Scientific Report on Tyre and Road Wear Particles, TRWP, in the aquatic environment

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Preface

In July 2018, the European Tyre & Rubber Manufacturers Association (ETRMA) took a proactive approach and launched the European Tyre and Road Wear Particles (TRWP) Platform, facilitated by CSR Europe, to explore a balanced and holistic method in addressing and understanding TRWP. This multi-stakeholder platform brought together experts from governments, academia, non-governmental organisations and industries. Through an open and inclusive dialogue, the Platform aimed to share scientific knowledge, achieve a common understanding of the possible effects of particles generated during normal tyre use and wear, and co-design mitigation options to reduce TRWP.

This report presents a summary of the present scientific knowledge regarding TRWP and the recognised knowledge gaps. It is based on the available literature, with an emphasis on recent studies, publications, reports and presentations of participants at the high-level and technical meetings of the European TRWP Platform. The report is mainly directed at the nature of TRWP and their transport and fate from generation on the road to the freshwater systems.

In recent years, some very interesting and high-quality scientific reviews have been published and recent experimental studies and modelling efforts on TRWP have become available. These promise useful developments in the understanding of the issue. This report is not a typical and complete scientific review, but an evaluation and interpretation of results of others in view of an integrated consideration of the fate of TRWP from “road to ocean”. Thus, only major references were used, and their results are reported in short paragraphs and open questions and challenges are derived. Since TRWP have been generated for many decades, it is somewhat surprising that sound scientific knowledge of this topic is comparatively limited, compared to the emissions of heavy metals or organic trace contaminants from various sources (for example, pharmaceutical residues).

It is to be expected that considerable progress will be achieved in the next years on the understanding of the fate of TRWP in aquatic systems; several research projects have started and will help to support or reject a number of hypotheses or assumptions. They will contribute to the evaluation of the effects of TRWP in the environment and will be the basis for decisions on optional countermeasures.

1. General introduction

Since the introduction and use of rubber tyres for all kind of vehicles, mainly for cars and trucks, more than a century ago, the abrasion of the tyres is an essential process during their use. Abrasion and wear are a consequence of the multiple tasks of tyres, namely the good performance on all kinds of roads in view of safe driving during all weather conditions and under variable driving behaviour. The particles formed during the interaction of tyres and roads are named here **TRWP, Tyre and Road Wear Particles**, as they are mostly a conglomerate of tyre and road materials. Tyre wear particles can also be formed as evaporative emissions due to heating of the tyre followed by condensation and coagulation. This process forms particles in the nanoscale size and contributes much less to the total mass emissions. Due to their size below 1 mm in the largest dimension and their rubber (elastomer) content, they are presently included within the definition of **microplastics, MP,** in most publications and reports of the last decade**.** However, this inclusion is not universally accepted by all actors concerned or recognised in scientific and policy papers.

TRWP were investigated quite early in view of environmental aspects, for example their emission into the air as fine particulate matter (PM10 and PM2.5 as fine dust) or in pollution of soils near roads. The recent interest is mainly concerned with the emission into fresh and marine waters, in the same way as for plastic debris and for other primary and secondary microplastic particles. Concerns regarding their presence are based on findings of plastic and microplastic in biota and in the food chain and the presumed environmental and human health effects.

In recent years, numerous international research activities have started on large and micro-sized plastic in all relevant aspects, including early modelling of mass balances, but rarely validated by real-world monitoring data. Thus, we encounter extensive shortfalls in knowledge about plastic pollution and especially regarding the sub-topic of TRWP. This report is produced to summarise the existing knowledge and the major knowledge gaps for a comprehensive evaluation of TRWP in the aquatic environment (“from the road to the ocean”). This analysis includes consideration of some very recently available review publications.

Remark: This report does not include any studies and the state of knowledge about ecotoxicological effects in freshwaters, in estuaries and in the oceans and it does not cover human health risks via the food chain.

2. Definitions and Acronyms:

Plastic and microplastic:

A very recent paper by (Hartmann et al. 2019) appears to be the most comprehensive effort to recommend definitions and a categorization framework for plastic debris and microplastic. Definition by size is widespread, commonly particles below 5 mm (Thompson et al. 2004), but this classification is not universally accepted. The seven criteria proposed are:

I Chemical composition

Tyre wear (and road) particles are specifically listed in the proposed framework. They are described as a special case, as the tyre contains about 40 – 60 % synthetic polymers (styrene-butadiene-rubber, SBR, or/and butadiene rubber, BR). The authors are aware that TRWP contain road materials and that the rubber content will decrease considerably due to weathering. However, there are no proposals on the minimum percentage content of any polymer in mixed material particles. TRWP are included as microplastic because synthetic polymers are an essential ingredient of tyres.

II Solid State

Melting and glass temperatures are above 20 ^{0}C

III Solubility

Solubility is less than 1 mg/l at 20 ^{0}C

IV Size

Nanoplastic: 1 – 1000 nm

Microplastic: 1 – 1000 µm

Mesoplastic: 1 – 10 mm

Macroplastic: above 1 cm

The authors state that the 5 mm size criterion for microplastic has no clear scientific justification.

V Shape and structure

Spheres, irregular particles, fibres and films

VI Colour

Any kind of colour

VII Origin (optional)

Primary and secondary particles. Primary particles are either used as such or are formed during intentional use, while secondary particles are due to fragmentation during weathering.

The definition of microplastic in a recent ECHA report (2018)

The European Chemicals Agency, ECHA, issued a recent report "Note on substance identification and the potential scope of a restriction on uses of microplastics" (Version 1.1 16/10/2018). It is based on a stakeholder workshop on the intentional uses of microplastic particles, held at ECHA on May 30-31, 2018. Polymers are included in the REACH process for chemicals, but microplastics as such are not. Thus, the ECHA report tries to redefine the term "microplastic" by several criteria, including the polymeric nature of MP.

It covers mainly the primary microplastic particles and their intentional release and it states that the microplastic concern is principally limited to common polymer-based synthetic plastics. However, it may also exist if other persistent particles would contribute to the concern (e.g. "elastomeric materials)". The paper does not contain any section or words on tyre wear particles. However, the "elastomeric materials" could cover the tyre wear particles which fall into the category of unintentional releases (which will be included in future work of ECHA). The report contains a large table with 17 indicative uses of substances, but none of them is related to tyres and tyre wear.

A newly published report (as SAM Scientific Opinion Paper, April 2019) on "Environmental and Health Risks of Microplastic Pollution" by the Group of Scientific Advisors of European Commission includes tyres as a source of microplastic. The currently available evidence suggests that microplastic pollution at present does not pose a widespread risk to humans and the environment, but that there are significant grounds for concern and precautionary measures. In one of the recommendations the scientific advisors mention improved drainage-system interceptors for tyre abrasion microplastics. The nanoplastic issue is mentioned, as it has the largest knowledge gaps.

