Review

Tyre and road wear particles (TRWP) - A review of generation, properties, emissions, human health risk, ecotoxicity, and fate in the environment

Beate Baensch-Baltruschat a,⁎, Birgit Kocher b, Friederike Stock a, Georg Reifferscheid a

a Federal Institute of Hydrology, Am Mainzer Tor 1, 56068 Koblenz, Germany
b Federal Highway Research Institute, Brüderstraße 53, 51427 Bergisch Gladbach, Germany

HIGHLIGHTS

• Current knowledge on TRWP is reviewed, environmentally relevant data is compiled.
• Data on annual tyre wear emissions estimated for various countries is reported.
• Entry paths into the environment and environmental fate are discussed.
• Degradation and ecotoxicological studies seldom apply real-world conditions.
• Environmental monitoring of TRWP is still hampered by challenges for analytics.

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ABSTRACT

In this paper, the current knowledge on tyre and road wear particles (TRWP) is compiled regarding all environmental and health aspects. TRWP generated on roads during driving processes contribute to airborne non-exhaust emissions and are discussed in connection with the microplastic pollution. The major amount of TRWP consists of coarser heterogeneous particles released to road surface, soils and aquatic compartments. The extensive compilation of annual emissions of tyre wear for numerous countries shows per-capita-masses ranging from 0.2 to 5.5 kg/(cap*a). Ecotoxicological studies revealed effects on aquatic organisms, but test concentrations and materials do not reflect environmental conditions. Contribution of tyre wear to PM10 accounts for up to approx. 11 mass %. A recent thorough risk assessment indicates the risk for human health via inhalation to be low, but no information is available on the risk caused by intake via the food chain. Data on degradation is scarce and most studies do not use realistic materials and conditions. The only published degradation study performed under environmental conditions implies a half-life of tyre rubber particles in soils of 16 months. For truck tyres, which mainly contain natural rubber, shorter periods were observed under optimum conditions in laboratory tests. Concentrations of tyre wear compiled from environmental monitoring studies show highly variable concentrations in road runoff, road dust, roadside soils, river sediments and river water, with a general decrease following the transport paths. However, the behaviour of TRWP in freshwater referring to transport, degradation, and sedimentation is still unclarified. Environmental monitoring of TRWP is still hampered by challenges for analytics. Thus, data on environmental concentrations is rare and has mainly exemplary character. Further research
is needed with regard to emission factors, development of analytical methods for environmental matrices, long-period monitoring, fate in surface waters and soils, (eco)toxicological impacts and degradation under realistic conditions.

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1. Introduction

Besides other components as fillers, softeners, vulcanization agents and various additives, tyre wear contains about 50% natural and synthetic polymers (Sommer et al., 2018) and is probably the most dominant source of synthetic polymer-based material released to the environment (Lassen et al., 2015; Magnusson et al., 2016; Sherrington et al., 2016; Sundt et al., 2014). For instance, the contribution of tyre wear to the total MP emissions is reported to be >50% in Denmark and Norway and about 30% in Germany (Bertling et al., 2018; Lassen et al., 2015; Sundt et al., 2014). According to Wagner et al. (2018) about 1.3 million tons of tyre wear are generated on roads in Europe per year. With regard to these figures the topic ‘tyre wear emissions’ deserves attention.

Most authors refer to tyre wear as microplastics (MP) due to its (semi-)synthetically produced polymer structure, solid state, insolubility and particle size included in the size range defined for MP (1–1000 μm, see Hartmann et al. (2019)). Properties and environmental behaviour of elastomer particles from tyres, which are formed directly at the emission source ‘road traffic’, probably differ substantially from plastic items referring to the latters’ partly crystalline structure, chemical compositions, shapes and surface. In the environment, pure tyre wear particles are rarely found. Instead, particles with a mixed composition consisting of tyre and other road related wear particles are present (Dall’Osto et al., 2014; Sommer et al., 2018). However, in scientific literature the term ‘tyre wear’ is often used even if the heteroaggregates of ‘tyre and road wear particles’ are meant. In the present article, we apply the term ‘tyre and road wear particles’ (TRWP) as far it is suitable whereas we name TRWP found in environmental compartments ‘real-world TRWP’.

Besides coarse TRWP, which are deposited on the road, transported to soils near the roadside or to surface waters, a fraction of smaller particles compatible with aerial transport is generated during driving processes. With regard to human health risk, this fraction was for a long time the main topic concerning tyre wear. Its percentage amounts to max. 10% by mass of total tyre wear emissions (Panko et al., 2013a; Park et al., 2018; Wik and Dave, 2009). Traffic-related so-called non-exhaust emissions, which include tyre, road and brake wear as well as road dust resuspension, are broadly discussed as a contribution to particulate matter (PM) in ambient air (e.g., Amato et al., 2014a; Denier van der Gon et al., 2013; Panko et al., 2019). In general, traffic emissions are the main source for exceeding the European Union annual limit value for PM10\(^1\) of 40 μg/m\(^3\) (Denier van der Gon et al., 2013). The contributions of exhaust emissions and non-exhaust traffic emissions to the PM10

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concentrations in ambient air are nearly in the same range. For instance, in urban areas close to roads, Bukowiecki et al. (2010) found a contribution of non-exhaust emissions to the traffic-related PM10 burden about 60%, Weinbruch et al. (2014) determined a percentage of 73%. Since exhaust emissions have been decreasing due to various mitigation measures, the percentage of non-exhaust emissions is growing (Denier van der Gon et al., 2013). The transition of motorized individual transport to e-mobility will lead to a further reduction of exhaust emissions, but not to a decrease of tyre wear emissions (Sommer et al., 2018). The latter are actually expected to increase in absolute terms due to growth of traffic volume (Denier van der Gon et al., 2013). However, the contribution of tyre wear to traffic-related emissions referring to PM10 is relatively low, amounting up to 11% according to Gehrig et al. (2001), Bukowiecki et al. (2010) and Harrison et al. (2012).

As to aquatic environments, TRWP have been addressed in connection with MP pollution in recent years (e.g., Verschoor et al., 2014). The pollution of aquatic ecosystems, especially of the marine environment with MP has been the topic of numerous studies (e.g., Andrady, 2011; Thompson, 2016), but MP in freshwaters also got into the focus of discussion (e.g., Baensch-Baltruschat et al., 2017). Due to their relative high density it can be expected that TRWP play a minor part in the marine environment compared to other polymers of lighter specific weight, but limnic surface waters might be affected even more. Since MP particles are ingested by various aquatic species including e.g. bivalves and fish (O’Connor et al., 2019; Roch et al., 2019; Zhao et al., 2019) the ecological risks as well as the risks for human health via food chains are under discussion. This concern may also apply to tyre wear.

With this article, we intend to give an overview on the generation of TRWP, their release, distribution and occurrence in the environment, environmental degradation and ecotoxicological and human health risks. It is intended to serve as an information source explaining the key issues of TRWP. Since numerous studies have been published on MP in general including reviews and monographies in which information on various issues of MP is compiled, this article is focused on the topic of TRWP. Exceptions are made for those items for which only data on MP, but not on tyre wear and TRWP are currently available. To our knowledge, this is the first literature review which compiles data on all environmental aspects with regard to tyre wear and TRWP.

2. Methods

A comprehensive research was conducted in two literature databases, i.e. ‘Web of Science’ and the online database maintained by the German Federal Institute of Hydrology (BfG). ‘Tyre’ and ‘tire’ as well as ‘tyre wear’ and ‘tire wear’ were used as search terms in combination with other key words (AND operations): microplastic*, particle, environmental, degradation, biodegradation, effects, ecotoxicological, ecotoxicity, toxicity, health, biota, surface water*, runoff, river*, ocean, sea, WTTP, wastewater, soil*, field study and monitoring. In addition, the term ‘non-exhaust emissions’ AND ‘tyre/tire wear’ OR microplastic* was applied. Studies were taken into account as far as they refer to the topics mentioned above. For instance, studies on tyres were excluded if they focused on their functional properties or on options to dispose or recycle used tyres. Besides peer-reviewed publications we reviewed other types of scientific literature like monographies and reports edited by institutions such as the European Commission and national competent authorities. Doctoral theses were taken into account as far they provided relevant information which was otherwise not available. Except monitoring studies, we did not set a limit regarding the year of publication. As to monitoring studies we focused on literature published since 2000. Literature published in languages other than English or German was excluded.

3. Generation and properties of tyre and road wear particles

3.1. Ingredients of tyre tread materials

Sommer et al. (2018) summarize the most common components of tyre tread as well as other compounds which determine lifespan and driving characteristics:

- Basic material (40–50 mass %): natural rubber (NR) (polysoprene \(\text{C}_x\text{H}_{y}\)) and synthetic rubber, e.g. styrene butadiene rubber (SBR) or butadiene rubber (BR) (Eisentraut et al., 2018).
- Filler (30–35 mass %): typically soot/carbon black (C), silica (SiO₂), and chalk (CaCO₃).
- Softener (15 mass %): consists of oil and resin,
- Vulcanization agents (2–5 mass %): sulfur (S) and zinc oxide (ZnO),
- Additives (5 to 10 mass %): preservatives (halogenated cyanohaloalkanes), anti-oxidants (amines, phenols), desiccants (calcium oxides), plasticizers (aromatic and aliphatic esters), processing aids (mineral oils, peptizers) (Wagner et al., 2018).

Eisentraut et al. (2018) estimate from literature data the average content of SBR in tyre tread material to be roughly 11% whereas heavy duty vehicle tyres usually contain little to no SBR. Concerning the MP discussion, it should be kept in mind that tyre tread possesses a polymer content of about 30% and that wear particles found on roads are conglomerates of TRWP with varying composition (see Section 3.2).

3.2. Formation and properties

TRWP are generated by contact between tyre and road surface during driving, acceleration and braking. Their properties and accordingly their behaviour in the environment depend not only on the components of the tyre tread but also on the generation process. The formation of TRWP is determined by numerous factors (see Section 5.1.2), however, their influence on the amount and properties of the particles is not yet quantified in detail. Shear and friction forces, which impact on tyre tread through contact with the road surface during driving processes, causes abrasion of predominantly coarser particles (Kreider et al., 2010; Pant and Harrison, 2013; Sommer et al., 2018). Heat and friction as a result of interaction between tyres and road surface alter the chemical composition of the generated particles compared to the original tyre tread (Kreider et al., 2010; Sommer et al., 2018). Moreover, material from the road surface is incorporated into tyre wear particles (Kreider et al., 2010; Panko et al., 2013a) resulting in TRWP. Real-world TRWP collected at roads consist of an inseparable heterogeneous composition of rubber, mineral particles and fine dust from other traffic-related wear particles such as brake wear and resuspension of wear particles (Adachi and Tainosho, 2004; Kreider et al., 2010; Sommer et al., 2018). Some authors mention that pure tyre wear particles exist in very low quantities in the environment while most tyre wear particles are associated with road material (Dall’Osto et al., 2014; Eisentraut et al., 2018; Grigoratos and Martini, 2014).

Some inconsistency exists in literature referring to the terms used for the different types of wear particles described above. Kreider et al. (2010) differentiate between tyre tread particles (TP), tyre wear particles (TWPs), and roadway particles (RP). TP are particles shaved from unused tyre tread and crushed while TWPs are generated on simulated laboratory driving courses. RP are collected at test vehicles during outdoor driving. Panko et al. (2013a) and Unice et al. (2013) use the term ‘tyre and road wear’ (TRWP) for particles present in the environment whereas they assume a contribution of 50% each for wear of tyre tread and of road surface. Sommer et al. (2018) define original emissions of pure tyre tread as ‘tyre-core particles’ while they describe ‘tyre-wear particles’ or ‘tyre-abrasion particles’ as contaminated and encrusted
tyre-core particles. This definition may include the incorporation of particles deposited on the road from other sources besides tyres and road surface. In the present article, we use the term TRWP for heterogeneous particles consisting of abraded tyre and road material according to Panko et al. (2013a) and Unice et al. (2013). TRWP containing contributions of tyre and road wear as well as road dust including contributions from other sources are denoted as real-world TRWP in this paper. For material abraded through contact between tyres and road excluding road wear contribution we chose the term ‘tyre wear’.

