House of Representatives Climate Change Energy Environment and Water Committee
Inquiry into plastic pollution in Australia's oceans and waterways

Questions on notice from CSIRO appearance 31 March 2023

Question 1

Dr Hardesty: Our work and our survey method absolutely include anything that one can see from standing human height. I think your question is particularly focused on the very small ash, if you will, that comes off tyres.
CHAIR: Yes.
Dr Hardesty: We do not have a body of research that is specifically focused on that one component.
CHAIR: Do you know if anybody does?
Dr Hardesty: Within Australia?
CHAIR: Yes.
Dr Hardesty: Within Australia there is no major research effort focusing explicitly or solely on that. It is encompassed in some aspects of certain projects that may be undertaken, in a more ancillary way, rather than it being the core focus.
CHAIR: Do you know if there's any international work being done on that?
Dr Hardesty: There has been recently—it's probably two or three years ago now—a major report that came out that listed microplastics as primary wear or loss from tires into the environment. In terms of ongoing research on that topic, there is a group within the United States and possibly one in Canada, and I don't believe there are any in Europe, but I could follow up on that and come back to you to provide that information if desired.
CHAIR: I'd certainly be very interested if there were any updated information about the impacts of tyre waste on our environment in both the air and water.
Dr Hardesty: To address that specifically, those studies actually focus on the occurrence, which is different from the impact. If you're wanting information on the impact per se, that's a little bit more tricky, but I'm happy to have a look at it. I think that most of the work has demonstrated the occurrence or presence, if that is still of interest.
CHAIR: Yes.

CSIRO response

See two peer reviewed research articles and a report provided as Attachments 1 -3.

Question 2

Ms STEGGALL: In that respect, I know the UK have introduced, for example, a tax or a levy on virgin plastic. Is that something that we should be looking out? Should we put a levy on the import into Australia, for example, of virgin plastic to create a market preference to go to recycled plastics?
Dr Hardesty: It's not my place to ever say that governments should or should not do anything. What I can tell you is that there was an industry voluntary scheme in South Africa. There was an amount of money from industry that came, and what it did was actually raise the floor; the idea was to raise the floor, to make it economically viable given the changes in the petroleum market to encourage or to provide the support required for materials recovery in South Africa. That was with waste pickers for example. They were always ensured enough economic benefit to make that a value proposition for them. But, as a scientist, it's not my place to say what government should or shouldn't do.
Ms STEGGALL: But you're aware of some jurisdictions like that South African one. Is it possible to get some information on that in writing to the committee? I'll put that one on notice.

CSIRO response

CSIRO is aware of programs in various countries such as South Africa, through industry bodies such as Plastics SA (South Africa), https://www.plasticsinfo.co.za/.

However, this question is best addressed to the South African Government.

Question 3

Mr REPACHOLI: Where are those technologies up to? I understand there are more coming online. We are on a trend with this. Where are we with continuing that trend?
Dr Lau: If we're talking about mechanical recycling, the technology exists. The rate-limiting factor there is the number of businesses that are operating that. In mechanical recycling, there's probably not a lot of technological or engineering advancement that's needed to transition that industry. It's really around the volume and the scale. For other technologies like advanced recycling, there are commercial plants. Many of those—I'm estimating in the dozens—are operating in the Northern Hemisphere. We know they're operating. We know they exist. They can process in the order of hundreds of thousands of tonnes per annum. There are plans that have been announced for the Parkes region in New South Wales, with Brightmark, to embark on construction. There's consideration of further growth. We know that Licella are scaling up their capacity and ability to process using hydrothermal catalytic decomposition. Those industries are, I suppose, in the growth phase. The technology is there. Some of the other decomposition technologies that were at earlier stages are now at more of a pilot stage. They're yet to grow to the commercial phase. Because there's a range of different approaches, they're all at different levels of maturity.
Mr REPACHOLI: What's the main limiting factor holding them back? You mentioned scaling up and product. What are the main factors that are holding them back?
Dr Lau: I think it's really the stage they're at. As was mentioned previously, the awareness and action around transitioning our plastics economy are really only just starting now. I couldn't speak to what's holding any individual business back, but I would say that we are really at the beginning of the transition that's needed.
Mr REPACHOLI: This is a question on notice. Would you be able to get us a list of those businesses that are currently doing it in Australia so that we could possibly touch base with them at some stage?
Dr Lau: Yes.

CSIRO response

Through our Ending Plastic Waste Mission, CSIRO is aware of the following businesses who are operating or developing facilities and infrastructure for Advanced Recycling in Australia. We are also aware of over 40 businesses and facilities operating internationally.

This should not be considered a complete list of businesses active in Australia but is indicative, as new businesses are at various stages of development and scale-up.

- **APR Plastics** is a sister company to Australian Paper Recovery and APR Kerbside. APR operate a pilot scale pyrolysis plant for post-consumer plastic waste.
- **Licella** operates a commercial-scale plant in Australia, which uses a hydrothermal catalytic upgrading process to convert non-recyclable plastic waste into a synthetic crude oil.
- **Plastics Pirate** is intending to conduct a pilot in Far North Queensland in 2023 to demonstrate small scale plastic pyrolysis.
- **Brightmark** operates a commercial-scale plastics-to-fuel plant in Indiana, USA, which converts mixed plastic waste into fuel and other products. Their plant in the Parkes Special Activation Precinct, NSW is expected to be in operation in 2025.

International companies **Plastic Energy,** which uses a Thermal Anaerobic Conversion (TAC) process, and **Axens,** which has developed the Rewind®Mix petrochemical purification process, are supporting the Qenos Circular Plastics Project (QCCP) for circular polyethylene production in Australia through Australian operator **Qenos** and partners in Altona, Victoria.

IUCN

Primary Microplastics in the Oceans:

a Global Evaluation of Sources

Authors: Julien Boucher, Damien Friot

INTERNATIONAL UNION FOR CONSERVATION OF NATURE

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Authors: Julien Boucher, Damien Friot

Published by: IUCN, Gland, Switzerland

Citation: Boucher, J. and Friot D. (2017). *Primary Microplastics in the Oceans: A Global Evaluation of Sources*. Gland, Switzerland: IUCN. 43pp.

ISBN: 978-2-8317-1827-9

DOI: dx.doi.org/10.2305/IUCN.CH.2017.01.en

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TABLE OF CONTENTS

FOREWORD

The invention of plastic based on a synthetic polymer in 1907 changed our lives forever – for better and for worse. Plastic is one of the most versatile materials ever produced and has revolutionised the way we package, eat, travel and dress.

The plastic revolution has however come at a cost. Plastic pollution in our marine environment is taking place on a staggering scale with 9.5 million tonnes of new plastic waste flowing into the ocean each year. This is impacting our planet's precious biodiversity and damaging the fragile ecosystems upon which we all depend. The widespread contamination of our oceans is also fast becoming a worldwide human health risk as plastic enters our food and water supplies.

Despite a growing body of work on plastic debris and a heightened global awareness of its global impact, remedial efforts to combat this pollution have been hamstrung by a lack of research and knowledge surrounding the original sources of the waste matter.

Primary Microplastics in the Oceans : a Global Evaluation of Sources helps fill this knowledge gap and provides an important reference point for decision makers as they design and implement the transformative policies and production practices we need to transition towards a circular global economy.

Following in the footsteps of the IUCN "Close the plastic tap" project and the "Plastic debris in the ocean" publication, this report provides a global estimate and mapping of the sources and quantities of primary microplastics – plastics that enter the oceans in the form of small particles released from household and industrial products.

Although mismanaged plastic waste is still the main source of marine plastic pollution globally, this report shows for the first time that, in some countries, more plastic may be released from our driving and washing activities than from the mismanagement of our waste.

Tackling the multitude of sources of marine plastics requires a holistic approach that addresses the problem at its source. The findings in this report must drive new thinking around the way we design, produce, consume and dispose of plastics. Action to turn off the plastic tap could come in the form of new engineered materials and smart design, such as clothes that shed fewer fibres or washing machines equipped with filters. These efforts must be supported by legislation and on-the-ground policies that force real change.

This report also reminds us that, as consumers of plastic, we have a responsibility to educate ourselves and adapt our behaviour in order to protect our blue planet.

We have a major challenge ahead. But with a collaborative global effort, we can reverse the grim forecasts for plastic pollution, and return to a world with healthy oceans.

Inger Andersen,
IUCN Director General

This report owes its existence to the outstanding support from *MAVA Fondation pour la Nature*. The *Swedish Postcode Foundation* and the *Gallifrey Foundation* are also acknowledged for supporting the launch of this report.

1. Abstract

Plastic has penetrated everyday life: from clothing to coatings and from transport vehicles to cleaning products. Plastic is cheap, durable, lightweight and malleable, resulting in a practically unlimited number of possible applications. The disadvantages of plastics however are becoming more and more visible. Large quantities of plastics leak into rivers and oceans, with adverse effects to marine ecosystems and related economic activities.

Plastic wastes include all size residues, from large visible and easily removable items, to small invisible particles. This report investigates the sources of primary microplastics i.e. microplastics that are directly released into the environment as small plastic particles (< 5 mm size). This contrasts with secondary microplastics that originate mostly from the degradation of large plastic waste into smaller plastic fragments once exposed to the marine environment. Primary microplastics can be a voluntary addition to products such as scrubbing agents in personal care products (shower gels, creams, etc.). They can also originate from the abrasion of large plastic objects during manufacturing use or maintenance such as the erosion of tyres when driving or the abrasion of synthetic textiles during washing.

This report is one of the first of its kind to quantify primary microplastics leakage and to demonstrate that these primary microplastics are globally responsible for a major source of plastics in the oceans. The model developed for this analysis enables us to conclude that between 15 and 31% of all of the plastic in the oceans could originate from primary sources. This is a significant but as-of-yet unrecognised proportion. In some countries benefitting from advanced waste treatment facilities, primary microplastics releases even outweigh that of secondary microplastics.

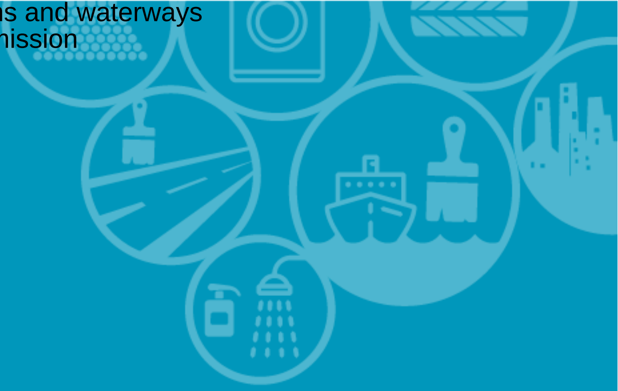
The global release of primary microplastics into the ocean was estimated at 1.5 million tons per year (Mtons/year). The estimate ranges between 0.8 and 2.5 Mtons/year according to an optimistic or pessimistic scenario. The global figure corresponds to a world equivalent per capita release of 212 grams or the equivalent of one empty conventional plastic grocery bag thrown into the ocean per person/per week worldwide.

The overwhelming majority of the losses of primary microplastics (98%) are generated from land-based activities. Only 2% is generated from activities at sea. The largest proportion of these particles stem from the laundering of synthetic textiles and from the abrasion of tyres while driving. Most of the releases to the oceans are occurring from the use of products (49%) or the maintenance of products (28%). The main pathways of these plastics into the ocean are through road runoff (66%), wastewater treatment systems (25%) and wind transfer (7%).

The study reviewed seven regions – Africa and Middle East, China, East Asia and Oceania, Europe and Central Asia, India and South Asia, North America, and South America. It revealed comparable releases per region in absolute value – ranging from 134 to 281 Ktons/year. The per capita releases, however, are very different between regions – ranging from 110 to 750 grams/person/year. Further, most regions are expected to have increased releases of primary microplastics in the next decades. This is due to improvements in per capita income without improvements in systems to prevent the releases.

Importantly this report is based on modelling sources and leakages from economic and household activities, using exclusively publicly available data and not on field measurements. The model could be further improved by using available fee-based proprietary data on regional plastic quantities. It could also be further strengthened by improving underlying regional assumptions on behaviours. Furthermore, confrontation of our predictive model with empirical data from the field would be beneficial in order to validate the model. This is however not feasible yet, given the status of literature and lack of adequate experimental set-up to perform this comparison. Nevertheless, the range of pessimistic/optimistic scenarios considered throughout our study allows for sufficient confidence in the orders of magnitude we put forward.

This report stresses the contribution of primary microplastics to the plastic pollution of oceans on a global scale. It opens the way to a new stream of actions to tackle the issue of plastics in the ocean beyond the traditional focus on waste management. Shaping these solutions will require approaches based on product eco-design and lifecycle thinking, as well as the involvement of key stakeholders from the private (e.g. textile and automotive industry) and public (water treatment and urban infrastructure planning) sectors.



2.

Introduction

Nowadays plastics have penetrated all aspect of everyday life from clothing to coatings and from transport vehicles to cleaning products. Plastic is a cheap, durable, lightweight, malleable material. It has a practically unlimited number of possible applications.

The disadvantages of plastics however are becoming more and more visible. Large quantities of plastics are released into rivers and oceans with various adverse effects to ecosystems and related economic activities.

Littering and mismanaged wastes are often referred to as the main source of plastics entering the ocean. However behind the headlines primary microplastics are becoming another major source of concern. Their release is much less visible, resulting from the voluntary addition of microbeads in products such as cosmetics or from the abrasion of larger plastic items such as textiles or tyres.

The purpose of this report is to provide one of the first global estimate and mapping of sources and quantities of primary microplastics released into the world ocean. It aims to provide new information to decision-makers and stakeholders to help them shape actions to close the plastic tap.

2.1 Plastics & microplastics contaminate the world ocean

Marine environments all over the world are contaminated with plastics (GESAMP, 2015). Plastics can be encountered in two forms: large plastic wastes, and small plastic particulates below 5 mm in size named microplastics (Thevenon et al., 2014).

Recent studies place the total amount of plastic produced since its invention at 8.3 billion tons (Geyer et al., 2017). Of this, an estimated 9% has been recycled (Geyer et al., 2017). Between 4.8 to 12.7 million metric tons of plastic waste are estimated to enter the ocean each year (Jambeck et al., 2015). Given these recent estimations and attempts to accurately quantify the problem, it is vital to understand the relationship between macro and microplastics when citing these numbers.

The estimate of between 4.8 to 12.7 million MT of plastic entering the ocean each year is an estimation of macroplastics (Jambeck et al., 2015). This is based on the mass of waste generated per capita annually, the percentage of plastic waste within that, and finally, the percentage of mismanaged plastic waste that has the potential to enter the ocean as plastic pollution. Therefore, the figures discussed in this report about primary microplastics are *in addition* to the estimations made in the Jambeck report, putting the overall quantity of both micro and macroplastics in the ocean at higher than the average and commonly-quoted value of 8 million metric tons.

Large plastic waste is readily visible. Studies have demonstrated negative social, economic and ecological impacts. These range from the ingestion, injury, entanglement or suffocation of wildlife to economic drawbacks for tourist areas and maritime industries (GESAMP, 2015; Raynaud, 2014). For a review of the many potential negative impacts of plastics on ecosystems, see Thevenon et al. (2014).

Microplastics are not easily visible to the human eye. While potential negative impacts are less obvious, their release into the oceans may also have far reaching consequences. Human health concerns are suspected through the accumulation of microplastics in the food chain and/or sorption of toxicants to plastic while traveling through the environment (Eriksen et al., 2014).

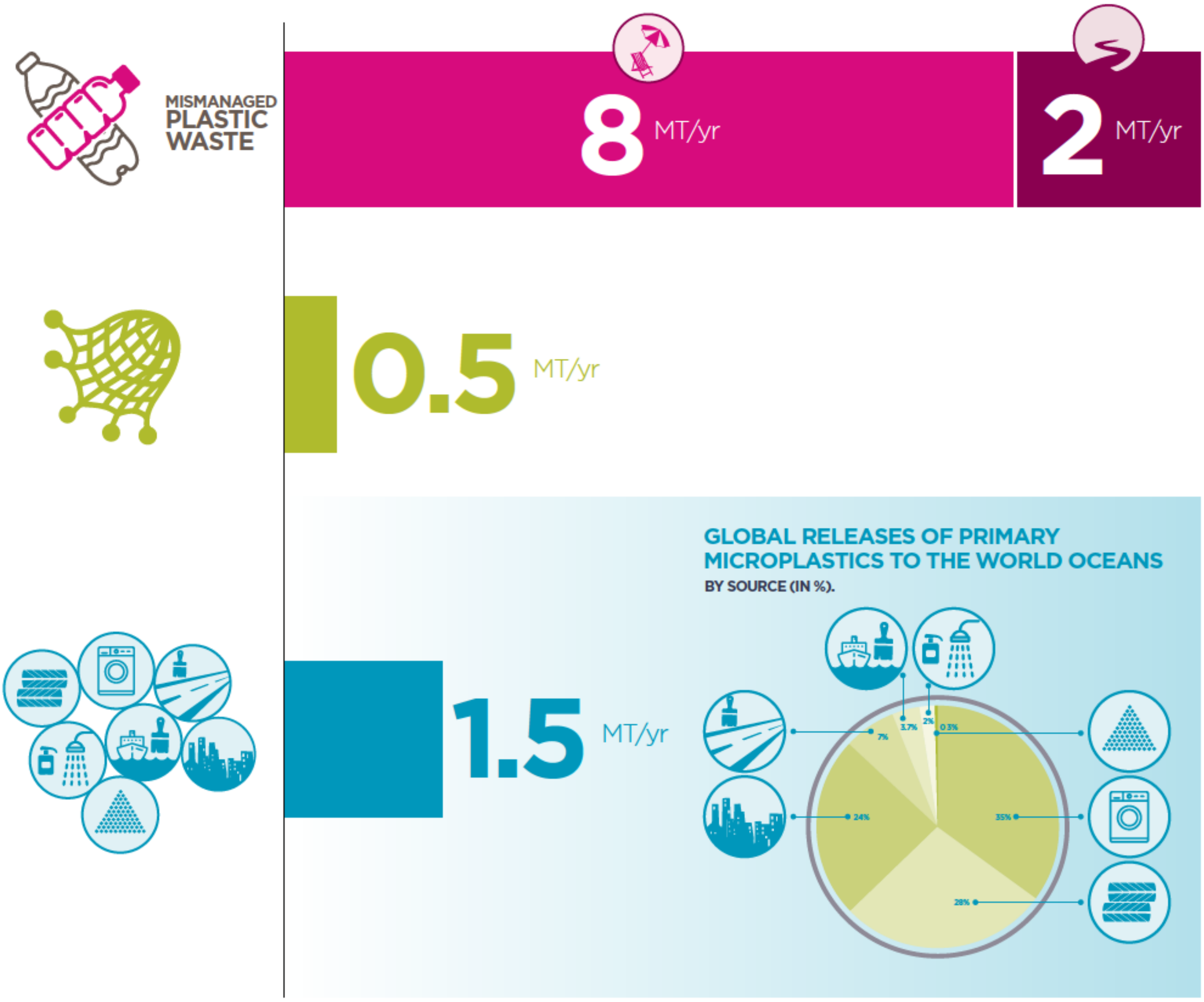
Two types of microplastics are contaminating the world ocean: primary and secondary microplastics. Different definitions have been used in the literature (Lassen et al., 2015) and we adopted the following as proposed by a Norwegian study (Sundt et al., 2014):

- **Primary microplastics** are plastics directly released into the environment in the form of small particulates. They can be a voluntary addition to products such as scrubbing agents in toiletries and cosmetics (e.g. shower gels). They can also originate from the abrasion of large plastic objects during manufacturing, use or maintenance such as the erosion of tyres when driving or of the abrasion of synthetic textiles during washing.

- **Secondary microplastics** are microplastics originating from the degradation of larger plastic items into smaller plastic fragments once exposed to marine environment. This happens through photodegradation and other weathering processes of mismanaged waste such as discarded plastic bags or from unintentional losses such as fishing nets. Given that the origins of secondary microplastics are difficult to trace given their degradation, it is difficult to meaningfully assess how much of the figures of macroplastics have now converted to microplastics. It is for this reason that the report seeks to focus on quantification of primary microplastics, as it is achievable within current data sets.

Once in the oceans, microplastics can either float or sink. Microplastics lighter than seawater such as polypropylene will float and disperse widely across the oceans. They eventually accumulate in gyres resulting from oceanic currents. Eriksen et al. (2014) and Sebille et al. (2015) estimate that 93 to 268 ktons of these microplastics are currently floating in the oceans. Other microplastics such as acrylic are denser than seawater and most probably accumulate on the ocean floor, which means that a significant amount of microplastics may eventually accumulate in the deep sea (Woodall et al., 2014) and ultimately in food chains (Seltenrich, 2015).

This ubiquitous contamination of oceans by microplastics is becoming a major concern. Given the magnitude and uniqueness of this global ocean contamination, some refer to the current period as the *plasticene* (Reed, 2016) and describe the world's ocean as a *plastic soup*[1].



2.2 Knowledge gaps about microplastics releases

There is a common belief that most of the plastics contaminating the world's ocean originate from mismanaged plastic wastes. Most recent reports on microplastics focus almost exclusively on the quantification of these secondary sources and on waste reduction and management (EUNOMIA, 2016a; Jambeck et al., 2015; McKinsey & Company and Ocean Conservancy, 2015; World Economic Forum et al., 2016). Jambeck (2015) reported that between 4.8 and 12.7 Mtons of plastic are released globally into the oceans every year because of mismanaged waste.

Knowledge about the global releases of primary microplastics into the world ocean is howeve rgenerally lacking. While many sources of primary microplastics have been identified in the literature, the global quantities released yearly have not yet been assessed. Consequently, the relative importance of primary versus secondary sources of microplastics is still unknown, and the plastic debate generally does not look outside of the waste management arena.

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[1] See: http://www.plasticsoupfoundation.org

The discovery of high levels of microplastic in the lakes and rivers in Europe, North-America and Asia (Eerkes-Medrano et al., 2015) might indicate that primary sources represent a significant release of microplastics into the oceans. One study showed that plastic abundance in the Austrian Danube was even higher than that of drifting larval fish, mostly in the form of industrial raw materials such a pellets and flakes (Lechner et al., 2014). Two other studies found that much of the plastic found in surface water originates from cosmetic products such as facial cleaners or from textiles (Browne et al., 2011; Driedger et al., 2015). In the recent assessments performed at country scale in Europe, Essel et al. (2015), Lassen et al. (2015), Magnuson et al. (2016), RIVM (2014), Sundt et al. (2014) identified and quantified around fifteen sources of primary microplastics. Tyres are often cited as the main contributor to the releases.

2.3 Rapidly increasing use of plastics

Today plastic is a common material that can be found in almost all parts of everyday life. This includes packaging, buildings and construction, vehicles, electrical and electronic equipment, agriculture production, clothes and footwear, householder and personal cleaning products. A practically unlimited number of applications are possible thanks to its unrivalled properties of durability, malleability, lightweight and low cost.

Plastic use has increased exponentially since synthetic organic polymers were developed in the mid-20th century. Over 300 million tons of plastic are currently produced yearly to manufacture objects in plastic. This quantity contrasts with only 1.5 million tons produced in 1950. The long-term average annual growth rate has been roughly 4% (PlasticsEurope, 2015). We can then add the plastics for other uses that are not accounted in these statistics[2] such as synthetic fibres for textiles (37.2 million tons) or synthetic rubber for tyres (6.4 million tons)[3]. Also, according to PlasticsEurope (2015), plastic production requires around 4% of the world's annual petroleum production while a similar amount of petroleum is used to provide the energy for plastic manufacturing.

Plastic use varies widely across regions. In North America and Western Europe, for example, the average plastic consumption *per capita* for plastics objects reached approximately 100 kg per year in 2005 and was expected to increase to 140 kg by 2015. In Asian countries, the individual consumption for plastics objects is much lower. It was about 20 kg per year per person in 2005 with an estimated increase to 36 kg by 2015. It is even lower in Africa with an estimation of 16 kilos *per capita* by 2015 (PlasticsEurope, 2009).

Due to the excellent functional properties of this material, it seems clear that plastic use will increase in the future, particularly in lower income regions as their economies grow. Solutions are thus required to close the plastic tap and to reduce the detrimental effects of plastic use on the world environment and potentially human health.

2.4 Objective of this report

The objective of this report is to provide one of the first global quantitative assessments of the direct releases of primary microplastics of petrochemical origin into the world's ocean. This report seeks to contribute to a better identification and prioritisation of the sources and pathways of microplastic pollution. With this information, decision-makers and key stakeholders can make informed decisions and undertake targeted actions to address the problem.

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[2] Other uses than plastic products are not accounted by plastic branch associations (personal communication from Plastics Europe).

[3] References are provided in the methodological appendix.

3.

Description of issues & methodology

Seven major sources of primary microplastics are identified and evaluated in this report: Tyres, Synthetic Textiles, Marine Coatings, Road Markings, Personal Care Products, Plastic Pellets and City Dust. Losses in the environment and releases to the world ocean are quanitifed and presented in the following three scenarios: optimistic, central and pessimistic, and for 7 geographic regions.

3.1 Literature review of key sources

Plastics are used in many activities performed by businesses and by households on land or at sea. The main known primary microplastic sources are reported and classified in Table 1, based on data recently published from Denmark (Lassen et al., 2015), Sweden (Magnuson et al., 2016), Norway (Sundt et al., 2014), and Germany (Essel et al., 2015).

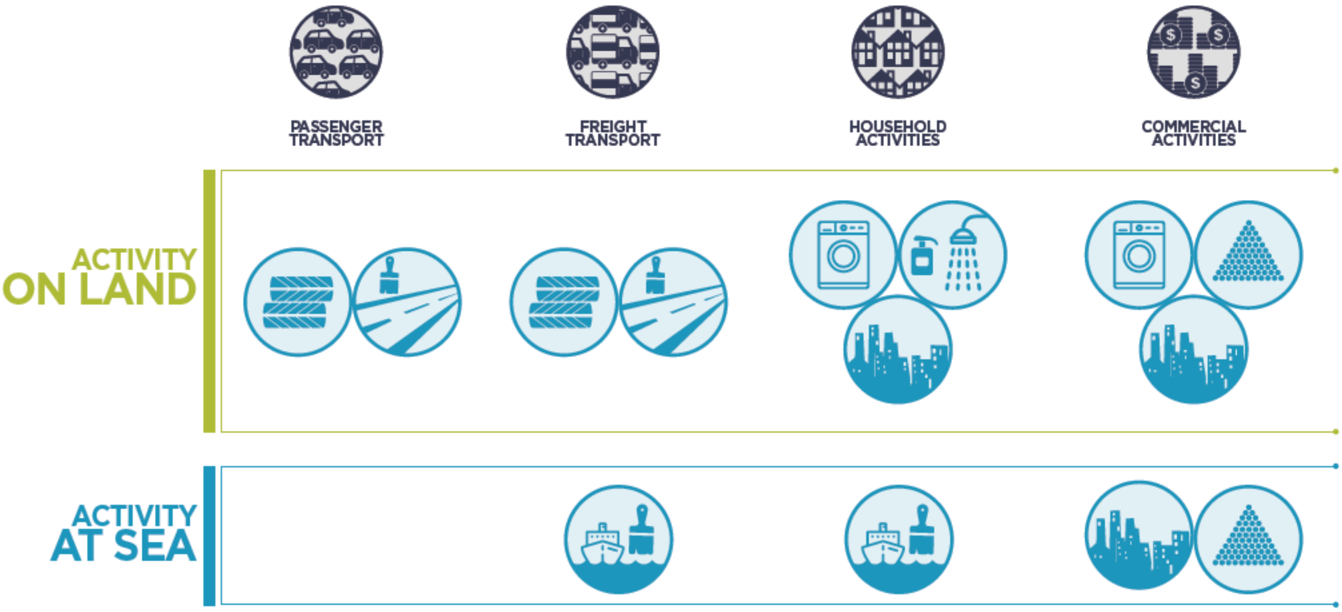
In our study losses and releases of primary microplastics are quantified at global scale for the seven sources identified as dominant in preceding studies. These are as follows:

These different sources stem from different household or commercial activities both on land and at sea, as presented in Table 1.