Conclusion*: Overall, the definition of tyre wear particles under the generic term "microplastic" is not yet fully clarified. However, an increasing number of papers (also cited below) are using this definition. The elastomers in tyres are considered to be a type of the different polymers which are the main ingredients of other plastics and microplastics.*

Specific definitions for particles from tyres and roads.

Tyre: rubber tyre containing natural rubber, NR, butadiene rubber, BR, styrene-butadiene-rubber, SBR, carbon-black, silica and various kinds of additives, including organic Zinc.

Tyre wear: The loss of tyre material in driving giving rise to the formation of particles.

Tyre particles, TP: Particles originating from tyre tread by different kind of processes, like cryogenically grinding, shaving, cutting, shredding, abrasion etc.

Tyre wear particles, TWP: Tyre particles formed by wear during driving on roads or in road simulators. It is a term formerly used, like in Kreider et al. (2009) or in other publications. It should not be used in further studies, as it has been applied with variable definitions.

Tyre and road wear particles, TRWP: Tyre wear particles containing particles from the road or road dust. It is the term recommended for further use, as it is more accurate than other definitions, namely TWP.

Road particles or road dust, RP or RD: Pure road particles formed by various processes, with or without interaction with tyre wear.

Road associated micro particles, RAMP: All kinds of particles originating from driving and from the road. This is a definition used by Vogelsang et al. (2018) in Norway and includes asphalt, concrete and fillers, polymers from low noise roads, road markings, road paintings, collected exhaust particles, brake wear particles etc..

Non-exhaust particles: All kind of particles produced by traffic, excluding the particles in the exhaust due to the combustion processes in the engine and remaining after exhaust treatment (like fine carbon black).

Primary microplastic type B: A new definition in a report (not peer-reviewed) of 2018 by Bertling et al. of the German Fraunhofer-Institute UMSICHT has been proposed, with the type A for microplastic particles intentionally added to consumer products and type B for those microplastic particles formed during the use of them (like tyre abrasion, textile fibres from washing or about 50 other listed sources of microplastic). Secondary microplastic is formed by degradation and weathering processes in the environment.

Conclusion: *A correct definition is vital for any scientific data. In this report, only two definitions will be used:*

TW or TWP: *Tyre Wear or Tyre Wear Particles are the preferred terms for the mass losses of tyres in driving and the mass balances and modelling of tyre mass flows in the environment, not including roadway particles.*

TRWP: *If a tyre is used for driving on roads, the generated particles are called TRWP being aggregates of TWP with road particles. If some of the generated TWP particles are not containing road wear particles (which seems to be a rare case), they may also be called TRWP, as they are resulting from the interaction with the road surface.*

3. Composition of tyres – an overview

As stated in a recent review on tyre wear emissions by (Panko, Kreider and Unice 2018), the manufacturing of a tyre is a complex process, requiring the use of a wide variety of chemicals, fillers and polymers. The main components of tyre tread are unreactive chemicals (polymers, fillers, oils, waxes, processing aids and antioxidants) and reactive chemicals (sulphur compounds, accelerators and retarders, adhesives and activators). Most of the reactive chemicals are consumed in the tyre manufacturing process, particularly in the vulcanization and curing steps.

Table 1 is derived from a review paper by (Wagner et al. 2018) and provides ranges for major chemicals in tyres.

Table 1. Composition of tyres

Compounds	Content in %	Ingredients
Rubber/Elastomer	40 - 60	Poly-butadiene (BR), styrene-butadiene (SBR), neoprene isoprene (NR), polysulphide
Fillers	20 - 35	Carbon black, silica, silanes
Process oils	12 - 15	Mineral oils
Vulcanization agents	1 - 2	ZnO, S, Se, Te, thiazoles, organic peroxides, nitrocompounds
Additives	5 - 10	Preservatives, anti-oxidants, desiccants, processing aids
Textile and metal reinforcement	5 - 10	

The content of SBR and Zn are useful for identification and mass quantification of TWP and TRWP, collected either on road simulators or in real driving conditions (see chapter 5).

Tyre tread composition varies between applications: car tyres are different from truck tyres. Truck tyres do not contain the SBR, which limits the analytical methods based on SBR in pyrolysis-GS-MS. The content of the tracer elements Zn and S are relatively constant and may serve as indicators for TWP (Kloeckner et al. 2019).

4. Formation of TWP and TRWP

Factors of influence

The use of tyres on vehicles driving on roads leads to tyre tread abrasion, which is a complex physico-chemical process at the interface of tyre tread and the road pavement. Large variations of wear rates are reported. (Panko et al. 2018) listed the factors affecting the tyre wear:

Tyre characteristics: Size, tread depth, construction, tyre pressure and temperature, contact patch area, chemical composition, accumulated mileage, wheel alignment.

Road surface characteristics: Pavement construction, aggregate rocks, binder (bitumen, cement), porous asphalt, macro and micro texture, porosity, condition, road surface humidity, road dust loading in surface texture.

Vehicle operation: Speed, linear and radial acceleration, frequency and extent of braking and cornering.

Vehicle characteristics: vehicle weight and distribution of loads, location of driving wheels, engine power, power/unassisted steering, electronic braking systems, suspension type and condition.

Tyre wear rates

The extensive list of influencing parameters above does not allow the provision of exact wear rates or wear masses, rather a wide range of values. Wear rates are available in many references and are used in mass estimations for emissions of TWP. Unice et al. (2019a) listed the following ranges of wear rates per vehicle by road type:

Urban areas: 60 – 850 mg/km
Rural areas: 39 – 546 mg/km
Highways: 47 – 668 mg/km

A summary report of CEDR (2018) lists published data on wear rates for different vehicles:

Table 1: Emission factor for tyre wear from roads (CEDR-report, 2018, references cited there)

Source	Emission factor in mg/vehicle km
Passenger cars	50 - 132
Light commercial vehicles	102 - 320
Trucks, commercial vehicles	546 - 1500
Buses	267 - 700
Motorcycles	39 - 47
Tyre wear, average	**90 - 270**

Figure 1 below is based on a review by Boulter (2005) and derived from a study by Councell et al. (2004) and shows the ranges of emission factors for a light duty vehicle. As a rule of thumb, the typical emission factor for all passenger cars is around 100 mg/vkm.

