to collect the microplastics that are inside the porous network. Another uncertainty is how the microplastics are recovered from inside the instruments since the reduced size of the particles and their tendency to stick to surfaces makes their recovery difficult.

4.3. Analytical methods for microplastics detection and quantification

Once the road dust samples are obtained, the analysis of the particles is done at the laboratory. In the literature, different analytical methods are proposed. Some of them for a qualitative assessment, such as Kovochich et al. (2021a, 2021b), Roychand and Pramanik (2020), while others allow for a more quantitative analysis, although still associated with a high degree of uncertainty (Miller et al., 2021; Rauert et al., 2021).

Table 3 shows the most used analytical methods for the analysis of tyre and road wear particles, including microscopy, micro-spectroscopy and gas chromatography-mass spectrometry.

Micro-spectroscopy methods measure the spectrum of samples when exposed to infrared radiation (μ -FTIR) or a beam of electrons (SEM) (Andersson-Sköld et al., 2020). Both techniques require extensive sample preparation and a time-consuming analysis. In the case of SEM, only visual differentiation of polymer material is allowed and the presence in the sample of unknown materials hinders the analysis. However, if combined with x-ray detection, quantitative information about the elemental composition of the sample is possible. As for the μ -FTIR, which is an improvement version of FTIR, spectrum of the particles can be obtained and compared to reference spectra of common polymers, but this method is not used for TWP detection because its black pigments absorbs the infrared light, as noted in Baensch-Baltruschat et al. (2020), Kang et al. (2022), Ziajahromi et al. (2020). To solve this issue, some studies (Käppler et al., 2016; Lange et al., 2022; Leads & Weinstein, 2019) have used Fourier transforms infrared spectroscopy with attenuated total reflectance, but a recent study (Ribeiro et al., 2021) has demonstrated the potential for under-reporting of microplastics when only FTIR was applied. Another issue is that these methods work better with pure substances, and TWP usually contain a mix of substances.

For the compositional analysis of multicomponent materials, as is the case of TWP, standard tools such as Thermal Gravimetric Analysis (TGA), Simultaneous Thermal Analysis (STA) and Differential Scanning Calorimetry (DSC) (Dümichen et al., 2017; Fernández-Berridi et al., 2006) have been explored. However, to differentiate TWP from other constituent materials in the sample, the use of markers is required. Markers are additives which are incorporated into the tyres, such as zinc or benzothiazoles used in the vulcanisation process, or even rubber itself as the main constituent of tyres (Baensch-Baltruschat et al., 2020; Chae et al., 2021; Klöckner et al., 2019; Wagner et al., 2018). In this sense, the analysis of microplastics via the identification and quantification of specific markers has been done

Table 3. Analytic methods used to analyse tyre and road wear particles, based on Andersson-Sköld et al. (2020).

| | Micro-spectroscopy methods | | Gas chromatography-mass spectrometry methods (GC/MS) | |
|---|---|---|--|---|
| | Scanning electron microscopy with X-ray detection (SEM-EDX) | μ -Fourier transform infrared spectroscopy (μ -FTIR) | Pyrolysis GC/MS | TED-GC/MS (Thermal extraction and desorption GC/MS) |
| Able to detect | Morphology, composition, number of particles, size | Composition | Composition, mass | Composition, mass |
| Detection limit | 10 μ m | 20 μ m | < 1 μ g | 0.5–2.5 μ g |
| Can be used for tyre and road wear particles | Tyre and road wear particles can be identified, and development is underway | Yes, and development is underway | Yes, with reference spectrum/library | Yes, with reference spectrum/library |
| Time to perform analysis incl. sample preparation | Hours | Days – weeks | Days – weeks | Hours |
| Research using it | Kreider et al. (2010), Park et al. (2018), Roychand and Pramanik (2020), Sommer et al. (2018) | Käppler et al. (2016), Lange et al. (2022), Mengistu et al. (2019, 2021), Kang et al. (2022), Ribeiro et al. (2021), Ziajahromi et al. (2020) | Miller et al. (2021), Panko et al. (2013), Rauert et al. (2021), Rødland et al. (2022b), Unice et al. (2013), Youn et al. (2021) | Eisentraut et al. (2018), Müller et al. (2022) |

using thermal gas chromatography with mass spectrometry (GS/MS) combined with pyrolysis (Pyr-GC/MS) (Miller et al., 2021) or with Thermal Extraction and Thermal Desorption (TED-GC/MS). The latter is a more recent technique that requires less sample preparation (Müller et al., 2022) and allows the analysis of larger samples than Pyr-GC/MS; but a study (Eisentraut et al., 2018), reported that NR could not be analysed by TED-GC/MS in environmental samples since dimers and higher oligomers originating from plant decomposition interfered with the analysis. In this case, the TWP of trucks with natural rubber as a major component would be underestimated.