Table 1

MAIN SOURCES OF PRIMARY MICROPLASTICS

IDENTIFIED IN THE LITERATURE



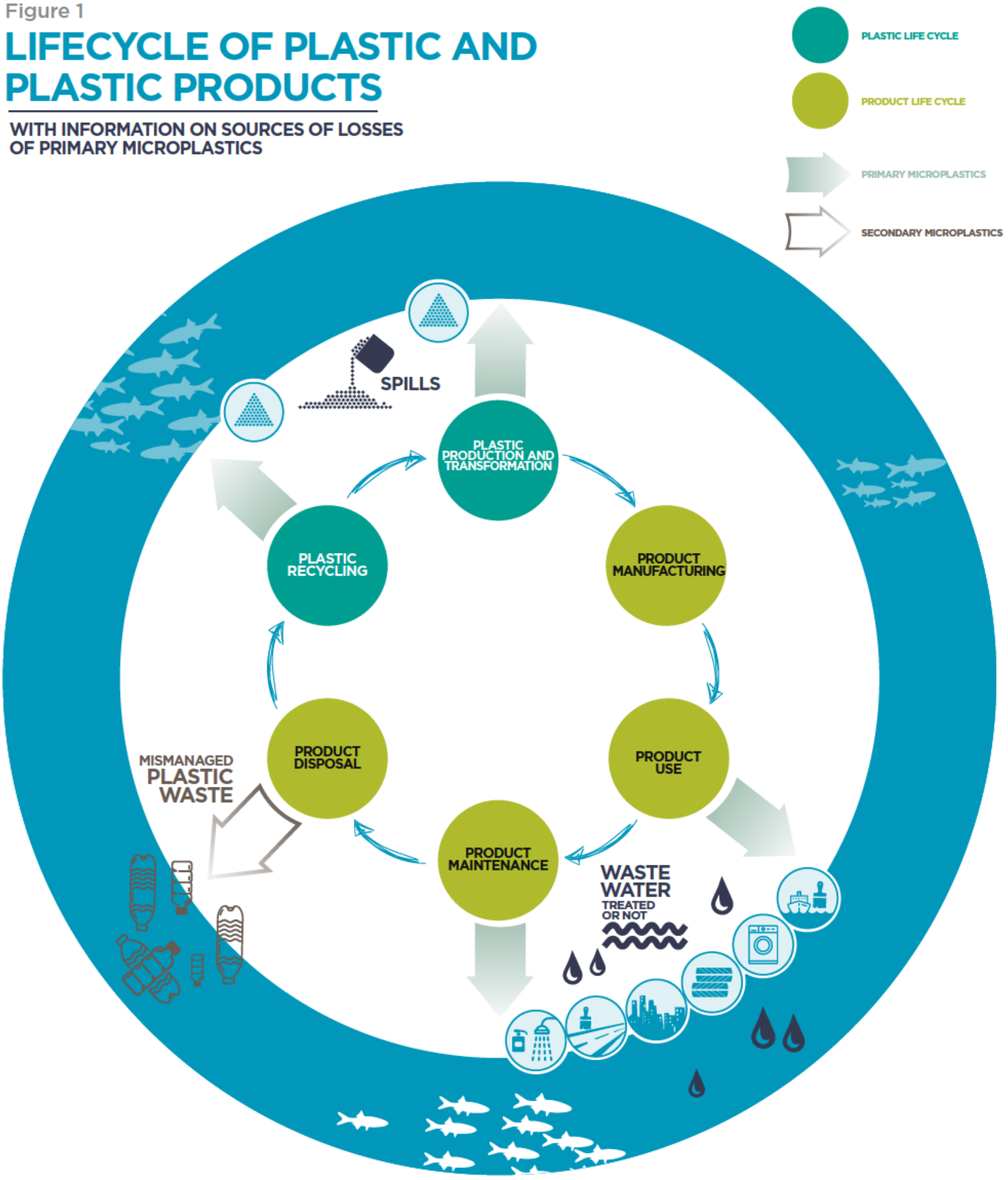
3.2 A lifecycle perspective of losses

Losses of primary microplastics occur at various stages of the lifecycle of plastic and plastic products (Figure 1). Plastic pellets are the only losses occurring during the production, transport or recycling stages of plastics. Most losses mainly occur during the use phase of products containing plastic such as driving a car or during their maintenance such as washing clothes. This contrasts with secondary microplastics that mostly originate from mismanaged waste during the disposal of products containing plastics.

Figure 1

LIFECYCLE OF PLASTIC AND PLASTIC PRODUCTS

WITH INFORMATION ON SOURCES OF LOSSES OF PRIMARY MICROPLASTICS



3.3 Description of seven key sources

The microplastic sources considered in this report stem from a global consumption of plastics of just over 300 Mtons, as presented in Table 2. Main uses are for plastic products which start their life as pellets (85%), for synthetic textiles (12%), and for synthetic rubber in tyres (2%).

Losses from Personal Care Products are the only losses that can be considered as *intentional losses*. The former is intentional because a product containing primary microplastics is poured on purpose into wastewater. By contrast other sources generate *unintentional losses* through abrasion, weathering or unintentional spills during production, transport, use, maintenance or recycling of products containing plastic.

Table 2

CONSIDERED SOURCES

YEARLY WORLD CONSUMPTION AND TYPE OF LOSS

SOURCES	WORLD CONSUMPTION KTONS / YEAR OF PLASTIC	INTENTIONAL LOSS	REFERENCES
PLASTIC PELLETS	257,000	NO	Plastics Europe (2007)
SYNTHETIC TEXTILES	42,534	NO	FAO/ICAC (2013)
TYRES	6,431	NO	ETRma (2010)
ROAD MARKINGS	588	NO	Grand View Research, Inc. (2016)
MARINE COATINGS	452	NO	Coatings world (2012)
PERSONAL CARE PRODUCTS	42	YES	Leslie, H.A. (2015)

1. Plastic Pellets: incidents during manufacturing, transport and recycling

In their primary form, many plastics are in the form of pellets – typically 2-5 mm in diameter – or powders. Pellets are transported to plastic transformers that generate the plastic products. During manufacturing, processing, transport and recycling, pellets can be spilled into the environment through small or large incidents along the whole plastic value chain (Essel et al., 2015). Many field studies are reporting the occurrence of plastic pellets in the environment. These are also known as nibs, nurdles or mermaid tears (Sundt et al., 2014).

Type of loss: unintentional
Lifecycle stage: primary plastic production, primary plastic transport, plastic recycling

2. Synthetic Textiles: abrasion during laundry

Washing synthetic textiles, in industrial laundries and households creates primary microplastics through abrasion and shedding of fibres. Fibres are then discharged in sewage water (Browne et al., 2011) and potentially end up in the ocean (Magnuson et al., 2016).

Significant amounts of these textile fibres have been observed in many *in situ* sampling studies both in open water and marine sediments (Browne et al., 2011). These fibres are typically made of polyester, polyethylene, acrylic or elastane (Essel et al., 2015).

Type of loss: unintentional
Lifecycle stage: product maintenance

3. Tyres: abrasion while driving

Tyres get eroded when used. The particles are formed from the outer parts of the tyre and consist of a matrix of synthetic polymers, namely Styrene Butadiene Rubber (approximately 60%), in a mix with natural rubber and many other additives (Sundt et al., 2014). Tyre dust will then either be spread by the wind or washed off the road by rain. In this study, losses of synthetic rubber are considered but losses of natural rubber are not.

There is no reliable information on the transfer of microplastics from tyres to the world's oceans. Both Norwegian and Swedish researchers have pointed out that a large fraction of particles found in the sea seem to originate from car tyres (Essel et al., 2015; Sundt et al., 2014).

Type of loss: unintentional
Lifecycle stage: product use

4. Road Markings: weathering and abrasion by vehicles

Road markings are applied during the development of road infrastructure and its maintenance. Different types of markings (paint, thermoplastic, preformed polymer tape and epoxy) are applied, with a global dominance of paint (45%) (Grand View Research, Inc., 2016). In most European countries, thermoplastics are the most commonly used material (Lassen et al., 2015).

Loss of microplastics may result from weathering or from abrasion by vehicles. As for tyres dust will either be spread by wind or washed off the roads by rain before reaching surface waters and potentially the oceans.

Type of loss: unintentional
Lifecycle stage: product use

5. Marine Coatings: weathering and incidents during application, maintenance and disposal

Marine coatings are applied to all parts of vessels for protection including the hull, the superstructure and on-deck equipment. They include solid coatings, anticorrosive paint or antifouling paint. Several types of plastics are used for marine coatings including mostly polyurethane and epoxy coatings and also vinyl and lacquers (OECD Series on emissions documents, 2009).

Primary microplastics are released from commercial and leisure boats during building, maintenance, repair or use. The key activities that may lead to releases are surface pre-treatment, coating application and equipment cleaning (OECD Series on emissions documents, 2009).

Type of loss: unintentional
Lifecycle stage: product use, maintenance and disposal

6. Personal Care Products: pouring during product use

Plastic microbeads are used as ingredients in personal care and cosmetic products for a variety of purposes such as sorbent phase for delivery of active ingredients, exfoliation or viscosity. Some products contain as much plastic added as ingredients as the plastic in which they are packaged (Leslie, 2015). These represent up to 10% of the product weight and several thousand microbeads per gram of product (Lassen et al., 2015).

Classical use of personal care products results in the direct introduction of the plastic particles into wastewater streams from households, hotels, hospitals, and sport facilities including beaches. Microbeads from cosmetics have been observed in field studies in different areas of the world (Driedger et al., 2015).

Type of loss: intentional
Lifecycle stage: product use

7. City Dust: weathering, abrasion and pouring

City Dust is the generic name given to a group of nine sources identified in recent country assessments, that are most often occurring in urban environments (Essel et al., 2015; Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014). City Dust includes losses from the abrasion of objects (synthetic soles of footwear, synthetic cooking utensils), the abrasion of infrastructure (household dust, city dust, artificial turfs, harbours and marina, building coating) as well as from the blasting of abrasives and intentional pouring (detergents). These sources are grouped together because their individual contribution is small. However together they account for a considerable amount of losses in the country studies. In contrast to other sources that have been subject to specific and regionalised modelling, city dust assessment is based on more basic extrapolation from the Nordic countries studies (Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014).

Type of loss: mainly unintentional but partly intentional
Lifecycle stage: product use or maintenance

3.4 Modelling activities, losses & releases

This report is based on modelling sources and leakages from economic and household activities, using exclusively publicly available data. This report is not based on field measurements.

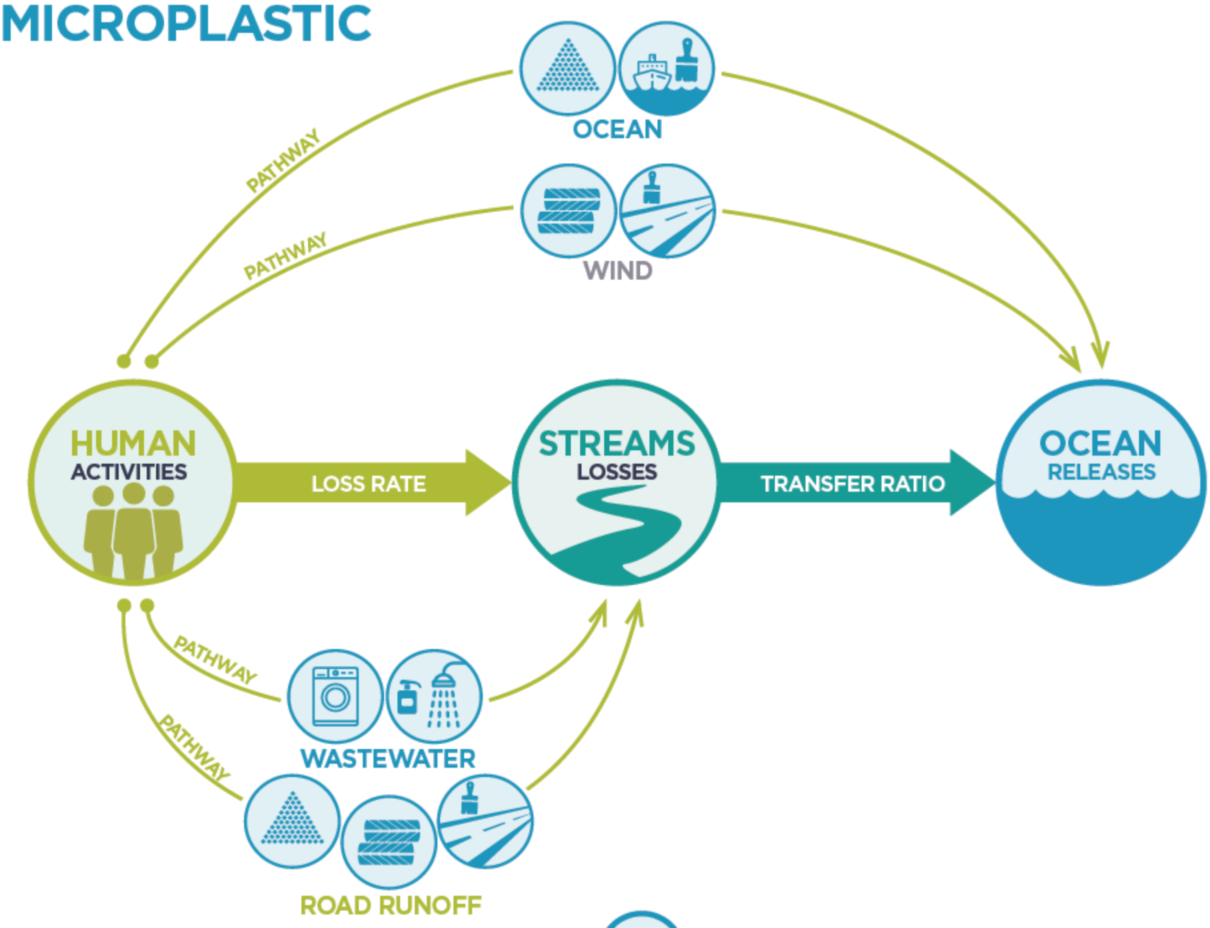
A general description of the approach applied to compute activities, losses and releases is presented in Figure 2. The detailed approach and references are presented in the methodological appendix of this report.

For each activity, the quantity of plastic used has been first computed at global and regional scales. Then the quantity of microplastics entering the environment – the "losses" – has been quantified by applying loss rates specific to each activity and region. In a final step, the fraction of the loss ultimately released into the oceans – the "releases" – has been computed applying appropriate transfer ratios. Transfer ratios are based on four different pathways presented below, and are also specific for different activities and regions:

- Ocean: when losses occur in the ocean (Plastic Pellets, Marine Coatings), 100% of the losses become releases.
- Wastewater: when losses are to wastewater streams (Synthetic Textiles, Personal Care Products) the transfer ratio depends on the regional coverage and efficiency of the wastewater treatment system.
- Road runoff: when losses are on roads (Plastic Pellets, Tyres, Road Markings), a share of the loss is transferred by wind while the other goes through road runoff. In the last case, the transfer ratio depends on the regional shares of roads connected to a separate sewer and to a combined sewer (hence treated in the wastewater treatment system).
- Wind: once the microplastics have been lost, they may be released into the oceans by wind.

Figure 2:

GENERAL DESCRIPTION OF THE MODELLING OF ACTIVITIES, LOSSES AND RELEASES OF PRIMARY MICROPLASTIC



The computations of activities, losses and releases have been performed in an iterative way. In a first step, the global relevance of each of the sources has been estimated at a regional scale. For all sources except City Dust, the estimation is based on the multiplication of generic loss and transfer ratios with regional populations and GDP (Gross Domestic Product).

For City Dust a global value has been estimated for losses based on existing quantified country studies (Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014). This global value has then been distributed to each region proportionally to previously computed losses. Different approaches for modelling have been tested and compared as described in Appendix 1, and allowed for testing the robustness of our model.

In a second step refined computations have been performed applying more detailed data and at the country level when available. A larger set of assumptions has also been tested to generate minimum and maximum bounds for the results using two complementary approaches: a first one based on yearly activities and a second one based on a lifecycle perspective.

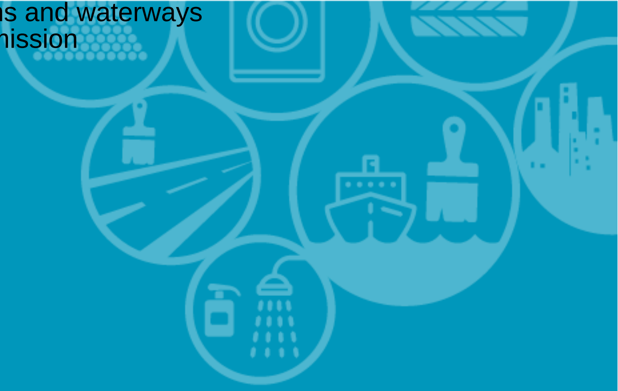
3.5 Geographic regions considered

The grouping of countries has been done according to a classification in 7 regions: Africa and Middle East, China, East Asia and Oceania, Europe and Central Asia, India and South Asia, North America, and South America, as detailed in Appendix 3.

3.6 Three scenarios for presenting the results

In this report, the results are presented using three scenarios – optimistic, central and pessimistic – for each of the seven sources both at global and regional scales.

These scenarios correspond to the most credible set of results based on the application of an extended set of assumptions during the study (cf. Appendix 1). The optimistic and pessimistic scenarios are built by selecting the minimum and maximum results after getting rid of extreme and unrealistic combinations of assumptions. The central scenario corresponds to a central value based on the distribution of remaining results.



4.

Results & Analysis

The global release of primary microplastics in the world ocean was evaluated in the order of 1.5 Mtons/year. i.e. between 0.8 and 2.5 Mtons/year. This global figure corresponds to a world equivalent per capita of 212 g or a plastic quantity equivalent to every human tossing one conventional light grocery plastic bag per week into the ocean.

From these results we conclude that between 15% and 31% of all plastic in oceans could originate from primary sources. This is a significant but as-of-yet unrecognised proportion. In higher income countries benefiting from adequate waste treatment facilities, the primary microplastics represent the main contribution to plastic release into the oceans.

The overwhelming majority (98%) of the losses of primary microplastics are generated from land-based activities. Only 2% is generated from activities at sea. The largest proportion of these particles stem from the laundering of synthetic textiles and from the abrasion of tyres while driving.

4.1 Primary microplastics are a significant source of plastic in the oceans

We estimate the following scenarios of releases into the oceans:

- Optimistic – 0.8 Mtons/year
- Central – 1.5 Mtons/year
- Pessimistic – 2.5 Mtons/year

These releases are a fraction of the estimate losses of primary microplastics into the environment presented below:

- Optimistic – 1.8 Mtons/year
- Central – 3.2 Mtons/year
- Pessimistic – 5.0 Mtons/year

The gap between losses and releases under the central scenario means that around 48% of the losses of primary microplastics from activities are released into the world ocean. The remaining primary microplastics are presumably trapped in soil, or sewage sludge. Their fate and effect over time is unknown and depends on conditions and practices in each region.

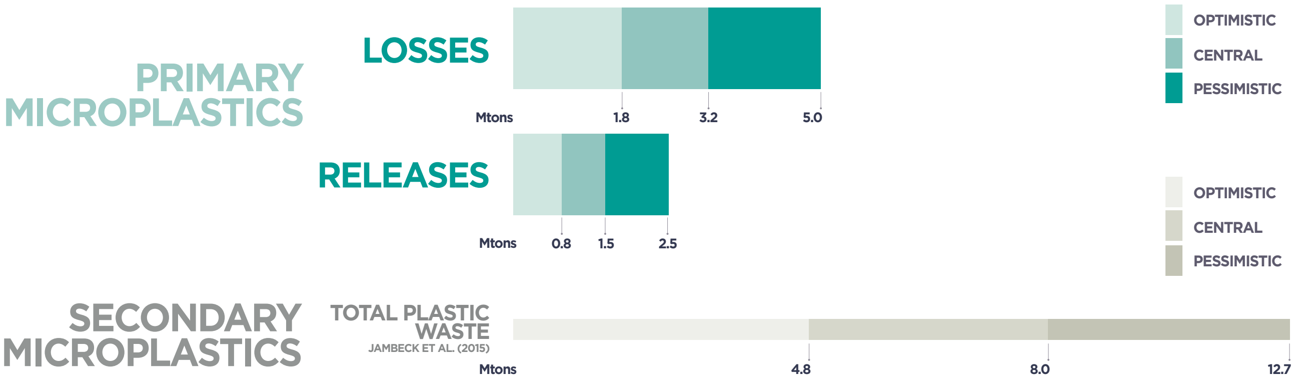
Comparing these releases of primary microplastics to known sources of secondary microplastics – i.e. from plastic waste and from lost fishing nets – indicates that the contribution of primary sources might be significant. The contribution of primary microplastics largely outweighs that of lost fishing nets[4] (0.6 Mtons/year) (Circularocean, 2015; Macfadyen et al., 2009). Considering values from Jambeck (2015), the potential releases from plastic waste range from 4.8 Mtons/years to 12.7 Mtons/year with an average value of 8.0 Mtons/year. This means that between 15% and 31% of the microplastics could be from a primary source, comparing central values in the first case and the pessimistic value from this report with the lowest bound for waste in the second one.

We have decided throughout the study to report microplastic losses and releases exclusively for microplastic from petrochemical origin. Plastics such as natural rubber are not accounted for. Extending the definition and assuming, as in some studies for Europe (Essel et al., 2015; Lassen et al., 2015; Magnuson et al., 2016), that natural rubber is also a concern for the world ocean, global releases from primary microplastics would increase by 45%, 33% and 26% respectively for the three scenarios. The resulting central value for releases would thus be 2.0 Mtons/year and the potential contribution of primary microplastics sources could be up to 37%.

Figure 3

GLOBAL RELEASES OF MICROPLASTICS TO THE WORLD OCEANS:

PRIMARY AND SECONDARY SOURCES (IN KTONS)



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[4] This value is the only one available but has not been scientifically validated. It should thus be taken with caution.

Our results are consistent with a recent research performed by Eunomia applying a very basic extrapolation of European releases of primary microplastics (from a former proprietary Eunomia report) scaled to a global level using GDP *per capita* in Purchasing Power Parity. Eunomia estimates that primary microplastics releases are between 0.5 and 1.41 million tons/year with a central value of 0.95 million tons/year annually (EUNOMIA, 2016b).

4.2 Two-thirds of the releases are from the erosion of synthetic textiles & tyres

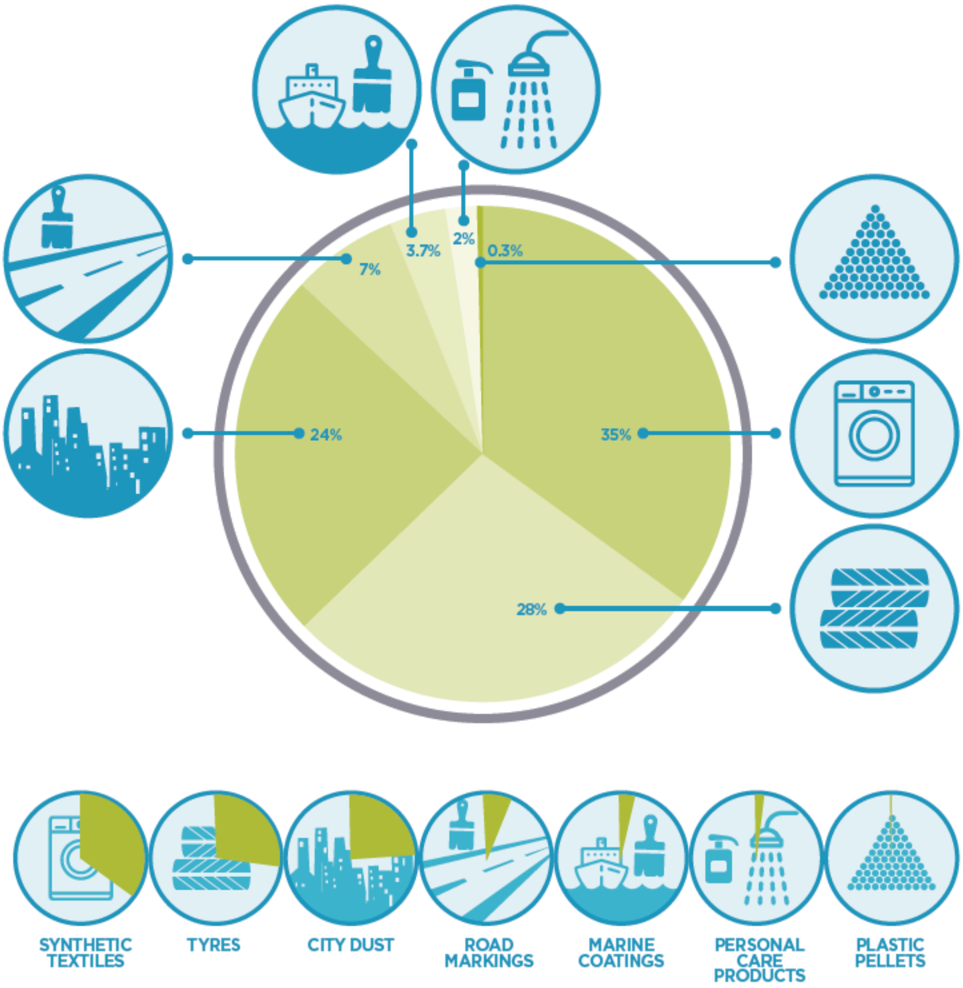
As shown in Figure 4 for the central scenario, close to two-thirds (63.1%) of the releases are due to first the laundry of synthetic textiles (34.8%), and second to the erosion of tyres while driving (28.3%). The order is the same in the pessimistic scenario. In the optimistic scenario the joint contribution is similar but tyres erosion dominates (38%). Note again that these results only consider synthetic rubber. Should natural rubber be considered the erosion of tyres would contribute to almost half of the releases (46.2%) in the central scenario.

The third important contribution (24.2%) is the source City Dust which has been computed with a simplistic approach. Further research should be performed on City dust to better understand the contributions per region. Personal care products only account for 2% of the global release of primary microplastics to the world ocean.

Figure 4

GLOBAL RELEASES OF PRIMARY MICROPLASTICS TO THE WORLD OCEANS

BY SOURCE (IN %).



Textiles

The yearly consumption of fibre for apparel amounts to 69.7 million tons globally – around 11.0 kg *per capita* (2010). Synthetic fibres represent almost two-thirds (60.1 %) of this consumption (FAO/ICAC, 2013).