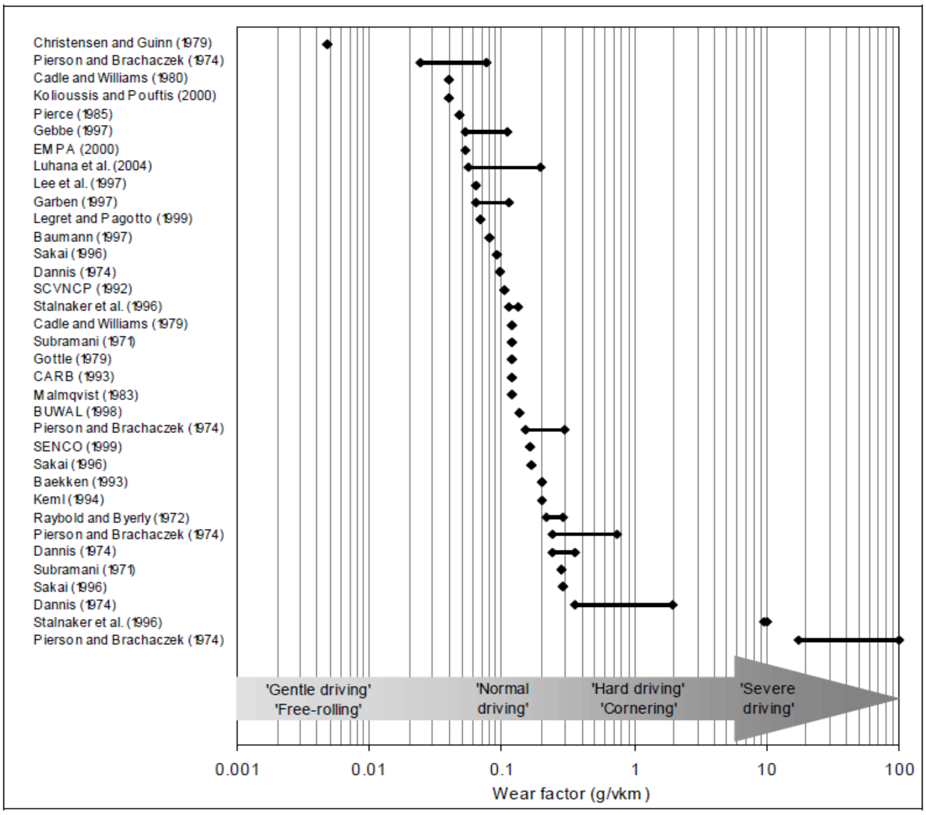


Fig. 1: Wide spread of tyre wear factors with 4 wheels. The range covers five orders of magnitude indicating the strong influence of driver behaviour (Boulter, 2005)

The relative influences of different factors on tyre wear are summarised in the Table 2 below. Factors with a high or very high influence can be associated with a large range of wear rates, with more than a factor of 10 between minimum and maximum values (as in Fig. 1). Thus, these strongly influential factors would offer the promise of significantly reducing the wear rates per vehicle and km driven. It is evident from Table 2 that all stakeholders are involved in tyre wear generation and every stakeholder category has factors with high or very high influence. Thus, achieving significant reductions of tyre wear would seem to be a multi-stakeholder task. However, in view of market trends with more and heavier vehicles and the growing market for electric cars with high instantaneous torques, it can be expected that aggregate tyre wear generation will increase, but to a yet unknown annual rate.

Table 2: Influential factors on tyre wear generation and relative importance. Summary of presentations in the CSR Technical Meetings.

Influencing factors	**Impact on the tyre generated by**	**Impact level on the tyre wear**	**Stakeholder category**
Tyre characteristics	Tyre type, tyre tread and rubber compounds	Very high	Tyre makers

	Tyre size	Medium	
	Mileage and Age	Medium	
Vehicle characteristics	Vehicle weight	High	Vehicle makers
	Suspension type: toe angle (single angle)	High	
	Suspension type: camber angle	Low	
	Vehicle Control	Medium to high	
Road surface characteristics	Road surface	Very high	Road makers
	Road and tyre interaction	High	
	Humidity and seasonal effects	Medium	
Driving behaviour characteristics	Speed	Medium	Drivers
	Acceleration, braking, cornering	Very high	
	Inflation pressure	High	
	Tyre maintenance (storage etc.)		

Some tyre wear mass balances

The annual total tyre wear in a given area (like a watershed) can be divided by the number of inhabitants, providing a specific emission in the range of about **0.28 – 4.7 kg/cap and year** (the highest value for the USA), with a global average value of 0.81 kg/cap and year (CEDR-Microproof report, deliverable 1.2, 2018). Unice et al. (2019a) found about 1.8 kg/cap and year for the Seine water catchment.

Another relative wear rate value can be derived from the total length of roads in a selected area. For Germany, the annual emissions of TWP are about 133.000 tonnes/year (Wagner et al. 2018) for about 670.000 km of paved roads. This amounts to an annual average specific tyre wear rate of about **1.6 kg/cap and year** (similar to the value of the Seine catchment) and to about **200 grammes/metre road**.

It has to be stated that these values above are all for **TWP only**, not including the road wear materials. Due to geographical variations, differences in climate, road surface characteristics, the fleet composition, driving behaviour and the types of brakes, the emission factors will vary widely.

The emissions of tyre-specific additives as a result of tyre wear are several orders of magnitudes lower than those of tyre wear particles, from 0.09 to 150 µg/vehicle-km (Panko et al. 2018). They are not covered in this report.

Conclusion: *The present knowledge of tyre wear rates and national/international masses of TWP generated seems to be relatively satisfactory for passenger cars, but remain uncertain for the heavy-duty vehicles (wide variation in numbers of axles and of tyres on each axle). There is an increasing trend in the use of these latter for freight transport. There is a need for forecasting TWP generation in view of some recognised developments:*

- *The trend to more powerful and heavier cars and trucks*
- *Increasing sales of cars and trucks*
- *Potential increase in distances driven*
- *The increasing trend towards electric or hybrid cars may also increase tyre wear, caused by heavier cars (battery weight) and their high instantaneous torque.*

Therefore it is recommended to develop a forecasting tool for predicting tyre wear in the next 5 – 15 years, including different scenarios.

5. Analysis of tyre and road wear particles in the aquatic environment

Introduction

In general, the qualitative and quantitative analysis of particles in size ranges from about 10 nm to 5 mm (covering the ranges of microplastic particles) in fresh waters, oceans, soils, biota and all kind of freshwater, estuary and ocean sediments is one of the most complex issues in modern environmental analysis. Analysis of these particles in air samples is comparatively easier and has been developed several decades ago, since atmospheric particulates became of great interest (“smog”). Air analysis is now a standardised methodology and in frequent use (such as PM10 and PM2.5 for particles below 10 and/or 2.5 µm). Similar approaches for the aquatic environment are still in their infancy and will need years of development and application checks.

The analysis of particles includes the sampling from the environmental compartments, sample handling and treatment and final analysis. All steps are sources of many errors and deficiencies. A new review paper (Stock et al. 2019) summarizes the sampling methodologies for microplastics in waters and sediments as well as sample treatment including density separation, chemical treatment and enzymatic sample preparation. There are no harmonised methods available at this date and it is the task of many groups to develop standard operating procedures or even standardised methods. The cut-off size in filtration is one of the determining factors, as many researchers use very different filter pore sizes, from below 1 µm (by membranes) up to meshes with 333 µm openings. Thus, many results reported are limited to a certain size range. The most difficult sampling is for particle sizes below 10 µm, a range which needs pressure driven membranes or filters.