TRWP possess elongated, cylindrical forms (Kreider et al., 2010; Sommer et al., 2018), which are similar to the morphology of wear particles collected at simulated laboratory driving courses while ground wear particles from unused tyre tread are more jagged in form (Kreider et al., 2010). For wear particles collected at a test vehicle during outdoor driving, Kreider et al. (2010) found a size distribution of 4 to 265 μm with a maximum at about 25 μm referring to particle number.

Sommer et al. (2018) report that TRWP attract fine road dust once they are deposited on the road surface. Due to their rounded cross section they roll over the road surface collecting these other road-dust particles like a rolling snow ball. This leads to a partial or complete encrustment of the TRWP with mostly inorganic smaller particles. Sommer et al. (2018) analysed 171 of these encrusted ‘super-cosmic’ airborne TRWP (10–80 μm). The results display a range of 10–50% volume content of the coating meaning a 50–90% volume content of rubber. The non-rubber components forming the coating appear to be a mixture of minerals from the road surface and metals. Kreider et al. (2010) analysed the chemical composition of different wear particles. Real-world TRWP were found to consist of 53% minerals and 23% polymers, other compounds are plasticizers and oils (13%) and carbon black (11%). This composition differs from that of wear particles generated at a road simulator and that of particles produced from original tyre tread. Polymer contents of these particles are 16% and 46%, respectively, the mineral contents amount to 61% and 16%, respectively.

The abrasion processes also result in a significant increase in density from 1.13 to 1.16 g/cm3 for tyre tread material to 1.5–2.2 g/cm3 for TRWP (Kayhanian et al., 2012; Rhodes et al., 2012). Based on current experimental data, Sommer et al. (2018) calculated a density of 1.26 g/cm3 for some real-world TRWP. Especially density, but also the change in chemical composition exerts serious influence on the environmental behaviour of TRWP. Except alkyd resin, polyester and polytetrafluoroethylene, most plastic materials possess a density of ≤1.6 g/cm3 (Duis and Coors, 2016). Thus, TRWP will probably tend to be deposited in sediments more than other MP types which will hamper further transport. Changes in the chemical composition can be expected to impact degradation and (eco)toxicity compared to the original tyre tread material.

Laboratory studies indicate that tyre wear may also contribute to nanoparticle emissions (Kumar et al., 2013). For instance, Dahl et al. (2006) detected particles with mean diameters between 15 and 50 nm referring to particle number in a road simulator study. Under harsh driving conditions (tyre slip), Park et al. (2018) observed the generation of a large amount of ultrafine particles (<100 nm). The authors suggest that evaporation and condensation of volatile compounds from the tyre material may be a plausible explanation for the formation of these small particles.

4. Behaviour and impact of TRWP in the environment

4.1. Ecotoxicological effects and effects on human health

Presence of synthetic particles as MP in the gastrointestinal tract of organisms possesses the potential of physical and chemical effects. Due to the volume which the particles occupy, a reduction of food intake and thus a loss of energy and vitality can be caused. Furthermore, polymer additives can be leached out in the digestive tract leading to toxic impacts (see Baensch-Baltruschat et al. (2017)). The question is how far this also holds true for TRWP. An overview on effect studies which have been published since 2000 was compiled by Wagner et al. (2018). In most studies, the effects of tyre wear leachates on various aquatic species such as daphnids, insects, fish, algae and bacteria were investigated in vivo. Toxic effects of leachates were connected with Zinc (Zn), other metals and organic compounds, e.g. benzo(b)fluoranthene, phthalates and resin acids (Wagner and Lambert, 2018; Wik and Dave, 2009). Tyre wear artificially produced by road simulators or with raps, ground tyre material and worn tyres, whose composition differs from real-world TRWP generated on roads (see Kreider et al. (2010)), were used as test materials. In earlier studies mostly leachates of whole tyres were used (Wik et al., 2009) which is even less representative since tyre tread can have a different chemical composition than other parts of the tyre.

In some studies, TRWP which were generated by road simulators in laboratory and possess more morphological and chemical similarity to real-world TRWP than other wear particles (see Kreider et al. (2010)) were used as test materials. In two studies the toxicity of TRWP in sediments were investigated by Marwood et al. (2011) and Panko et al. (2013b) in which TRWP were generated at a road simulator and particles ≤150 μm were removed. Marwood et al. (2011) investigated the acute toxicity of elutriates of a sediment-water-TRWP mixture with TRWP concentrations ranging from 10 to 10,000 mg/L. No significant acute toxic effects were observed on algae (Pseudokirchnerella subcapitata), fish (Pimephales promelas) and Daphnia magna when the elutriates were prepared at standard temperature (22 °C). Panko et al. (2013b) examined the chronic toxicity of TRWP spiked sediments and elutriates (mixture ratio 1:4) in four test species at TRWP concentrations similar to those realistic for surface water sediments (10 g TRWP/kg) (see Table 3). The test results showed no chronic toxicity for Hyalella azteca and Ceriodaphnia dubia and slight effects for Chironomus dilutus (growth inhibition) and Pimephales promelas (survival in larvae). Wik et al. (2008) used real-world samples from sites where relatively high TRWP concentrations are expected. The authors tested acute and chronic toxicity of sediment and water samples from 18 different road runoff detention systems in different nonvertebrates. For instance, the observed effect rates referring to 72 h-mobility range from 20 to 100% for Hyalella azteca. However, the impacts from other traffic-related sources cannot be excluded in this study. In other studies, test materials were produced by grinding tyre material or shaving particles from the tyre or the tyre tread whereas scrap and new tyres were used (see Wagner et al. (2018)). Wagner et al. (2018) report that acute toxic effect concentrations range from 25 to 100,000 mg TWP/L in studies in which crushed tyres are used to prepare test leachates. The highest sensitivity was observed by Turner and Rice (2010) who tested the toxicity of tyre particle leachate to a marine macroalga (Ulva lactuca). The highest effect concentration was found by Gualtieri et al. (2005a) in an amphibian species (Xenopus laevis). Wik and Dave (2009) assume that probable reasons for the wide range of toxicity data are differences in rubber recipes for tyres, in test designs and in species sensitivity. For comparison: Acute toxic effect concentrations of Cd and Cu in aquatic organisms range from 0.06 to 5 mg Cd/L and from 0.1 to 0.8 mg Cu/L according to Merian et al. (2004), EPA (2016) reports 0.005 to 50 mg Cd/L for a thoroughly tested study set of a wide range of organisms. An average Cd content in tyre tread is 1.66 mg Cd/kg (Rocher et al., 2010b). According to Wik and Dave (2009) leachates of tyre particles also show sublethal effects such as teratogenicity, mutagenicity and estrogenicity.

Wik et al. (2009) tested the chronic toxicity of leachates prepared with worn tyres in green algae, two crustacean species and fish eggs. The highest sensitivity was observed in Ceriodaphnia dubia, with chronic toxicity values of 50–6000 mg/L for survival and 10–1800 mg/L for reproduction. In a recent study, Redondo-Hasselerharm et al. (2018) investigated the ingestion and egestion of tread particles from second hand tyres by the amphipod Gammarus pulex as well as chronic toxicity effects on four freshwater benthic macroinvertebrates in spiked
sedi ments at tyre tread particle concentrations ≤10 mass % referring to dry weight. Although particles were retained by the assessed organisms no toxic effects were observed in the four investigated species Gammarus pulex, Asellus aquaticus, Tubifex spp. and Lumbriculus variegatus. The authors assume that the reason for these results is the low bioavailability of Zn observed in sediments.

Experimental design in the cited studies does mostly not reflect environmental conditions. First, test materials generated by crushing tyres differ from real-world TRWP in morphology and chemical composition and therefore very likely also in toxicity (see Kreider et al., 2010). To confirm the previous results application of TRWP collected in the environment would be useful to prepare leachate and spike sediments. Another point is that concentrations of tyre particles in leachates or sediments were mostly much higher than TRWP concentrations found in surface waters (Wagner et al., 2018). Wik and Dave (2009) estimated average concentrations in river water between 0.5 and 3.6 mg TWP/L based on data given by Reddy and Quinn (1997), Kumata et al. (2000), and Ni et al. (2008). For river sediments concentrations up to about 12 mg/g dw were reported in literature (see Table 3). Little is known on the terrestrial toxicity of TRWP. Only few studies are available in which the use of ground tyre rubber in potting soil showed growth reduction for different species (Wik and Dave, 2009).

Risk for human health is primarily related to the inhalation of TRWP emitted to the atmosphere. In principle, adverse effects can occur due to the physical characteristics of the particles depending, e.g. on size and shape, to surface chemistry and to toxic components such as heavy metals and organic compounds (Amato et al., 2014a; Kole et al., 2017). The toxicity of tyre wear was investigated in various in vitro studies. Karlsson et al. (2008) exposed cultures of human lung cells and human macrophages to tyre wear particles. The authors observed DNA damage and inflammatory effects. Gualtieri et al. (2005b) exposed human alveolar lung cells to organic extracts of tyre wear whereas at higher concentrations of the extracts (60 μg/mL for 72 h) a significant increase in cell mortality and DNA damage occurred. The effects of particles generated by the contact of studded tyres and pavement on different human cell cultures including macrophages, nasal epithelial and bronchial epithelial cells were examined by Lindbom et al. (2006) and Karlsson et al. (2011). Inflammatory effects were observed for macrophages and bronchial epithelial cells. TRWP containing granite pavement had a significantly higher capacity to induce the release of cytokines than particles generated at the surface of quartzite pavement (Lindbom et al., 2006). The same difference between these pavement types were also observed when TRWP were exposed to murine macrophage cells (Lindbom et al., 2007).

Some authors published in vivo studies in which effects on mice or rats were investigated. Gottipolu et al. (2008) applied particles of ground recycled SBR and scrap tyres to rats through intratracheal instillation. Acute pulmonary toxicity of these particles was observed, but no effects on cardiac enzymes were noticed. Denier van der Gon et al. (2013) suggest that heavy metals contained in tyre material such as Zn may be relevant regarding the toxicity of tyre wear. From their in vivo tests with Zn and Cu solutions, Gottipolu et al. (2008) assume that these metals may induce cardiac oxidative stress at higher concentrations.

Mantecca et al. (2009) applied a suspension of ≤2.5 μm and ≤10 μm cryo-fractured tyre scrap to mice through intratracheal instillation. The effects were studied 24 h after application. The ≤0.5 μm fraction evoked an inflammatory response while strong cytotoxic effects were observed when the test organisms had been exposed to particles ≥2.5 μm. In a following study, Mantecca et al. (2010) investigated the effects occurring 3 h after intratracheal instillation of both size fractions applying the same test material as in the previous study. The results showed that the ≤2.5 μm fraction reached the alveolar spaces and is able to deeply penetrate and rapidly induce inflammatory events in the parenchyma whereas particles ≥10 μm are mainly distributed in the bronchial district. Kreider et al. (2012) exposed rats to TRWP generated at a road simulator by nose-inhalation 6 h per day during a 28-day test. In this study, no TRWP-related effects were observed on survival, clinical observations, body or organ weights, gross pathology, food consumption, immune system endpoints, serum chemistry, or biochemical markers of inflammation or cytotoxicity. Only weak, non-significant subacute inflammatory effects were found in five organisms of which four were exposed to the highest concentration (100 μg/m3) and one to the mid concentration (40 μg/m3). From these results, Kreider et al. (2012) derived a No-Observable-Adverse-Effect-Concentration (NOAEC) of 112 μg/m3 for respirable TRWP.