The global yearly consumption of fibres for apparel has steadily increased over the last two decades (+79.3% between 1992 and 2010). This growth is almost exclusively due to the increased consumption of synthetic fibres with an increase close to 300% (from 16 to 42 million tons) over the same period (FAO/ICAC, 2013).

The majority (62.7%) of synthetic fibres are nowadays consumed in developing economies. In these economies, consumers buy a larger proportion (68.0%) of synthetic textiles than in developed economies (48.2%).

Averaging at 6.1 kg globally, the *per capita* consumption patterns vary largely between regions: going from 0.7 kg *per capita* in Africa and 5.7 kg in Asia, and up to 17.8 kg in North America, i.e. a 25 times range (FAO/ICAC, 2011).

Tyres

Around 1,412.6 million vehicles are in use globally – around 0.3 vehicle *per capita* (2010) (ETRma, 2011). More than two-thirds (71.2%) of these vehicles are personal cars and light commercial vehicles, 25.6% are motorbikes and 3.1% are medium and heavy commercial vehicles. This global vehicle count has steadily increased over the last fifteen years (78%) between 2000 and 2015. Most vehicles are in Asia (36%), with 13% in China, in Europe (27%) and in North America (21%) (ETRma, 2011).

To equip these vehicles with tyres, rubber is needed. Synthetic rubber in tyres represents around 57% of all synthetic rubber uses in 2010. The global consumption of synthetic rubber has increased by 27.3% between 2002 and 2010, mainly driven by China. This is a lower increase than the growth of the global consumption of natural rubber over the same period (+41.2%) (ETRma, 2009).

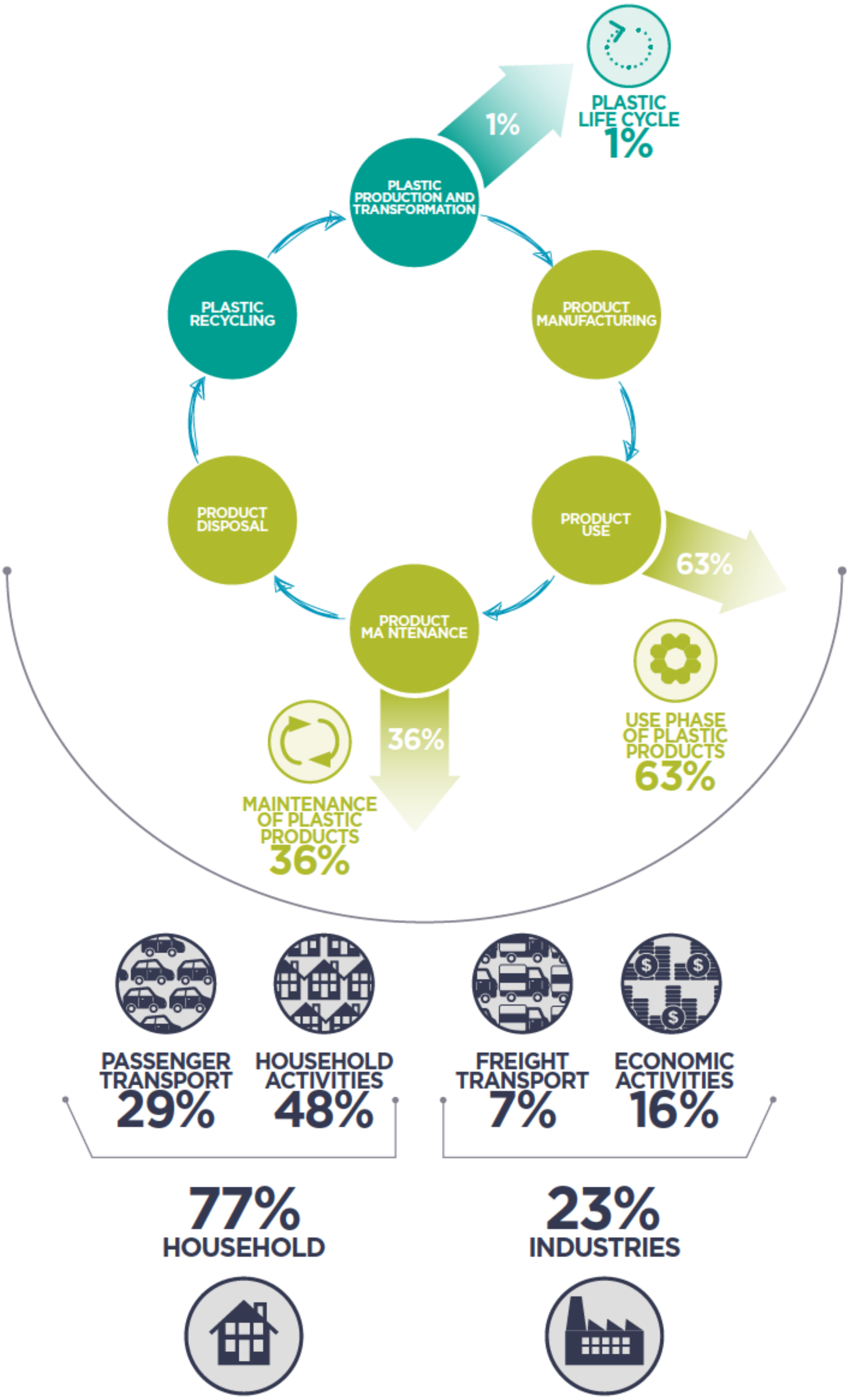
The yearly sales of rubber for tyres were around 13.9 Mtons in 2010, of which 46% (6.4 Mtons) was synthetic rubber (ETRma, 2011). Averaging at 0.93 kg of synthetic rubber globally, the *per capita* consumption patterns vary largely between regions.

4.3 Household activities generate almost three quarters of the releases during the use phase and maintenance of plastic products

Figure 5

GLOBAL RELEASES OF MICROPLASTICS TO THE WORLD OCEANS:

SPLIT PER TYPE OF ACTIVITIES AND LIFECYCLE STAGES



As shown in Figure 5 households generate directly through their activities about three quarters (77%) of the microplastics releases. The rest are generated by economic activities. Most of these household releases occur during the use phase of products (49%) and the rest (28%) during the maintenance of products. Transportation activities correspond to one third (38%) of the sources of households' releases (passenger transport)[5] due to tyres and road markings, i.e. mainly due to the use of personal cars. In Europe for example 90% of the total driven distance is with personal cars (EU JRC/NL PBL, 2010). The rest (62%) of the sources are related to other household activities – laundry of synthetic textiles, use of personal care products and leisure boats coatings.

Releases from economic activities are thus much lower (23%) than releases from households. Most of those are also occurring during the use phase of products and the share due to the plastic part of the lifecycle only accounts for around 0.5% to 1.5%. The share of releases due to commercial transport is similar but slightly lower (30%) than the share due to transportation by households. The road transportation system by itself is thus generating around one third (35.1%) of the total releases.

4.4 Two-thirds of the losses are through road runoffs

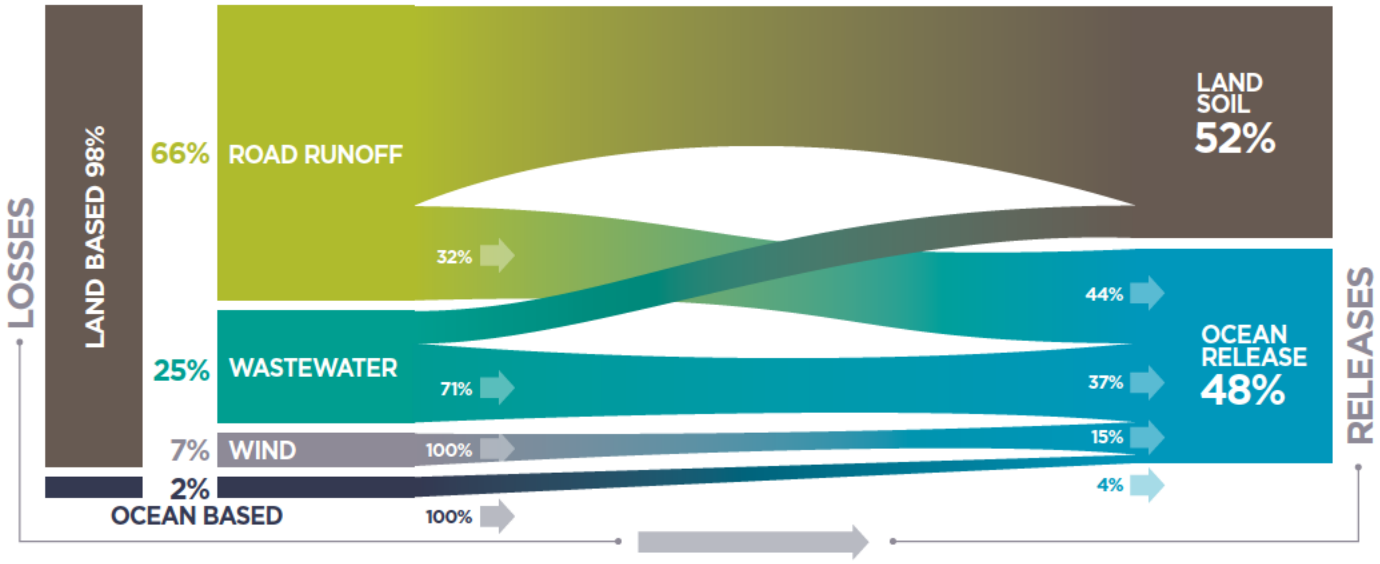
The overwhelming majority (98%) of the losses of primary microplastics are generated during land-based activities. As illustrated in Figure 6, the main pathway is road runoff (tyres, road markings and pellets incidents on land) (66%) followed by wastewater treatment systems (25%) and by wind transfer (7%). Marine activities only generate around 2% of the losses.

The distribution is slightly different in terms of releases. At the global level, around one third (29.5%) of the population is connected to a wastewater treatment system. Accounting for overflows, this means that for this pathway more than two-thirds (71%) of the microplastics are on average released to the oceans. For road-runoff, only 32% of the losses end up as releases. This concerns mainly the losses going through separate sewers. All losses occurring in the ocean and all losses transported by wind become releases[6]. Thus, 44% of the releases are along the road runoff pathway, 37% along the wastewater pathway, 15 % are transported by wind and 4% are direct releases to the oceans.

Figure 6

GLOBAL RELEASES TO THE WORLD OCEANS:

CONTRIBUTION OF DIFFERENT PATHWAYS TO THE RELEASE OF MICROPLASTICS



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[5] For tyres erosion and for road markings, the split of the global values between households and commercial activities is based on the global driven distance per type of vehicle.

[6] The proportion of particules transported by wind to soil is not known.

4.5 All regions contribute significantly to releases

The regional distribution of releases of primary microplastics, regional GDP and population, are presented in Figure 7. All regions contribute significantly to the releases. The largest regional releases are in India and South Asia (18.3%) and North America (17.2%), followed closely by Europe and Central Asia (15.9%), China (15.8%) and East Asia and Oceania (15.0%). Releases are lower in South America (9.1%) and Africa and the Middle East (8.7%).

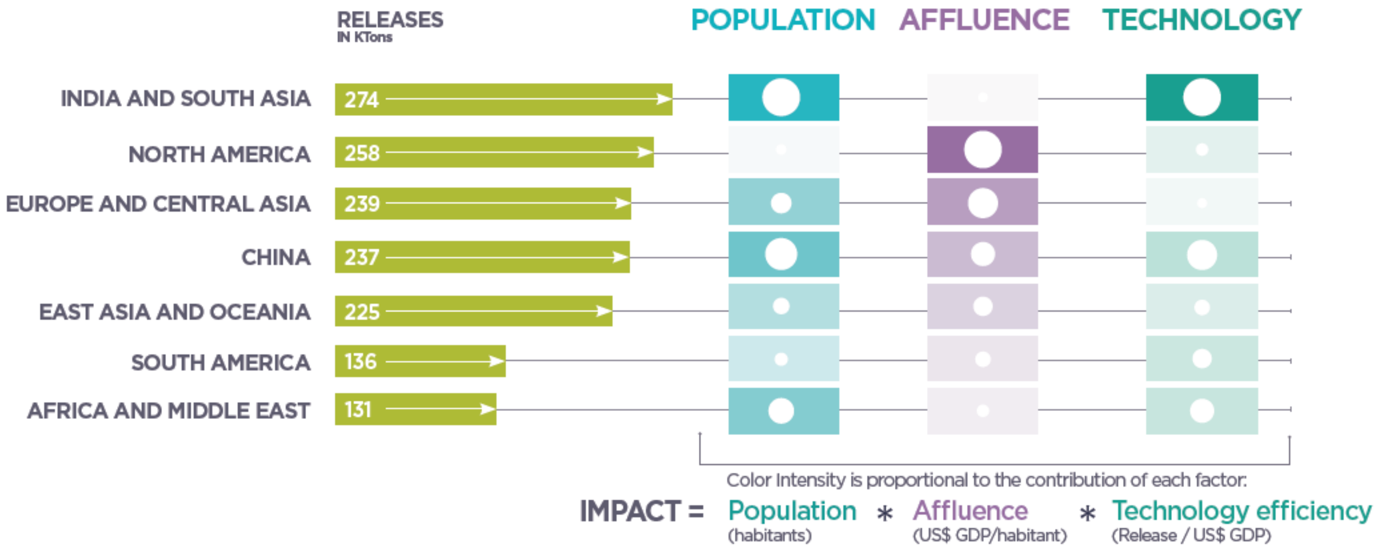
Releases can be explained with a combination of three factors according to the classical IPAT (Impact = Population x Affluence x Technology efficiency) formula (Ehrlich and Holdren, 1971), i.e. in our case the size of a population, the portfolio of activities generating losses (GDP *per capita*), and the capacity of the treatment systems to catch the losses.

In India and South Asia, China and Africa and Middle East, *per capita* losses are below the global average (212 grams/year *per capita*). Large populations combined with a low share of population connected to wastewater treatment systems, e.g. 6.2% in India and South Asia, results in significant releases. Significant releases also occur in Europe and Central Asia as well as North America, but for the opposite reason. In these regions *per capita* losses are larger than the global average, but a relatively smaller regional population is combined with a high share of the population connected to wastewater treatment systems, e.g. 71.4% in North America. South America and East Asia and Oceania sit somewhere in-between.

Figure 7

GLOBAL RELEASES TO THE WORLD OCEANS:

CONTRIBUTION OF THE DIFFERENT REGIONS OF THE WORLD

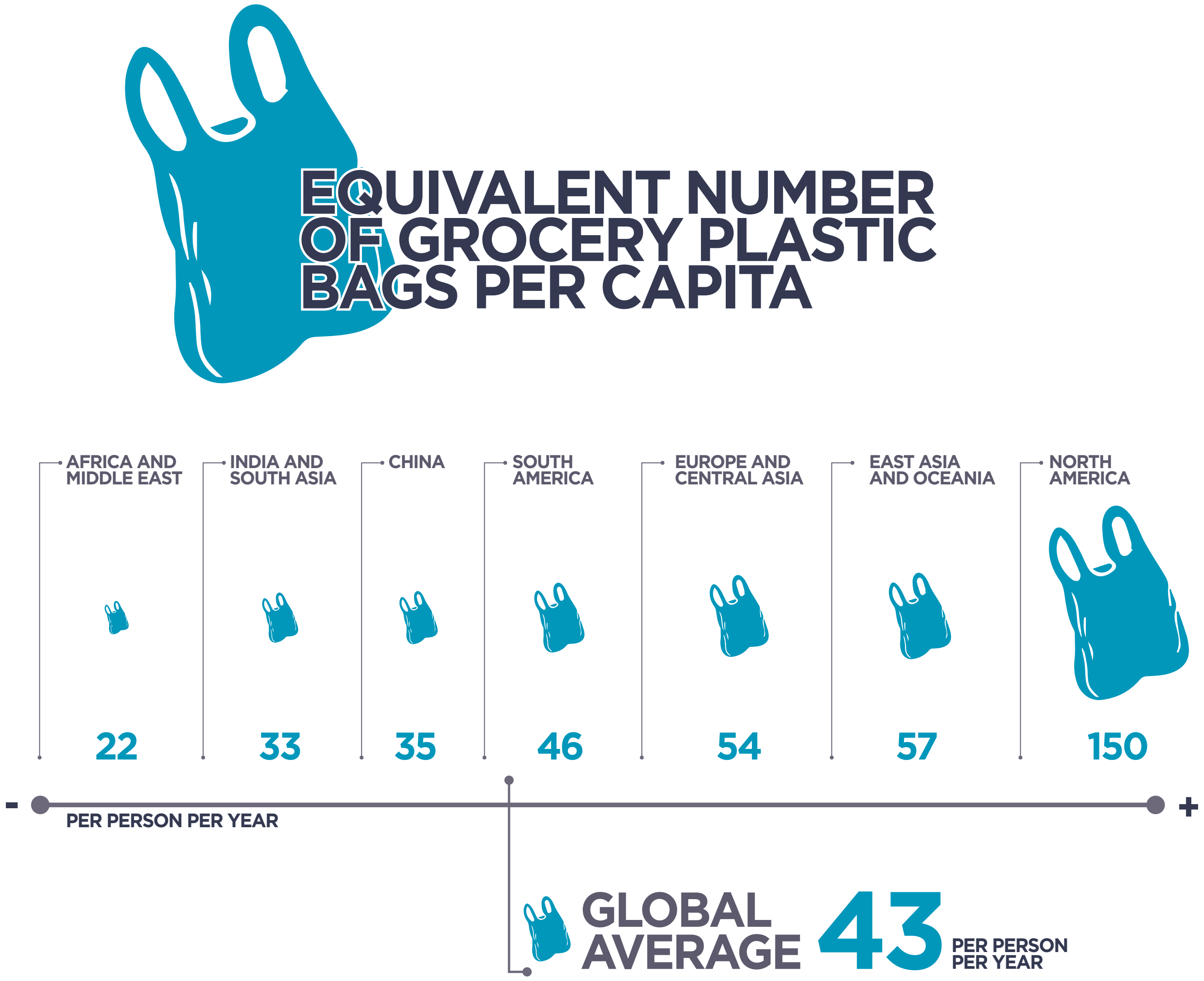


4.6 Releases average one plastic bag per person per week

The release of 1.53 Mtons/year of primary microplastics corresponds to an average release of 212 grams/year *per capita*[7]. This is equivalent to 43 light plastic grocery bags[8] thrown into the world ocean per person or roughly one per week.

This number varies however widely across regions as shown in Figure 8. Going from 22 equivalent grocery bags *per capita* in Africa and the Middle East, this goes up to 150 bags in North America – a seventh fold difference.

Figure 8



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[7] Global population for 2012.

[8] Assuming a weight of 5 grams per grocery bag.

4.7 Key sources of releases differ among regions

The global importance of each regional source is shown in Figure 9. Synthetic textiles are the main source of primary microplastics in Asia, Africa and the Middle East. In these regions the share of synthetic clothes is larger than the global average, the share of the population connected to wastewater treatment systems is lower than the average and the distance driven are lower than in the Americas and in Europe and Central Asia.

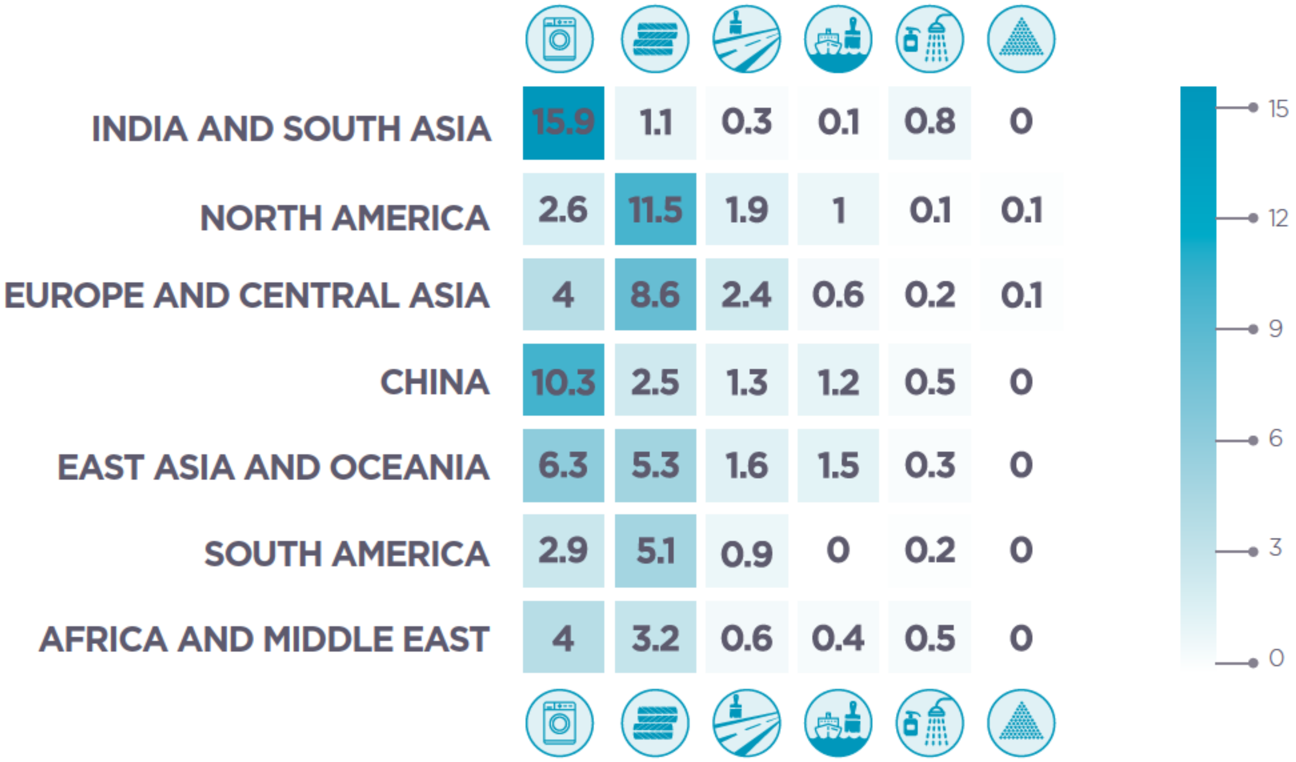
In the Americas, Europe and Central Asia tyres dominate. In these regions regional distances driven are much higher than in the rest of the world. Moreover wastewater treatment systems are more common, thus capturing a higher share of the microplastics going through non-runoff or wind pathways, e.g. from synthetic textiles. For Europe, these results are coherent with the results already reported from Northern European countries where tyres dominate losses and releases (Essel et al., 2015; Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014).

At global scale, the key issues are, by order of importance (in % of global releases, without the source City Dust): textiles in India and South East Asia (15.9%), tyres in North America (11.5%), textiles in China (10.3%) and tyres in Europe and Central Asia (10.3%).

Figure 9

GLOBAL RELEASES TO THE WORLD OCEANS:

KEY SOURCES AMONG REGIONS (TOTAL AMOUNT TO 100%)



INDIA AND SOUTH ASIA	15.9	1.1	0.3	0.1	0.8	0
NORTH AMERICA	2.6	11.5	1.9	1	0.1	0.1
EUROPE AND CENTRAL ASIA	4	8.6	2.4	0.6	0.2	0.1
CHINA	10.3	2.5	1.3	1.2	0.5	0
EAST ASIA AND OCEANIA	6.3	5.3	1.6	1.5	0.3	0
SOUTH AMERICA	2.9	5.1	0.9	0	0.2	0
AFRICA AND MIDDLE EAST	4	3.2	0.6	0.4	0.5	0

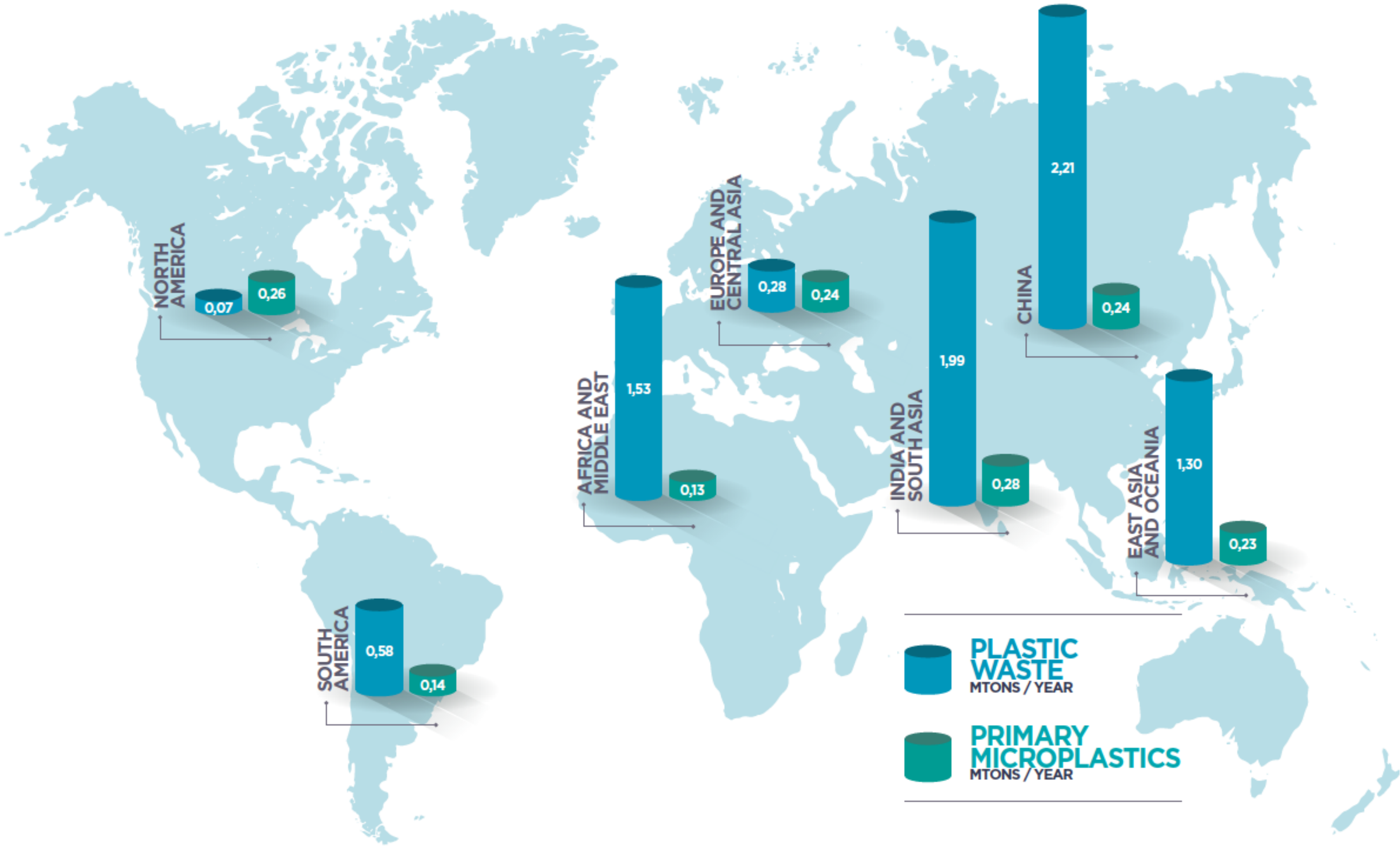
4.8 Microplastic releases are high in Europe & North America

Outside of the dominance of primary microplastics from tyres in the Americas, Europe and Central Asia, the second characteristics of these regions is that releases from primary microplastics are equivalent or outweigh that of secondary microplastics from mismanaged waste for Europe and North America (Figure 10). Such is not the case for the other regions.