The author is coordinating a German research team on microplastic analysis and has experienced a slow development process, leading to false positive and false negative results. This is confirmed by a new publication by Koelmans et al. (2019) on the assessment of data quality for microplastic in freshwaters and drinking waters. Their evaluation of 50 publications is based on rigorous quality criteria. They state that only 4 out of the 50 papers received positive scores for all proposed quality criteria, implying a significant need to improve quality of sampling from waters, sample treatment and final detection. The data quality of TRWP will be even worse, due to larger gaps in analytical procedures.

If tyre wear particles are the target, some of the methods for conventional microplastic do not work (like infrared, IR or Raman spectroscopy), due to strong interferences with the other compounds in tyre materials. This may explain why only a few researchers have tried to determine TRWP in environmental samples, either for qualitative or for quantitative data.

Qualitative analysis of TRWP/TWP

The task of qualitative analysis of TRWP is the identification of TRWP in samples, but not their mass or particle sizes and numbers. The results are: presence or absence. The microscopy methods (light and scanning electron microscopy) are most suited, but the particles can be only identified if they show clear characteristic features like shape, size, colour or any ingredient which can be seen by activation. The main problem in real world samples is the presence of other particles, mostly present in excess. Thus, a pre-step for separation of TRWP is needed, like density separation. Assuming a density of around 1.8 g/cm^3 (Unice et al., 2019a), the lighter and heavier particles can be separated by liquids of higher and lower density. This has been done in a recent paper by Kloeckner et al. (2019), with a salt solution of about 1.9 g/.cm^3.

The optical identification of TRWP is based on the assumption of an elongated shape in the size range from 5 – 250 µm (Kreider et al, 2009), the black colour and the observed inclusion of mineral road particles in the agglomerates. However, black particles are frequently found, not being TRWP and the agglomerates may be not stable in the aquatic samples, due to disintegration, a process not yet studied.

Kloeckner et al. (2019) proposed the use of laser ablation with the detection of Zinc as a marker for tyre material. Thus, a particle emitting Zinc into the mass spectrometer is identified as a TWP. The method has been applied only to a small number of samples where high portions of TRWP were expected (highway run-off treated in a settler and artificial wetland).

Scanning electron microscopy with the addition of elemental analysis are other approaches to identify and characterise the TRWP. However, the chances for the determination of the mass of TRWP/TWP in samples as well as the size distribution are low or are very time consuming, even for enriched environmental samples.

Quantitative analysis of TRWP/TWP

The quantitative analysis of particles in general needs a well-defined target system:

- Mass determination together with the identity of materials (thus concentration or content in environmental samples)
- Particle sizes and size distribution, particle numbers together with the identity for the different materials in the particles.

The present experience for microplastic analysis in the environment indicates that the different methods are not congruent for the same samples: The calculation of masses of MP out of particle types and sizes does not lead to similar results, due to severe limitations of all present methods.

Mass determination methods:

Rubber analysis

The specific mass determination for TRWP/TWP is the combination of pyrolysis with gas-chromatography and mass spectrometry, known as pyr-GC-MS. This relatively old method has been applied to tyre materials, especially for the three elastomers. The method has been used by several authors and is based on the pyrolytic formation of specific and volatile products which are separated in GC and detected and quantified in the MS. The sample mass is rather small, in the range of 10 – 100 µg absolute, as the GC-MS-system would otherwise be overloaded. The limits of detection are satisfactory, but it must be assured that the small sample mass is representative for a collected environmental sample.

(Eisentraut et al. 2018) showed recently in a new method development that out of the elastomers in tyres: NR, BR and SBR, only SBR offers two well-defined tracers which are useful for quantification. NR and BR either interfere with natural solid matter or do not offer clear decay products. An internal standard has to be added, a deuterated polystyrene. His method is a modification of the pyr-GC-MS, replacing the pyrolysis with a thermo-analytical treatment, gas phase extraction and subsequent desorption for the GC-MS. It is abbreviated as TED-GC-MS (Thermoanalysis-Extraction-Desorption-Gaschromatography-Mass-Sepectrometry) and is now in the market for microplastic identification and mass determination of the single polymer types. The limit for detection for SBR is about 0.2 µg, out of solid sample masses in the range of 50 mg. The solids are filtered from water samples or are from dried sediments, with a subsequent milling for homogenisation. SBR is, however, a rubber not used in truck tyres, thus they had to estimate the SBR content of all tyres to be roughly 11.3 % (based on their literature study).

Analysis of TRWP/TWP additives as markers

The inclusion of a number of additives in rubber production offers chances for environmental analysis, if the additives are specific for TWP. Wagner et al. (2018) summarized a number of studies on the analytical capabilities and limitations of various methods. They specified six criteria for markers in TRWP, either metals or organic compounds. Elements suitable for TRWP are reduced sulphur and zinc, if interfering sulphur and zinc are removed in sample treatment. Benzothiazole markers are another organic group, but they are leached off and are found in the aqueous phase in contact with TRWP. Organic zinc appears to be not suitable either. Wagner et al. (2018) list an additional number of candidate tracers, but state that satisfactory analytical approaches to determine the amount of TWP are not readily available. The challenges increase with increasing distance from the emission source. Thus, the analysis of rubber as such seems to be more promising, rather than searching for novel markers.

Particle sizes and size distributions for TRWP/TWP

The determination of particle sizes and size distributions is a standard analytical method for powdered products and in all kinds of environmental samples. For water and sediments, quick and automatic size determinations are commercially available and cover the range from about 0.5 to 1000 µm and more. They are based on mechanical sieving of dry or suspended samples or on optical or electro-physical measurements of separated single particles in flow-through capillaries or by particle movement in a sample cell. The methods are all not selective in a mixture of different particle materials. They are more or less either used in sample pre-treatment (sieving) or as a comparative measurement in addition to the specific detection of target particles (infrared or Raman spectroscopy, like for polymeric MP).

Single particle analysis for TRWP has been applied for “pure TRWP” but in environmental samples it is the topic with the lowest state of development. It is, however, needed for all kinds of scientific studies on the fate of TRWP. The task is similar to the analytical needs for other microplastic particles in view of their sources, transport processes, ageing and final sinks. Unfortunately, the basic methods of infrared and Raman microscopy (in development for MP) are not suitable for TWP and TRWP, due to matrix and fluorescence effects (Ivleva, Wiesheu and Niessner 2017).

The recent work of Kloeckner et al. (2018) may have the potential for a microscopic identification of TRWP by sample treatment with density separation, elution of mobile zinc and laser ablation of solid zinc from single particles and detection by ICP-MS (a standard method for elemental analysis). The zinc determination can identify the rubber content and thus, the identity of the particle. Sizes and shapes could be directly measured optically under the microscope. The analysis of particle by particle is probably quite time-consuming and needs some automation before it can be applied in practice.

Conclusions:

The present methodology for sampling from waters, for sample treatment and for final analysis of masses and single particles of TWP/TRWP is insufficient and neither harmonised nor standardised. There are some recent developments which appear somewhat promising.