During the tyre wear, several particles are generated at the same time, not only from the tyre but also from the road and other components of the vehicle, as well as from the environment. Due to this, the analysis of TWPs in isolation is difficult. A simpler method to differentiate and quantify the different particles would be desired. However, in this moment there is not a unique method to do so. Some advances can be found in Rødland et al. (2022b), where a new method based on the Pyr-GC/MS analysis is proposed, which aims to measure the combined mass of rubbers in the sample by using multiple marker compounds and includes an improved step to calculate the amount of tyre and road particles based on the measured rubber content and site-specific traffic data. According to the study, TRWP concentrations reported in road samples are likely overestimated due to SBS concentrations. Additionally, using the Pyr-GC/MS, two ISO technical specifications (International Organization for Standardization, 2017a, 2017b) have emerged, which were used in some studies such as Youn et al. (2021). However, a number of assumptions are made that need to be further improved in order to obtain a solid standard, as highlighted by Rauert et al. (2021). An example of such assumptions is the rubber content of the different tyres depending on the model and brand, which are critical in calculating the total mass of TWPs in an environmental sample and the different formulations used by each manufacturer, which are not usually disclosed. All these assumptions lead to error-prone quantifications (Rauert et al., 2021). Emphasise that these are not establish standards but technical specifications, which are intended to address work that is still in the technical development phase with the objective of feedback and improvement for eventual publication as a standard.

Concerning the use of markers for the identification and quantification of TWP, it should be noted that its presence is not a positive proof for the existence of tyre particles; however, contrariwise, the absence of benzothiazoles or rubber, for example, is a strong hint for their absence (Baensch-Baltrusch et al., 2020). One of the disadvantages of using markers is that the original tyre compounds may change over time due to aging processes, making it difficult to use them (Johannessen et al., 2021; Wagner et al., 2022). So, commercially available analytical standards are needed to advance in this area of research. Some authors (Klöckner et al., 2021a) have investigated the use of markers of organic origin, but to function as a quantitative marker, it should be tyre rubber specific and be present in all tyres, regardless of brand or model, not easily leachable, easily and accurately detectable and stable to environmental conditions. The study identified three promising marker compounds although it suggested that more studies are needed to validate their use.

Recently, some researchers (Mengistu et al., 2019) chose the STA and combined it with FTIR and Parallel Factor Analysis (PARAFAC), demonstrating that the method was capable of detecting and quantifying tyre particles from controlled sediments. PARAFAC has proven to be very useful in this respect, as it offers a reliable basis by which to estimate component quantities using Beer-Lambert's law for multi-component matrices. Using this process, a study of TWPs in the gully pot was carried out to verify it with real sediments, quantifying between 0.1 and 15% TWP among the sediments collected at various points (Mengistu et al., 2021).

Although there are several possible analytical method for the analysis of TWP, in general, the sample preparation is long and tedious (Goßmann et al., 2021; Järlskog et al., 2020, 2021; Klöckner et al., 2019; Oh et al., 2022) and very specific and costly equipment is requested. On the other hand, there is no a single method offering a complete analysis of microplastics. To obtain a whole characterisation including the number of particles, their shape, composition and mass, a combination of methods is needed. Thus, more research is still needed to define a standardised methodology for a more efficient and accurate detection and quantification of TWP.

5. Conclusions

This paper summarises the composition of microplastics from tyre wear (TWP), the current status to estimate the amount that is generated, their pathways to the environment and the different methods and techniques for sampling, detection and quantification of these microplastics in the road environment. The most relevant conclusions are described next:

- According to several researchers, TWPs are considered as the largest source of microplastics in the environment. Their generation depends on several variables and the high number of these variables complicates the study and subsequent analysis of the samples increasing the uncertainty in the results.
- The road is considered an intermediate step between the particle's generation and its final destination in the environment; therefore, a good design and maintenance would help to avoid the dispersion of microplastics and thus reduce environmental pollution. There are several forms of dispersion, the most common being wind, passing vehicles and runoff generally caused by rain. Although specific studies are lacking, the physical properties of the microparticles, as well as the pavement and weather conditions are considered the factors that contribute the most to dispersion of TWP in the environment. In this sense, studies on the physical properties of these microparticles, in terms of size, shape and density, generally show great heterogeneity in the results, likely due to the differences in the sampling method.
- The collection of samples in the road is carried out in different ways, such as vacuum or WDS, but there is not a standardised method. This leads to difficulties in comparing different research studies. Furthermore, sampling of road dust is limited to that existing on the surface. According to some of the analysed studies, the use of porous asphalt mixtures could contribute to reduce the contamination by TWP, since the particles are thought to be trapped in the asphalt mixture porous network; however, measurements to confirm this hypothesis have not been found. So, the collection and quantification of the particles retained by a porous asphalt mixture need to be investigated.
- So far, the analytical methods studied have some limitations, including the tedious pre-processing required for their analysis and the difficulty resulting from the variety of components present in the road samples. These methods are still under development and research is being carried out in order to improve and standardise a definitive method for the accurate detection and quantification of TWPs.

This review shows that spite the large number of research studies carried out in the last years, there are still many gaps that need to be solved. In this regard, there are significant knowledge gaps that require to be solved in future research. Thus, very little information is available concerning the dependence between the wear of the tyre and the type of existing road surfaces (i.e. asphalt concrete, porous asphalt, etc.). Another area of study is the size of the microparticles that are generated by tyre wear, as well as their way of dispersing and the potential modifications that they can undergo in the environment. Finally, the possibility of improving and validating the methods for sampling, detection and quantification of this type of microplastics should be further studied in order to achieve a practical and standardised method.

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