A comprehensive assessment of the cited studies which concern the risk of human health via inhalation of RTWP/tyre wear particles would surpass the scope of this paper. In general, results of studies in which TRWP generated at a road simulator are applied as test material, are more reliable than data from effect tests using materials such as particles shaved from tyres or fractured scrap tyres (see Kreider et al. (2010)) as applied by Gualtieri et al. (2005b), Gottipolu et al. (2008), and (Mantecca et al., 2010; 2009). A comprehensive and quantitative risk assessment was elaborated by Kreider et al. (2019) referring to human health concerning cardiopulmonary effects associated with the inhalation of airborne TRWP. Literature on effects of airborne TRWP in mammalian and on concentrations of TRWP in ambient air was evaluated by the authors. Kreider et al. (2012) was identified as the most appropriate study concerning human health risk. Based on the results of Kreider et al. (2012), Kreider et al. (2019) determined an human equivalent NOAEC (NOAECHEC) of 55 μg/m3 referring to cardiopulmonary effects. The authors elaborated a risk characterization for TRWP considering NOAECHEC and exposure concentrations in air whereas indoor, transit and outdoor exposure of different age groups were taken into account. For exposure, the study on TRWP in air by Panko et al. (2013a) was assessed as the most appropriate one (see Section 7.2). Kreider et al. (2019) conclude that the ratio of NOAECHEC and exposure indicate a low risk potential for TRWP. The authors remark that their exposure assessment may not be representative for all regions of the world, especially not for developing countries, and that particularly sensitive or highly exposed population groups were not considered. Since the study focuses on cardiopulmonary effects, other effects such as cancerogenicity and reproductive toxicity cannot definitively be excluded according to the authors. In addition, it has to be noted that risk assessment is based on only one effect study and one monitoring project as a source of exposure data. Further monitoring studies including other countries and additional reliable effect data are necessary to confirm the conclusion of a low risk for human health.

Currently, there is no information available on human health risk related to the potential uptake of TRWP via the food chain while the possible threat posed by MP particles in human food is widely discussed. MP was detected in various fish and seafood species (Lusher et al., 2017; Wright and Kelly, 2017). Especially consumption of seafood such as bivalves can lead to a significant uptake of MP with food (Wright and Kelly, 2017). Eating of non-gutted fish may also be a source of MP intake (Lusher et al., 2017; Wright and Kelly, 2017). Wright and Kelly (2017) report that outcomes from studies in model mammalian systems indicate that MP particles can pass the intestinal wall. Eldridge et al. (1989) investigated the dissemination of biocompatible microspheres (≤10 μm) which were composed of the biodegradable polymer poly(DL-lactide-co-glycolide) after oral application to mice. The authors found microspheres ≤5 μm in mesenteric lymph nodes, blood circulation and spleen. Jani et al. (1992) who fed rats with polystyrene latex particles of different size (≤1 μm), observed the occurrence of these particles in lymph nodes and liver. Volkheimer (1975) applied larger PVC particles (5–110 μm) into the digestive tracts of dogs and other mammals and found particles in blood and other body fluids, but does not report a size threshold for the mobility through membranes. Except this early study, no data on the translocation of coarser MP particles or TRWP are currently available.
4.2. Degradation of tyre wear in the environment

With regard to environmental behaviour a central question is how fast TRWP are degraded in environmental compartments. Two degradation paths seem to be relevant, photo- and biodegradation. The following section gives an overview on the information currently available. The susceptibility of organic compounds to photochemical reactions is well known, but concerning rubber degradation, no references giving quantitative information are available. For instance, Wypych (2015) reported that rubbers such as polybutadiene, poly-styrene-butadiene and poly-isoprene rubber undergo degradation by photo-oxidation without providing quantitative data on the photodegradation processes.

Linos and Steinbüchel (2001) report that only cis-1,4-isoprene containing rubbers, i.e. natural rubbers and synthetic isoprene rubbers, have reliably been found to be biodegradable. In addition, cis-1,4-isoprene is sensitive to abiotic oxidative aging. The authors assume that the degradation of the polymer occurs most probably also in the absence of molecular oxygen since no evidence for the accumulation of cis-1,4-isoprene in anaerobic environments is reported in literature. cis-1,4-polybutadiene, the polymer component of butadiene rubber, is resistant towards biotic and abiotic oxidation processes (Linos and Steinbüchel, 2001).

Stevenson et al. (2008) report that biodegradation of tyre wear is hampered by toxic additives and stabilisers contained in the tyre material. Tsuchii and Tokiwa (2001) examined the microbial degradation of tyre rubber particles under laboratory conditions (stirred test solution) to find a feasible option for the treatment of ground tyres. Under optimum conditions (stirring rate: 40 rpm at 30 °C), the authors observed weight losses of 57, 50 and 36%, respectively, of particles cut from truck tyre tread with smooth surfaces and mean diameters of about 0.8, 1.1 and 2.3 mm after 8 weeks whereas the mineralization rate was estimated to be 40–50%. Berekaa (2006) investigated the microbial degradation of natural and different synthetic rubbers using a bacterium which had been isolated from old tyre rubber material. After 3 weeks of incubation, dense microbial masses had developed on natural rubber granulates. As to synthetic rubbers, cis-1,4-polyisoprene rubber proved to be most susceptible to degradation (weight loss after 6 weeks: 11%) while other synthetic rubber types were more resistant (polybutadiene rubber: 2% weight loss, styrene rubber: 6% weight loss).

Data on degradation of tyre wear from field studies has hardly been published. Cadle and Williams (1978) analysed samples from a highway before and after opening for traffic. The total removal rate of rubber was 0.67% per day whereas the rate of environmental degradation was found to be 0.15% per day. The authors assume that wind erosion and water runoff also contribute to the total removal rate. Cadle and Williams (1980) also investigated the degradation of tyre tread wear generated by a tyre emission test facility in a long-term test under environmental conditions. When applying extraction pyrolysis GC they found that the average degradation rate of the polymer in the tread wear exposed in soil was 52% after 16 months. Atmospheric oxidation appeared to be more important than biodegradation. Currently, no studies are available on the degradation of TRWP in the aquatic environment. However, scrap tyres are used to build up breakwaters and artificial reefs at coastlines which requires high persistence of the solid material in the marine environment (Collins et al., 2002). The authors assume that photo-degradation of the tyre material is hampered by light absorption in the water layer.

4.3. Summary

Effects on various aquatic species were investigated especially in leachates of tyre wear or TRWP, in some studies ecotoxicity of spiked sediments was examined. For fractured tyre material, acute and chronic aquatic toxicity values were found ranging from 25 to 100,000 mg/L and 10–6000 mg/L, respectively. Effect studies under environmental conditions are lacking, regarding TRWP concentrations and real-world TRWP as test materials. As to human health risk via inhalation, adverse effects such as DNA damage and inflammatory effects were observed. In a comprehensive risk assessment, which is based on two studies identified as most appropriate, the risk of TRWP to human health was evaluated as low. Gaps of knowledge exist referring to human health risk via the food chain and degradation of TRWP in the environment.

5. Emissions of tyre and road wear

5.1. Approaches in literature

5.1.1. General

Two approaches are applied to estimate annual tyre (and road) wear emissions on a regional or national scale.

1. The estimation is based on emission factors (EF), i.e. mass of generated tyre wear or TRWP per vehicle km, and total annual mileage.
2. Emissions are estimated by taking into account total consumption of tyres and their weight loss due to abrasion per year.

Some authors used both approaches for their calculation. It has to be noted that the second approach does not take into account the contribution of road wear. But this is also the case for the derivation of most EF.

5.1.2. Emission factors

Emission factors, which should reflect the generation of tyre (and road) wear as realistic as possible, depend on a variety of features: tyre characteristics (e.g., type, age, size and chemical composition), properties of road surfaces (e.g., material, porosity, surface dressing, wetness and temperature), characteristics of vehicles (e.g., weight, braking system) and vehicle operations (speed, acceleration, extent and frequency of braking, cornering) (Denier van der Gon et al., 2013; Luhana et al., 2004; Wagner et al., 2018). EF reported in literature were derived from studies at test benches, with test vehicles, and in road tunnels, or were calculated based on weight loss of tyres or annual tyre replacement, respectively, and mileage (Rauterberg-Wulf, 1998). EF were compiled by various authors (see Table 1). Partly, the original source is not given or is not publicly available. Therefore, the approach or test procedure applied to determine the EF is not always comprehensible. A number of authors give EF exclusively for the airborne fraction of tyre wear which represents only a small contribution to the entire emissions.

Ten Broeke et al. (2008) present EF for coarse and fine particulates (the latter referring to the PM10 and PM 2.5 fraction, respectively) which were lastly amended in 2016 (DELTARES and TNO, 2016). They differentiate extensively between different vehicle and road types. Since the original data sources are not given the estimation procedure cannot be retraced. Furthermore, it is not clear if the given EF relate especially to the conditions in the Netherlands (e.g., speed limit and porous road surface on highways) or whether they are more generally applicable. Luhana et al. (2004) estimated an average EF for passenger cars based on tyre consumption and weight loss (data source: tyre usage statistics in the UK, report year, 1999). Furthermore, the authors determined EF for passenger cars by gravimetric measurement for 5 in-service vehicles. Both studies include no differentiation of road types.

Hillenbrand et al. (2005) compiled EF from various studies which were published from 1992 to 2002 comprising data from different countries (Germany, Switzerland and California, USA) (Baumann and Ismeier, 1997; Buwal, 2000; Buwal, 1992; CARB, 1993; Gebbe and Hartung, 1997). In BUWAL (1992), EF for passenger cars and lorries were estimated regarding the weight loss of tyres at average mileage. Since the report of the Californian Environmental Agency (CARB, 1993) is not available it cannot be clarified how the EF presented therein were determined. Gebbe and Hartung (1997) determined the wear rate of tyres by manual measurement of tread depth whereby they investigated several hundred vehicles registered in the City of Berlin.
According to Grigoratos and Martini (2014), a fraction of the coarse fraction takes into account this fact. However, the atmospheric emissions which is emitted to the atmosphere. Not every study in which EF are applied takes into account that a fraction of these data sets for tyre wear rates were applied. In most studies a difference was made between vehicle types. Most used data on EF were those given by Hillenbrand et al. (2005), Luhana et al. (2004) and Anonymous (2012) and the different versions elaborated by DELTARES and TNO (original work: Ten Broeke et al. 2008, last updated version available: DELTARES and TNO (2016)). In some cases, different data sets were combined. EF provided by TNO and DELTARES allow a differentiation between tyre wear emissions generated on urban and rural roads and on highways provided that mileage data for different vehicle and road types is available. Verschoor et al. (2016) elaborated such an estimation of the annual tyre wear emissions in the Netherlands. Kole et al. (2017) also estimated tyre wear emissions based on these data sets for the Netherlands. Additionally, the authors recalculated the emissions estimated by Lassen et al. (2015) (Denmark) and Yamashita and Yamanaka (2013) (Japan) and updated the results of EA UK (1999) (United Kingdom) and Councell et al. (2004) (USA). For the updates they took into account the population growth since the report year of the original estimations. Based on the emission data for seven European countries and six countries outside Europe, in which 57% of the world’s vehicles are registered, they extrapolated the world’s tyre wear emissions amounting to 3,369,698 t/a.