The different methods under development for microplastics are only partly suitable. The analytical results are needed for validating modelling efforts and for the risk assessment of TRWP in the environmental compartments.

6. Characteristics of tyre (and road) wear particles

Sizes of TRWP/TWP

The determination of tyre and road wear particle characteristics is a complex scientific task and depends on the methodology of their generation (and many influential factors as described above). Studies by Kreider et al. (2019) are cited frequently and included different modes of TRWP generation, including on-road collection, laboratory generated particles under simulated driving conditions and cryogenic grinding of tread rubber. The particles from on-road collection and by laboratory generation exhibited the elongated shape morphology of tyre wear particles ("sausage like morphology"), whereas cryogenically-generated tread particles were more angular. The chemical composition also differed, with the particles from on-road collection containing chemical contributions from other sources, such as pavement or particulates from the traffic-related sources. Particle size distributions are presented as well as chemical data on metallic content and polycyclic aromatic hydrocarbons (PAH).

The sizes range for TWP and TRWP from about 4 to 250 µm, as determined by two different methods. The particle size distribution of TRWP was bimodal, with maxima at about 6 and 25 µm, while the volume distribution had a mono modal shape and a maximum near 100 µm. It should be noted that the volume and mass distributions are severely influenced by the larger particles. However, environmental behaviour is usually more important for smaller particles.

Wagner et al. (2018) summarized the literature data on size distributions of TWP and TRWP and listed results from 1 nm to hundreds of µm for road simulators, from about 0.4 to 20 µm for on-road driving and above 1 µm to 200 µm from road run-off. The authors state that the discrepancies in the obtained particle size distributions can result from different conditions of particle formation, in sampling and in the size analysis method. The fine particles below 10 µm or 2.5 µm are most relevant for the air-borne TRWP, making up some share of the fine dust air pollution (Grigoratos et al., 2014, Grigoratos et al., 2018). The percentage ranges of these small fractions, related to the total formation of TWP are estimated at about 0.1 – 10

%, which is a wide range (Wagner et al, 2018). The ultrafine particles below 0.1 µm are heterogenic and seem to originate from organic constituents of tyres (maybe also sulphur). The air-borne particles within the topic of TRWP/TWP are not further considered in this report, but were already subject of many studies several decades ago and in recent work (in air pollution research). A new publication by (Foitzik et al. 2018) demonstrates influential parameters on the size distribution and number of ultrafine particles generated in a tyre-road simulator.

Depending on the sampling procedure, the smaller fraction may be lost (passing the pores of filters), as observed in microplastic sampling. The larger particles are relatively rare, thus a high-volume sampling is needed for representative values. Overall, the reported size distributions have to be considered with great care.

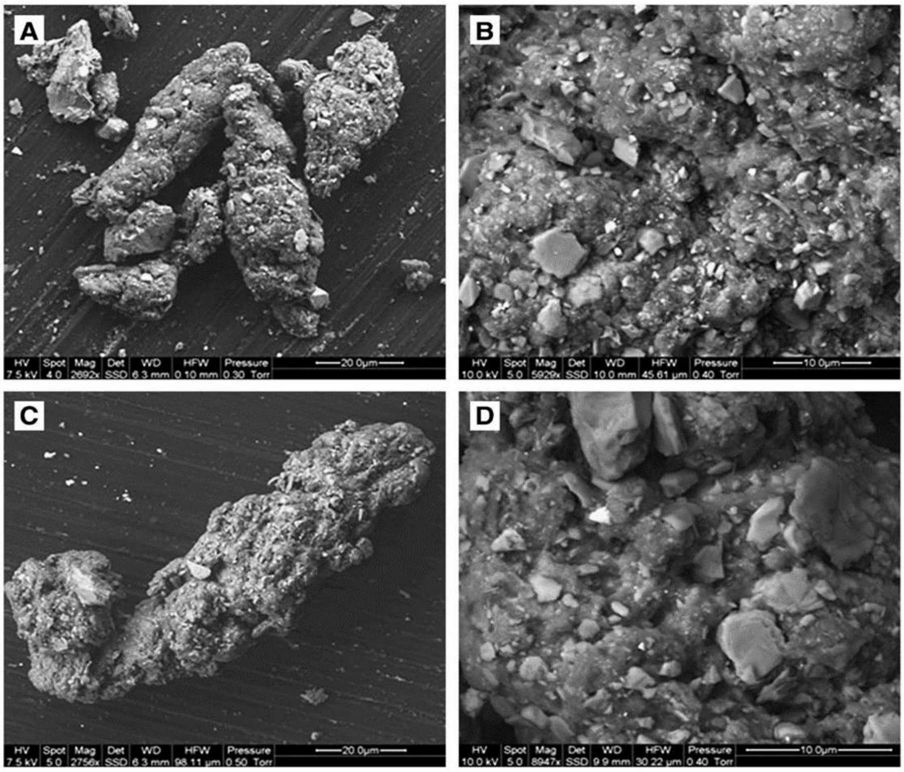
Composition of TRWP

The microscopic pictures below (Figure 2) show TRWP as produced in a previous TIP-project and published by (Kreider et al. 2009).

The main contributors to the chemical compositions for TRWP are the tyre wear and the road materials, like asphalt, and concrete. The relative mass shares are estimated to be about 40 – 60 % of road material, with a high density of about 2.4 g/cm^3, while the tyre has a density of about 1.1 - 1.2 g/cm^3, giving an average density of the agglomerate of 1.8 g/m^3 with 50 % mass contributions each (Unice et al, 2019a).

Figure 2: TRWP photos under the microscope by Kreider et al. (2009)

- **Scanning electron microscope images of particles collected on road (A, B) and particles collected in BASt (C, D).**
- **Mineral incrustations are evident in the photos of greater magnification (B, D).**



(Sommer et al. 2018) collected coarse particles above 10 µm by passive aerosol sampling in the air near three German highways and found by particle analysis strong indications of combined materials in the TRWP. The data are only qualitative but support the hypothesis of agglomerated tyre and road particles generated on roads, but do not allow the derivation of mass fluxes of TRWP.

There are as yet no experimental density measurements available for TRWP, which is a major prerequisite for environmental modelling and fate analysis. The difference of particle density to the water density is one of the major factors of influence for settling rates, as described in the classical Stoke´s equation for spheres (for laminar settling of particles in the lower size ranges).

The stability of TRWP in view of their content of tyre and road materials is another unknown factor. If we assume that the road particles can be released from the agglomerates, the density will approach the lower TWP value of about 1.2 g/cm^3 or even below, due to bound water in the pores. Low particle densities near the density of water will lead to low or negligible settling rates and thus to long ranges of transport in surface waters, including the transmission into estuaries and the receiving oceans.

Due to the agglomeration with road dust, the density of TRWP may vary, depending on the type of road material, the size of road dust and ageing effects of road surfaces.