Figure 10

GLOBAL RELEASES TO THE WORLD OCEANS:

COMPARISON WITH PLASTICS ORIGINATING FROM MISMANAGED WASTES



4.9 Closing the plastic tap requires different sets of solutions

The present study clearly demonstrates that primary microplastics are a globally significant source of plastics into the oceans. Consequently actions to close the plastic tap should focus not only on implementing better waste management in some regions, but also on finding solutions to reduce diffuse loss over the lifecycle of some products. Depending on the region, priority setting could thus be very different.

For the regions where plastic releases are dominated by mismanaged wastes, priority should be given to implementing better waste management and behaviours. Beyond a classical end-of-pipe approach, reducing the quantity of plastic waste generated (e.g. optimizing, reducing and re-using packaging) as well as increasing the recycling rates (e.g. by increasing the value of plastic waste) could be valid options.

For the regions where primary microplastic releases equal or overweight secondary microplastics from wastes, very different set of solutions should be designed either focusing on product design, infrastructure design, consumer behaviours, or all the three together:

- In the case of intentional losses, such as for personal care products, solutions could be based on finding substitutes to the plastic microbeads and removing them for the product. Several countries and brands are currently in the process of phasing out these microbeads, which hopefully will cancel this source of plastic release into the oceans within a few years.[9]

- In the case of diffusive loss during use phase or maintenance of the product, solutions should be developed through a dialogue between the many actors of the lifecycle of the products, from product designers to water infrastructure engineers. Examples for textiles and tyres are given below.

Potential solutions to reduce microplastic releases from synthetic textiles

Plastic transformation and product manufacturing (chemist/designer): design textiles/textiles fibres to reduce the shedding of fibres

Product manufacturing (designer): pre-wash textiles to reduce heavy loads from first wash

Product maintenance (designer of washing machines): install filtering devices on washing machines

Water infrastructure: understand and increase treatment efficiency.

Product use (consumer): Reduce the share of synthetic textiles by changing individual behaviours or purchasing policies. Note that using recycled fibres instead of virgin fibers may reduce the number of fibres shed. Indeed, using recycled fibres and repurposing wasted plastic for a useful purpose can potentially prevent some mismanaged waste from entering the ocean.

Potential solutions to reduce microplastic releases from tyres

Plastic transformation and product manufacturing (chemist/designer): Ecodesign of rubber polymers and tyres to reduce abrasion

Product use (road/asphalt designer): design road pavement to reduce abrasion

Product use (water infrastructure designer): Ensure water run-off collection and appropriate separation of plastics

Water infrastructure: understand and increase treatment efficiency.

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[9] Regarding the UK, for example, see: http://www.bbc.com/news/uk-37263087

5.

Conclusion

This study is one of the first to demonstrate that primary microplastics are globally a major source of plastics into the oceans with between 0.8 and 2.5 Mtons/year global releases (central value 1.5), regionally outweighing that of secondary microplastics from mismanaged wastes.

If reducing mismanaged plastic waste remains a priority at global scale, for many regions and sectors, solutions need to be found to also reduce primary microplastic releases. Shaping these solutions require a systemic lifecycle management approach rather than a purely waste management approach. Eco-design of the product and dialogue with all stakeholders from product design to urban infrastructure planning both from private and public sectors will be key to close the plastic tap.

5.1 Take home messages

It is very likely that:

- Losses of primary microplastics from commercial and household activities into the environment is in the order of 3.2 Mton/year.
- The greatest contributors to these losses are abrasion of tyres while driving and abrasion of synthetic textiles while washing, i.e. diffusive losses during use/maintenance phase.
- Release of primary microplastics is a significant source of plastic into the ocean.
- In high income countries with adequate waste management, primary microplastic release equals or overweighs the releases from mismanaged plastic wastes. However in lower income countries plastic releases from mismanaged wastes still is the main source of plastic release into the oceans.

It is more likely than not that:

- Releases of primary microplastics from commercial and household activities into the oceans is in the order of 1.5 Mton/year. This represents 48% of the losses ending up in the ocean.
- The losses and releases from primary sources are going to increase in next decades, due to high population and increasing living standard (affluence) in Asiatic and African countries, unless action is taken to reduce loss rate (Impact = Population * Affluence * Technology efficiency) (Ehrlich and Holdren, 1971). While waste management practices are being improved worldwide, the contribution of primary microplastics for less developed countries may thus increase.
- Main pathways are through sewage water and urban run-off waters, from which microplastic transit to the oceans through rivers.
- About 52% of the microplastic loss is trapped in soils when waste water treatment sludge is used as fertilizer and / or when particulates are washed from the road pavement. Fate and effect of these microplastics in soils is still unknown.

5.2 Shaping action to close the plastic tap

If at global scale reducing mismanaged plastic waste remains a priority, for many regions and sectors, solutions need to be found to reduce primary microplastic releases also.

Reducing mismanaged plastic waste mainly requires implementing adequate infrastructure and waste management practices as well as educating behaviours of consumers. Technologies are readily available and the challenge is more a political and financial one.

Solving the primary microplastic release into the world's oceans requires a very different set of solutions. Apart from personal care products, where microbeads are included intentionally in the product (and thus easy to remove or ban as the trend shows), most losses are unintentional, diffusive losses that cannot be easily solved with end of pipe solutions. The losses are from product use and maintenance, mostly from households, and must be tackled with a global producer-consumer perspective. The banning of microbeads from cosmetics is an illustrative action but will not solve the wider problem. Attention must be paid not to overlook other sources, such as textile and tyres, as our study shows that cosmetics only contribute for 2% of the releases of primary microplastics to the ocean at global scale.

Closing the plastic tap will require design and implementation of both technological, behavioural and policy solutions considering plastics and products over their whole lifecycle to reduce plastic losses during production, use, maintenance or end of life of products and releases to the world ocean.

This eco-design approach requires a systemic lifecycle management approach and dialogue with all stakeholders from product design to urban infrastructure planning both from private and public sectors, as already well documented elsewhere (UNEP SETAC, 2009).

Based on the principle that you can improve only what you can measure, metrics and indicators should be developed to set targets and monitor progress. This should include integration in target settings frameworks and policies (e.g. Sustainable Development Goals), as well as in more operational tools such as Life Cycle Assessments. This is the way business developers and product designers will have the microplastic issue under the radar.

5.3 Next steps

An immediate next step is to further develop the analysis presented in this study. This will require additional data collection and a more in depth understanding of regional and sectorial impacts. We also need to better understand the cumulative implication of these releases over time. Furthermore, confrontation of our predictive model with empirical data from the field would be beneficial in order to validate the model. This is however not feasible yet, given the status of literature. Adequate experimental set-up should be developed in order to perform this comparison.

While further research is underway, this study can play a very critical role in opening discussions about how to address primary microplastics beyond traditional plastic waste management approaches. The growing presence of microplastics in the oceans requires new thinking about how to mitigate both primary and secondary releases across the supply chain.

Several organisations including IUCN have started multi-stakeholder dialogue processes to explore how we can close the plastic tap. We need now to ensure that the issue of primary microplastics is not being overlooked in these processes vis-à-vis plastic from mismanaged waste origin. This will require engaging new stakeholders and developing innovative life cycle management approaches.

6.

Appendix 1: Sources

The assumptions for the computation of the optimistic, central and pessimistic scenarios are presented here per source in descending contributing order. These scenarios represent the most credible selection among an extended set of results in this study. The complete set of assumptions, scenarios and results will be published elsewhere.

6.1 Synthetic textiles: abrasion during laundry

Activities and losses are computed in two ways. The first approach combines the estimated number of wash cycles per region with reported losses of microplastics per wash as measured in the effluent of washing machines. The second approach combines data on yearly global and regional synthetic textiles sales with the typical losses over the lifecycle of a synthetic textile cloth.

Approach 1

Activities: Annual number of laundry cycles *per capita* (55), and load per standard wash (4 kg) derived from Pakula and Stamminger (2010). Regional population (2007) from (UNDP, 2015). The regional synthetic shares are from the own-computed regionally-extended dataset based on (FAO/ICAC, 2011) (see approach 2).

Losses: Pessimistic/optimistic: 300/1500 mg of microplastics loss per kg of synthetic textiles per wash, as reported in (Lassen et al., 2015). The central value is set to 900 mg/kg.

Releases: See wastewater pathway in appendix 2.

Approach 2

Activities: Global and regional consumptions (2007) of textiles from (FAO/ICAC, 2011). Data correction and extrapolation to sub-regions based on *per capita* consumption and on population from UNDP (UNDP, 2015).

Losses: Optimistic/pessimistic: 0.74/5% of microplastics loss over the lifecycle (Essel et al., 2015; Lassen et al., 2015). The central value is set to 2%.

Releases: See wastewater pathway in appendix 2.

Comparison of the two approaches

Releases in the optimistic scenario based on the low value for losses in approach 1 match the lower value applied in approach 2 (± 13%). The central scenario is based on an average of the central scenarios in approach 1 and 2: both approaches differ at global scale by only ± 9% but result in different distribution across regions.

6.2 Tyres: abrasion while driving

Activities and losses are computed in two ways. The first approach combines the estimated driven distance covered by all vehicles in a region with reported particulate matter emissions from tyres per km per type of vehicle. The second approach combines data on yearly global and regional sales of synthetic rubber for tyres with the typical particulate matter emissions over the lifecycle of a tyre.

Approach 1

Activities: Annual distance driven in countries (2011) for goods and for people traffic from OECD (OECD, 2013). Extrapolation of distance to other countries based on a linear regression model (OLS) using carbon emissions from transportation (2005) (EU JRC/NL PBL, 2010). The share of goods traffic outside of OECD countries is set to the OECD average (13%). Share of heavy trucks (vs light trucks) in goods traffic: optimistic: 0%, central: 50%, pessimistic: 100%. The selected share of synthetic rubber share per tyre is equivalent to the global share of synthetic rubber in tyres in 2010 (46%) (ETRma, 2011). The urban/rural split is 50% (optimistic scenario) or based on the urban share of the population (central and pessimistic scenarios) from UNHSP (United Nations Human Settlements Programme, 2016).

Losses: Optimistic/central/pessimistic: 0.033/0.051/0.178 gram per tyre-km for cars, light and heavy trucks from (GRPE, 2013; Lassen et al., 2015; Sundt et al., 2014).

Releases: See road runoff pathway in appendix 2.

Approach 2

Activities: Global consumption of synthetic rubber for tyres in 2010 from (ETRma, 2011). Allocation to regions based on (a) the number of vehicles per type (motorcycles, cars and light commercial vehicles, medium and heavy commercial vehicles) per region in 2010 from (ETRma, 2011), (b) the number of wheels per type of vehicle, respectively 2, 4 and 6 and, (c) an average tyre weight per type, respectively 10, 16 and 50 kg (selected values based on literature), and (d) a 25% share of rubber (tread pattern only) per tyre (WRAP, 2006). The proportion of synthetic rubber per tyre is like approach 1.

Losses: Optimistic/pessimistic: 10/25 % of microplastics loss over the lifecycle (Essel et al., 2015; Magnuson et al., 2016; Sundt et al., 2014). The central value is set to 20%, which is equivalent to the global apparent loss of rubber (quantity of synthetic rubber losses computed in approach 1 over the global sold quantity of synthetic rubber for tyres).

Releases: See road runoff pathway in appendix 2.

Comparison of the approaches for the selected scenarios

Releases in the optimistic scenario based on the low value for losses in approach 1 match the lowest value applied in approach 2 (± 1.5%). In the central scenario, based on the central values for losses in approach 1, releases match the central value applied in approach 2 (± 9%). In the pessimistic scenario, the high value for losses in approach 1 are 37% lower than when applying the high value in approach 2.

6.3 City dust: spills, weathering & abrasion

City Dust has been modelled in a different and less specific manner than other sources.

City Dust is the generic name given to a group of nine sources, identified in recent country assessments that are most often occurring in urban environments (Essel et al., 2015; Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014). These sources are grouped together because their individual contribution is small but they account together for a considerable amount of losses in the country studies.

Activities: City Dust includes losses from the abrasion of objects (synthetic soles of footwear, synthetic cooking utensils), the abrasion of infrastructure (household dust, city dust, artificial turfs, harbours and marina, building coatings) as well as from the blasting of abrasives and intentional pouring (detergents). In contrast to other sources that have been subject to specific and regionalised modelling, for city dust assessment is based on more basic extrapolation from the Nordic countries studies. A more detailed assessment should be undertaken in the future to precise the relevance of this source.

Losses: Losses from other sources were extrapolated (+ 29% at global scale) from (Lassen et al., 2015; Magnuson et al., 2016; Sundt et al., 2014) proportionally to the losses computed for the six key sources.

Releases: 50% roadrunoff pathway and 50% wastewater pathway (own hypothesis)

6.4 Road markings: weathering and abrasion by vehicles

Activities and losses are computed by combining data on yearly global and regional consumption of traffic road marking coating with the typical plastic content per segment of paint (paint, thermoplastic, preformed polymer tape, epoxy) and reported loss rates from existing studies.

Description: The global traffic road marking coating consumption is around 1'200 ktons (2014). The use of road marking coating is related to the development of the road infrastructure, its maintenance and to security aspects. The largest market is Europe (31.9%) followed by the USA. Different types of paints (paint, thermoplastic, preformed polymer tape) are applied, with a dominance of paint (44.7%) (Grand View Research, Inc., 2016).

Activities: Global, US and EU consumption (2014) of marking paint for road from (Grand View Research, Inc., 2016). Allocation to other regions, i.e. 43% of road marking paints, based on GDP from (The World Bank, 2016). Global share of segment 'paint' from (Grand View Research, Inc., 2016). Proportion by types of segments of road marking paints for USA from (Grand View Research, Inc., 2016). Proportion of plastic per segment of paint: 10.5/40% for the optimistic/central and pessimistic scenarios (Lassen et al., 2015; Sundt et al., 2014), 25% for thermoplastics (Lassen et al., 2015; Sundt et al., 2014) and 100% for the other segments (tape). The share of the segments is based on the US shares and respect the global 'paint' share. The urban/rural split is based on the urban population (United Nations Human Settlements Programme, 2016).

Losses: Optimistic/central/pessimistic: 23/43/100% from Grand View Research (2016) of microplastics loss (Sundt et al., 2014) (Lassen et al., 2015).

Releases: See road runoff pathway in appendix 2.

6.5 Marine coatings: weathering, application & maintenance

Activities and losses are computed based on the global and regional quantities of marine coatings applied yearly accounting for the plastic content of the paint and reported losses over the whole lifecycle of boats from existing studies.

Activities: Global and regional applications of marine coatings (2012), as well as global shares per type of boats (containers, leisure, commercial) and per reason of application (new build or maintenance, repair and overhaul) from (Wright, 2009). Regional split between types of boats based on (Wright, 2009) and complemented for Asia based on GDP (The World Bank, 2016) and on the number of boats in US and in EU[10] for leisure (Laaksonen, 2012). Proportion of plastic in paint: 50% from (Lassen et al., 2015).

Losses: Optimistic/central/pessimistic loss rates for commercial boats: 3/6/9% based on (OECD Series on emissions documents, 2009) for the central value. Optimistic/central/pessimistic loss rates for leisure boats in developed economies (North America and Europe): 10/30/50%. Optimistic/central and pessimistic loss rates for leisure boats in developing economies 50/90% derived from (Sundt et al., 2014).

Releases: See ocean pathway in appendix 2.

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[10] http://www.europeanboatingindustry.eu/facts-and-figures. Accessed on 23/08/16.

6.6 Personal care products: loss during use

Activities and losses are computed based on regional populations combined with yearly losses *per capita* in Europe reported in existing studies.

Activities: Regional population from (UNDP, 2015).

Losses: Optimistic/central/pessimistic: 1/6/13g *per capita* per year from (Gouin et al., 2015; Leslie, 2015; Essel et al., 2015).

Releases: See wastewater pathway in appendix 2.

6.7 Plastic Pellets: manufacturing, transport & recycling

Activities and losses are computed based on the global and regional quantities of primary plastics produced yearly and accounting for the losses due to production, land and water transport as well as end-of-life of plastic and plastic products as reported in existing studies. Releases are allocated to the region using the plastic products, i.e. based on a consumer perspective (plastic footprint).

Activities: Global and regional production of primary plastics (2007) from (PlasticsEurope, 2009). Allocation of primary plastics to sub-regions and countries is based on GDP. Computation of regional footprints (consumption perspective) using an extended global Multiregional Input-Output Model (MRIO) based on Exiobase 2.0 (Tukker et al., 2014).

Losses: Losses are computed at four stages: production of primary plastics, manufacturing of plastics, transport on land (for domestic uses of plastics products) and water (for interregional trade of plastics products), as well as plastic end-of-life. Optimistic/central/pessimistic: 0.00003/0.0001/0.001 % of microplastics losses per stage from (Cole and Sherrington, 2016).

Releases: Land transport of primary plastics and plastics for domestic use: see wastewater pathway in appendix 2. Sea transport of imported primary plastics and plastics: see ocean pathway in appendix 2.

7.

Appendix 2: Pathways

The assumptions for the computation of the pathways from losses to releases are presented here. For each of the six main sources once activities are computed a loss rate is applied to estimate the primary microplastics losses. Then losses become so-called “releases” into the world ocean through four possible pathways.

7.1 Road runoff pathway

Where: land-based losses and releases
Sources: tyres, road markings, plastic pellets on land

When losses are on roads, part of them is transferred by wind (see wind pathway). The remaining part is washed by rainwater. In rural areas, it is considered that few roads are connected to sewers: A global average value equivalent to 3.5% of losses in rural areas is assumed to end up in the oceans (Lassen et al., 2015; Ten Broeke et al., 2008). In urban areas, two cases are possible: a drain to a separate sewer or a drain to a combined sewer. In the first case, 80% of the releases are assumed to end up in the oceans (Lassen et al., 2015). In the second case, releases depend on an additional assumption, the share of roads connected to a combined sewer. For the optimistic scenario, it is assumed that 50% of roads are connected. For the central and pessimistic scenarios, it is assumed a proportionality between the proportion of the population connected to wastewater treatment systems and the proportion of the roads with a sewer system to collect water from roads. See more on the wastewater pathway below.

7.2 Wastewater Pathway

Where: land-based losses and releases
Sources: synthetic textiles, personal care products, plastic pellets on land

When losses are to wastewater streams, the release ratio depends on the regional coverage and efficiency of the wastewater treatment system. It is assumed that the share of water treated is proportional to the proportion of the population connected to wastewater treatment systems. The regional share of the population connected to wastewater treatment systems is based on an own-computed dataset extending data from UNSTATS (UNSD, 2011) for missing countries and for missing regions with data from the literature. Global treatment efficiency is set at 85%. This number accounts for the non-retained share of microplastics in wastewater treatments systems (3-6%) for fibres according to Lassen et al. (2015) and for overflows (10%) in Europe according to Phillips et al. (2012). Overflow is probably much higher for some developing economies but values are scarce.

7.3 Wind Pathway

Where: land-based losses and releases
Sources: tyres and road markings

When losses are on roads, a part of them is spread by the wind. This part is set at 10% based on Wang et al. (2015). 100% of these losses become releases.

7.4 Ocean Pathway

Where: sea-based losses and releases
Sources: plastic pellets, marine coatings

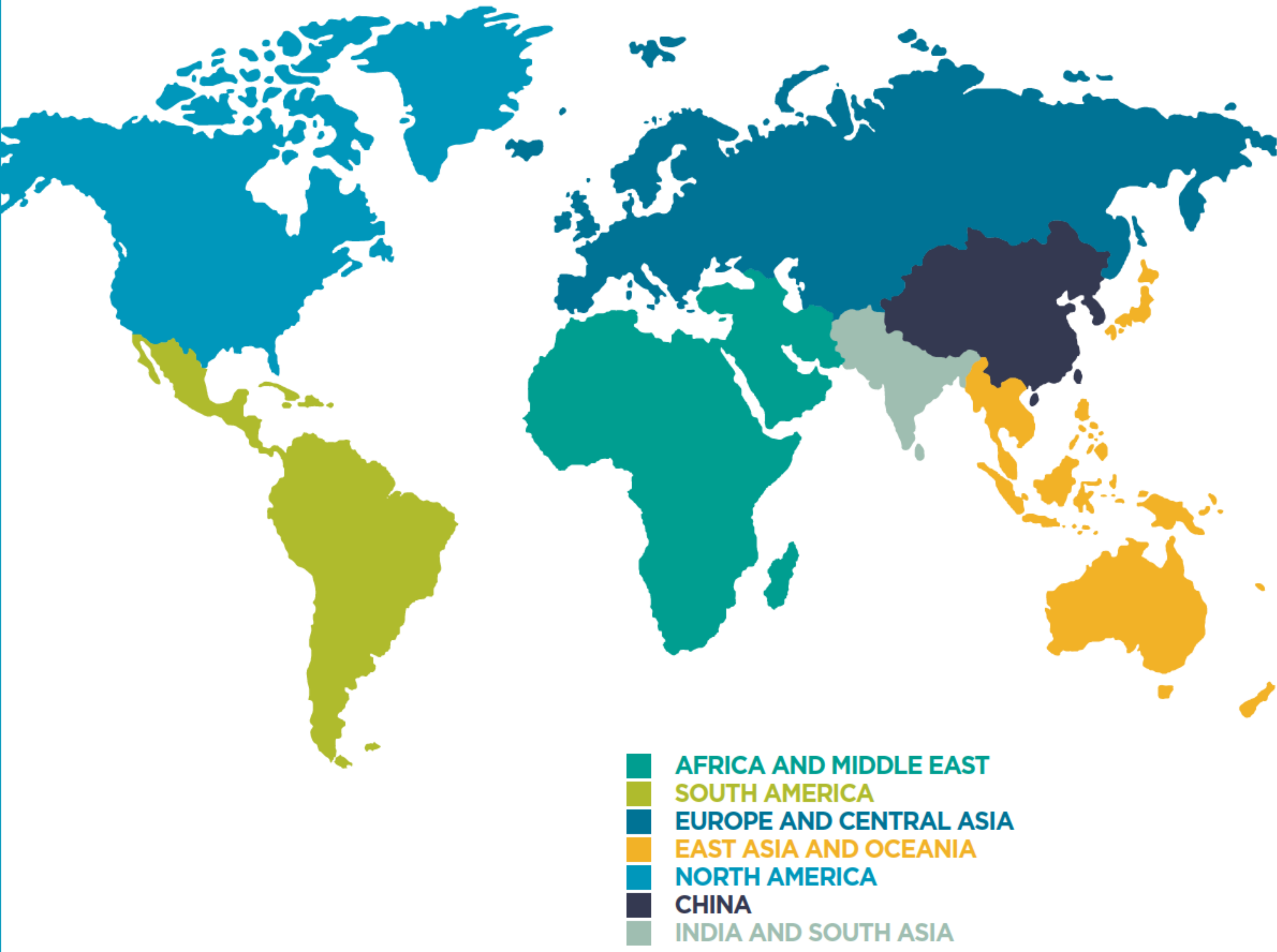
When losses occur in the ocean, 100% of the losses become releases.

8.

Appendix 3: Regions

The grouping of countries has been done according to a classification in 7 regions: Africa and Middle East, China, East Asia and Oceania, Europe and Central Asia, India and South Asia, North America and South America.

THE SEVEN REGIONS CONSIDERED THROUGHOUT THE STUDY



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ARTICLE

https://doi.org/10.1038/s41467-020-17201-9 OPEN

Atmospheric transport is a major pathway of microplastics to remote regions

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In recent years, marine, freshwater and terrestrial pollution with microplastics has been discussed extensively, whereas atmospheric microplastic transport has been largely overlooked. Here, we present global simulations of atmospheric transport of microplastic particles produced by road traffic (TWPs – tire wear particles and BWPs – brake wear particles), a major source that can be quantified relatively well. We find a high transport efficiencies of these particles to remote regions. About 34% of the emitted coarse TWPs and 30% of the emitted coarse BWPs (100 kt yr^{-1} and 40 kt yr^{-1} respectively) were deposited in the World Ocean. These amounts are of similar magnitude as the total estimated direct and riverine transport of TWPs and fibres to the ocean (64 kt yr^{-1}). We suggest that the Arctic may be a particularly sensitive receptor region, where the light-absorbing properties of TWPs and BWPs may also cause accelerated warming and melting of the cryosphere.

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Global annual plastic production reached 359 million tonnes in 2018[1] and, consequently, plastic pollution in freshwater[2], marine[3] and terrestrial[4] ecosystems has received a lot of attention recently. Plastics are released into the environment as macroplastic (>5 mm)[5], microplastic (1 µm to 5 mm)[6] and nanoplastic (<1 µm)[7] particles that can fragment into smaller sizes via photodegradation, physical abrasion, hydrolysis and biodegradation[8]. Plastics can affect coral reefs[9], marine[10] and terrestrial animals[11], as well as human health[12,13].

An important source of plastics is road traffic emissions[14,15]. Kole et al.[14] reported global average emissions of tyre wear particles (TWPs) of 0.81 kg year^{1} per capita, about 6.1 million tonnes (~1.8% of total plastic production). Emissions of brake wear particles (BWPs) add another 0.5 million tonnes. TWPs and BWPs are produced via mechanical abrasion and corrosion[16,17].

Tyres consist of a mix of elastomers such as rubber (natural and synthetic)[18], carbon black, steel cord, fibres, and other organic and inorganic components used to improve their stability[19]; TWPs are produced by shear forces between the tread and the road pavement, generating coarse particles[20], or by volatilization generating submicronic particles[21]. The wearing process depends on the type of tyre, road surface and vehicle characteristics, as well as on the vehicle's state of operation[22].

Most car braking systems consist of a disc or drum with either a pair of shoes or pads mounted in callipers. Brake linings consist of binders, fibres, fillers, frictional additives or lubricants and abrasives[23 25]. Thus, BWPs are a complicated mixture of metal and plastic. BWP emissions depend on the bulk friction material[23,26], on the frequency and severity of braking[27], speed, weight, condition and maintenance of the automobile[28] and the environmental conditions[23,29,30].

Transport of TWPs and BWPs via runoff and wash out to marine and/or freshwater ecosystems has been studied[31,32]. However, very little is known about their dispersion in the atmosphere[33 35] and where they are deposited, despite their health impacts in animals[9 11,36] and humans[12,37], possibly enhanced by adsorbed toxic organic compounds and heavy metals[38]. Greater use of plastics results in more extensive consumption of fossil fuels and, in turn, in larger emissions of greenhouse gases[39] such as methane and ethylene[40]. Since TWPs and BWPs can be present at sizes <10 µm[41], they can remain airborne for long periods of time; different types of microplastics have been detected already in remote areas[42 47]. Considering that they are colourful particles[48], they also absorb light and thereby decrease the surface albedo of snow and ice accelerating melting, similar to black carbon (BC)[49].

Here, we examine atmospheric transport and deposition of TWPs and BWPs on a global scale (see "Methods"). For simplicity we often refer to these particles jointly as road microplastics, although TWPs and BWPs are not the only microplastics that are emitted by traffic (other sources include polymer modified bitumen used for road pavement or road marking paint). Even though TWPs and BWPs contain components other than plastics (e.g., metals), plastics are the dominant component, especially for TWPs. We also speak of microplastics[50], since we only consider the particles of mean size 0.5 9.5 µm, which can remain airborne for long periods of time.