To enable the comparison between different countries, Table 2 also displays the annual emissions per capita. The data comprises less than two orders of magnitude ranging from 0.2 to 5.5 kg/(cap*a) with an average global value of 0.8 kg/(cap*a). For some countries different results have been published (e.g., for Germany or the Netherlands) which show that the estimated emissions depend on diverse input data. An own estimation of the annual total tyre wear emissions for Germany was performed based on EF and total vehicle km (see Baensch-Baltruschat et al., 2020). Total annual emissions of coarse particles were estimated to be 0.9 kg/(cap*a) (EF derived by Gebbe and Hartung (1997)) and 1.2 kg/(cap*a) (EF compiled by DELTARES and TNO (2016)), respectively. These figures are consistent with the results of Hillenbrand et al. (2005) and Essel et al. (2014) and somewhat lower than the emissions estimated by Wagner et al. (2018).

Taking into account the variation of input data some statements can be made. The highest emissions per capita occur in the USA where transport is mainly focused on roads. The lowest value has been estimated for India where many inhabitants’ economic status is relatively low compared to the standard of living in industrial countries. Kole et al. (2017) estimated relatively low emissions for the Netherlands taking into account that 95% of the Dutch highways are paved with open asphalt concrete. It was assumed that 95% of the tyre wear emissions are captured in the pores of the road surface and that the highways are regularly cleaned. For the Seine river basin a higher value was calculated than for entire France based on the higher density of population and road network.

### 5.3. Summary

Emissions of tyre wear can be estimated either by emissions factors (EF) and mileage or by consumption and average weight loss of tyres. Only a small fraction of tyre wear is airborne (0.1–10 mass %).

### 5.2. Estimations of tyre and road wear emissions for different countries

Annual emissions of tyre (and road) wear have been estimated for numerous European and non-European countries (see Table 2). Most authors calculated the emissions based on EF and total annual vehicle km at national or regional scale whereas different data sets for tyre wear rates were applied. In most studies a difference was made between vehicle types. Most used data on EF were those given by Hillenbrand et al. (2005), Luhana et al. (2004) and Anonymous (2012) and the different versions elaborated by DELTARES and TNO (original work: Ten Broeke et al. 2008, last updated version available: DELTARES and TNO (2016)). In some cases, different data sets were combined. EF provided by TNO and DELTARES allow a differentiation between tyre wear emissions generated on urban and rural roads and on highways provided that mileage data for different vehicle and road types is available. Verschoor et al. (2016) elaborated such an estimation of the annual tyre wear emissions in the Netherlands. Kole et al. (2017) also estimated tyre wear emissions based on these data sets for the Netherlands. Additionally, the authors recalculated the emissions estimated by Lassen et al. (2015) (Denmark) and Yamashita and Yamanaka (2013) (Japan) and updated the results of EA UK (1999) (United Kingdom) and Councell et al. (2004) (USA). For the updates they took into account the population growth since the report year of the original estimations. Based on the emission data for seven European countries and six countries outside Europe, in which 57% of the world’s vehicles are registered, they extrapolated the world’s tyre wear emissions amounting to 3,369,698 t/a.

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### 5.3. Summary

Emissions of tyre wear can be estimated either by emissions factors (EF) and mileage or by consumption and average weight loss of tyres. Only a small fraction of tyre wear is airborne (0.1–10 mass %).
The major fraction is emitted to water and soils. Annual emissions were calculated for numerous countries. The annual tyre wear emissions are estimated to be 1.3 million t/a in Europe and 1.1–1.8 million t/a in the USA while world’s annual emissions are approx. 3.4 million t/a. The highest emissions per capita is reported for the USA (>5.5 kg/cap*a), the lowest for India (0.25 kg/cap*a).

### Table 2

Annual emissions of tyre wear for different countries and regions.

| Country/Region | Tyre wear emissions in total (referring to tyre tread) [t/a] | Tyre wear emissions per capita [kg/(cap*a)]¹ | Calculation method | EF applied | Remarks | Reference |
|---------------|-----------------------------------------------------------|---------------------------------------------|-------------------|-----------|---------|-----------|
| Europe        |                                                           |                                             |                   |           |         |           |
| EU            | 1,127,000                                                 | 2.6                                         | b                 |           |         | Wagner et al. (2018) |
| DEU           | 61,000                                                   | 0.8                                         | b                 | Own data  |         | Baumann and Ismeier (1997) |
| DEU           | 111,420                                                  | 1.4                                         | b                 |           |         | Hillenbrand et al. (2005) |
| DEU           | 60,000–111,000                                           | 0.7–1.4                                     | c                 |           |         | Essel et al. (2014) |
| DEU           | 133,000                                                  | 1.7                                         | b                 |           |         | Wagner et al. (2018) |
| DEU           | 75,200                                                   | 0.9                                         | b                 |           |         | Own calculation (Baensch-Baltruschat et al., 2020) |
| DEU           | 98,400                                                   | 1.2                                         | k                 |           |         |             |
| DNK           | 6514–7660                                                | 1.1–1.3                                     | b                 |           |         | Recalculation of data estimated by Lassen et al. (2015) |
| DNK           | 4200–6600                                                | 0.7–1.1                                     | d                 |           |         | Lassen et al. (2015) |
| DNK           | 7310                                                     | 1.3                                         | d                 |           |         | Unice et al. (2019) |
| FRA (entire)  | 37,646                                                   | 0.6                                         | b                 |           |         | Unice et al. (2019) |
| Seine river basin | 13,804                                                  | 0.9                                         |                 |           |         |             |
| ITA           | 50,000                                                   | 0.8                                         | n/a              |           |         | Milani et al. (2004) |
| NLD           | 15,452 (total)                                           | 0.9                                         | b                 |           |         | Sherrington et al. (2016) |
| NLD           | 7726 (polymer)                                           | 0.5                                         | b                 |           |         |             |
| NLD           | 17,300 (only tyre wear)                                  | 1.0                                         | b                 |           |         | Verschoor et al. (2016) |
| NLD           | 15,030                                                   | 0.9                                         | b                 |           |         | Köle et al. (2017) |
| NLD           | 8768 (corrected for amounts trapped in open pore road surface) | 0.5                                         |                 |           |         |             |
| NOR           | 7500 (total)                                             | 1.4                                         | b                 |           |         | Sundt et al. (2014) |
| NOR           | 4500 (polymer)                                           | 0.8                                         |                   |           |         |             |
| NOR           | 9600 (total)                                             | 1.8                                         | f                 |           |         | Sundt et al. (2014) |
| NOR           | 5700 (polymer)                                           | 1.1                                         |                   |           |         |             |
| SWE           | 13,000                                                   | 1.3                                         | b                 |           |         | Magnusson et al. (2016) |
| GBR           | 38,000–76,000                                            | 0.6–1.2                                     | d                 |           |         | EA UK (1999) |
| GBR           | 42,000–84,000                                            | 0.6–1.3                                     | d                 |           |         | Köle et al. (2017) |
| Countries outside Europe |                                                             |                                             |                   |           |         |           |
| AUS           | 20,000                                                   | 0.9                                         | n/a              |           |         | Milani et al. (2004) |
| BRA           | 254,011                                                  | 1.4                                         | b                 | m         |         | Köle et al. (2017) |
| CHN           | 756,240                                                  | 0.5                                         | b                 | m         |         | Köle et al. (2017) |
| IND           | 292,674                                                  | 0.2                                         | b                 | m         |         | Köle et al. (2017) |
| JPN           | 239,762                                                  | 1.9                                         | x                 |           |         | Köle et al. (2017) |
| USA           | 1,000,000                                                | 3.0                                         | b                 | s         |         | Counsell et al. (2004) |
| USA           | 1,110,000                                                | 3.4                                         | d                 |           |         |             |
| USA           | 1,524,740                                                | 4.6                                         |                   |           |         | Update of data given by Counsell et al. (2004) |
| USA           | 1,797,480                                                | 5.5                                         | b                 | m         |         | Köle et al. (2017) |
| USA           | 1,120,000                                                | 3.4                                         | b                 | i         |         | Wagner et al. (2018) |
| World total   | 5,917,518                                                | 0.8                                         | b                 |           |         | Köle et al. (2017) |

¹ Own calculation, data source for number of inhabitants: total Europe (EUROSTAT, 2018), others (CIA, 2018).

b Estimation based on emission factors and total vehicle km.

c Data based on Gebbe et al. (1997) and WDK (Association of German Rubber Manufacturing Industry, reporting year 2005).

d Estimation based on consumption of tyres and abrasive loss (weight loss during use).

e Derived from data for entire France based on population density and lengths of urban and rural roads in the Seine river basin.

f Estimation based on the number of tyres collected for retreading and weight loss during use.

x Estimation based on the number of registered vehicles, life expectancy of tyres and loss during life time.

h Extrapolated from the emission data for DEU, DNK, GBR, ITA, NLD, NOR, SWE, AUS, BRA, CHN, IND, JPN, and USA, and the world’s number of vehicles.

i Emissions factors compiled by Hillenbrand et al. (2005).

j Emissions factors compiled by Gebbe and Hartung (1997).

k Emission factors by DELTARES and TNO (2016).

l Luhana et al. (2004).

m Anonymous (2012).

n DELTARES and TNO (2014).

o Klein et al. (2015) (Dutch Pollutant Release and Transfer Register).

p Van Duijnhove et al. (2014).

q Gustafsson et al. (2008).

r Aatmeeyata et al. (2009).

s Unified EF: 50 mg/km.
6. Paths of tyre and road wear into environmental compartments

6.1. Particles leaving the road system

The coarse fraction of TRWP generated by vehicle motions is first deposited on the road surface. During rainfall events TRWP will be washed off to the roadside. According to Unice et al. (2019) a precipitation rate of at least 2 mm/d is necessary to mobilize the particles. Even high rain and runoff intensities do not lead to complete washoff, however, at 5 mm/d the main share of the particles will move from the road surface. Thus, runoff from roads will come in pulses and will strongly depend on drought and rain periods. At roads without artificial drainage, road runoff will infiltrate in a small zone directly adjacent to the paved road surface with a width of 0.75–1.5 m according to Blok (2005) and Kocher et al. (2010c). Due to air turbulence caused by vehicles at high speed, wind and spray water (aerial dispersion, see POLMIT (2002)), particles can also be transported to the verge besides the road (Kaufmann et al., 2007). The tyre and road wear material accumulated in soil near road does not contribute to the particle transport with rainwater runoff to the aquatic environment. Instead, these particles are integrated into soil as the contaminants they contain. Many data proves that a corridor of some meters close to a road is polluted by undissolved substances and heavy metals (e.g., Werkenthin et al., 2016). This probably holds true for TRWP. The width of the affected verge will depend on the traffic density and, using zinc as a marker, will range from about 6 m at highways to 1–3 m at other roads (e.g., Kocher et al. 2010a, c).

From the results of eight field studies, Blok (2005) concluded that on average one third of solids emitted on roads are removed from the road system by drift while the remaining two third are transported by runoff. Most authors differentiate between runoff in urban areas, on rural roads and on highways. Kaufmann et al. (2007) assume a runoff rate of 50–60% at urban roads, 20–30% at rural roads and 10% at highways. Lassen et al. (2015) applied a runoff rate of 40% for areas with sewage systems to estimate the paths of the annual tyre wear emissions into the aquatic environment for Denmark. Ten Broeke et al. (2008) recommend to consider that 60% of TRWP are released from urban roads into sewers while outside urban areas 10% are transported into surface waters and 90% remain in soil (see also application by Verschoor et al. (2016) to model the paths of TRWP in the Netherlands). Unice et al. (2019) assumed a proportion of 49% for runoff, 49% for soil and 2% for aerial transport as parameters to model the paths of TRWP in the Seine river basin (FRA).