TRWP are not the only particulates on roads, as we also find many natural and man-made particles (depending on location, season, wind etc.). They may be included in the TRWP materials, but no investigations have been found in this regard. In addition, the homo-coagulation as well as hetero-coagulation of TRWP may occur on the road, which may increase the effective diameter and sizes of aggregates, before they are washed off the road surfaces or from the roadside verges. During longer dry periods, the TRWP deposited may undergo more mechanical stress by the traffic and be milled down to smaller size particles (a process not yet studied).

Conclusions:

The composition of TRWP is very complex, as agglomerates are formed on the road with different particles from the road material and from other dust. Exact knowledge of the constituents of TRWP would require new methods and representative sampling programmes for the major conditions of generation. It is questionable whether these specific efforts would provide more and useful knowledge on TRWP. It may be more reasonable to sample the TRWP emitted from the roads into air, soils and run-off and determine their gross parameters, like size, shape, density and TWP-content, as these are more influential on the fate of TRWP along their path-ways.

The mitigation of TRWP formation by low-wear tyres has to be investigated carefully in view of the particle sizes formed in relation to conventional tyres. A lower mass loss may be associated with smaller TRWP sizes, which are less effectively removed in rivers by settling (see later chapters).

7. Processes and Fate of TRWP

Wash-off from road surfaces

Establishing which size fractions are mobile from the road surface is an essential aspect in determining TRWP particle sizes and their fate in the further transportation into soils and run-offs. The data in the references ((Kreider et al. 2009) and (Wang et al. 2017) are not conclusive, but it is hypothesised that small particles (say below 1 - 10 µm) are not transported by splash and spray or washed-off, as the shear forces (including washing water in wet street cleaning) are not high enough for detachment. On the other hand, small size fractions which are not firmly attached to the pavement texture may be more easily transported away.

The mass fractions of TRWP transported via run-off are estimated at about 50 % with a large range of 25- 75 % (Unice et al., 2019a), based on other studies. The storage of about 1 % on the road surface appears small but it could include the very small fractions which are most mobile in the aquatic environment.

This topic definitely needs more study by real road experiments and analytical methods for single TRWP size measurement and the mass flows in run-off. The permanent deposition of small TRWP fractions on the road will influence the overall environmental fate.

According to Stoke´s equation the settling rate is proportionate to the square of the particle diameter, thus 10 µm particles will settle at the rate of only 1 % of 100 µm particles with the same density.

The particle size distribution of the TRWP formed on the dry road surface has been studied in various settings, while very few data are available for the particle sizes in the run-off. However, the detachment from the road surface is again a rather complex process, depending on the preceding dry time, the traffic, the roughness and porosity of the road surface, the accumulated solid matters, the characteristics of the rain events etc. It can be predicted that experimental studies directly in the run-off will deliver quite variable values. Representative sampling during rain events is rather difficult for suspended solids. Thus, investigations and sampling in drainage systems equipped with settling ponds (at highways) or any kind of filters (retention soil filters, artificial wetlands etc.) will provide integrated samples and information on the particles sizes collected over longer times.

In summary, the knowledge on particles sizes, particle morphology and the effective density are major determining factors for the further fate of TRWP in aquatic systems.

Conclusions:

The state of methodology is not satisfactory for major questions, leaving room for more studies, but with reliable methods for TRWP particle size analysis and mass determinations via the SBR or zinc content. In relation to the general fate of TRWP in aquatic systems, it seems to be more important to know the particle size distribution and the mass flows in the run-offs than the TRWP generated on the different kinds of roads. Another important and overlooked aspect for the environmental transport by water is the mechanical stability of TRWP and effects of shear forces on the separation of TWP and RWP from TRWP This would lead to smaller sizes and especially to a much lower particle density (1.10 – 1.20) instead of about 1.8 g/cm^{3}*.*

Fate of TRWP in road-side soils

Major fractions of TRWP are transported via splash or via air to the road-side soils, where they are deposited and eventually degraded or bound into the soil matrix (as bound residue, known for hydrophobic persistent organic pollutants (POPs)). The percentages of TRWP to this transport pathway seem to be in the range of 25 to 75 % (average 50 %) according to Unice et al. (2019a). The measured TRWP in road-side soils decreases rapidly with the distance from the edge of the road. The soil is one of the important sinks for TRWP and offers possible chances for degradation of TRWP or the fixation of TRWP in the soil matrix as bound residue. However, these important processes were rarely studied, due to lack of analytical methods. An older publication of Cadle and Williams (1980) on the degradation of the SBR in test soils is the only available information. It states a half-life of 490 days, which is a significant time period for removing TRWP from the soil. It is surprising that no other and more recent studies are available on TRWP degradation in soils, as this compartment offers multiple options for degradation processes by bacteria and especially by fungi. Fungi can produce oxidizing extracellular enzymes which are known to attack even lignins in wood or man-made pollutants. The erosion from road-side soils including from land that received activated sludge with TRWP was estimated by Unice et al. (2019a) to be a negligible pathway in the watershed of the Seine River. Thus, the soil compartment is the most dominant sink for about 50 % of all TRWP generated on the roads.

Conclusions:

The road-side soil is one of the important sinks for TRWP but studies on the fate and the rates of degradation are very rare. It is recommended to initiate sound studies on TRWP in different soils under realistic conditions with an appropriate analytical coverage of the physico-chemical and biological reactions in soils. Biodegradation and fixation of TRWP in the soil matrix may include soil fungi, besides bacteria.

The study times are expected to be longer than usual, in view of the reported half-life of about 500 days. An alternative could be the measurements of TRWP in road-side soils for some selected sites where the history of traffic is documented and the emissions of TRWP could be back calculated (although with larger error margins).

TRWP in drainage systems with and without treatment

Run-off from highways, urban and rural roads is frequently collected via installed drainage systems with the out-flows

- directly into receiving water bodies (streams or lakes or oceans without treatment) or
- into specific treatment units (for all kinds of road run-off)
- into separate sewers on urban roads (with or without treatment) or
- wastewater treatment plants (typically designed for dry weather and moderate rainfalls) and
- into the combined sewer overflows, CSO, during heavy rains.

The partition between these five pathways is highly variable and depends on location, rainfall events and the rural and urban drainage design. If no treatment is installed, removal of TRWP will be close to zero on a long-term, while on short time frames, the settling in the collection systems may remove at least some coarser materials.

The present knowledge of the efficiencies of some major treatment systems is summarized below (Unice et al. 2019a):

Infiltration basins and ditches for rural runoff: 60 – 90 %

Storm water treatment for highways: 40 – 60 %

Wastewater treatment (not CSO): 90 – 100 %

The high efficiency of wastewater treatment plants reflects the general removal of all particles (including the microplastic and all suspended solids), leaving the combined sewer overflow (a more or less diluted raw sewage) as a major source of surface water pollution, including TRWP (as they are washed-off specifically during heavy rains, the cause of CSO events). It is surprising that many wastewater operators do not know much about the share of CSO in their annual wastewater flows. Berlin has estimated the CSO share at about 3 %, with some decreases due to retention basins in the collector systems, but only about 20 % of the city area has a combined sewage system. For Paris, about 40 % of CSO were reported, due to a full coverage of combined sewers. It can be predicted that climate changes will induce more intensive rains and thus an increase of CSO flows.