Results

Annual global emissions of road microplastics. TWP emissions were calculated using two different approaches, (a) one based on detailed information of TWP emissions in Northern Europe and extrapolation using a CO$_2$ ratio method[51], and (b) one based on the GAINS (Greenhouse gas Air pollution Interactions and Synergies) model[52] (see "Methods"). The two methods were compared in detail (Supplementary Fig. 1) showing that TWP emissions are very similar (0.25 32 t (tonnes) year^{1} per grid cell); therefore, we report the geometric average. Uncertainties were calculated based on different assumptions on the airborne fraction of total emissions (see "Methods"). The use of different methods in emission calculations was, in addition, a tool to cross validate whether the emissions are realistic or not. As seen in "Methods", the CO$_2$ ratio method is based on country statistics of returned tyres. Unfortunately, similar statistics for brake wear do not exist, and therefore, only emissions from the GAINS model were used for BWPs.

Emissions of road microplastics are concentrated in the eastern US, Northern Europe and large urbanized areas of Eastern China, Middle East and Latin America where vehicle densities are highest (Fig. 1, Supplementary Fig. 1). Annual total global TWP emissions were 2907 kt (kilotonnes) year^{1} (3434 kt year^{1} from the CO$_2$ ratio method and 2380 kt year^{1} from the GAINS model), while BWP were 175 kt year^{1} (Fig. 1a, b). For the particulate matter 2.5 (PM2.5) and PM10 size fractions, TWP emissions were 29 kt year^{1} (12 75 kt year^{1}) and 288 kt year^{1} (113 826 kt year^{1}), respectively (Table 1). The highest emissions were calculated for Asia (excluding Russia) (PM2 5 4.8 30 kt year^{1}, mean 12 kt year^{1}; PM10 85.0 167 kt year^{1}, mean 113 kt year^{1}) and North America (PM2.5 2.6 16 kt year^{1}, mean 6.4 kt year^{1}; PM10 46 90 kt year^{1}, mean 64 kt year^{1}) (Fig. 1). The annual global emissions of BWP were 98.2 kt year^{1} (63.4 152 kt year^{1}) for

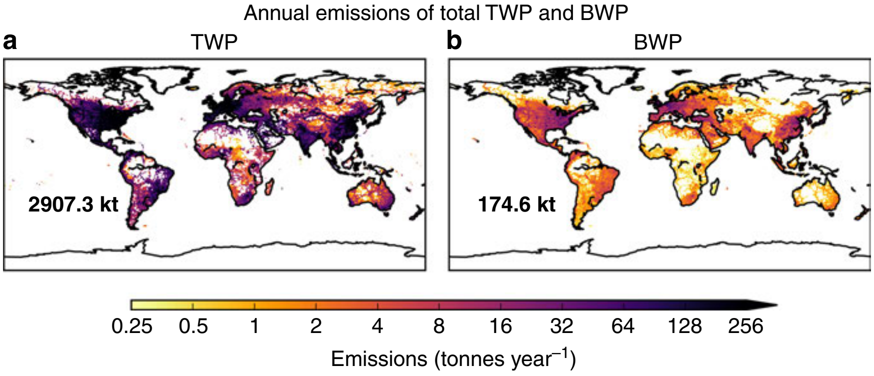

Fig. 1 Annual gridded emissions of road microplastics. Global annual emissions of total road microplastics (tyre wear particles, TWPs, in **a**, and brake wear particles, BWPs, in **b**). TWP emissions are the average of the calculated emissions using the CO$_2$ ratio method and the GAINS model. Bold numbers at the left bottom of each panel represent the annual emissions of total TWPs and BWPs from road vehicles for 2014, which were estimated to be 2907 and 174.6 kt, respectively.

Table 1 Annual road microplastic emissions.

	Europe	Asia	Russia	North America	Central America	South America	Africa	Oceania	Total
PM2.5 TWP	2.3-15 (5.8)	4.8-30 (12)	0.26-1.6 (0.64)	2.6-16 (6.4)	0.32-2.0 (0.80)	0.8-5.0 (2.0)	0.56-3.5 (1.4)	0.19-1.2 (0.48)	12-75 (29)
PM10 TWP	42-82 (58)	85.0-167 (113)	4.6-9.0 (6.4)	46-90 (64)	5.7-11 (8.0)	14-28 (20)	10-20 (14)	3.4-6.8 (4.8)	113-826 (288)
PM2.5 BWP	13-32 (21)	26-62 (40)	2.5-6.0 (3.9)	11-26 (17)	1.7-4.0 (2.6)	4.4-10 (6.8)	3.4-8.1 (5.2)	0.97-2.3 (1.5)	63.4-152 (98.2)
PM10 BWP	28-37 (32)	50-67 (58)	6.9-9.1 (7.9)	22-29 (25)	3.2-4.2 (3.7)	8.4-11 (9.7)	6.5-8.6 (7.5)	1.9-2.5 (2.2)	85.8-248 (146)

Annual continental emissions (in kt) of road microplastics (tyre wear particles, TWPs, and brake wear particles, BWPs) in PM2.5 and PM10 size modes averaged for the two different methodologies used (CO_2 ratio and GAINS mode emissions). Corresponding ranges are variations of continental geometric standard deviations from geometric means (presented within parentheses) following a log-normal distribution (see "Methods"). The airborne PM10 fraction was assumed to be 2.5, 5, 10, 20 and 40% of the total TWP emissions, while the PM2.5 was assumed to be 0.25, 0.5, 1, 2 and 4% of the total TWP emissions. For BWPs, it was assumed that 30, 40, 50, 60 and 70% of total BWPs are PM2.5 and 60, 70, 80, 90 and 100% of the total BWPs are PM10. Note that Russia has been excluded from both Europe and Asia and is listed separately, while America has been divided into three parts (north, central and south).

PM2.5 and 146 kt year ^{1} (85.8 248 kt year ^{1}) for PM10 (Fig. 1, Supplementary Fig. 2). BWP emissions were very similar in Europe and North America, for both PM2.5, but highest in Asia (Table 1, Supplementary Fig. 2).

Atmospheric transport and deposition of road microplastics. Surface concentrations of TWPs range between a few ng m ^{3} and 20 ng m ^{3} for PM2.5 and up to 50 ng m ^{3} for PM10 (Supplementary Movie 1). BWP surface concentrations reach 50 ng m ^{3} at maximum (Supplementary Movie 1). The highest concentrations were calculated for eastern USA, Europe and South eastern Asia. All concentrations (TWPs 0.4 μg m ^{3} for PM2.5, 1.8 μg m ^{3} for PM10; BWPs 0.8 μg m ^{3} for PM2.5, 1.4 μg m ^{3} for PM10) were far below air quality limits for PM (annual mean 10 μg m ^{3} for PM2.5, double for PM10[53]) and lower than typical BC concentrations in remote regions[54]. The annual mean modelled lifetime of PM2.5 TWPs was estimated to be 28 ± 2.7 days (range 18 37 days), while for PM10 it was equal to 8.3 ± 1.0 days (range 5.5 11 days). Accordingly, for BWPs, the annual mean modelled lifetime was calculated as 28 ± 2.8 days (range 17 37 days) for the PM2.5 size class and 1.3 ± 0.16 days (range 0.94 1.6 days) for PM10. The large calculated lifetimes for PM2.5 road microplastics are due to the small scavenging coefficients for in cloud used in the model, as it was assumed that road microplastics should be rather hydrophobic (Methods). For comparison, a typical lifetime for atmospheric black carbon in the PM2.5 size mode is 3 11 days[49].

Annual deposition maps (Fig. 2) show that smaller road microplastic particles (PM2.5) are dispersed more widely than larger ones (PM10). PM10 road microplastics are deposited mainly close to the hotspot emission regions (North America, Europe and South eastern Asia) (Fig. 2). Of the annual global TWP PM2.5 emission of 29 kt year ^{1} (12 75 kt year ^{1}), ~1.7 kt year ^{1} (0.60 4.8 kt year ^{1}) were deposited over Europe, 4.3 kt year ^{1} (1.5 12 kt year ^{1}) over Asia, 3.3 kt year ^{1} (1.1 9.6 kt year ^{1}) in America, and much lower amounts in Africa and Oceania (<4% of the total deposited mass). Overall, ~43% (4.3 34 kt year ^{1}, mean 12 kt year ^{1}) of the total deposited mass of PM2.5 TWPs was deposited on land and ~57% (5.3 48 kt year ^{1}, mean 16 kt year ^{1}) in the ocean (Table 2). About 8.1 kt year ^{1} (2.8 23 kt year ^{1}) of PM2.5 TWPs were deposited on ice and snow surfaces (polar regions, mountains, etc.). Accordingly, annual total deposition of PM10 TWPs was 284 kt year ^{1} (102 787 kt year ^{1}) with ~65% (68.1 497 kt year ^{1}, mean 184 kt year ^{1}) deposited on land. Around 28 kt year ^{1} (10 76 kt year ^{1}) of the TWPs were deposited on snow and ice. The vast majority (~60%) was deposited in Europe, America, Russia and Asia (Table 2). Although deposition of TWPs to Antarctica was very small compared to other regions (0.03 kt year ^{1} for PM2.5, 0.01 kt year ^{1} for PM10), other forms of microplastics have been determined there likely transported via sea and/or air[55].

Of the 97 kt year ^{1} (59.2 162 kt year ^{1}) annual total deposition of PM2.5 BWPs, 45 kt year ^{1} (30 68 kt year ^{1}) were deposited on land (~46%) and 52 kt year ^{1} (29 94 kt year ^{1}) in the World Ocean (~54%) (Table 2). About 14 kt year ^{1} (11 18 kt year ^{1}) were deposited in Asia, 12.9 kt year ^{1} (8.6 19 kt year ^{1}) in America, 6.1 kt year ^{1} (4.7 7.9 kt year ^{1}) over Europe and 7.2 kt year ^{1} (6.0 8.6 kt year ^{1}) in Russia. A significant amount (~31%) of 30 kt year ^{1} (20 45 kt year ^{1}) was deposited on snow and ice surfaces. As regards to PM10 BWPs, half of the deposition (~53%) occurred in Asia, Europe and North America. About 72% (78.4 133 kt year ^{1}, mean 102 kt year ^{1}) were deposited on the land and the rest in the ocean, and only 20 kt year ^{1} (11 36 kt year ^{1}) on snow and ice surfaces (~14% of global deposited mass). Similar to TWPs, transport to Antarctica was small compared to other continents (0.04 kt year ^{1} for PM2.5, 0.01 kt year ^{1} for

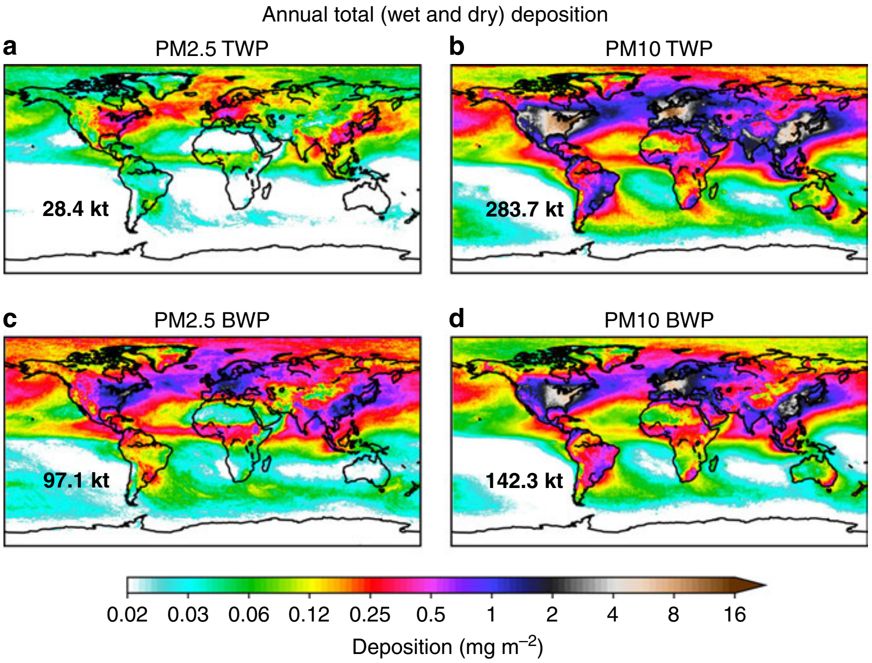

Fig. 2 Wet and dry deposition of road microplastics. Annual total (wet and dry) deposition of tyre wear particles (TWPs) and brake wear particles (BWPs) in PM2.5 and PM10 size classes, respectively. The projected deposition has been calculated as the geometric mean of all simulations using TWP emissions estimated using the CO_2 ratio method and the GAINS model and using BWP emissions calculated from the GAINS model, respectively. The simulations comprise 120 ensemble members with different assumption for the airborne fraction (five for each of the PM2.5 and PM10 fractions), particle size distribution (eight for each of the PM2.5 and PM10 fractions) and CCN/IN (cloud condensation nuclei/ice nuclei) efficiency (three different sets of scavenging coefficients per fraction) following a log-normal distribution (see "Methods"). Bold numbers at the left bottom of each panel represent the annual total deposition of TWPs and BWPs from road vehicles in PM2.5 and PM10 sizes for year 2014.

PM10). The slightly smaller relative deposition of BWPs over the ocean compared to TWPs in both particle sizes (Table 2) can be attributed to the higher particle density of BWPs (see "Methods"), which leads to more rapid deposition.

Road microplastics in snow-covered land and ice surfaces. TWP concentrations in the Arctic snow ranged between 1 and 10 ng kg^{1} of snow for PM2.5 and between 4 and 80 ng kg^{1} for PM10 (Fig. 3). Modelled concentrations of BWPs were 2 30 ng kg^{1} for PM2.5 and 2 70 ng kg^{1} for PM10 (Fig. 3). For comparison, note that these values are almost three orders of magnitude lower than those of BC in Arctic snow[56,57]. It is seen that Northern Europe (e.g. Scandinavia), on one side, and North America, on the other, present higher TWP/BWP snow concentrations than the Arctic. This is a combination of the proximity to source regions and the fact that the calculation was performed only for pixels with substantial snowfall as compared to total precipitation (Supplementary Movie 2). The largest Arctic snow concentrations were predicted on the sea ice between Northern Greenland and Europe. This area receives road microplastics emitted both in North America and Europe (Supplementary Movies 3 and 5). Transport of microplastics into the Arctic occurs particularly in winter and spring (Fig. 3e, f, Supplementary Fig. 3) and is likely enhanced during positive phases of the North Atlantic Oscillation (NAO)[58]. It is known from previous work[58] that pollution transport from North America and Europe is enhanced during positive phases of the NAO and that this effect is strongest in winter and spring. Since 1990, there have been 7 years with high NAO indexes, a negative phase around 2010 and again positive phase between 2014 and 2019[59]. Another hotspot region, in terms of snow concentrations, is Northern Eurasia (Fig. 3). This region is affected by air transport from high emission regions further south (Supplementary Movies 3 and 5).

The uncertainty of road microplastics deposition was calculated from 120 model ensemble members, each comprising different size distribution characteristics, different coefficients for in cloud and below cloud scavenging and variable airborne fraction with respect to total emissions (see "Methods"). The uncertainty was calculated as the geometric standard deviation of deposition resulting from all ensemble members (see "Methods", Supplementary Fig. 4, Supplementary Table 1 and Supplementary Table 2).

The relative uncertainties in deposition both for PM2 5 and PM10 road microplastics are high, with a geometric standard deviation of up to 3. This is mainly due to the great uncertainty in the size distributions of emitted TWPs and BWPs, which controls the fraction of the total mass that can become airborne and the removal properties within that fraction. Dannis[60] found that the mean particle diameter of TWPs decreases, while vehicle speed increases, which may contribute to the large differences in reported size distributions. In an effort to explain the size distribution, Cadle and Williams[61] suggested that the formation of sub micron TWPs may be due to the thermal degradation of tyre polymer, with the larger particle mode being generated by mechanical abrasion. Deposition of PM2.5 road microplastics is more uncertain closer to the highest emitting continents (North eastern USA, South eastern China and Europe) and in tropical regions where precipitation is intense (Fig. 4a, c, e). On the contrary, the highest uncertainties for road microplastic deposition of PM10 occur in remote regions (Fig. 4b, d, e). This is related to the large sensitivity of long range transport efficiency to gravitational settling and below cloud scavenging of larger particles, which is relatively more important for PM10 than PM2.5.

Discussion

To examine the susceptibility of different remote regions (e.g. oceans, Arctic, etc.) to TWP and BWP emissions, we computed

Table 2 Annual deposition of road microplastics.

	Europe	Asia	Russia	North America	Central America	South America	Africa	Oceania	Antarctica	Land	Ocean	Snow/ice	Total
PM2.5 TWP	0.60–4.8 (1.7)	1.5–12 (4.3)	0.53–3.6 (1.5)	0.76–6.4 (2.2)	0.11–0.90 (0.32)	0.27–2.3 (0.80)	0.38–3.2 (1.1)	0.03–0.2 (0.08)	- (0)	4.3–34 (12)	5.3–48 (16)	2.8–23 (8.1)	9.6–82 (28)
PM10 TWP	11–84 (31)	24.7–180 (66.8)	4.4–32 (12)	15.5–113 (41.9)	1.9–14 (5.1)	5.1–37 (14)	4.4–32 (12)	0.70–5.1 (1.9)	- (0)	68.1–497 (184)	34.4–290 (100)	10–76 (28)	102–787 (284)
PM2.5 BWP	4.7–7.9 (6.1)	11–18 (14)	6.0–8.6 (7.2)	5.9–13 (8.8)	0.80–1.8 (1.2)	1.9–4.4 (2.9)	2.6–6.7 (4.2)	0.18–0.40 (0.27)	- (0)	30–68 (45)	29–94 (52)	20–45 (30)	59.2–162 (97.2)
PM10 BWP	13–22 (17)	27–46 (35)	6.7–11 (8.7)	18–30 (23)	1.8–3.0 (2.3)	5.6–9.5 (7.3)	5.4–9.1 (7.0)	0.92–1.6 (1.2)	- (0)	78.4–133 (102)	23–68 (40)	11–36 (20)	101–201 (142)

Annual global (wet and dry) deposition (in kt) of road microplastics (tyre wear particles, TWPs, and brake wear particles, BWPs) in PM2.5 and PM10 size bins estimated with FLEXPART version 10.4 model. TWP deposition (average values are presented within parentheses) is the geometric mean of the two simulations with emissions calculated with the CO_2 ratio method and the GAINS model (IIASA) each including 120 ensemble members with different assumption for the airborne fraction (five for each of the PM2.5 and PM10 fractions), particle size distribution (eight for each of the PM2.5 and PM10 fractions) and CCN/IN (cloud condensation nuclei/ice nuclei) efficiency (three different sets of scavenging coefficients per fraction). BWP deposition was calculated in the same way (120 ensemble members), but only using emissions from the GAINS model. Uncertainties of TWP and BWP deposition are expressed with the geometric standard deviation taking into account all the simulations (120). The results are given in ranges based on the variation of the geometric standard deviation from the geometric mean (see "Methods").

the probability of road microplastics emitted globally to reach these remote regions via the atmosphere. We define the transport efficiency as the ratio between the mass of microplastics deposited in a remote area and the total mass of microplastics emitted globally. We calculate these efficiencies by masking several geographical regions (Table 3).

About 15% of the PM2.5 road microplastics were transported to the Atlantic Ocean (Table 3), whereas coarse particles were less efficiently deposited there (TWPs 10%, BWPs 11%). Due to the smaller size of PM2.5, their transport efficiency to the Pacific Ocean was even more strongly enhanced relative to PM10 deposition (Table 3). The South China Sea receives ~2% of airborne road microplastics, at maximum, a large amount considering its relatively small surface. This is due to the fact that South eastern Asian emissions of microplastics tend to travel towards the South China Sea and the Western Pacific before they turn to the north, all the way to the Arctic (Supplementary Movies 3 and 5). The calculated transport of PM10 road microplastics shows a relatively high efficiency over Greenland (TWPs 1.7%, BWPs 2.3%) and over the Arctic Ocean (TWPs 6.8%, BWPs 4.3%) and much smaller to the Southern Ocean (TWPs 1.4%, BWPs 0.5%). Negligible transport to Antarctica was simulated.

A notable point here is the fact that in areas surrounded by road microplastic emissions sources, PM10 particles are more efficiently deposited than PM2.5 particles. For instance, in the Alps, the Mediterranean, Baltic and South China Seas, transport efficiencies of coarse particles were up to twice of those of the fine particles. Short exposure to PM10 particles has been highlighted as a major cause of respiratory diseases (e.g. asthma), especially considering that such regions are heavily populated[53]. The opposite is the case in remote areas that are far from emission sources such as the Arctic and Greenland, where deposition of fine particles is greater than that of coarse ones (Table 3).

Another important receptor region of global microplastic emissions is the Arctic (Table 3). It is well known that aerosols can be transported efficiently to the Arctic, in particular during the Arctic Haze season in late winter and spring[62]. We find a transport efficiency of almost 3.6% for the Arctic (excluding Greenland) and a similar transport efficiency for Greenland. These transport patterns may potentially intensify the climatic risk of microplastic pollution with respect to their ability to decrease the albedo in the Arctic and enhance snow and ice melting. In addition, road microplastics may concentrate in Arctic melt pools, with unknown ecological consequences. TWPs and BWPs constitute ~1.8% of total plastic production[14], hence the anticipated impact of all microplastics arriving to snow and ice surfaces might be greater.

One aspect that is missing from our simulations is potential resuspension of road microplastics. Strong winds may re mobilize deposited microplastic particles both from the land and the ocean surface, allowing secondary transport of these particles and thus enhancing efficiency of airborne transport to remote areas such as the Arctic, somewhat similar to the well known grasshopper effect of persistent organic pollutants[63]. Another important aspect is the fact that emissions from non road vehicles (tractors, mining trucks and equipment, construction and forestry machinery, and even military) have not been included in our emission inventories. While these vehicles are fewer, they work in difficult conditions, are heavier and carry heavy loads leading to enhanced tyre and brake wear.

There is a lack of measurement data that could be used to validate our results. However, Bergmann et al.[43] reported that the mean number concentration of plastic fibres detected in snow from Svalbard were 1.38 ± 1.10 ml^{-1}, while in Bavaria they were 1.43 ± 0.32 ml^{-1}. Most particles identified were between 11 and 25 μm, while those <11 μm were not identified due to

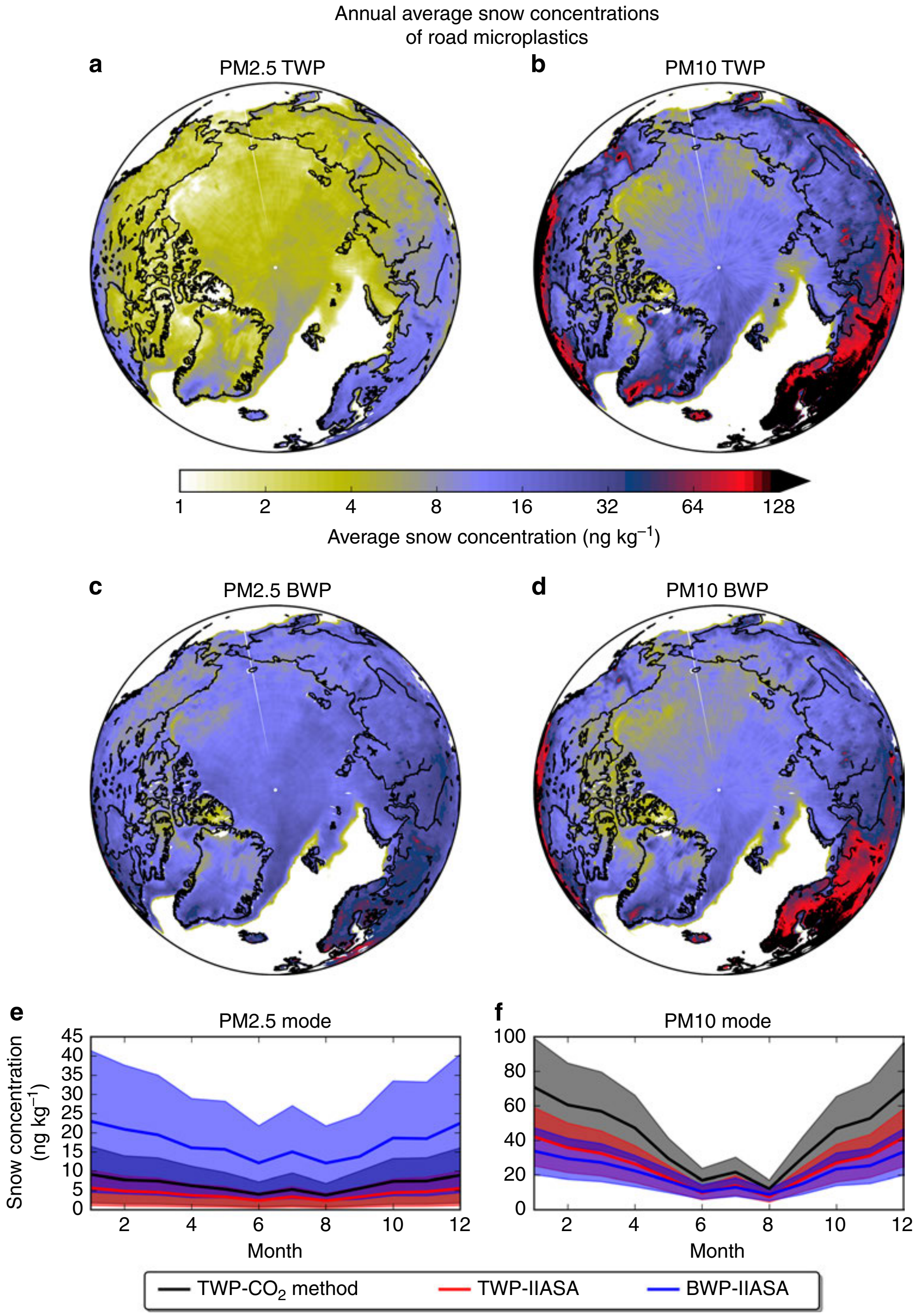

Fig. 3 Road microplastics in snow and ice. Annual average concentrations of road microplastics in Arctic snow (**a**–**d**) in ng kg^{-1}. Snow concentrations were calculated using daily fields of sea-ice area fraction and total snowfall from European Centre for Medium-Range Weather Forecast (ECMWF) combined with daily modelled deposition. The latter includes results from 120 simulations that accounted for different airborne fractions (five members for each of the PM2.5 and PM10 fractions), particle size distribution (eight members for each of the PM2.5 and PM10 fractions) and CCN/IN (cloud condensation nuclei/ice nuclei) efficiency (three different sets of scavenging coefficients per fraction) following a log-normal distribution (see "Methods"). Monthly variation of concentrations of road microplastics in the Arctic snow in both sizes (PM2.5 and PM10) are presented in (**e**, **f**). For the latter, model results using emissions from both methods are presented. Tyre wear particles (TWPs) and brake wear particles (BWPs) uncertainties have been calculated as the geometric standard deviation of all the 120 simulations with different assumption (airborne fraction, size distribution and CCN/IN efficiency, see "Methods"). Note that the smallest concentrations occur in mid-summer (**e**, **f**).

methodological limitations. According to Stylios[64], a microfibre is a fibre with <1 decitex (dtex) per filament, with the most common types being from polyesters and polyamides (1 dtex = 1 mg per 10 m). Since the majority of the fibres was <25 µm size, these number concentrations can be converted to mass concentrations of 3.4 ± 2.8 and 3.6 ± 0.80 ng g ^{1} in Svalbard and Bavaria, respectively. Materić et al.[65] reported PET (polyethylene terephthalate), PPC (polypropylene carbonate) and PVC (polyvinyl chloride) concentrations in Alpine snow of 5.6 23, 11 16 and 6.9 ± 0.2 ng g ^{1}, respectively. These concentrations are

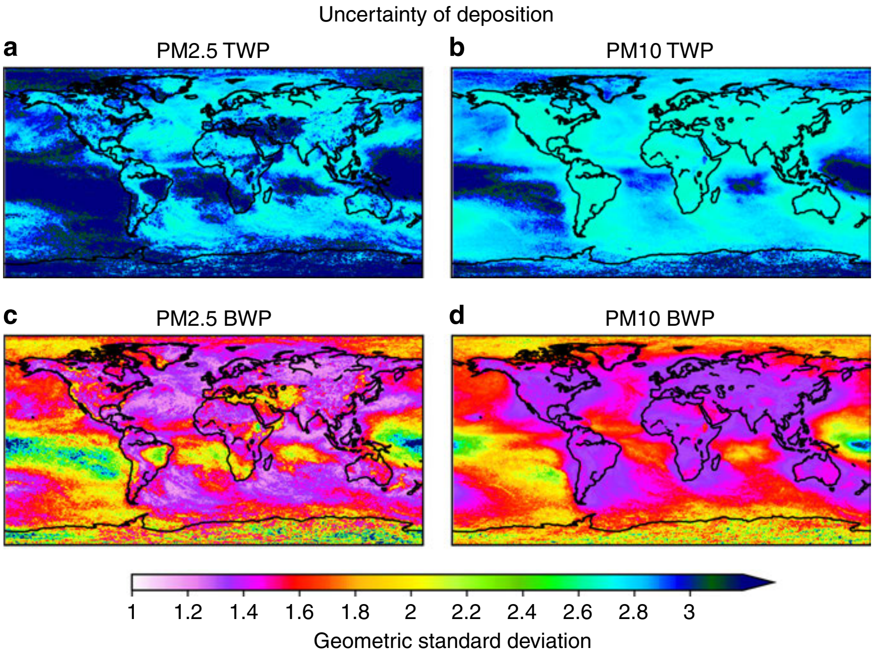

Fig. 4 Model uncertainty. Calculated model uncertainties of deposition. Uncertainties were calculated from a model ensemble of 120 members for each of the PM2.5 and PM10 sizes, both for tyre wear particles (TWPs) and brake wear particles (BWPs). The ensemble accounts for (**a**) airborne PM10 TWP fraction to be 2.5, 5, 10, 20 and 40% of the total TWP emissions, (**b**) PM2.5 TWP to be 0.25, 0.5, 1, 2 and 4% of the total TWP emissions, while (**c**) PM2.5 BWP fraction was assumed to be 30, 40, 50, 60 and 70% of total BWPs and (**d**) PM10 BWP fraction of 60, 70, 80, 90 and 100% of the total BWPs, different wet scavenging coefficients that define CCN/IN (cloud condensation nuclei/ice nuclei) efficiency, and different assumptions on the airborne fraction in the emissions (see "Methods"). Uncertainties are given as the geometric standard deviations, since sensitivity scenarios followed a largely log-normal distribution (see "Methods").