6.2. Transport and treatment

6.2.1. Transport and treatment at rural roads and highways

Rural roads and highways are in general not connected to sewage systems. Thus, the emitted TRWP are mainly deposited in soil close to the roads where infiltration and retention of particles and contaminants occur (e.g., Baensch-Baltruschat et al., 2020; Werkenthin et al., 2016). For constructional reasons, drainage systems are mainly installed at road sections leading over bridges and viaducts and through tunnels (Blok, 2005). At highways, sections in curves can also be equipped with drainage systems in the median strip (BASt, 2019). Roads which run through flat terrain without curves do mostly not have a drainage system. At sections with an artificial drainage system, treatment facilities can be installed including sedimentation basins, skimmers, retention soil filters and grass swales. In other cases, untreated runoff water is released to surface waters (Blok, 2005). With regard to the Water Framework Directive (WFD, 2000), the Conference of European Directors of Roads (CEDR, 2016) states that polluted road runoff has to be cleaned before it is released to surface waters. Runoff from tunnel washes is considered as a pollution hotspot which should not be discharged untreated. Both demands are currently not implemented in every European country. According to CEDR (2016), decisions to treat runoff water at certain road sections are mainly based on traffic density in practice, normally above the range of 10,000–15,000 vehicles/day. The technical standard for runoff treatment is different in the various European countries and best developed in Austria, Germany and Switzerland (CEDR, 2016). According to German technical regulations, retention soil filters with an efficiency degree of 95% and sedimentation basins with an efficiency degree of 40–70% are state of the art with regard to new constructions (DWA, 2016; REWS, 2020). Performance of already existing treatment plants may deviate from these requirements (BASt, 2019). Treatment options in Germany and their efficiency are described in detail by Baensch-Baltruschat et al. (2020).

6.2.2. Transport and treatment at urban roads

In most European countries, urban roads are generally completely paved. Thus, most of the TRWP will be deposited on sealed surfaces. If streets are regularly cleaned a fraction of TRWP will be removed together with other components of road refuse. Another fraction of TRWP will be drifted to unsealed zones close to the road, e.g. to planted verges or beds along streets where it will be accumulated over time (Ten Broeke et al., 2008) (see Section 6.1). The remaining fraction will be washed off and transported to the roadside. In countries with abundant and frequent rainfall such as in western Europe, roads in urban areas are mostly connected with a sewage system which collects runoff from streets. It is expected that coarser TRWP are already retained in sewer inlets, gully pots and channels while the finer fraction is transported through the entire sewage system (Loganathan et al., 2013; Wagner et al., 2018). Sewage systems in urban areas are either constructed as separate or combined sewage systems. In separate systems runoff from streets and waste water from households and industry is collected in different channels. Road runoff water can be treated as described in Section 6.2.1 whereas treatment facilities in urban areas will mostly have smaller dimensions than at highways since space is often scarce. In Germany, decentralised rainwater installations for single street inlets are also in use. In cases, road runoff sewers are not connected with a treatment facility, the runoff water is discharged to surface water without prior purification.

6.2.3. Combined sewer systems and wastewater treatment in urban areas

In contrast to separate sewage systems, combined sewage systems collect both, waste water from households and industry as well as runoff from streets. During heavy rainfall the capacity of the sewage system and the wastewater treatment plant (WWTP) may not be sufficient to receive and treat the whole water volume. Combined sewer overflows (CSO) occur (Brombach et al., 2005). Their frequency depends on the design of the sewage system and on local climatic conditions. If no constructions exist to retain the overflow the wastewater is only partly treated and discharged to the surface water without sufficient purification. For Germany, Hillenbrand et al. (2005) assume an overflow rate of 44.2 vol% as average value for combined sewage systems to estimate the release of tyre wear into the aquatic environment. The approach of this estimation is not elucidated. Brombach et al. (2005) derived an overflow rate of 20% based on the long-term evaluation of 34 real sewage systems in Southern Germany including the WWTPs. In Germany, according to the state of the art, CSO should be stored either in rain overflow basins or in storage sewers. From there partial currents will be discharged to the WWTP, to retention soil filters and probably directly to surface water (DWA, 2013). DWA (2016) also provides CSO installations without storage capacity which divide the wastewater stream into a fraction flowing to the WWTP and a fraction which is directly discharged to the surface water with only partial treatment.

Currently, no specific information is available on the fate of TRWP in WWTPs. However, there are some studies on the environmental behaviour of MP in WWTPs. Mintenig et al. (2014) assume that MP of higher density (e.g. polyurethane with a density of 1.2 g/cm3) sediments in the sewage sludge or is captured in the sand trap. For shredded tyre materials, a density of 1.13 to 1.16 g/cm3 has been found (Rhodes et al., 2012); for TRWP a density between 1.3 and 2.2 g/m3 is reported.
performed a study including 12 German WWTPs where MP concentrations in effluents and sewage sludge were examined. At the WWTP equipped with a final filtration, a cleaning efficiency of 93% (referring to particle number) was found for MP < 500 μm. In other studies, efficiencies between 95 and 99% were found (Horton et al., 2017). Carr et al. (2016) report that MP particles are almost completely retained (99.9%) based on a study including 8 WWTPs in California. From results of a batch study, Mitrano et al. (2019) concluded that >98% of nanoplastic particles remain in the sludge. Breitbarth and Urban (2018) examined the retention of larger MP particles (size: 1–5 mm) in four different WWTPs in the German federal state of Hessen. They report percentages of 1.4–7.1% relating to the number of particles and 0.5–4.1% referring to particle mass in the effluents. Small amounts were also found in the sand trap and partly in the screenings. Thus, it can be expected that TRWP will predominantly be retained in WWTPs, mainly in the sludge. Sewage sludge is either incinerated, disposed of in landfills or used as a fertiliser in agriculture (Duis and Coors, 2016). Hurley and Nizzetto (2018) report that about 50% of sewage sludge are spread on farmland in Europe and North America. In Germany, 23.9% of sewage sludge were utilised in agriculture and 9.6% used in landscaping in 2016 (Rokosch et al., 2018). By spreading of sewage sludge on agricultural areas, MP including TRWP and other pollutants are released into the terrestrial environment. MP present in the sewage sludge may remain in the soil, be mobilised and distributed by wind, or be transported with surface runoff to the aquatic environment (Duis and Coors, 2016). Rather probably, this also applies for TRWP, if not degraded meanwhile.

6.3. Releases of TRWP into environmental compartments

It can be expected that TRWP are released into different environmental compartments. A small fraction is emitted into the atmosphere while much larger proportions will reach soils close to roads and aquatic compartments, respectively. In case that roads are equipped with a drainage system including runoff treatment or transport to a WWTP, TRWP are more or less retained and will not or only to a low extent reach surface waters. In some studies the tyre wear masses which are annually released into surface waters were estimated whereas partly rough estimations were made. For German highways, Wagner et al. (2018) estimated the mass fractions which are released to surface waters and which remain in roadside soil assuming two scenarios with different runoff (15 and 50%, respectively, according to Hillenbrand et al., 2018). For runoff treatment a general efficiency of 50% was assumed. Corresponding to the two scenarios in this study, 6 and 23%, respectively, of the tyre wear generated on highways are released to surface waters while 77 and 45%, respectively, remain at the roadside. Lassen et al. (2015) calculated the annual tyre wear masses, which are generated in the entire road network in Denmark and released into the aquatic environment, taking into account the runoff from roads and the percentage of the different stormwater treatment systems. According to their results, 8 to 40% of the tyre wear formed on roads reaches surface waters. A mass balance including the entire paths from the generation on roads to the Seine river and its estuary located in France was modelled by Unice et al. (2019). The likewise predicted sediment concentrations appeared to be in good agreement with the experimental results of a previous study (Unice et al., 2013) (see Section 7.2). A comprehensive calculation including the entry paths into soils and surface waters was performed by Baensch-Baltrusch et al. (2020) for the entire German road system. The results show that most of the TRWP are deposited into soil near roads (66–76% of the coarse TRWP generated on roads), a smaller proportion is released to surface waters (12–20%) and only a small amount is spread on agricultural areas (2–3%). Since data on the degradation in soil are scarce it is currently unclear if and how the particles will accumulate over a long period.

Little is known on the behaviour of TRWP in rivers regarding sedimentation, further fragmentation, degradation and transport. Nizzetto et al. (2016) and Besseling et al. (2017) calculated the transport of MP in the rivers Thames (United Kingdom) and Dommel (Netherlands), respectively, finding high rates of retention for larger particles (particles ≥200 μm, if heavier than water, and ≥50 μm, respectively). Unice et al. (2019) modelled the releases of TRWP into surface waters for the Seine (France) and Scheldt (France, Belgium and Netherlands) river basins. According to this case study, only 2% of the TRWP generated on roads reaches the sea.

It is questionable whether results for MP are transferable to the behaviour of TRWP in rivers due to different density, shapes and other properties of the particles. Likewise, results for individual river basins are not generally applicable to other surface waters with different hydrological conditions.

7. Results from monitoring studies concerning tyre and road wear particles

The first monitoring studies on the occurrence of TRWP in the environment were performed in the 1970s (Cadle and Williams, 1978; Pierson and Brachaczek, 1974). The presence of TRWP is reported in air, road dust, soil, snow, street runoff, wastewater treatment systems, river water and sediments as well as in air. An overview on publications up to 2008 is given by Wik and Dave (2009). Table 3 presents more recent field studies from 2000 up to present. As far as possible, their results are converted into tyre wear particle concentrations to enable the comparison between the different studies.

7.1. Challenge for analytics

Environmental monitoring of TRWP is hampered by analytical problems. TRWP cannot be analysed using FTIR or Raman spectroscopy, which is commonly used for other types of MP (Baensch-Baltrusch et al., 2017). Since tyre tread as well as TRWP have a complex chemical composition and TRWP are difficult to separate from environmental matrices markers are needed to detect the particles in environmental samples. In general, chemical compounds which are contained as additives in tyre tread materials in sufficiently high concentrations are used as markers.