Conclusion:

The data available for drainage systems (separate and combined sewers) and run-off and sewer overflow treatment are yet insufficient for a sound evaluation of current practice. The mass fluxes of TRWP in combined sewer over-flows and run-off are probably more important than the treated wastewater effluents, thus they deserve more studies based on different installations. The developments for CSO and run-off treatment systems need to cover in addition different target pollutants (bacteria, nutrients, suspended solids, trace organics), not only TRWP.

TRWP in receiving freshwater bodies

Freshwater bodies are roughly separated into lakes and flowing rivers, with some differences in the expected fate of particles in general and TRWP in particular. The properties of TRWP in size and density favour the sedimentation to aquatic sediments, while photolytic and biological degradation or floating and volatilisation in the water phase are presumably not important processes. Sediments are habitats for many organisms, including fish, which may accumulate compounds and particles present in sediments. However, no studies exist on this pathway, due to the absence of a sensitive analytical method for biota.

Degradation may be expected in sediments but with quite long half-lives. Unice et al. (2019a) estimated a half-life of about 5000 days, ten times of the half-life in soils, but there are no experimental proofs. There are no data available on the kinetics of degradation in sediments of fresh or salt waters. Fine river sediments undergo hydraulic transport downstream by flood events, while lakes bury the particles for long periods.

The settling rates of particles in liquids are a well-known process for spherical particles, but are more complex for non-spherical particles. Some authors have developed empirical formulae for different shapes and sizes of particles, providing more accurate estimations of settling rates.

Density and size are the most important influential parameters:

- the settling rate is proportional to the density difference of the particle and the liquid: The correct density includes the water-filled pores of a particle, not the pure material density, and a coverage layer of particles, such as a biofilm with its own density
- the settling rate for small particles is proportional to the square of the diameter, which is a strong dependency. The settling rate is thus 100 times faster for two particles with a size difference of a factor 10 (like 10 and 100 µm). The correct diameter has to include the film layers often found on particles (biofilm or an organic layer of natural organic matter).

(Unice et al. 2019b) could show by modelling of the fate of TRWP in rivers Seine and Scheldt that these parameters have the highest influence on the mass balances in the rivers, where about 90 % of the inflowing TRWP mass is removed by settling to the sediments. The experimental check on the TRWP content of Seine sediments indicated a reasonable modelling result. The particle fractions not prone to settling are in diameters below about 20 µm and with a density closer to the density of water.

The average contents of TRWP in sediments (by models for erosion and TRWP settling) were 0.24 % (2.400 ppm) for the Seine sediments and 0.32 % (3.200 ppm) for the Scheldt sediments. If an analytical method is applied for sediments, its limit of quantification should be lower, around 0.01 % or 100 mg/kg or 100 ppm. The method of (Eisentraut et al. 2018) has a limit of quantification for SBR only of about 4 ppm (50 mg of solid sample). Taking a factor of about 9 for SBR to TWP, the limit of quantification is 36 ppm as TWP, which appears to be sufficient for sediment analysis.

Conclusions:

Overall, the river and lake sediments are natural sampling systems for capturing an average content of TRWP and they integrate spatially over the upstream watershed. They are easy to sample and analyse and may be a very informative compartment, while sampling from the running waters is much more difficult, providing only spot samples with high variations in the results (considering the high flow variances of rivers). Lake waters are more mixed and the monitoring results appear to be more constant and provide an average information on the influents from the watershed.

TRWP in estuaries and oceans

Data and knowledge about TRWP in estuaries and oceans are not available due to complex analytical issues. Up to now only mass balance modelling has been applied to estimate the release of TRWP from freshwaters to the estuaries and oceans. The most comprehensive and integrated model system by (Unice et al. 2019a) ends at the interface of freshwater and estuaries of the rivers Seine and Scheldt. Their prediction of TRWP outflows amounts to about 2 % of the mass generated on the roads, quite similar for the 2 watersheds. The parameter variations applied in about 1000 model runs indicate a release in the range of 1.4 – 4.9 % by mass for the 25 -75 percentiles of the modelling variants. The predicted range widens to 0.97 – 13 % for the percentile range of 10 – 90 %. The authors show the need for in-situ characterization of TRWP sizes and densities at the interface of freshwater and estuaries to validate their model results.

Despite the low expected percentage transfer from terrestrial and aquatic freshwaters to the oceans, the annual mass flow per capita will be in the range of about 20 – 40 grams of TWP. The concentrations of TWP in the Suspended Particulate Matter, SPM, can be calculated from the data by dividing the TWP mass flows by the SPM mass flows on an annual basis and give values of 300 ppm for the Seine and 500 ppm for the Scheldt basin. These relative contents for TWP are about a factor of 10 lower compared to the river sediments (see above: 2400 ppm for Seine, 3.200 ppm for the Scheldt basin). The reason for this difference is probably the lower particle sizes of SPM compared to TRWP and thus, lower settling efficiencies in rivers.

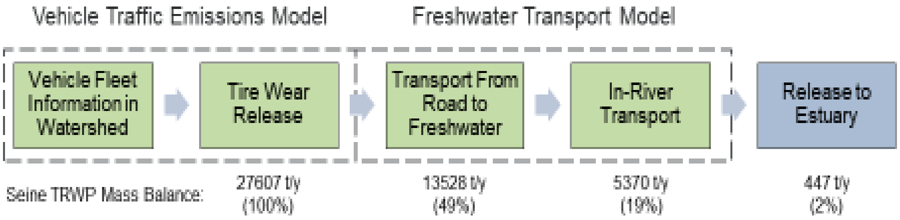
Conclusions:

The further fate of SPM and TRWP in estuaries is an open topic for research. The hydrodynamic, physico-chemical and biological processes in estuaries are extremely complex. However, it can be predicted that they have important influences before the open ocean is reached. The estuaries are well-known for their particle removal effects due to the salt increases in the mixing zones during the tidal changes. Raising salt concentrations will induce the destabilisation and coagulation of charged particles (all are negatively charged), forming larger aggregates which settle faster than the single particles. Thus, we expect a strong gradient in the turbidity and SPM content from the freshwater body to the open ocean and vice versa, an enrichment of SPM and TRWP in the estuarine sediments. The sediment sampling along transects of selected estuaries with a sensitive method for TRWP is recommended.

8. Modelling of TRWP in the aquatic environment

The integrated model approach by (Unice et al. 2019a, Unice et al. 2019b) is the best available effort for estimating mass fluxes for TRWP from the road to the entrance to estuaries. Their study is based on two watersheds of Seine and Scheldt Rivers and provided the following simplified results:

Figure 3: Mass fluxes of TWP in the Seine river basin. Data on Scheldt River are very similar.