~100 times higher than those estimated here for TWPs and BWPs in snow (Fig. 3), which is likely realistic considering the larger usage of these polymers as compared to TWPs and BWPs. A measurement strategy for both atmospheric concentration and deposition (e.g. in snow samples) measurements is highly recommended at different distances to major source regions in order to facilitate validation of our model results.

We calculated that out of 102 787 kt (mean 284 kt) of PM10 TWPs emitted, 34.4 290 kt (mean 100 kt) were deposited in the World Ocean. In the most recent study on riverine transport from land to ocean, van Wijnen et al.[66] reported a total annual global export of microplastics to the ocean of 47 kt, 80% (37.6 kt) of which was produced by macroplastic degradation, and 20% (9.4 kt) from direct discharges of TWP and laundry fibres. The total annual releases of TWP in their model were assumed to be 426 kt (Table 3 in van Wijnen et al.[66]). If we assume that all microplastics are transported from land to the World Ocean over time (wash out and runoff processes) and scale the van Wijnen et al.[66] TWP emissions to match ours (2907 kt, Fig. 1), we calculate that 64 kt of TWP may be washed out from the land in a year. This suggests that direct deposition of airborne road microplastics is likely the most important source for the ocean.

Methods

TWP emission calculations based on a CO_2 ratio method. Top-down estimates of total annual tyre wear emissions of 5700 10,000 and 100,000 tonnes, respectively, have been reported for Norway[67], Sweden[68] and Germany[69] based on measurements of lifetime weight loss of returned tyres. For the rest of the globe, we did not have access to such data. To obtain global emissions (Fig. 1), we assumed a constant ratio of TWP emissions to CO_2 emissions from the road transport sector (0.49 mg TWP g^{-1} CO_2), using CO_2 emissions from the CMIP6 (Coupled Model Intercomparison Project phase 6)[70] inventory ($0.5° \times 0.5°$ resolution) for the year 2014. The TWP/CO_2 emission ratio is the average value of the ratios obtained for Norway, Sweden, and Germany, which were all very similar: 0.43, 0.50 and 0.55 mg TWP g^{-1} CO_2, respectively.

While the total TWP emission is relatively well constrained, the fraction of total TWP and BWP emissions that becomes airborne, assumed to be particles <10 µm (PM10), is highly uncertain. Values reported in the literature range from ~1 to 40%, while those for the PM2.5 fraction (particles <2.5 µm) are ~1%[23,30,32,41,71–74] (Table 6 in Grigoratos and Martini[75]). We examined how sensitive the calculated concentrations of TWP are with respect to this fraction. For that, we created five scenarios that assumed that 2.5, 5, 10, 20 and 40% of the total TWP emitted are PM10 and 0.25, 0.5, 1, 2 and 4% are PM2.5 (Supplementary Table 1). We report TWP emissions as a range (geometric mean and geometric standard deviation) based on the derived emissions from the five ensemble members, each one with a different assumed fraction for the PM2.5 and PM10 mode.

TWP and BWP emission calculations with the GAINS model. The GAINS (http://gains.iiasa.ac.at) model[76] is an integrated assessment model where emissions of air pollutants and Kyoto gases are estimated for nearly 200 regions globally considering key economic activities, environmental regulation policies and regionally specific emission factors. For emissions of PM, GAINS provides size speciated PM discriminating PM1, PM2.5, PM10 and total PM, as well as carbonaceous particles (BC, organic carbon); detailed description of the methodology can be found in Klimont et al.[52]. Emissions of non-exhaust PM in GAINS include TWPs, BWPs and road abrasion, and the calculation is based on region-specific data and estimates of distance-driven (km vehicle $type^{-1}$ $year^{-1}$) and vehicle-type-specific emission rates (mg km^{-1}). Distinguished vehicle types for road transport include motorcycles, cars, light duty vehicles, buses and heavy-duty vehicles. The estimates of distance driven for 2015 are derived using data on fuel use in road transport (from https://www.iea.org) supported by national data on vehicle numbers and assumptions of per-vehicle mileage travelled. Considering explicitly vehicle-type-specific emission rates and respective activity data allows for better reflection of often significant regional differences in fleet structure, that is, large number of motorcycles in South and South-East Asia and generally lower car ownership numbers in parts of the developing world. GAINS emissions are distributed globally ($0.5° \times 0.5°$) using road network data, assumptions about road-type vehicle density and population data.

The vehicle-type-specific TWP and BWP emission factors draw on review of several measurement papers (Klimont et al.[77]) that were recently updated[52] using primarily Denier van der Gon et al.[78], EEA[79] and Harrison et al.[41]. There are large uncertainties in emission factors including the PM size distribution. GAINS assumes that PM10 TWPs represent ~10% and PM2.5 ~1% of total TWPs, whereas

Table 3 Annual transport efficiencies of road microplastics.

	Arctic	Alps	Himalayas	Greenland	Atlantic Ocean	Pacific Ocean	Indian Ocean	Southern Ocean	Arctic Ocean	Mediterranean Sea	Baltic Sea	South China Sea
PM2.5 TWP	1.2–10 (3.5)	0.11–0.84 (0.30)	0.048–0.45 (0.15)	1.2–9.5 (3.4)	5.4–42 (15)	6.3–57 (19)	1.9–15 (5.3)	0.43–3.4 (1.2)	5.0–39 (14)	0.080–0.72 (0.24)	0.018–0.14 (0.051)	0.19–1.4 (0.52)
PM10 TWP	0.46–3.1 (1.2)	0.22–1.4 (0.56)	0.046–0.31 (0.12)	0.65–4.4 (1.7)	3.7–27 (10)	4.3–34 (12)	1.3–11 (3.9)	0.52–3.8 (1.4)	2.4–19 (6.8)	0.16–1.1 (0.42)	0.024–0.15 (0.061)	0.32–2.0 (0.78)
PM2.5 BWP	2.4–5.4 (3.6)	0.22–0.49 (0.33)	0.078–0.25 (0.14)	2.3–5.1 (3.4)	10–22 (15)	9.1–36 (18)	3.2–8.1 (5.1)	0.60–2.0 (1.1)	10–20 (14)	0.13–0.48 (0.25)	0.041–0.069 (0.053)	0.41–0.59 (0.49)
PM10 BWP	1.0–1.7 (1.3)	0.50–0.85 (0.65)	0.078–0.15 (0.11)	1.4–3.7 (2.3)	6.9–18 (11)	5.2–23 (11)	1.2–5.1 (3.2)	0.27–0.94 (0.52)	2.7–6.7 (4.3)	0.35–0.58 (0.45)	0.055–0.092 (0.071)	0.65–1.3 (0.91)

Transport efficiencies (%) of road microplastics over remote areas. Transport efficiency is defined as the ratio between the mass of microplastics deposited in a remote region divided by the total mass of microplastics emitted globally (see "Discussion"). Results are given as geometric means (presented within parentheses) that are based on simulations from different scenarios for the airborne fraction in the emissions (five for each of the PM2.5 and PM10 fractions), different particle size distribution for transport (eight for each of the PM2.5 and PM10 fractions) and different CCN/IN (cloud condensation nuclei/ice nuclei) efficiency for deposition (three different sets of scavenging coefficients per fraction). Uncertainties have been calculated as the geometric standard deviation of all simulations (120) using the aforementioned assumptions and they are expressed as ranges of the geometric standard deviation from the geometric mean (see "Methods").

PM10 BWPs is ~80% and PM2.5 is 40–50% of total BWPs independently on vehicle type[77]. Here, we assumed that 2.5, 5, 10, 20 and 40% of the total TWPs and 60, 70, 80, 90 and 100% of the total BWPs is PM10 (Supplementary Table 1) and then calculated the geometric mean and geometric standard deviation. Accordingly, 0.25, 0.5, 1, 2 and 4% of total TWPs and 30, 40, 50, 60 and 70% of total BWPs were assumed PM2.5 (Supplementary Table 1), based on the range of values reported in the literature (Table 3.96 and 3.97 in Klimont et al.[77] and references therein).

Atmospheric transport modelling of road microplastics. The gridded TWP emissions were adopted to the Lagrangian particle transport model FLEXPART (FLEXible PARTicle Dispersion Model) version 10.4[80–83]. The model was set to run in forward mode for year 2014 with a spin-up period of 1 month (December 2013). Boundary layer turbulent mixing and convection processes affecting particle transport in clouds are parameterized in the model[80,84]. The model was driven by 3-hourly 1° × 1° operational analyses from the European Centre for Medium-Range Weather Forecast, the spatial output resolution of concentration and deposition fields was set to 0.5° × 0.5° in a global domain with a daily temporal resolution.

FLEXPART assumes a spherical shape of particles[80] and, as such, the dispersion of road microplastics was modelled. One of the most uncertain aspects of the TWP and BWP emissions is their size distribution. It depends on different properties of the tyre, driving operation and composition and texture of the pavement[21]. Mathissen et al.[85], Sanders et al.[74] and Kumar et al.[73] reported that TWP and BWP can be even emitted as ultrafine particles due to thermomechanical processes. A bimodal size distribution for TWP has been suggested with one maximum in the fine mode and another in the coarse mode[86–89]. On the contrary, an unimodal size distribution has been reported for BWP with maxima ranging between 1.0 and 6.0 μm[23,30,41,74,90] (Supplementary Fig. 4).

Model simulations were carried out for each of the above emission scenarios (five). However, since also the size distribution within the PM10 and PM2.5 modes is uncertain, we simulated particle transport for three different particle sizes in the PM2.5 (0.5, 1.0 and 2.1 μm) and five in the PM10 mode (0.5, 2.1, 3.2, 6.0 and 9.5 μm) and applied a range of different a posteriori weightings of these size classes. The respective eight mass fractions for TWPs and BWPs used in FLEXPART are shown in Supplementary Fig. 4.

Yet another source of uncertainty is the efficiency with which particles are scavenged by precipitation. Plastics are generally hydrophobic and should therefore be rather inefficient cloud condensation nuclei (CCN)[91,92]. However, as known for BC, coatings may make the particles more hydrophilic with time in the atmosphere[49]. The efficiency of aerosols to serve as ice nuclei (IN) is also not well known. To bracket this type of uncertainty in our simulations, we accounted for three different in-cloud scavenging properties (low, medium and high CCN/IN efficiency, Supplementary Table 2) in each of the aforementioned particle sizes. We report simulated concentrations and deposition amounts as the geometric mean values of the 120 ensemble members (five assumptions of the airborne fraction, eight for the size distribution, three for the CCN/IN efficiency) and quantify their uncertainty as their geometric standard deviation.

The simulations also accounted for below-cloud scavenging and dry deposition, assuming a particle density for TWPs of 1234 kg m^{-3}, which is in the middle of the densities of 945 kg m^{-3} for natural rubber and 1522 kg m^{-3} for synthetic rubber[93,94]. This density is within the reported range for microplastics (940–2400 kg m^{-3})[95]. For BWPs a higher density was assumed (2000 kg m^{-3}) considering that BWP may also contain metals[22]. These values were held constant for all ensemble members.

Atmospheric lifetimes of road microplastics. The modelled lifetime (τ) of of road microplastic particles in each grid cell is identical to the lifetime due to transport (t_{trans}) in and out of the aforementioned grid cell, chemical loss (t_{chem}) and deposition (t_{depo})

$$\frac{1}{\tau} = \frac{1}{t_{\mathrm{trans}}} + \frac{1}{t_{\mathrm{chem}}} + \frac{1}{t_{\mathrm{depo}}}.$$

For road microplastics, we assume no chemical interactions of TWPs and BWPs. The modelled lifetime in FLEXPART can be written by the species mass balance equation of Croft et al.[96] as follows:

$$\frac{\mathrm{d}\mathbf{C}(t)}{\mathrm{d}t} = \mathbf{S}(t) - \frac{\mathbf{C}(t)}{\tau(t)},$$

where $\mathbf{C}(t)$ is the atmospheric burden of road microplastics at time t, $\mathbf{S}(t)$ is the time-dependent source emission fluxes and $\tau(t)$ is the removal timescale. Assuming steady-state conditions and considering that emission fluxes are continuous, there is a quasi-equilibrium between sources and removals; hence, the modelled lifetime τ_{mod} can be defined as:

$$\tau_{\mathrm{mod}} = \mathbf{C}_{\mathrm{mp}} / \mathbf{L}_{\mathrm{mp}}^{\mathrm{trans,chem,depo}},$$

where $\mathbf{C}_{\mathrm{mp}}$ is the atmospheric burden of road microplastics and $\mathbf{L}_{\mathrm{mp}}^{\mathrm{trans,chem,depo}}$ is the total loss due to any process affecting TWPs and BWPs in the model (transport, chemistry, deposition).

Statistics and uncertainty calculations. We plot the probability density functions (PDFs) for deposition of TWPs and BWPs that resulted from all the ensemble members of our sensitivity in Supplementary Fig. 5. In the present case, five ensemble members represented the uncertainty in the emissions (Supplementary Table 1), eight that are in the size distribution (Supplementary Fig. 4) and three members that are in the CCN/IN efficiency (Supplementary Table 2), which gives a total of 120 ensemble members for each size (PM2.5 and PM10). Supplementary Figure 5 shows that deposition follows a log-normal distribution with a PDF that can be expressed as follows:

$$f\left(\chi,\mu_g,\sigma_g\right)=\frac{1}{\chi\sigma_g\sqrt{2\pi}}\exp\left(\;\frac{(\ln x\quad\mu_g)^2}{2\sigma_g^2}\right),$$

where χ is the random variable, and μ_g and σ_g are the mean and standard deviation of the distribution of $\ln\chi$. This relationship is true regardless of the base of the logarithmic or exponential function[97]. Thus, the results can be expressed by the geometric mean (μ_g) and the uncertainty by the geometric standard deviation (σ_g) of χ, which are given below:

$$\mu_g=\sqrt[N]{\mathbf{A}_1\mathbf{A}_2\ldots\mathbf{A}_\mathbf{N}}\text{ and }\sigma_g=\exp\left(\sqrt{\frac{\sum_{i\;1}^{N}\left(\ln\frac{A_i}{\mu_g}\right)^2}{N}}\right),$$

where $\mathbf{A}_1,\mathbf{A}_2,\ldots,\mathbf{A}_\mathbf{N}$ are the results from each ensemble member and N the size of the ensemble (120 members for the PM2.5 and PM10 mode, respectively, for each of the TWP and BWP).

The geometric standard deviation is a dimensionless multiplicative factor, also called geometric SD factor[98]. We present resulting concentrations and deposition here with geometric SD factor in conjunction with geometric mean as "the range from "the geometric mean divided by the geometric SD factor" to "the geometric mean multiplied by the geometric SD factor", rather add/subtract "geometric SD factor" to/from "geometric mean"[99].

Data availability

All primary sources (TWP and BWP emission data) are publicly available in https://doi.org/10.5061/dryad.qrfj6q5bx (temporary link: https://datadryad.org/stash/share/ dEIxj28-AHDSoIEPIRdufbljARA-NLyKYOs2n2CqqE). FLEXPART version 10.4 model is publicly available[80]. Operational meteorological data that were used in FLEXPART version 10.4 model can be downloaded directly from the European Centre for Medium-Range Weather Forecasts (ECMWF, https://www.ecmwf.int) following their rules and regulations. All FLEXPART version 10.4 simulation results can be found in https://doi.org/10.5061/dryad.qrfj6q5bx or upon request to N.E. The same dataset also contains land–sea, ocean, continental and country masks (Supplementary Fig. 6) that were used in the calculations of continental emissions, oceanic deposition and transport efficiencies, together with the ECMWF data of sea-ice area fraction, snow depth (for the definition of mountains), snowfall and total precipitation that were used in the calculations of snow concentrations.

Code availability

The Lagrangian particle dispersion model FLEXPART is open access and can be obtained from https://www.flexpart.eu. All figures were created using open access Python 3 programming language. The post-processing codes can be obtained from the corresponding author upon request.

Received: 16 March 2020; Accepted: 18 June 2020;

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Acknowledgements

This work was supported by COMBAT (Quantification of Global Ammonia Sources constrained by a Bayesian Inversion Technique) funded by ROMFORSK—Program for romforskning of the Research Council of Norway (Project ID: 275407), website: https://www.forskningsradet.no/prosjektbanken/#/project/NFR/275407.

Author contributions

N.E. performed the simulations, analyses, wrote and coordinated the paper. H.G. and S.L.-A. made the calculations of TWP emissions with the CO_2 ratio method, Z.K. and C.H. calculated TWP and BWP emissions with the GAINS model, S.E. helped in the FLEXPART version 10.4 simulations and A.S. wrote and commented on the manuscript. All authors contributed in the final version.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41467-020-17201-9.

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Peer review information *Nature Communications* thanks Deonie Allen, Sebastian Primpke, and other, anonymous, reviewer(s) for their contributions to the peer review of this work. Peer review reports are available.

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Tamis *et al. Microplastics and Nanoplastics* (2021) 1:10
https://doi.org/10.1186/s43591 021 00008 w

Microplastics and Nanoplastics

RESEARCH ARTICLE Open Access

Environmental risks of car tire microplastic particles and other road runoff pollutants

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Abstract

Tire wear represents a large source of microplastic entering the aquatic environment, however little is known about its environmental risks. Here, we provide the first assessment of the environmental risks of pollution with tire wear microplastic particles (TWP) and associated organic micropollutants present in road runoff in Europe, in one go. Besides microplastic TWP, the assessment focused on priority substances as defined by the Water Framework Directive (WFD). In addition, several other pollutants (mercaptobenzothiazole, tolyltriazole, diisodecyl phthalate and hexa (methoxymethyl)melamine) were included. The risk assessment comprised a hazard identification (selection of traffic related substances), an assessment of exposure (Predicted Environmental Concentrations, PECs), based on estimated and measured values, effect assessment (selection of Predicted No Effect Concentrations, PNECs, and effect values) and a risk characterization (PEC/PNEC and Species Sensitivity Distributions (SSDs)). Whole Effluent Toxicity (WET) tests on samples taken from road runoff, surface water and sediment were conducted as a retrospective approach to support the risk assessment. We demonstrate that risks exist for TWP and for several TWP associated chemical substances in surface water and sediment. In addition, WET tests of the runoff samples showed significant dose related effects for algae. However, WET tests of surface water showed no significant toxic effects. The present study provides opportunities to protect the quality of European waters from complex road runoff pollution, focusing on TWP microplastic, their associated WFD priority substances and other hazardous substances.

Keywords: Tire wear particles, Road runoff, Microplastic, Organic micropollution, Environmental risk assessment, Whole effluent toxicity test

Introduction

Microplastics are of increasing concern in the environment [1, 2]. Tire wear is estimated to be one of the largest sources of microplastics entering the aquatic environment [3–7]. The mechanical abrasion of car tires by the road surface forms tire wear particles (TWP) [8] and/or tire and road wear particles (TRWP), consisting of a complex mixture of rubber, with both embedded asphalt and minerals from the pavement [9]. In our study we focus on tire wear particles as a component of microplastics in the aquatic environment. Therefore the abbreviation TWP is used to indicate particles originating from road-generated tire abrasion. Small TWP are typically emitted into the air and prone to air dispersal, whereas large TWP will get deposited on the road surface where some parts will get trapped and other parts will be transported by rainwater runoff into soils, sewers and/or surface waters [4]. TWP are present in all environmental compartments, including air, water, soils/sediments, and biota [10]. Pure TWP are rarely found in the environment as it is mostly of mixed composition, i.e. consisting of tire and other road related wear particles [11]. Hence, the term TRWP may be more appropriate when referring to tire wear particles in the environment

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[12]. Recently, a nomenclature on anthropogenic polymer particulates has been proposed, introducing microrubber (MR) as an umbrella term covering all micronized rubber particles such as T(R) WP [13]. TWP contain rubber polymers (about 50%) and other components such as fillers and softeners [7, 11, 12]. Road runoff including TWP has been studied extensively over the past decades, addressing characteristics, environmental concentrations and effects of TWP [9, 10, 14–17], with an additional focus on (effects of) leaching of heavy metals and other trace elements [18–34]. Leachability of toxic compounds differs greatly among different tires (i.e. rubber formulas) [26] and different tires were found to vary in toxicity by 2 orders of magnitude [23]. Tire components may also form toxic transformation products. Recently, a previously unknown transformation product of a globally ubiquitous tire rubber antioxidant was identified as the primary causal toxicant for decades of stormwater-linked salmon acute mortality observations in the U.S. [35]. Furthermore, toxicity of TWP leachates is also related to the leaching methods used to prepare and extract the tire particles [15]. Harmful effects from TWP leachates were observed on aquatic organisms including algae, crustacean and fish, although significant effects were not always determined [9, 15, 17, 21, 26]. The acute effect concentrations of TWP leachates (TWP removed) in aquatic media, including marine environments, were found to cover a range of 25 to 100,000 mg TWP/L [8, 36], while chronic effect concentrations vary from 10 to 3600 mg TWP/L [8]. A Predicted No-Effect Concentration for water (PNEC water) of 3.9 mg TWP/L was derived based on TWP leachate [10]. Although expressed in TWP mass, these effect values are all based on the chemicals leaching from TWP and not on the particles themselves. Components of leachates found to be largely responsible for the toxicity were zinc and organic compounds [17, 26].

The briefly described literature above indicates an extensive knowledge base is available for TWP leachates. For TWP as a component of microplastics, knowledge on the concentrations, degradation, retention and toxicity of in the aquatic environment is however limited [12, 37, 38]. Due to interactions with other particulate matter, TWP are subjected to changes in morphology and composition, which makes it difficult to track and quantify them in the environment [8]. Environmental concentrations of TWP have been measured using markers [16, 39], i.e. tire constituents which occur in specific concentrations in tire material [8]. This approach is limited due to multiple sources of most available marker substances and variations of the marker content in the tires [8]. The presence of actual TWP was first detected as airborne particles, e.g. [11], indicating tire wear as a significant contribution to the flux of microplastics into the environment. Recently, TWP have been measured in stormwater [40], sediment and surface water [41], confirming that road runoff likely is an important pathway of microplastic pollution. Therefore, there is a need to evaluate the effect of TWP on biota and to assess potential ecological risks posed by TWP.

Effects in the aquatic environment may stem from TWP itself or from compounds released from TWP [8, 42–44]. Furthermore, other traffic related sources (brakes, lubricants, coolants, asphalt, road marking and vehicle parts) may also release microplastics and other pollutants [4, 29, 45–59]. A wide range of species are capable of ingesting microplastics and 'food dilution' has been found to be the effect mechanism with the highest weight of evidence [60, 61]. Considering TWP, ingestion by aquatic species has recently been confirmed and a dissimilar mechanism of toxicity of TWP and leachate was suggested [44]. Therefore, when addressing risks of microplastics from road runoff, one needs to consider the toxicity of the microplastic particles themselves, the toxicity of tire compounds which may or may not leach out of the particles, as well as the toxicity of other pollutants in road runoff. With a few exceptions [42–44], available research on TWP has been limited to forced TWP leachates, thus disregarding the actual particles [8]. To date, we are not aware of studies that assess the risks of TWP including both the chemical as well as the particle effect modes of action in one go.