A reliable marker should i) be specific to tyre rubber polymer, ii) be present in comparable concentrations preferably independent of tyre brands, iii) not easily leach out or be transformed under environmental conditions and iv) be easily and precisely detectable (Klöckner et al., 2019; Unice et al., 2013; Wagner et al., 2018). Quite often Zn has been used as a marker, e.g. by Harrison et al. (2012) for airborne particles and by Kocher et al. (2008) for soil profiles. However, inorganic Zn, a marker applied in earlier studies, is regarded as not very specific to tyre wear in general since it is also emitted from other traffic-related sources such as, for example, road markings, galvanized metal in automobiles, brake wear and road markings (Unice et al., 2013; Wagner et al., 2018). But all these sources may contribute to the composition of real-world TRWP. However, Zn is also released from buildings to the aquatic environment (Hillenbrand et al., 2005) and emitted from industry (Padoan et al., 2017). On the other hand, Kocher et al. (2010b) analysed 62 car tyres of broad origin and found an average Zn concentration of 11.3 g/kg in the tyre tread. This is more than the 100-fold content compared to typical natural topsoils in Germany (LABO, 2017). Thus, it can be concluded that Zn is at least a significant indicator for TRWP in soil or sediment samples taken close to emissions on roads and in absence of other relevant sources, as the comparison between road environment and background values confirms (e.g., Fauser, 1999; Kocher et al., 2010a, c). Fauser et al. (1999) developed an analytical method for tyre tread in environmental samples based on the measurement of extractable organic Zn using dichloromethane as extractant. In this regard, Unice et al. (2013) remark that impurities of inorganic Zn cannot be excluded.
reliably if dichloromethane is utilised for extraction. Klöckner et al. (2019) applied Zn as a marker after density separation. This method allows separating TRWP from other particulate metal species in environmental samples. The concentrations of TRWP found by this method are in good agreement with those of a second method applied by the authors, a thermo-analytical procedure (see below). Other often used markers are benzothiazoles such as 2-(4-morpholinyl)benzothiazole (24MoBT) and N-cyclohexyl-2-benzothiazalolamine (NCBA), both components of vulcanization accelerators. Kumata et al. (2000) assessed the application of both substances as markers for environmental

### Table 3

| Environmental compartment | Location of sampling sites | Country | Environmental concentrations (exemplary) | Analytical method applied | Reference |
|---------------------------|---------------------------|--------|------------------------------------------|---------------------------|-----------|
| Cross-media studies       |                           |        |                                          |                           |           |
| Road dust:                | Tokyo                     | Japan  | Tyre wear concentrations<sup>a</sup>        | GC/FPD/MS; Marker: NCBA, 24MoBT | Klöckner et al. (2000) |
| - inside tunnel           |                           |        | 7.7–204 mg/g dw<sup>b</sup>                |                           |           |
| - outside tunnel          |                           |        | 4.5–37 mg/g dw<sup>b</sup>                |                           |           |
| River water SPM (during storm flow) |             |        | 5.2–92 mg/g dw<sup>b</sup>                |                           |           |
| River sediment-           |                           |        | 2.2–52 mg/g dw<sup>b</sup>                |                           |           |
| - Sumidagawa river       |                           |        |                                          |                           |           |
| - Tamagawa river         |                           |        | 0.8–3.9 mg/g dw<sup>b</sup>                |                           |           |
| Sediment core (moat)     |                           |        | 0.4–2.3 mg/g dw<sup>b</sup>                |                           |           |
| atmospheric aerosols    |                           |        | 0.9–7.8 mg/g dw<sup>b</sup>                |                           |           |
| Runoff river sediments   |                           |        | 39–116 μg/m<sup>3</sup>                    |                           |           |
| Road dust                |                           |        |                                          |                           |           |
| Pinang and Klang         |                           | Malaysia | 0.6–17 mg/L<sup>c</sup>                  | GC/FPD/MS; Marker: NCBA, 24MoBT | Kumata et al. (2002) |
| Riverine and estuarine   |                           |        | 0.4–2.8 mg/g<sup>e</sup>                  |                           |           |
| sediments                |                           |        | 6–19 mg/g<sup>e</sup>                     |                           |           |
| Kuara Lumpur             |                           |        | 0.092–0.59 mg/g<sup>e</sup>               | GC/FPD; Marker: NCBA     | Zakaria et al. (2002) |
| Surficial sediments      | Seine, Chesa-peake estuary, Yodo river basin - Lake Biwa | France/USA/Japan | 1.4–8.4 mg/g<sup>b</sup> | Pyrolysis GC–MS | Monitoring study: Unice et al. (2013) Analytical method: Unice et al. (2012) |
| Seine                    | Seine                     |        | Mean: 2000; median: 840 μg/g dw          |                           |           |
| Chesa-peake              |                           |        | 95th percentile: 8400 μg/g dw           |                           |           |
| Yodo-Biwa                |                           |        | range: 62–11,600 μg/g dw                 |                           |           |
|                         |                           |        | Mean: 910; range: 50–4400 μg/g dw        |                           |           |
| Roadside soil (<15 cm depth) | Seine                  |        | Mean: 770; range: 26–4600 μg/g dw        |                           |           |
| Ambient air              | Seine                     |        |                                          |                           |           |
| River water aqueous phase |                           |        |                                          |                           |           |
| River water SPM          | Pearl River Delta         | China  | n.d.–0.8 mg/L<sup>c</sup>                | GC–MS; Marker: 24 MoBT   | Ni et al. (2008) |
| Sediments from two intertidal estuaries | Charleston Harbor and Winyah Bay (North Carolina) | USA | n.d.–85 mg/g<sup>b</sup> | GC–MS; Marker: 24 MoBT and five other benzothiazoles | Gray et al. (2018) |
| Street runoff (grab sample) | Berlin Halensee           | Germany| 35–70 mg/g<sup>h</sup> (particle fractions) | Thermal extraction desorption (TED)/GC/MS; Markers: cyclohexenylbenzene and phenyl[4,4.0]bicyclodecene for SBR oligomers of isoprene for NR | Eisenraut et al. (2018) |
| Sedimentation basin (sludge from the bottom) | Berlin Halensee           | Germany| 82 mg/g<sup>h</sup> | Inductively coupled plasma mass spectrometry (ICP-MS); Marker: Zn (applied after density separation) | Klöckner et al. (2019) |
| Lake sediment            | Lake Tegel, Berlin        | Germany| <0.73 mg/g | Validation by TED–GC–MS; Markers: 3-phenylcyclo-hexene a.o. |           |
| Sludge from two road runoff treatment systems | Berlin Halensee Leipzig | Germany| 16–150 mg/g |                           |           |
|                         |                           |        | 0.4–2.0 mg/g (Data relates to TRWP assuming a tyre wear content of 50% and a rubber content of 25%) |                           |           |

(continued on next page)
| Environmental compartment near road, one site | Location of sampling sites | Country | Environmental concentrations (exemplary) | Analytical method applied | Reference |
|---------------------------------------------|----------------------------|---------|------------------------------------------|--------------------------|-----------|
| Ambient air close to roads                  | Copenhagen, Riso (short term) | Denmark | Content of tyre wear in PM10<sup>i</sup> Absolute concentration of tyre wear<sup>j</sup> About 5.8% referring to mass of total PM 2.81 µg/m<sup>3</sup> 8% referring to mass | Atomic absorption spectrometry Marker: extractable organic Zn | Fauser (1999) |
| Ambient air at an urban road and a background site | Berlin (long-term: >1 year) | Germany | Thermography Marker: ratio of elemental and semi-volatile organic carbon 7.5% referring to mass 1.9% referring to mass | Gehrig et al. (2001) |
| Ambient air in two city centres             | Bern (kerbside) Zurich (court yard) (long-term: >1 year) Long Beach, Los Angeles, Azusa, Claremont (California) (short term) | USA | GC–MS Marker: NCBA ≤3% referring to fine particulate mass ≤2.0 µg/m<sup>3</sup> | Schauer et al. (2002) |
| Ambient air at an urban street canyon and at an interurban highway | Zurich, Reiden (short term) | Switzerland | No significant contribution to PM10 Rotating drum impactor sampling followed by synchrotron radiation X-ray fluorescence spectrometry (RDI-SR-XRF) PMF Carbon: light absorption | Bukowiecki et al. (2010) |
| Ambient air at a residential and an industrial site | Kolkata (long term: 1 year) | India | FID-GC, carbon analyser principal component analysis - multiple linear regression (PCA-MLR) 8% referring to mass (industrial site) | Karar and Gupta (2007) |
| Ambient air at a site including both sides of a highway | Near Bonn (long term: >1 year) | Germany | ICP-MS Carbon: detection in an IR oven PMF 3% referring to mass | Quass et al. (2008) |
| Ambient air in an urban street canyon and at an interurban highway | Granada Norte (long term: 4 years) | Spain | Pyrolysis GC–MS markers: butadiene and isoprene dimers | Panko et al. (2013a, 2013b) |
| Ambient air at one roadside - on one rooftop | Hachiogi City (single sampling) | Japan | GC–MS Markers: hydrogenated resin acids 0.68% referring to mass 0.83% referring to mass | Kumata et al. (2011) |
| Ambient air at a busy roadside | Chennai City (short term) | India | Inductively Coupled Plasma with Optical Emission Spectroscopy (ICP-OES) PMF 4.1% referring to mass | Srimuruganandam and Shiva Nagendra (2012) |
| Ambient air at an urban street canyon and at an urban background site | London (5 campaigns) | United Kingdom | ICP-MS Marker: Zn 10.7% referring to mass (street canyon) (Contribution to PM0.9–11.5) Average: 0.84 mass % range: 0.14–2.8 mass % referring to TRWP mass | Harrison et al. (2012) |
| Ambient air at a kerbside in a street canyon and an urban background station | Areas of Seine, Chesapeake and Yodo river basins (single sampling) | France/USA/Japan | Pyrolysis GC–MS markers: butadiene and isoprene dimers X-ray fluorescence spectroscopy (XRFS) PMF | Panko et al. (2013b) |
| Ambient air at bus road | Granada Norte (long term: 4 years) | Spain | ICP-MS, ICP atomic emission spectrometry (ICP AES) PMF 8% referring to mass 3.4 µg/m<sup>3</sup> | Amato et al. (2014b) |
| Ambient air at 60 residential sites in 3 cities | Chicago (Illinois), St. Paul (Minnesota), Winston-Salem (North Carolina) (short term) | USA | X-ray fluorescence spectroscopy (XRFS) PMF 1.6–7.0% referring to mass (Contribution to PM2.5–10) | Sturtz et al. (2014) |
| Ambient air, sites at roadides | London Barcelone (short term) | United Kingdom, Spain | Aerosol Time-Of-Flight Mass Spectrometry (ATOFMS) 2.5% referring to particle numbers 2.0% referring to particle numbers (Contribution to fine particle fraction: ≤3 µm) | Dall'Osto et al. (2014) |
| Ambient air, sites at a kerbside in a street canyon and an urban background station | Cities of Essen and Mülheim, Ruhr region (short term) | Germany | SEM/EDX 7 mass % at the road kerbside, 6 mass % at the urban background (contribution of total traffic-related abrasion) | Weinbruch et al. (2014) |
| Air close to two highways and one urban road | Cologne area, Freiburg (long term: 4 years) | Germany | TLM, SEM-EDX 33% referring to number of particles 54% referring to volume of identified particles (Contribution to coarse fraction PM10–80) | Sommer et al. (2018) |
| Ambient air, sites at a traffic junction, a street canyon, urban background | Leipzig (repeated campaigns) | Germany | Total reflection x-ray fluorescence spectroscopy (TXRF), marker: inorganic Zn<sub>1</sub> 1.7–2.9 mass % 0.3–0.5 µg/m<sup>3</sup> (at the roads) 0.2–0.4 µg/m<sup>3</sup> (at the urban background station) | Fomba et al. (2018) |
| Ambient air near road, one background site | London, Los Angeles and Tokyo (short term) | USA, Japan | Pyrolysis GC–MS; markers: 4-vinylcyclohexene (SBR, BR) and dipentene (NR) 0.45–2.48 mass % (referring to TRWP mass) 0.095–1.91 µg/m<sup>3</sup> (Referring to TRWP concentrations) | Panko et al. (2019) |
samples. The authors observed that NCBA appears to be less stable than 24MoBT and that benzothiazoles are biologically transformed under aerobic conditions. Besides tyre wear, leakage of antifreeze in automobile radiators was found as another possible source of both substances (Kumata et al., 2002). Unice et al. (2013) state that benzothiazoles are not specific to tyre wear and can separate from the TRWP by leaching in aqueous environments. Kumata et al. (2011) assessed hydrogenated resin acids can enable semi-quantitative source apportionment in runoff, in soil, on the road and in residential areas.