Both watersheds are similar in view of the mass fluxes. About half of the TWP generated is released to freshwaters and close to 20 % are transported in the rivers. About 90 % of the TRWP in the rivers settle, while about 2 % of the formation of TRWP (or 447 t/a) are left for transport to the marine environment. The data are based on a central estimate of a number of important parameters. The details of this balance were described above in the subsections on the fate of TRWP. Taking the estimates of more than 500,000 t TRWP annually in the EU-28, the 2 % would be about 10.000 t/a (if the balances are comparable in all watersheds).

The model has been checked partly by former experimental measurements of TWP by Pyr-GC-MS in Seine sediments, with an average content of about 2.200 ppm, which is in the range of the expectations based on parameter variations. However, efforts for the model validation would be very helpful to confirm a number of assumptions and in decisions on the major and dominating processes, while other aspects could be deleted as negligible effects.

Conclusions:

The best available modelling efforts show clear trends in view of sources and sinks for TWP, regarding also the changes in properties from TWP to TRWP. Road-side soils, drainage treatment systems and the river sediments are the dominating sinks for TRWP, while a few percent (below 5 %, central estimate of 2 % in two watersheds) of the mass seem to enter the estuaries. The influential factors are surprisingly limited to mainly the particle diameter, the density and the possible biofilm cover (changing size and density).

The modelling results need more validation efforts by monitoring soils, sediments and freshwater flows to estuaries by sensitive methods (low partition of TRWP in other solids, usually below 0.1 %).

The fate of TRWP-fractions inflowing into estuaries is widely unknown and needs sound studies, mainly on some cases with a well-documented watershed. TRWP could become part of the estuary sediments, thus are not transported into the open oceans.

9. Mass fluxes of TRWP in comparison with other microplastic

Several research groups tried to estimate mass fluxes of TWP/TRWP as a part of all microplastic emitted (primary MP) or formed in the environment (secondary MP). The main problems in evaluating these estimates are different data bases and different locations in the environment where the fluxes are calculated or estimated.

A report by the Danish Environmental Protection Agency (Lassen et al., 2015) balances the total emissions of microplastics and the ultimate emissions to the aquatic environment. The share of tyre wear on the total emissions is estimated at 56 % and the share on emissions into the aquatic environment at 60 %

Boucher and Friot (2017) estimated the global evolution of the primary microplastics to the oceans and reported a share of about 28 – 46 % for tyre wear, depending on the scenarios pessimistic, optimistic and central. The report by IUCN is not a peer-reviewed publication and it does not cover the secondary formation of microplastic from larger plastic debris.

The EUNOMIA study (Hann et al, 2018) was directed to emissions of microplastic into the aquatic environment in Europe. About 0.5 million tonnes of tyre wear (about 1 kg/inhabitant per year) are generated in Europe, being the most important primary source of microplastic. About 20 % of the generated tyre wear is emitted annually to the surface waters. This estimate is close to the detailed modelling of Unice et al. (2018a) for the watersheds of Seine and Scheldt rivers (17 and 19 %).

In a peer-reviewed publication (Siegfried et al. 2017) established a mass balance approach for export of microplastic to the sea. They claim a share of 42 % of all MP flows to the sea to have an origin from tyre wear. The absolute mass flows presented are 1,600 tonnes/year for the Atlantic Ocean, 500 tonnes/year for the North Sea and 400 tonnes/year for the Baltic Sea.

Bertling et al (2018) published a report by Fraunhofer Association on macro- and microplastic in the environment (in German, not peer-reviewed). Their mass balances are best estimates from a number of participating partner organizations (consortium study) and includes tyre wear as one of about 50 different sources of plastic debris. The share of tyre wear has been estimated at about 30 – 42 %, but the sources of their data are unclear. The mass balance is for the generated masses only, not the transported masses or emissions into freshwaters or oceans.

Conclusions:

The studies cited above show in general that the tyre wear has a significant share in general microplastic emissions, even though the percentages in these reports are not comparable due to different assumptions and target points in the environment. It can be expected that new publications will become available in the next years with similar approaches. The databases are unfortunately not the same and the authors seem to define microplastic in different ways. It is a task of environmental research to refine the mass balances together with the validation of existing or newly developed models for integrated balances of tyre wear and micro- and macroplastic.

10. Knowledge gaps

Despite the longstanding emissions of tyre and road wear particles, the present scientific and practical knowledge on the whole pathway from the road to the receiving oceans is quite limited and a number of knowledge gaps can be identified. Some topics, already studied, need confirmation studies, other topics are still open for starting research projects. The integration of all relevant aspects of TRWP is essential for a sound evaluation of TRWP in the aquatic environment and will need several years. The topic of TRWP appears to be more complex than of other microplastic particles, where the present knowledge is also rather limited.

The major knowledge gaps are listed below:

- The need for a reliable and representative tyre abrasion test, including the preparation of reference TRWP for research
- Influence of road parameters on TRWP generation, particle size distribution and particle properties
- Influence of tyre design parameters on TRWP generation and particle size distribution
- Influence of driving conditions on TRWP generation and particle size distribution
- Influence of environmental conditions, especially ambient temperature, on TRWP generation and particle size distribution
- The analytical tools for TRWP are insufficient, namely on the particle properties and their changes in the pathway to the ocean
- TRWP in real run-off from roads are the primary source for aquatic emissions, but the present data on the particle sizes and mass flows are insufficient.
- Soils are probably important sinks for TRWP, but very few studies exist on the fate processes and TRWP contents
- Capture systems for TRWP on the road, in the run-off and in wastewater appear to be suitable for reducing the outfall into rivers
- Settling of TRWP in the rivers appears to be an important process, but needs more detailed field studies
- The estimates for TRWP emissions into the estuaries and oceans indicate a low percentage (2 – 5%), which must be validated for different watersheds

- The processes in the estuaries from different watersheds are unknown, as well as in the oceans and on the beaches.
- Integrated mass and particle balance models are very useful, if their input data are validated and if their estimations are confirmed by specific monitoring.

List of abbreviations

BASt	Bundesanstalt für das Strassenwesen, German Institute for Roads
BR	Butadiene rubber
CEDR	Conference of European Directors of Roads
CSO	Combined sewer overflow
CSR	European Business Network for Corporate Social Responsibility
ECHA	European Chemicals Agency
ETRMA	European Tyre & Rubber Manufacturers Association
ICP-MS	Inductively coupled plasma with mass spectrometry
MP	Microplastic
NR	Natural rubber
PM10	Particles below 10 µm in air
PM2.5	Particles below 2.5 µm in air
ppm	Parts per million
pyr-GC-MS	Pyrolysis coupled to gas-chromatography and mass spectrometer
RAMP	Road associated micro particles
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
RWP	Road wear particles
SBR	Styrene-butadiene rubber
SPM	Suspended particulate matter
TP	Tyre particles
TWP	Tyre wear particles
TRWP	Tyre and road wear particles

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