This study aims to provide insight in risks of pollution in road runoff for European waters by conducting a risk assessment for the components in road runoff that can be considered hazardous. Following Diamond et al. [62] the assessment comprised of a generic prospective assessment using conservative assumptions to identify potential ecological risks (Predicted environmental concentration (PEC) / Predicted no-effect concentration (PNEC) ratio), followed by a higher resolution prospective assessment (Species Sensitivity Distributions (SSDs) used to estimate the Potentially Affected Fraction (PAF) of species) and then by a retrospective assessment (Whole Effluent Toxicity (WET)-tests). In relevance to European water quality, the prospective risk assessment was focused on WFD priority substances (Directive 2013/39/EU), supplemented with other hazardous substances found in road runoff. As the number of different compounds potentially present in road runoff is high (306 compounds [29]), a selection was made of 10 substances and TWP itself to be included in the risk assessment. Estimated and measured exposure concentrations (PECs) of these pollutants reported in literature were compared to the sensitivity of the environment as represented by 1) PNECs and 2) SSDs [63]. Due to lack of effect data for TWP as a specific subcategory of microplastic, we used generic microplastics data as a proxy to estimate their particle related effects and hence use the term TWP_{MP} (Tire Wear Particles - Microplastic). Whole Effluent Toxicity (WET)-tests using bacteria, algae and crustacea were conducted to support the risk assessment in retrospect [62]. With this retrospective assessment realistic field samples are tested and combination toxicology as well as yet unidentified substances are covered.

Methods

To provide insight in risks of pollution in road runoff for European waters, a risk assessment of individual substances in road runoff was conducted. This risk assessment comprised hazard identification (selection of substances), exposure assessment, effect assessment and risk characterization [64]. Following Diamond et al. [62], this prospective risk assessment was combined with a retrospective assessment in the form of Whole Effluent Toxicity (WET)-tests on representative environmental samples. The prospective risk assessment is used to identify potential ecological risks whereas the retrospective assessment provides additional information that integrates the complexity of real mixtures.

Selection of substances

The group of microplastics, or more specific TWP_{MP}, was selected as primary focus of this study. We use the term microplastics to refer to the group of microplastic particles in general, with particle sizes ranging from 1 to 5000 μm [61, 65, 66], whereas TWP refers to the particles from tires with sizes within the range of microplastics [14]. Besides microplastic particles, tire compounds and other pollutants in road runoff needed to be identified for inclusion in the risk assessment. To select a most relevant and also manageable number of TWP-associated compounds for the risk assessment, 10 compounds were selected from a list of organic micropollutants that are reported to be released from traffic and roads (Supporting information: Table S1 to S3, page 4–12). Selection criteria were as follows. First, compounds listed as priority substance under the Water Framework Directive (WFD 2000/60/EC) were selected: Benzo(a) pyrene (BaP); Fluoranthene; Nonylphenol (NP); 4-tert-octylphenol (OP); Di (2-ethylhexyl) phthalate (DEHP). One substance, Bisphenol A (BPA), is considered to be a potential candidate for eventual priority substance status under article 16 of the WFD and was also selected. Four other substances were selected based on the following criteria (1) concentrations in runoff are reported in literature, (2) the pollutants originate from a variety of sources and (3) the pollutants are expected to dominate risk, based on the assessment by Baun et al. [67] and Markiewicz et al. [29]. The latter assessment prioritizes pollutants with properties such as: low volatility; persistent; risk for bioaccumulation; risk for toxicity; and long-term adverse effects. These four substances are Mercaptobenzothiazole (MBT); Tolyltriazole (TT); Diisodecyl phthalate (DIDP); and Hexa (methoxymethyl) melamine (HMMM).

Exposure assessment

Measured values reported in literature in runoff or storm water were taken as the Predicted Environmental Concentration (PEC) in runoff (all values found in literature are provided as Supporting information, Table S4 on page 13–15).

For the PEC in surface water, both estimated (extrapolated from runoff concentrations) and measured values (reported in literature) were used. For the estimated values it was assumed that the concentration in a water body equals the runoff concentration divided by 100. When more than one PEC in runoff was available, the maximum (worst case) value was taken for the derivation of the PEC in surface water. The dilution factor of 100 is derived for a typical small surface water body (2 m wide and 1 m depth) with a flow of 1 m^3/sec, combined with a typical discharge from a rain event of 30 mm in 1 h on a road of 100 m long and 12 m wide (resulting in a discharge of 0.01 m^3/sec). For relatively small and/or stagnant surface water bodies that are mainly loaded with road runoff water, the dilution factor of 100 may be too high. For those water bodies, the risk assessment based on runoff concentrations is more relevant. The estimation was only applied for the 10 substances, as the dilution assumption does not apply for particles (TWP). The estimated TWP concentration is based on a conceptual model of TRWP fate [37].

For the PEC of sediment, the same approach was used as for surface water. The estimated value for each of the 10 substances was extrapolated from measured values in the solid fraction of runoff by using a dilution factor of 100. In case no concentration in runoff sediment was available, a fixed ratio of 10 between the PEC surface water and PEC sediment was assumed, based on the ratio between surface water concentrations and sediment concentrations of phenols and phthalates in Björklund et al. [68]. A list of estimated and measured values in surface water and sediment is provided as Supporting information (Tables S4 and S5 on page 13–16).

Effect assessment

PNEC selection or derivation

The PNEC is defined as the concentration of the substance below which adverse effects in the environmental sphere of concern are not expected to occur (Regulation (EC) No. 1907/2006). For each substance, a PNEC water and PNEC sediment was selected from (in order of priority):

- Existing EU standards, i.e. Environmental Quality Standards (EQS). EQS are available for priority substances (Directive 2008/105/EC and related documents);
- PNEC values reported in EU Risk Assessment Reports;
- PNEC values reported on the ECHA website (https://echa.europa.eu/substance-information/);
- PNEC values reported in literature.

In case there was no PNEC value available from the above sources, a PNEC was derived using available toxicity information in combination with an appropriate safety factor as indicated by the EU Technical Guidance Document [64]. Therefore a search was conducted in the US-EPA ECOTOX database (https://cfpub.epa.gov/ecotox/), which is a comprehensive, publicly available knowledgebase providing single chemical environmental toxicity data. If toxicity data was still limited, a search was performed in peer-reviewed literature using the search engine SCOPUS (www.scopus.com) and, if necessary, in grey literature (e.g. using google-scholar). In case a PNEC sediment was not available in literature and toxicity data for benthic species was lacking, the PNEC sediment was estimated based on equilibrium partitioning [64] of the organic compounds. All PNECs and their literature sources or derivation (including chemical equilibrium equations) are provided and explained in detail as Supporting information (page 17–22).

Effect data search

A search for No Observed Effect Concentration (NOEC) values was conducted to provide input for the SSD approach (see Risk characterization below). To enlarge the availability of ecotoxicological data we also searched for effect concentrations up to 10% (EC_0 to EC_{10}, with EC_x as the effect concentration at which x% effect (mortality, inhibition of growth, reproduction, etc.) is observed compared to the control group) to represent NOEC values, and for acute EC_{50} and LC_{50} (Lethal Concentration 50%) values. For the latter values, a pragmatic acute to chronic ratio of 10 was used [69] to represent the NOEC. As toxicity values for sediment are scarce, we limited our search to the water phase.

The US-EPA ECOTOX database (https://cfpub.epa.gov/ecotox/) was searched using the CAS numbers of the 10 selected substances. The search results (Table S15) were checked for relevance according to the following criteria:

- Only relevant endpoints (NOEC; $EC/LC_{0\ \text{to}\ 10}$; EC_{50}, LC_{50});
- Concentration must be expressed as environmental concentration (i.e. exposure: mg/l, μg/l) and not in food or organism (i.e. dosage: mg/kg bw etc.);
- Only exact numerical values were selected, in other words, effect concentrations reported as: 'NR'; greater than ('>'); smaller than ('<'), or approximate ('~') were not included.

Risk characterization

The risk assessment followed two approaches: 1) calculation of the PEC/PNEC ratio, indicating whether unacceptable effects on organisms are likely to occur and; 2) comparing the PEC with an SSD, to indicate the probability that a specific fraction of species is exposed above their no effect value; the Potentially Affected Fraction of species (PAF) [70]. The SSD approach uses the lowest available NOEC per species [71–74]. When more than one value was found for the same species under similar conditions, i.e. same end-point and an analysis of the test conditions used cannot explain the difference in observed response, the geometric mean of these values was used. The minimum number of species required when using the SSD method is 10 (preferably more than 15) covering at least 8 taxonomic groups [64]. Because of these requirements and acknowledging the scarcity of sediment toxicity values, the probabilistic risk assessment was focused on the water phase only.

When sufficient NOECs were available for a substance, an SSD was constructed, by using the software ETX 2.1 [75] which is freely available at https://rvs.rivm.nl/risicobeoordeling/modellen-voor-risicobeoordeling/ETX. ETX 2.1 applies a cumulative log-normal distribution, where sensitivity values for species are fitted to a logarithmic scale. The SSD was used to estimate the PAF, see e.g. European Commission [64] and Aldenberg & Slob [76]. Using the SSDs, the PAF at the exposure concentration (PEC) is estimated as a median estimate (50% confidence), plus lower estimate (5% confidence) and upper estimate (95% confidence) of the fraction affected.

Whole effluent toxicity (WET)-tests

Additional to the risk assessment, WET-tests were executed to assess the toxicity of runoff and of surface water next to a highway. In contrast to our prospective risk assessment, the WET-tests constitute a retrospective, empirical approach [62].

Road runoff samples were taken in Germany and Sweden. In Germany, on 18 March 2019 runoff samples have been gathered from the highway A61 between Kreuz Meckenheim and Dreieck Bad Neuenahr-Ahrweiler (at the parking place 'Goldene Meile', Coordinates: 50.58 N, 7.06 E). On the sampling day, it was partly cloudy and there was some precipitation (0.4 mm in Köln-Bonn Flughafen) with a maximum temperature of 10 degrees Celsius. There was more precipitation registered in Köln-Bonn Flughafen on the days before the sampling day (2.3–18.2 mm per day in the 5 days before the samples were gathered). Highway A61 is a busy highway with 5 lanes in total and an emergency lane with an average traffic intensity of 73,310 vehicles per day. The asphalt consists of normal asphalt. In Sweden, on 14 June 2019 samples have been gathered from the Swedish National Road and Transport Research Institute (VTI) test site located at the highway E18 (Coordinates: 59.63 N, 16.86 E). Water samples were collected from a concrete storm water well containing road runoff water. On the sampling day, it was cloudy at first (no rain), and later sunny, with a temperature of 18–21 degrees Celsius. In the days before the sampling, there was some rain measured at weather station Enköping Mo (0–13 mm per day in the 5 days before the sampling). Highway E18 is a highway with 2 lanes in each direction (4 lanes in total) and no emergency lane, with an average traffic intensity of 21,300 vehicles per day and the asphalt consists of stone mastic asphalt. Surface water samples have been taken in the Netherlands, in a surface water body next to the A2 highway (Coordinates: 52.24 N, 4.98 E), on 17 April 2019. Samples were collected from the center of the water body; 10 l surface water just under the surface at approx. 30 cm depth. On the sampling day, the weather was partly cloudy (no rain) with a maximum temperature of 16 degrees Celsius. The last rain event occurred 2 weeks before the sampling. It should be realized that the surface water samples reflect time integrated concentrations from emission pulse loadings (runoff events) already diluted and transported in the receiving environment. This renders a longer term interpretation and does not include such short term weather dynamics. Highway A2 is a busy highway with 5 lanes in each direction (10 lanes in total) and an emergency lane with an average traffic intensity of 190,000 vehicles per day. The asphalt consists of porous asphalt, which reduces the amount of pollutants that could reach the surface water body. To maintain the in situ state of the compounds, samples were preserved as follows. All samples were stored in green/brown colored glass bottles or jars in the dark at 4 ± 3 degrees Celsius. This implies that degradation by UV and changes in toxicity profiles were prevented. Sample preservation by use of biocides was avoided because that would change the toxicity of the samples. During transport the samples were cooled with cooling elements. The samples arrived at the laboratory and were tested in August 2019. At arrival the samples were stored at 7 °C. Before being used in the WET-test, the temperature was gradually brought back to room temperature.

WET-tests enable the evaluation of combination toxicity and yet unidentified toxicants in a natural matrix. Fresh water WET-tests were performed for species from multiple tropic levels, namely bacteria (*Vibrio fischeri*), algae (*Raphidocelis subcapitata*) and crustacea (*Daphnia magna*). The samples contained some solids, which easily settled out. To avoid interference with the measurements, the overlying water was used (elutriate) after settling out for 24 h. The tests were done on dilution series of these overlying water samples. In the algal growth inhibition test (ISO guideline 8692 (2012)), adapted for micro volumes [77] and the *Daphnia* immobilization test (ISO guideline 6341 (2012)), the concentration series used (expressed as % of the chemical concentration in the original sample) was 0% (blank), 31.6%, 42.2%, 56.2%, 75% and 100%, assuming toxicity will be moderate to low. For the Microtox test with *Vibrio fischeri*, the range was adapted to meet the requirements of the Microtox test, with a maximum of 45% sample due to dilution of the sample with the bacterial suspension (ISO guideline 11,348–3 (2007)). Experimental details of the WET-tests are provided as Supporting information (page 2 and 3).

The NOEC is derived from the data noting that the effect at the NOEC should not exceed 10% of the tested population [64]. The EC_{50} was calculated using a 'sigmoidal dose-response curve' with variable slope and is based on the effect in the test concentrations relative to the blank condition.

Results

Exposure assessment

The average, minimum and maximum values of the reported concentrations in runoff found in literature were used as PECs runoff (Table 1). For the PEC in surface water and sediment, both estimated (extrapolated from runoff concentrations) and measured values (reported in literature) were used (Table 2).

Effect assessment

PNEC selection or derivation

For all organic substances, $PNEC_{water}$ and $PNEC_{sediment}$ values were taken from literature or were derived (Table 3). For five substances (BaP, Fluoranthene, NP, OP, DEHP), EQS were available, which apply under the WFD for surface waters in Europe [96]. For most other organic substances PNEC values were available in the literature. Exceptions are the $PNEC_{water}$ and $PNEC_{sediment}$ of DIDP and the $PNEC_{sediment}$ of HMMM, which were derived by using ecotoxicity values from literature combined with an assessment factor and/or the equilibrium method as described in the Supporting Information (page 18–20). For TWP, PNEC values including the particle effect modes of action were not available. Therefore PNEC values for microplastics (MP) available in the

Table 1 Predicted environmental concentrations (PECs) in runoff (water phase and solids) based on measured values (average, minimum and maximum) reported in literature (see Supporting information, Table S4)

Pollutant[b]	Runoff water (μg/l)	Runoff solids ($\mu g/g_{dw}$)	Reference
TWP	29,737 (975 58,500)	148,040 (520[a] 390000[a])	[78 80]
BaP	0.28 (0.0008 0.83)	0.58 (0.21 0.94)	[78, 81]
BPA	0.23 (0.03 0.55)	0.15 (0.06 0.24)	[78, 82]
DEHP	1.22 (0.66 2.27)	55.30 (2.44 98.00)	[78, 83]
DIDP	3.93 (0.60 8.60)	72.10 (4.61 139.59)	[78, 83]
Fl.	1.22 (0.003 3.65)	1.36 (0.30 2.41)	[78, 81]
HMMM	2.32 (0.88 3.89)	0.017 (0.002 0.032)	[78, 84]
MBT	0.043 (0.010 0.110)	0.60 (0.19 1.01)	[78, 85]
NP	0.13 (0.01 0.36)	1.03 (0.001 3.1)	[78, 82]
OP	0.091 (0.016 0.197)	0.99 (0.53 1.45)	[78, 82]
TT	0.90 (0.010 2.30)	0.57 (0.039 1.10)	[78, 85]

[a] Concentration in solids reported in literature in ww and here converted to dw using a factor of 2.6

[b] Pollutants: *BaP* Benzo(a) pyrene, *BPA* Bisphenol A, *DEHP* Di (2 ethylhexyl) phthalate, *DIDP* Diisodecyl phthalate, *Fl.* Fluoranthene, *HMMM* Hexa (methoxymethyl) melamine, *MBT* Mercaptobenzothiazole, *MP* Microplastics, *NP* Nonylphenol, *OP* 4 tert octylphenol, *TWP* Tire Wear Particles, *TT* Tolyltriazole

literature were used to represent the PNEC for TWP, i.e. TWP_{MP}.

Effect data search

A total of 63 effect values were gathered for microplastic (Supporting information, Table S15) which were used to derive the SSD for TWP_{MP} (Supporting information, Figure S1). For the organic micropollutants a total of 1823 effect values were gathered (Supporting information, Table S16) which were used to derive the SSDs for BaP, BPA, DEHP, Fluoranthene, NP, and OP (Supporting information, Figures S2-S7). Only a few values were found for DIDP and TT and for HMMM there were no values available. No SSDs could be derived for these substances.

Risk characterization

The PNEC values have been compared with concentrations, in runoff (Fig. 1) and in surface water and sediment (Fig. 2) in order to derive an indication of risk (PEC/PNEC ratio). A PEC/PNEC ratio higher than 1 indicates that unacceptable effects on organisms are likely to occur; the higher the ratio, the more likely that unacceptable effects may occur [64]. For road runoff water, the maximum PEC/PNEC is higher than 1 for TWP_{MP}, BaP, DEHP, DIDP, fluoranthene, NP and OP (Fig. 1). For solids in runoff, the PEC/PNEC ratios are higher than 1 for TWP_{MP}, BPA, DIDP, fluoranthene, MBT, OP and TT. For surface water, ratios exceeds 1 for TWP_{MP}, BaP and fluoranthene in water. For sediment, ratios exceeds 1 for TWP_{MP}, OP and TT.

For the probabilistic risk assessment, sufficient data for deriving an SSD was gathered for TWP_{MP}, BaP, fluoranthene, NP, OP, DEHP and BPA (SSDs provided as Supporting information (page 23–30). For MBT, TT, DIDP and HMMM the data availability did not meet the requirements for SSD derivation [64] and therefore the PAF could not be estimated for these substances. Using the SSDs, the PAF at the exposure concentrations (PECs) is estimated as a median estimate (50% confidence), plus lower estimate (5% confidence) and upper estimate (95% confidence) of the fraction affected [75]. All PAF estimates are provided as Supporting

Table 2 Predicted environmental concentrations (PECs) in surface water and sediment based on measured values (reported in literature) and estimated values (extrapolated from maximum runoff concentrations (Table 1)), see Supporting information, Table S5

Pollutant[c]	Surface water (μg/l) measured / estimated	Sediment ($\mu g/g_{dw}$) measured / estimated	Reference (measured value)
TWP	8.00 (6.00 10) / 120	552 (5.98[a] 1900[a]) / 1200[b]	[41, 78, 79]
BaP	0.0001 / 0.0083	0.073 / 0.0094	[78]
BPA	0.01 / 0.0055	0.001 / 0.0024	[78]
DEHP	0.98 / 0.023	10.30 / 0.98	[78]
DIDP	0.001 / 0.086	1.97 / 1.40	[78]
Fl.	0.001 / 0.037	0.17 / 0.024	[78]
HMMM	0.07 / 0.039	0.001 / 0.00032	[78]
MBT	0.010 / 0.0011	0.002 / 0.010	[78]
NP	0.0010 / 0.0036	0.021 / 0.031	[78]
OP	0.01 / 0.002	0.001 / 0.015	[78]
TT	0.010 / 0.023	0.0058 / 0.011	[78]

[a] Concentration in sediment reported in literature in ww and here converted to dw using a factor of 2.6

[b] Not an estimated value, but modelled by Unice et al. [37]

[c] Pollutants: *BaP* Benzo(a) pyrene, *BPA* Bisphenol A, *DEHP* Di (2 ethylhexyl) phthalate, *DIDP* Diisodecyl phthalate, *Fl.* Fluoranthene, *HMMM* Hexa (methoxymethyl) melamine, *MBT* Mercaptobenzothiazole, *MP* Microplastics, *NP* Nonylphenol, *OP* 4 tert octylphenol, *TWP* Tire Wear Particles, *TT* Tolyltriazole

Table 3 PNEC values for microplastics and organic micropollutants in road runoff

Pollutant[c]	PNEC Surface water (μg/l)	PNEC Sediment (μg/kg$_{dw}$)	Reference
TWP$_{MP}$	0.33[a]	100[a]	[65]
BaP	0.00017	1830	[86, 87]
BPA	1.5	63	[88]
DEHP	1.3	100,000	[87, 89]
DIDP	0.6	3300	Derived[b]
Fl.	0.0063	2000	[87, 90]
HMMM	54	133	[91], Derived[b]
MBT	4	147	[92]
NP	0.3	4620	[87, 93]
OP	0.1	1.61	[87, 94]
TT	8	3	[95]

[a] PNEC values for microplastics (MP) available in the literature were used to represent the PNEC for TWP, i.e. TWP$_{MP}$. It should be noted that the PNEC for MP has a limited reliability due to heterogeneity of the tested microplastic considering polymer type, size and shape

[b] PNEC is not available in literature and thus derived using available toxicological data and extrapolation factors (for surface water) or the equilibrium method (for sediment), see Supporting information (Table S6, page 17 22)

[c] Pollutants: *MP* Microplastics, *BaP* Benzo(a) pyrene, *BPA* Bisphenol A, *DEHP* Di (2 ethylhexyl) phthalate, *DIDP* Diisodecyl phthalate, *Fl.* Fluoranthene, *HMMM* Hexa (methoxymethyl) melamine, *MBT* Mercaptobenzothiazole, *MP* Microplastics, *NP* Nonylphenol, *OP* 4 tert octylphenol, *TWP* Tire Wear Particles, *TT* Tolyltriazole

information (Table S14). A PAF of 5% is considered as the threshold value for environmental protection [64]. The average PAF at PECs in runoff based on measured values reported in literature was above 5% for TWP$_{MP}$, BaP, DEHP and fluoranthene (Fig. 3). For surface water, the average PAF only exceeded the threshold for TWP$_{MP}$. This means that unacceptable effects cannot be ruled out for these cases.

WET-tests

WET-tests using bacteria, algae and crustacea were conducted to support the risk assessment, as realistic field samples are tested and combination toxicology as well as yet unidentified substances are covered. The acute luminescence inhibition test with the bacteria *Vibrio fischeri* showed a slight inhibition for the runoff samples from Germany and Sweden at the highest concentration tested (Table 4). However, this was within the normal variation for this test. Due to the need to suspend the bacteria in culture medium, 45% was the highest concentration that could be tested. Consequently, effects at higher sample concentrations could not be assessed using this procedure. The surface water sample from a water body near highway A2, the Netherlands, did not show any inhibition at any of the concentrations tested.

The algae growth inhibition test with *Raphidocelis subcapitata* showed significant dose-related growth inhibition when exposed to the runoff samples (Table 1). The NOEC for runoff from highway E18, Sweden, was established at 42.2% of the original sample concentration. Even more effect was found for highway A61, Germany, where even at the lowest concentration of 31.6%, 84% effect was observed. A NOEC could not be established. The surface water sample from a water body near highway A2, the Netherlands, did not show statistically significant growth inhibition.

The acute immobilization test with the freshwater crustacean *Daphnia magna* showed no effects for any of the three samples (Table 4). The highest effect of 10% was within the normal range of variation for this test. The test results indicate absence of significant toxicity of runoff and surface water samples to the freshwater crustacean *Daphnia magna*.

Discussion

We evaluated aquatic ecological risks of TWP and associated road runoff pollutants by combining a prospective risk assessment with a retrospective assessment in order to test whether the risk identified in the prospective assessment could have deleterious effects on aquatic life [62]. We assessed the prospective risk by comparing predicted exposure concentrations of TWP$_{MP}$ particles and 10 organic micropollutants with: 1) limits below which no adverse effects of exposure in the aquatic environment are expected (PEC/PNEC ratios), and; 2) by assessing the fraction of species exposed to these concentrations above their NOEC (PAF). This prospective risk differentiates between particle effects (TWP$_{MP}$) and chemical effects (organic micropollutants). For the retrospective assessment, combined particle and chemical effects were assessed by conducting WET-tests.

Overview of demonstrated effects and risks

The risk assessment (PEC/PNEC ratios and PAF) shows that for most of the selected substances in surface water and sediment, the risks from road traffic for the

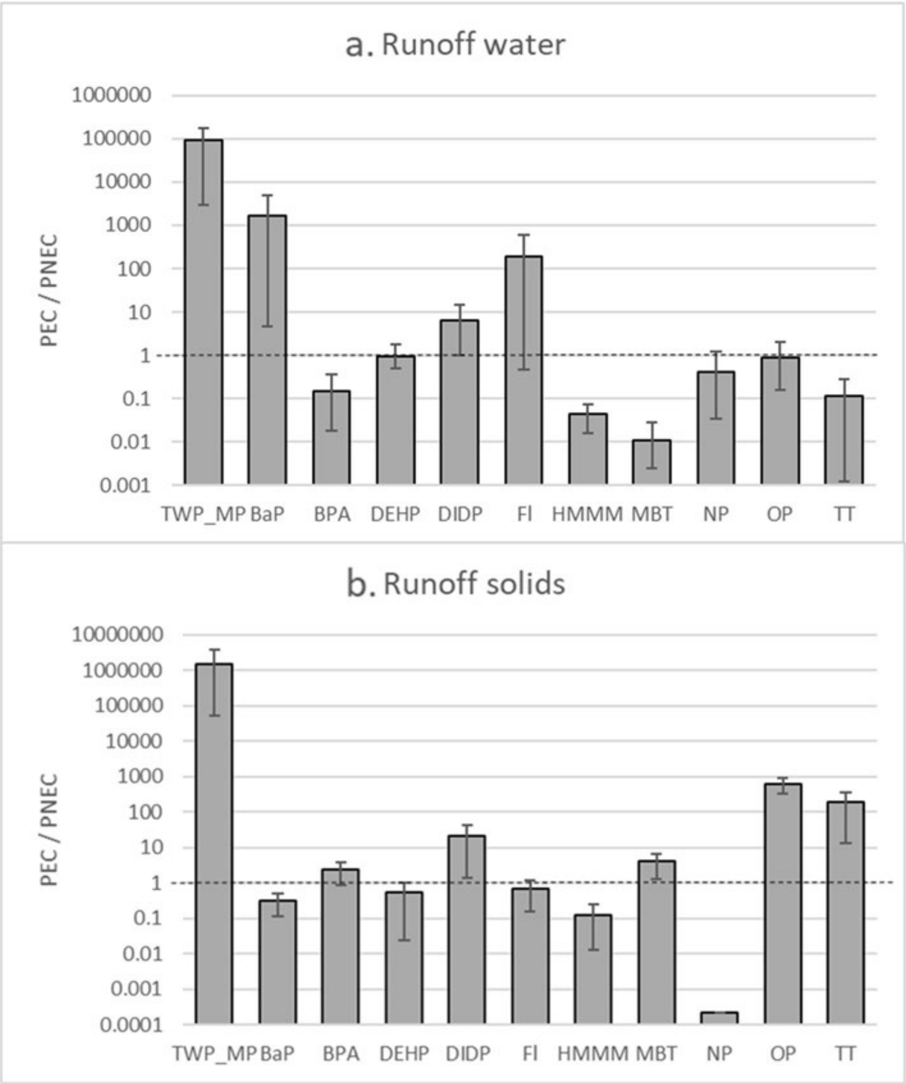

Fig. 1 PEC/PNEC ratios of selected substances in runoff water (**a** top) and solids (**b** bottom). Selected substances: tire wear microplastic particles (TWP MP), benzo(a) pyrene (BaP), bisphenol A (BPA), di (2 ethylhexyl) phthalate (DEHP), diisodecyl phthalate (DIDP), fluoranthene (Fl), hexa (methoxymethyl) melamine (HMMM), mercaptobenzothiazole (MBT), nonylphenol (NP), 4 tert octylphenol (OP) and tolyltriazole (TT). The bar chart shows the average PEC/PNEC based on measured concentrations reported in literature, with the minimum and maximum values represented by the error bars. PEC and PNEC values are provided in Tables 1 and 3, respectively. The dashed horizontal line in the figure marks the threshold indicating unacceptable effects on organisms are likely to occur for PEC/PNEC ratio's higher than 1

investigated European waters are within acceptable limits. However, for a few substances environmental risks have been identified. Based on estimated concentrations in surface water and sediment extrapolated from reported concentrations in runoff, we demonstrate that risks are above threshold values (PEC/PNEC > 1) for TWP_{MP}, BaP and fluoranthene in surface water and for TWP_{MP}, OP and TT in sediment. However, based on reported empirical data, that is, values reported in literature, risks only exist for TWP_{MP} in surface water and TWP_{MP} and TT in sediment. In retrospective, WET-tests of the surface water sample showed no significant toxic effects for bacteria, algae and crustacea.