In Table 3 the environmental concentrations found in monitoring studies are presented. Unice et al. (2013), Pankot et al. (2013a) and Klöckner et al. (2019) estimated concentrations of TRWP with the measured marker concentrations assuming that TRWP consist of 50% tyre wear and 25% rubber polymer. As far as calculation factors were available we derived the tyre wear concentrations from the marker concentrations found by other authors (Eisentraut et al., 2018; Kumata et al., 2000, 2002; Ni et al., 2008) to make the results of the various studies comparable.

As far as calculation factors were available we derived the tyre wear concentrations from the marker concentrations found by other authors (Eisentraut et al., 2018; Kumata et al., 2000, 2002; Ni et al., 2008) to make the results of the various studies comparable. Alternatively to the use of the markers as described above. Here, the structure of the rubber polymer is thermally decomposed by application of pyrolysis (py) or thermal extraction desorption (TED). Polymer specific degradation products are captured and detected by gas chromatography–mass spectrometry (GC–MS) or by a flame ionization detector (FID). Eisentraut et al. (2018) identified markers which are degradation products generated by TED. Cyclohexenylbenzene and phenyl[4.4.0]bicyclocdecene prove to be markers for SBR while oligomers of isoprene stand for NR concentrations. The authors state that analysis of NR in environmental samples requires the removal of all plant matter.

### Table 3 (continued)

| Environmental compartment | Location of sampling sites | Country | Environmental concentrations (exemplary) | Analytical method applied | Reference |
|---------------------------|---------------------------|---------|------------------------------------------|---------------------------|-----------|
| Road dust and soil        | Roadside soils (samples ≥10 cm depth, 0.5 m distance from road edge) | Germany | TWP-content: mean 20.2 mg/g dw, median 11.2 mg/g dw, range: 0.4–158 mg/g dw | Marker: Zn (11.3 g Zn/kg tyre tread) | (Kocher et al., 2008) |
| Dust from urban and rural roads, rural roads partially unpaved (12 sites in total) | San Joaquin Valley, California | USA | Concentration of benzothiazoles in total: 17.6–2242 ng/g | GC–MS: indicators: benzothiazole, 2-hydroxy benzothiazole | Rogge et al. (2012) |
| Dust from 4 roads         | London                     | United Kingdom | Content of tyre wear in fine particle fraction (0.3 μm): 4% referring to particle number | ATOFMS | Dall’Osto et al. (2014) |

As far as calculation factors were available we derived the tyre wear concentrations from the marker concentrations found by other authors (Eisentraut et al., 2018; Kumata et al., 2000, 2002; Ni et al., 2008) to make the results of the various studies comparable. Additionally, since the content of other contributions to real-world TRWP besides tyre wear is quite variable (Sommer et al., 2018) it seems sensible to report results of monitoring studies in terms of tyre wear. The authors observed that NCBA appears to be less stable than 24MoBT and that benzothiazoles are biologically transformed under aerobic conditions. Besides tyre wear, leakage of antifreeze in automobile radiators was found as another possible source of both substances (Kumata et al., 2002). Unice et al. (2013) state that benzothiazoles are not specific to tyre wear and can separate from the TRWP by leaching in aqueous environments. Kumata et al. (2011) assessed hydrogenated resin acids can enable semi-quantitative source apportionment in runoff, in soil, on the road and in residential areas.

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### Table 3 (continued)

| Environmental compartment | Location of sampling sites | Country | Environmental concentrations (exemplary) | Analytical method applied | Reference |
|---------------------------|---------------------------|---------|------------------------------------------|---------------------------|-----------|
| Road dust at 7 sites - close to busy roads | 721 samples from eight federal states, average daily traffic: mean 24,600, median 10,000, max. 160,000 vehicles/d | Hachioji City | Contribution to PM10: 0.14% referring to mass 0.07% referring to mass 2.0% referring to mass | GC–MS: Markers: hydrogenated resin acids | (Kumata et al., 2011) |
| Dust from urban and rural roads, road dust | San Joaquin Valley, California | USA | Concentration of benzothiazoles in total: 17.6–2242 ng/g | GC–MS: indicators: benzothiazole, 2-hydroxy benzothiazole | Rogge et al. (2012) |
| Dust from 4 roads | London | United Kingdom | Content of tyre wear in fine particle fraction (0.3 μm): 4% referring to particle number | ATOFMS | Dall’Osto et al. (2014) |

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ments. Since no data is given on the environmental concentrations of Kumata et al. (2002). Tyre wear was found in all investigated compartments. NCBA/24MoBT ratios vary from 0.59 to 41.3. Further results are presented in Kumata et al. (2000) (see Table 3). By this, a comparison is enabled with the results from other monitoring studies.

Unice et al. (2013) analysed TRWP in surficial sediments of three river basins located in France, USA and Japan assuming that the proportion of tyre wear in TRWP is about 50% and the proportion of rubber polymer 25%. The authors applied pyrolysis GC-MS using dipentene and vinyl cyclohexene as markers for NR and SBR/BR, respectively. A stratified approach was used to select sampling sites which should cover a wide variety of different areas and potential habitats. In total, 149 samples at 35 sites were collected. In addition, 69 surface soil samples were taken at road sides. TRWP was found in all investigated areas with a detection frequency of 97% in the river sediment samples. Mean values amount to 4500 μg/g dw in the Seine river basin, 910 μg/g dw in the Chesapeake and 770 μg/g dw in the Yodo river basin. The average concentrations on roadside soil are 9100 μg/g dw (Seine) and 1400 μg/g dw (Chesapeake and Yodo). When soil and sediment samples are grouped by population density as an indicator for traffic density the concentration in road-side soil is ca. 70% higher on average as in river sediments. In a concurrent project, Panko et al. (2013a) investigated the atmospheric concentrations of TRWP whereas sampling sites were located in the same three river basins. The 13 sample locations include residential, commercial/industrial, recreational and rural areas. The overall average concentration of TRWP is 0.16 μg/m³ with values ranging from <0.004 to 1.34 μg/m³ for individual sampling sites. On average, the contribution of TRWP to PM10 is 0.84%. The highest value (2.80%) was found in Troyes (France) whereas the authors consider the reasons for the difference to others sites as unclear.

7.3. Monitoring studies in the aquatic environments

Some studies have been published within the last twenty years which focus on the quantification of tyre wear concentrations in aquatic compartments. Ni et al. (2008) analysed 24MoBT and five other benzothiazoles in water and suspended particulate matter from eight inlets to the Pearl River Delta (China). The authors consider tyre wear as the main source of benzothiazoles. Based on the concentration of 24MoBT in tyre tread given by Kumata et al. (2000) we estimated the tyre wear concentrations in the samples amounting up to 0.8 mg/L in water and 85 mg/g in suspended particulate matter (see Table 3).

Eisentraut et al. (2018) applied thermal extraction desorption (TED) combined with GC-MS to analyse tyre wear and thermoplastic MP in samples from a street runoff treatment system at a highway in Berlin (Germany) including sampling in the influent channel and sludge from the sedimentation basin. SBR concentrations were determined for different particle size fractions in the sample from the channel and for the sludge sample in total. The highest SBR concentrations were found in the runoff fraction 100–500 μm (ca. 7.9 mg/g) and in the sludge sample (9.3 mg/g). Eisentraut et al. (2018) estimated the content of tyre wear in particulate matter from road runoff and sedimentation sludge to be 3.4–8.2% dw. This is 13–49 higher than the sum of thermoplastic MP present in these samples (0.07–0.62% dw). From the measured SBR concentrations we derived absolute tyre wear concentrations whereas we assumed an average SBR content in tyres of 11.3% referring to worldwide production and all types of tyres (trucks and passenger cars) as reported by Eisentraut et al. (2018). According to our estimation, the tyre wear concentrations in the different particle fractions of street runoff are 35–70 mg/g and 82 mg/g in the sludge sample.

Klöckner et al. (2019) assessed Zn as a marker whereby TRWP were removed from the environmental matrix via density separation prior to analysis. Zn concentrations were measured by using inductively coupled plasma mass spectrometry (ICP-MS). The developed method was applied to analyse sludge samples from road runoff treatment facilities at two sites at different highways (Berlin, Leipzig/Germany). The results were validated by applying TED-GC–MS. From analytical results for a background sample spiked with tyre wear, Klöckner et al. (2019) conclude that the method developed is more appropriate for samples with high traffic influence while Pyr-GC-MS or TED-GC–MS are more sensitive and therefore more suitable for samples from sites with low traffic influence such as river sediments. In Berlin, the highest concentrations were found in the sedimentation basin and the upper layer of the subsequent soil retention filter close to its inlet (130 and 150 mg/g, respectively). Significantly lower concentrations were recorded at the opposite side of the soil retention filter at the distance of ca. 35 m to the point of discharge (16 mg/g). According to the authors, the results show that the pre-treatment sedimentation basin does not retain all TRWP washed off from the road surface. Concentrations found at the highway in Leipzig were much lower with 2.0 mg/g at the first settling pond and 0.4 mg/g at the consecutive settling pond. This is explained by the lower traffic volume, more steady traffic flow without frequent accelerating and braking and the agricultural surrounding of Leipzig. The latter is supposed to cause higher deposition of soil particles on the road which may lead to dilution of TRWP in road dust. Moreover, at the site in Leipzig, the sediment collected in the treatment system is not regularly removed in contrast to the site in Berlin. The authors assume that aging of TRWP along with formation of larger and denser heteroaggregates with further mineral material may hamper detection as TRWP due to increase in density. Furthermore, Klöckner et al. (2019) state that biodegradation may lead to a decrease of TRWP over time.

Only few studies are available on tyre wear in marine compartments. In an early study Spies et al. (1987) analysed 24 MoBT in four coastal sediments from San Francisco Bay and a pristine lagoon. 24MoBT concentration range from 23 to 360 ng/g dw at San Francisco Bay and were below limit of detection at the background site. Based on the calculation factor for 24MoBT derived by Kumata et al. (2000) we estimated tyre wear concentrations of 10–155 ng/g for the sediments from San Francisco Bay. The high maximum value can be explained by the sample location near a large bridge with high traffic volume. Zakaria et al. (2002) investigated the presence of tyre wear in sediments from two estuaries at the Malacca Street in Malaysia. NCBA concentrations lie between 2.5 and 5.6 ng/g. Gray et al. (2018) found a high abundance of black MP fragments in sediment samples from two estuaries in South Carolina (USA). Their number accounts for 73% of the MP particles in total. Due to their shape and morphology these fragments are assumed to be TRWP.
7.4. Monitoring studies in ambient air

The tyre wear concentrations in ambient air close to road and the relative contribution of tyre wear to PM have been determined in a number of studies. The most important results of these studies are compiled in Table 3. Sampling sites were mostly located at busy roads in larger cities or close to highways. Sampling was done partly in long-term projects (≥1 year), partly during shorter measuring campaigns. Some authors used markers to measure the tyre wear concentrations in air samples. Substances which were applied as markers comprise Zn (Fauser et al., 1999; Fomba et al., 2018; Harrison et al., 2012), NCBA (Gehrig et al., 2001), hydrogenated resins acids (Kumata et al., 2011) as well as 4-vinylcyclohexene and dipentene (Panko et al., 2019) of which advantages and disadvantages are discussed in Section 7.1. The study with the greatest geographical coverage was performed by Panko et al. (2019) who collected PM2.5 samples at 17 sites in Los Angeles (USA), London (United Kingdom) and Tokyo (Japan) and PM10 samples at five sites in London and Tokyo. Assuming that the polymer content in TRWP is 25% they report a contribution of TRWP to PM10 of 0.45–2.48 mass % with absolute TRWP concentrations between 0.095 and 1.91 μg/m³. The contribution of TRWP to PM2.5 is lower: up to 0.68 mass % with absolute concentrations ≤0.29 μg/m³. In a previous study, PM10 samples from sites in France, Japan and USA had been analysed (see Section 7.2). Regarding all compiled studies with application of markers the contributions of tyre wear to PM10 ranges from 3 to about 11% referring to mass.

Other authors applied statistical modelling to determine the contribution to PM from different sources including tyre wear (source apportionment). Quite often the receptor model Positive Matrix Factorization (PMF) was used in combination with trace elemental analysis of airborne PM samples. In a first step PMF, which is based on the law of mass conservation, identifies a set of factors which can be taken to represent major emission sources. Subsequently, scores on these factors are regressed against the element concentrations to estimate the contributions from each source (IEHIA, 2020). For instance, during more than one year, Quass et al. (2008) examined the concentrations of airborne particles (size: ≤10 μm) and their components such as organic matter, metals and other elements at both side of a highway (upwind-lee-measurement) near the city of Bonn (Germany). Based on the results for characteristic components, they modelled the contributions of various traffic-related emissions to the total emissions of PM 10 amounting to 3% for tyre wear, 14% for brake wear and 7% for resuspension of road dust (with regard to mass percentages). The contribution of exhaust emissions was estimated to be about 60%. Another receptor model, principal component analysis – multiple linear regression (PCA–MLR), was used in combination with trace element analysis by Karar and Gupta (2007). Schauer et al. (2002) applied a Chemical Mass Balance receptor model using concentrations of organic compounds to determine source fingerprints which allow source apportionment. The relative contributions of tyre wear to PM10 determined in these studies range from 0 to 8% (see also Padoan and Amato (2018)).

A third option to investigate the presence of tyre wear in PM is to analyse single airborne particles applying electron or light microscopy combined with energy-dispersive X-ray spectroscopy or mass spectroscopy. Dall’Osto et al. (2014) report on the analysis of aerosols collected at road sides in London (United Kingdom) and Barcelona (Spain). Measurements were performed by application of Aerosol Time-of-Flight Mass Spectrometry (ATOFMS) which allows to determine the distribution of chemical species within individual particles. The number of particles identified as TRWP was 2.5% (London) and 2% (Barcelona), respectively.

Weinbruch et al. (2014) took samples of airborne particles at two sites in a metropolitan region in Western Germany at the kerbside of a busy urban road and an urban background station. Using scanning electron microscopy combined with energy-dispersive X-ray spectroscopy (SEM-EDX), 80 samples were analysed in detail whereas particles with a size of 0.2 to 10 μm were taken into account. The contributions of traffic abrasion and road dust resuspension to the mass concentration of particulate matter were determined. The absolute additional load of abrasion at the kerbside station in comparison to the background station is 0.8 μg/m³, the additional load for resuspension amounts to 4.2 μg/m³ referring to PM10. The authors do not give specific values for individual components of abrasion and resuspension such as tyre wear.

Sommer et al. (2018) analysed airborne particles collected in passive-sampler devices placed close to two highways located in the surrounding of Cologne (Germany) and one urban road in Southwestern Germany whereas they focused on particles with a size of 10–80 μm being remobilised by wind and passing traffic. Using single particle analysis via transmitted light microscopy (TLM) and scanning electron microscopy combined with energy-dispersive X-ray spectroscopy (SEM-EDX), they identified >500 particles and investigated their size, shape, volume and chemical composition. They found that 89% of the analysed particles could be assigned to traffic-related sources with a percentage of 33% for tyre wear. Other contributions are road wear (39%), brake wear (17%) and various non-traffic sources (11%). No pure tyre wear particle without any encrustment was found. The investigated coatings consist of relatively large road wear particles (1–10 μm) embedded in a matrix of smaller dust particles whereas the larger particles were identified as minerals and metals.

The above cited monitoring studies on tyre wear in ambient air provide data on its relative contribution to particulate matter, mostly to PM10, which gives information on the relevance of tyre wear as a traffic-related pollution source. Some authors also report data on absolute concentrations in ambient air which are required to assess exposure with regard to human health. Up to now, binding thresholds are missing to evaluate these results. However, the concentrations presented in Table 3 are below the NOAEC derived by Kreider et al. (2019).

7.5. Monitoring studies in soils

Studies on the presence of tyre wear/TRWP in soil are rather scarce. Instead, most publications focus on the concentrations of organic compounds, metals and other elements in soil and percolating water which are related to non-exhaust emissions (e.g., Beer et al., 2011; Kocher et al., 2010a; Kocher et al., 2008; Wessolek and Kocher, 2003). However, using own results, further evaluation of data from previous studies was possible. In Kocher et al. (2008) the Zn content of 721 soil samples collected from roadside soils in eight federal states of Germany is reported. Taking a mean Zn concentration of 11.3 g/kg in tyre tread (as given in Kocher et al. (2010b)) into account, tyre wear concentrations in the soil samples range from 0.4 to 158 mg/g dw with a median of 11.2 mg/g dw. This data lies in the same order of magnitude as the concentrations of tyre wear found by Unice et al. (2013) for the Seine river basin (0.1–10 mg/g dw). The higher values measured by Kocher et al. (2008) can be explained with the smaller sampling depth representing the soil layer with the highest traffic influence and the selection of sampling sites exclusively at roads with high traffic density in this study. Furthermore, the dataset of the German study includes sampling sites with Zn coated safety-barriers which may lead to an overestimation of the calculated tyre wear concentrations.

Kumata et al. (2011) used hydrogenated resin acids as markers to estimate the contribution of tyre wear to PM10 from seven sites located at busy roads, in tunnels and residential areas. Rogge et al. (2012) analysed about 200 organic compounds in dust samples collected from paved and unpaved roads at 12 sites in the San Joaquin Valley in California (USA). Detection of long-chained n-alkanes (up to C₄₀) and benzothiazoles (benzothiazole, 2-hydroxy benzothiazole) are reported as indicators for TRWP. Concentrations of tyre wear/TRWP in road dust were not estimated based on these data. Dall’Osto et al. (2014) report that dust from roads were collected at four sites in London. About 2800 particles originating from 48 samples were analysed by ATOFMS. On average, 4% of the particles were identified as tyre-related dust.
7.6. Summary

TRWP were detected in aquatic compartments, in ambient air, road dust and soils. Studies in which chemical markers were used such as Zn, NCBA, 24MoBT or hydrogenated resin acids are less reliable than studies applying thermo-analytical methods. The latter was used by Unice et al. (2013) and Eisenreut et al. (2018) for analysis of aquatic samples and by Panko et al. (2013a) and Panko et al. (2019) for analysis of ambient air samples. A more reliable approach referring to Zn as a marker was developed by Klöckner et al. (2019) who combined Zn analysis with previous density separation. Most studies in aquatic compartments are short-term measuring campaigns. In contrast, Unice et al. (2013) collected samples from aquatic compartments and road dust on a large spatial scale in three countries. Sampling sites were selected following a stratified approach, sampling was repeated several times. This has been the study in which the most extensive data set on TRWP in aquatic compartments was generated up to now. A number of studies were published in which the contribution of airborne tyre wear particles to PM10 was determined. Quite often receptor models such as the Positive Matrix Factorization model (PMF) were used in combination with trace elemental analysis of airborne PM samples. Other authors applied markers for detection of tyre wear. A third option is analysis of single airborne particles applying electron or light microscopy combined with energy-dispersive X-ray spectroscopy or mass spectrometry. Considering all cited studies, tyre wear contribution to PM10 was reported to be up to approx. 11 mass %. The highest absolute concentration of tyre wear in ambient air is reported to be 3.4 μg/m³ (without mass contribution from road wear). Only few studies were published on tyre wear in soils or road dust. For soils near roads, tyre wear concentrations ranging from 0.4 to 158 μg/g dw are estimated.

8. Conclusion, suggested prevention and mitigation measures

The annual emissions of tyre wear generated on roads have been estimated for numerous countries, however, it is often unclear whether tyre wear emissions pose the highest contribution to the generation of MP referring to mass. Concentrations of tyre wear compiled from several monitoring studies show highly variable concentrations in road runoff, road dust, roadside soils, river sediments and river water, with a general decrease following the transport paths. The highest concentrations were found in tunnel dust (max. 204 mg/g) and sediments of a treatment basin for road runoff (max. 150 mg/g). Concentrations of tyre wear in road runoff range from 5 to 92 mg/dw, in soil from 0.2 to about 160 mg/g dw. Little information is available on the entry paths of TRWP into the aquatic environments. Up to now, no specific data has been published on the behaviour of TRWP in WWTPs. The results of few modelling studies indicate that the predominant amounts of TRWP released into surface waters may remain there and are not transported to the marine environment. However, there is hardly any information about what happens in rivers carrying loads of TRWP with regard to sedimentation and degradation. If the latter is hampered by lack of sunlight in deeper water, sedimentation may be the major sink of TRWP. Under hydrological conditions different to those of the rivers which have already been object of modelling studies, higher amounts of TRWP may reach the marine environment. Only few studies have been published on the occurrence of TRWP in different environmental compartments until present in which modern analytical techniques are applied. Most of these studies focus on testing new analytical methods, especially the choice of appropriate markers, to detect TRWP in environmental samples whereas environmental monitoring is not the primary objective. To investigate environmental concentrations repeated sample collecting over longer periods would be necessary. Up to now, only few studies have been published on the presence of TRWP in marine sediments. The data available does not allow for confirming or disproving the assumption that only a small percentage of TRWP is released to the seas. Likewise, data on transport and degradation in soil is scarce.

Doubtless, huge amounts of TRWP are annually emitted to the environment. However, up to now, the extent of ecotoxicological effects is not clarified since most test studies were performed with artificially produced tyre wear particles, which differ from real-world TRWP, and under concentrations much higher than those found in aquatic compartments. No relevant data are available on the terrestrial ecotoxicity of TRWP. Contribution of airborne TRWP to PM10 amounts up to 11%. As to human health risk by inhalation, some in vitro and in vivo studies observed hazardous effects. Only in one study, a toxicological threshold value was derived (NOAEC: 112 μg/m³ for respirable TRWP). The results of this risk assessment indicate that the potential risk for human health concerning cardiopulmonary effects is low. Further monitoring and effect data are required to confirm this conclusion. For other health risks no current data is available. Health risks by exposure via the food chain are currently unclarified.

In view of the high TRWP emissions it is recommendable to close the existing knowledge gaps. Further research is required including:

- Determination of more precise, current EF for different types of roads, road surfaces, road conditions, and vehicles,
- Further development of analytical methods,
- Monitoring and field studies to investigate environmental concentrations and degradation of polymer compounds under environmental conditions,
- Modelling studies to estimate transport, degradation and retention of TRWP in rivers and soils,
- Ecotoxicological studies with environmental samples including freshwater and marine species.

Although the impacts on ecosystems and partly also on human health are unclarified, mitigation measures should be implemented according the principle of precaution:

- Installation of sufficient communal stormwater treatment systems and retrofitting of existing plants regarding the state of the art,
- Improvement of runoff treatment systems at rural roads and highways according to the state of the art, regular control and maintenance of drainage systems including disposal of sediment
- Incineration of sewage sludge containing large proportions of road runoff sediments instead of spreading on agricultural areas or natural soil,
- Regular maintenance of roads and successive optimization of roads surfaces,
- Promotion of lightweighter vehicles,
- Incentives for avoiding transport, use of the combined intermodal traffic, creating awareness, e.g. regarding driving behaviour,
- Implementation of speed limits,
- Optimisation of tyre materials with regard to higher durability and wear resistance taking into account safety and other essential requirements.

Traffic-related measures such as use of lighter vehicles, avoidance of transports or speed limits will also reduce other environmental impacts like emissions of CO₂, NOₓ and noise.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
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