Concentrations in runoff (water and solids) are much higher than in surface water and sediment and risks (PEC/PNEC > 1 and/or PAF > 5%) cannot be ruled out for most substances. A risk was indicated for TWP_{MP}, BaP, BPA, DEHP, DIDP, Fluoranthene, MBT, NP, OP and TT in road runoff. WET-tests of road runoff showed no significant toxic effects for bacteria and crustacean, but the algae growth inhibition test showed significant dose-related growth inhibition. The latter confirms the outcome of the prospective risk assessment, i.e. that

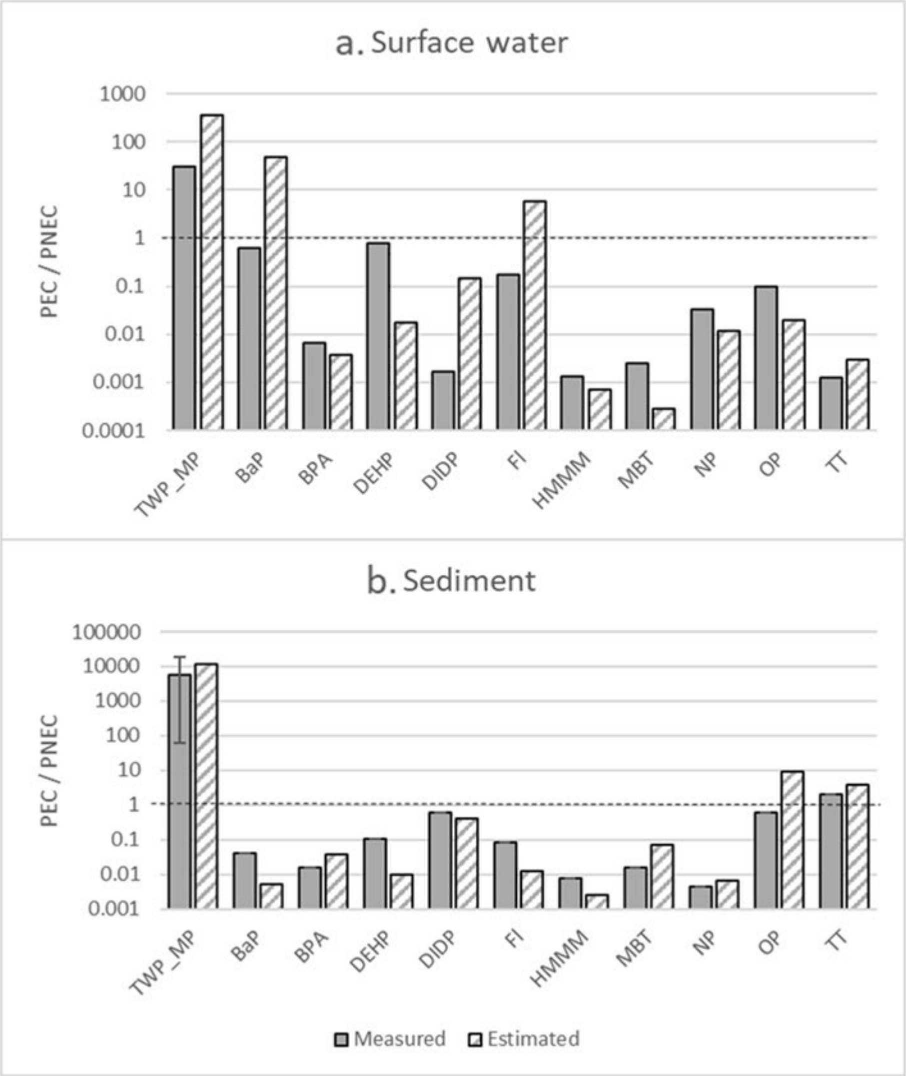

Fig. 2 PEC/PNEC ratios of selected substances in surface water (**a** top) and sediment (**b** bottom). Selected substances: tire wear particles (TWP MP), benzo(a) pyrene (BaP), bisphenol A (BPA), di (2 ethylhexyl) phthalate (DEHP), diisodecyl phthalate (DIDP), fluoranthene (Fl), hexa (methoxymethyl) melamine (HMMM), mercaptobenzothiazole (MBT), nonylphenol (NP), 4 tert octylphenol (OP) and tolyltriazole (TT). PEC and PNEC values are provided in Tables 2 and 3, respectively. PEC values are measured values reported in literature (measured) and extrapolated from values found in literature (estimated). For TWP, the estimated value is the result of modelling [37]. The dashed horizontal line in the figure marks the threshold indicating unacceptable effects on organisms are likely to occur for PEC/PNEC ratio's higher than 1

adverse effects cannot be ruled out for runoff. This could be caused by (the combination of) the substances included in the prospective risk assessment, as well as other runoff components. Other studies have found organic compounds (including a transformation product) and zinc to be largely responsible for the toxicity of TWP leachate [17, 26, 35]. In highway runoff, not only zinc but also copper was found to be the primary cause of toxicity [100]. These main pollutants identified in literature could play a role in the toxicity effects observed by the WET-tests in this study. Concentrations of copper in the runoff samples used for the WET-tests (8.3 and 9.1 μg/l in samples from Sweden and Germany, respectively [78]) are above the lowest NOEC value found for bacteria *Vibrio fisheri* (3.39 μg Cu/l [101]), but below the lowest NOEC value found for crustacea *Daphnia magna* (12.6 μg Cu/l [102] and for algae *Raphidocelis subcapitata* (15.7 μg Cu/l [102]). For *R. subcapitata*, however, a NOEC as low as 4.2 μg Cu/l was suggested under worst case conditions (i.e. considering metal toxicity modifying factors like pH, water hardness and dissolved organic carbon) [103]. For zinc, concentrations in runoff (72 and 230 μg/l in samples from Sweden and Germany, respectively [78]) are well above the lowest NOEC values found for all species used for the WET-tests, i.e. *V. fisheri* (10 μg Zn/l [101]), *R. subcapitata*

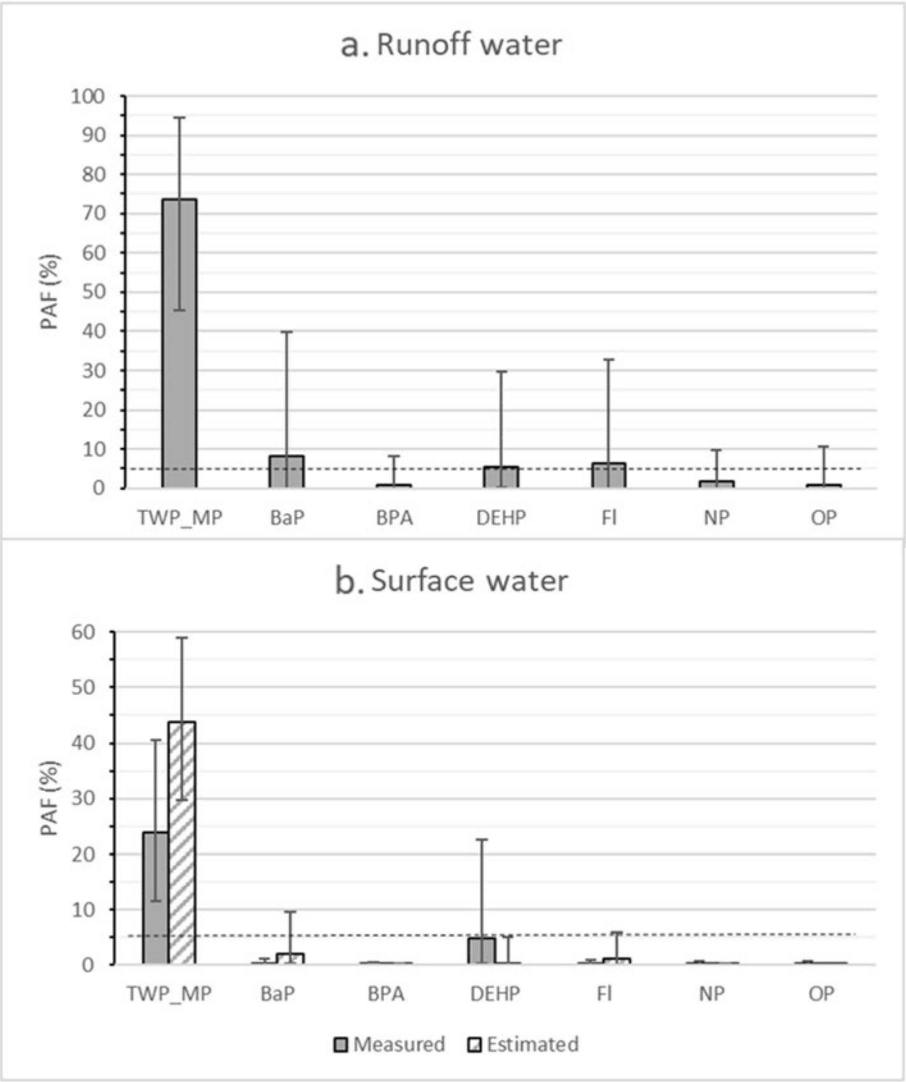

Fig. 3 PAF (%) of selected substances in runoff water (**a** top) and surface water (**b** bottom). Selected substances: TWP MP (Tire Wear Particles, with PEC represented by TWP and SSD by microplastics), BaP (benzo(a)pyrene), BPA (bisphenol A), DEHP (di (2 ethylhexyl)phthalate) Fl (fluoranthene), NP (nonylphenol), OP (4 tert octylphenol). The PAF (Supporting information, page 31 32) is derived by comparing the PEC (Tables 1 and 2) with the SSD (Supporting information, page 23 30). PAF up to 5% is considered acceptable [64], as marked by the dotted line in the figure. The bar chart shows the average of the median estimate PAFs (50% confidence), with the error bars representing the lowest value of lower estimates (5% confidence) and the maximum value of upper estimates (95% confidence)

(21 μg Zn/l [104]) and *D. magna* (86 μg Zn/l [104]), although for *D. magna* only the zinc concentration in the sample from Germany exceeds the NOEC.

Selection of substances

The selection of substances was focused on WFD priority substances and other substances with hazardous properties. For practical reasons, we limited the selection to 10 substances. Although we addressed relevant pollutants, this does not provide a complete risk profile of all runoff components. A total of 306 organic compounds [29] and heavy metals such as Zn, Cu, Ni, and Cd [31] have been identified in road runoff. A prospective risk assessment accounting for more chemicals could lead to a higher estimation of risk. However, by including the WET-tests of realistic field samples as retrospective assessment, the risk assessment covers all runoff components, including possible unidentified substances.

Exposure assessment

Estimated PECs in surface water and sediment were based on the highest reported concentration in runoff combined with a dilution factor (1:100) representative for a small surface water body. However, the measured

Table 4 Summary of WET-test results. Details on the method and results of the WET-tests conducted with runoff samples from highway E18, Sweden and highway A61, Germany and a surface water sample near highway A2, the Netherlands can be found in the technical reports [97 99]. Some endpoints are not available (n.a.), when effect parameters are too low to enable calculation. The concentration is expressed in % sample in test solution

Test	**Endpoint**	**Sample**		
		Runoff, Germany	**Runoff, Sweden**	**Surface water, the Netherlands**
Bacteria	Effect	8.68% effect[a] at highest conc. (45%)	7.06% effect[a] at highest conc. (45%)	no effects
	EC_{50}	> 45%	> 45%	> 45%
	NOEC	n.a.	n.a.	n.a.
Algae	Effect	84% effect at lowest conc. (31.6%)	35% effect at highest conc. (100%)	6% effect[a] at highest conc. (100%)
	EC_{50}	< 31.6%	> 96%	> 96%
	NOEC	< 31.6%	42.2%	n.a.
Crustacea	Effect	0% effect[a] in highest conc. (100%)	10% effect in highest conc. (100%)	10% effect[a] in highest conc. (100%)
	EC_{50}	> 100%	> 100%	> 100%
	NOEC	n.a.	n.a.	n.a.

[a] Within the normal range of variation for this test

PECs exceeded the estimated PECs in many cases (i.e. for all substances in either surface water and/or sediment). Thus, the estimated concentrations do not represent a worst case exposure scenario. This could be caused by the applied dilution factor, assuming surface water containing 1% road runoff. This dilution has however been applied by others to estimate road runoff concentrations in standing waters [105]. However, for small static water bodies receiving relatively high volumes of runoff, e.g. through splash and spray, dilution may be less. For such water bodies, undiluted runoff concentrations better represent a worst case exposure scenario. Other factors are the fact that the number of measurements in literature is limited for these pollutants (therewith hampering a representative comparison) and the input of other emissions besides traffic related sources. For example, other main sources of phthalates (DEHP, DIDP) and NP are roofing and cladding and flexible PVC [68, 106], while OP, BPA, HMMM and MBT are also released from industrial and/or residential wastewater [84, 107–109].

The PEC values of TWP were based on reported values in literature, which were based on measurements [41, 78–80] and modelling [37]. TWP concentrations in runoff were measured in the water phase at values of 975 µg/l and 58,500 µg/l [78] and in the solid phase at values of 13,000 mg/kg$_{\text{dry weight (dw)}}$, 150,000 mg/kg$_{\text{dw}}$ [78], 2000 mg/kg$_{\text{wet weight (ww)}}$, 150,000 mg/kg$_{\text{ww}}$ [79] and 70,000 mg/kg$_{\text{ww}}$ [80]. Liu et al. [110] have measured microplastics in runoff excluding TWP and reported concentrations of 0.231 (0.085–1.143) µg/l, which were not used as PECs in our study. These concentrations of microplastics without TWP are much lower than those measured of TWP, which is to be expected considering runoff as a main pathway of TWP entering the aquatic environment and TWP being a major source of microplastics [3–6]. In surface water, TWP were measured at 6 µg/l [78] and 10 µg/l [41] and modelled at 27 µg/l (annual average with ranges between 3.7 and 120 µg/l [37]). In sediment, TWP were measured at 2.3 mg/kg$_{\text{ww}}$ [41], 300 mg/kg$_{\text{dw}}$ [78] and 730 mg/kg$_{\text{ww}}$ [79]. A review of microplastic particle concentrations in river and lake water, groundwater, tap water, bottled drinking water and wastewater in Asia, Australia, Europe and North America found concentrations to range from 1×10^{-2} to 10^{8} particles/m^{3} [111]. Assuming a particle weight of 12.5 µg per particle [112], this corresponds to a concentration of 1.25×10^{-4} to 10^{6} µg/l. Our TWP PECs fall within this broad range including those of river and lake water.

Effect assessment

Effect data for TWP were limited and therefore we used effect data for microplastics as a proxy for physical particle effects. Given that particle-volume based 'food dilution' has been found to be the primary effect mechanism for low-caloric particles such as TWP and microplastics, this approach is legitimate [60, 61]. Preferably effect data for TWP should be based on real-world concentrations and material of TWP. However this is very scarcely available. Most effect data originated from experiments with leachates of whole tires or artificially produced tire wear (see compilation by Wagner et al. [8]) and some experiments with TWP dispersions [43, 44] and spiked sediments, e.g. Redondo-Hasselerharm et al. [42]. Novel data are presented here for the WET-test with three aquatic species exposed to environmental samples taken from road runoff and surface water near highways. However, more ecotoxicity testing is required to provide sufficient insight in the potential effects and concomitant risk of TWP in surface water. The main approach presented here is a risk assessment of selected substances as components of TWP based on published effect data on the ecotoxicity of single substances and microplastics. The chronic effect values used for the SSD were based on NOECs and EC_0 to EC_{10} values. This approach complies to the EU TGD [64]. Research has demonstrated that using either EC_{10} or NOEC values does not largely affect the results of effect assessment [113]. For BaP, OP and DEHP the number of chronic effect values did not meet the requirements for SSD derivation so acute effect values were also included (i.e. EC_{50} and LC_{50} values). A pragmatic acute to chronic ratio of 10 [69] was used to extrapolate chronic toxicity levels from acute toxicity values for the chemical substances. A more refined approach is to apply extrapolation factors depending on the exposure duration of the toxicity test, as applied for microplastic following Besseling et al. [65], Adam et al. [66] and Koelmans et al. [61].

The effect values for the SSD approach were based on exposure via the water phase only as sediment toxicity data is limited and the SSD approach requires at least 10 effect values. More toxicity data for sediment organisms are necessary in order to derive an SSD for TWP and related substances. Recently, standardized protocols for bioassays with tire particles are developed [42, 60], enabling the generation of sediment toxicity data with maximum applicability for risk assessments. With increasing data availability, a probabilistic risk assessment for exposure via sediment and solids could be possible. This is especially important considering microplastic particles with a density higher as well as lower than water can settle and be buried in the sediment [114], making sediments act as a sink for TWP [8, 10].

The software used for SSD derivation, ETX 2.1, applies a cumulative log-normal distribution, where sensitivity values for species are fitted to a logarithmic scale. The data is tested for normality by three statistical tests: the Anderson-Darling, the Kolmogorov-Smirnov and the Cramer von Mises test. SSDs should not be applied on a dataset when statistical tests for log normal distribution fail [64, 115]. This criterion has been partly applied for the underlying risk assessment. The Kolmogorov-Smirnov test for normality was accepted for all substances at a significance level of 0.005. The Anderson-Darling and Cramer von Mises goodness-of-fit tests were accepted for most substances but rejected for fluoranthene and NP. This adds uncertainty to the PAF for fluoranthene and NP. For BPA, the dataset was adjusted in order to achieve better results with respect to the normal distribution [64, 115]. As the type of effect (e.g. mortality, morphology, development, reproduction) has influence on the effect concentration and therewith adds to the uncertainty for species sensitivity, the dataset for BPA was limited to only include mortality effects. This dataset was found to be normally distributed (all tests were accepted), whereas the dataset based on all type of effects had a low probability of normal distribution (all tests failed).

Risk characterization

The risk assessment (i.e. PEC/PNEC ratio and PAF) applied in this study only considers individual substances and not the combined toxic pressure of multiple substances in runoff. Combined toxicity can be addressed by applying the multi species (ms) PAF [63, 116]. This is not elaborated in this study. However, the WET-tests cover combined toxicity and these showed significant toxic effects for algae when exposed to road runoff, whereas bacteria and crustacea showed no significant toxic effects. The risk assessment for runoff indicates potential effects of TWP_{MP}, BaP, fluoranthene, OP, DIDP and DEHP. The cause of the toxicity cannot fully be explained by the underlying risk assessment as none of the substances are more toxic to algae compared to other species (i.e. algae are not the most sensitive species group for the selected substances, see Tables S6 and S7). This suggests that the observed toxicity is caused by one or more currently unidentified toxicants. Additional approaches, such as the TIE (Toxicity Identification and Evaluation) approach, would be required in order to get insight in the type of compounds, or even to identify the individual compound(s) responsible for the toxicity in the runoff water.

Risk of TWP_{MP}

To assess the risk of TWP, environmental concentrations of TWP need to be compared to effect values of TWP. As aquatic effect data of microplastic particles from tires are scarce [43, 44, 117], we used microplastics in general as a proxy to assess the particle component of the effect of TWP_{MP}. This is considered a limitation because consistent risk assessment for microplastic particles requires alignment of exposure and effect data, i.e. whatever metric or unit is used to characterize exposure also is used for the effect assessment [61]. Koelmans et al. (2020) propose and test rescaling methods for exposure and effect assessment. In future, when exposure and effect data of TWP is sufficiently available, methods to correct for the differences in particle types and size ranges [61] could be applied to improve the risk assessment. The PNEC and SSD used in this study are based on data for a wide range of microplastics generated by Adam et al. [66] and Besseling et al. [65]. The "all-inclusive" SSD [65] can be considered as a proxy for TWP because the size and density of the particles used for the SSD (0.1 to 600 μm [65, 66] and 0.78 to 1.39 g/cm^3 [118, 119] are covering those of car tire particles (4 μm to

350 µm [14] and 1.2 g/cm^3 [79]). Recently, Redondo-Hasselerharm et al. [42] studied TWP toxicity to four benthic species and found that neither the particles themselves nor any of the associated chemicals were toxic at tire particle concentrations up to 10% sediment dry weight. That concentration is far (factor 10^6) above the PNEC$_{sediment}$ for microplastics (0.1 mg/kg dry weight [65]) we used in our study as representative for species sensitivity to TWP, in line with our approach for the PNEC$_{water}$. The PNEC from Besseling et al. [65] is based on the lowest effect value from six benthic species together with a safety factor of 1000 to derive a PNEC, whereas Redondo-Hasselerharm et al. [42] used four benthic species and no safety factor because a PNEC was not derived. Considering this safety factor, the difference is reduced to a factor 10^3. This suggests that we may have overestimated the risk posed by the tire wear particles in sediment and solids by using the conservative PNEC$_{sediment}$ for microplastics from Besseling et al. [65]. The same applies for the water phase, as recent studies [43, 120] observed effects of TWP suspensions at far higher concentration levels (NOEC is 0.26 g/l for *Hyalella Azteca* [43]) than the PNEC$_{water}$ used in this study (0.33 µg/l [65]).

The underlying preliminary risk assessment indicated that unacceptable effects of microplastics from traffic related sources are likely to occur. PEC/PNEC ratio's and PAF exceeded acceptable levels for all locations and matrices. For runoff, this might be expected, as this can be considered as the undiluted and untreated effluent from traffic. Since the environmental risk from microplastics in runoff has not yet been addressed in the scientific literature, we cannot compare our findings with other studies. In surface water and sediment, however, this is a different case. On the basis of evidence published to date, effects of microplastic at a population level appear unlikely [5], although for some hotspots, effects cannot be excluded [61, 65, 66]. As the scope of the underlying risk assessment is focused on hotspots, by extrapolating the highest reported concentrations in literature as estimated PEC and measurements in samples taken in close proximity to the source [78] as measured PEC, our findings are not contradictory. However, PEC/PNEC ratio's and PAF estimated in the risk assessment are relatively high compared to Adam et al. [66] and Besseling et al. [65]. As risk is determined by the comparison of environmental exposure (PEC) with a measure of sensitivity (PNEC or SSD (via PAF)), this could be the result of higher exposure, higher sensitivity, or both. Comparing the PNEC for microplastic used in this study (0.33 µg/L, corresponding to a 5% hazard concentration (HC$_5$) of 1.65 µg/L or 1015 particles/L [65]) with other PNEC and HC$_5$ values available in literature (HC$_5$ of 3500 particles/L [121, 122]; HC$_5$ of 3214 particles/L [123]; PNEC of 0.042 µg/L or 740 particles/L [66]; PNEC of 0.14 µg/L or 71.6 particles/L [124]; and HC$_5$ of 251 particles/L or 75.6 particles/L when corrected for the differences in size ranges, bioavailability and polydispersity [61]) shows that the PNEC is within the range of literature values. The SSD used in the present study (see Supporting information, page 23) is based on NOEC values from Adam et al. [66] and Besseling et al. [65] combined, and is thus also comparable to the SSDs in their studies. Besseling et al. [65] uses a freshwater PEC of 0.14 particles/L, corresponding to 1.77 µg/L when assuming a particle weight of 12.5 µg [112]. Adam et al. [66] used PECs mostly between 10^{-2} and 10^4 particles/m^3, corresponding to a range between 1.25 10^{-4} and 12.5 µg/L. The PECs used in this study for TWP in surface water (6 µg/L [78] and 10 µg/L [41]) are higher than PECs for microplastics used by Besseling et al. [65] but comparable with maximum values for microplastics used by Adam et al. [66].

Conclusions

This study provides opportunities to protect the quality of European waters from road runoff pollution, focusing on car tire microplastic particles, WFD priority substances and other hazardous substances. It emphasizes the relevance of addressing risks of microplastic particles originating from car tires. Results indicate that TWP occur in relatively high concentrations compared to microplastics in general and that the corresponding risk of TWP is above threshold levels. Because TWP exists both as anthropogenic particulates and as a source of a suite of chemicals, providing a risk assessment is challenging. This study provides a first risk assessment posed by particle effects (TWP$_{MP}$) as well as risks posed by chemical effects (organic micropollutants). Additional research is required to further address the risks of TWP, e.g. toxicity testing for environmentally realistic TWP material and aligning exposure and effect data. Furthermore, the risks of some micropollutants (TT, DIDP, MBT and HMMM) could be further investigated in future when more effect data might be available, as well as addressing the risks of exposure via sediment and identifying the main contributors to the toxicity of road runoff to algae.

Abbreviations

BaP: Benzo(a)pyrene; BPA: Bisphenol A; DEHP: Di (2 ethylhexyl)phthalate; DIDP: Diisodecyl phthalate; EC$_x$: Effect concentration at which x% effect (mortality, inhibition of growth, reproduction, etc.) is observed compared to the control group; EQS: Environmental Quality Standard; Fl: Fluoranthene; HC$_5$: Hazardous Concentration for 5% of the species; HMMM: Hexa (methoxymethyl)melamine; LC$_{50}$: Lethal Concentration 50%; MBT: Mercaptobenzothiazole; ms PAF: Multi species PAF; NOEC: No Observed Effect Concentration; NP: Nonylphenol; OP: 4 tert octylphenol; PAF: Potentially Affected Fraction of species; PEC: Predicted Environmental Concentration; PNEC: Predicted No Effect Concentration; SSD: Species Sensitivity Distribution; TIE: Toxicity Identification and Evaluation; TRWP: Tire and road wear particles; TT: Tolyltriazole; TWP: Tire wear particles, i.e. microplastic particles originating from car tires; TWP_{MP}: Tire Wear Particles Microplastic, where exposure is represented by TWP and effect by microplastics; WET: Whole Effluent Toxicity; WFD: Water Framework Directive

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s43591 021 00008 w.

Additional file 1.

Acknowledgments

We gratefully acknowledge the financial and technical support of the CEDR and the CEDR Programme Executive Board.

Authors' contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Funding

This study is a part of the Conference of European Directors of Roads (CEDR) Transnational Research Programme Call 2016 Water Quality: Environmentally Sustainable Roads (funded by Austria, Finland, Germany, Ireland, Netherlands, Norway and Sweden).

Availability of data and materials

All data generated or analyzed during this study are included in this published article and its Supporting information file. The Supporting information file contains information on: 1) WET tests; 2) organic micropollutants and sources released from traffic and roads; 3) selection of substances for the risk assessment; 4) PEC values; 5) PNEC values (including equilibrium partitioning); 6) SSDs; 7) PAF values; 8) effect values for microplastics and selected organic micropollutants.

Declarations

Competing interests

The authors declare that they have no competing interests.

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Received: 29 December 2020 Accepted: 27 May 2021

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