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Export of microplastics from land to sea. A modelling approach

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ABSTRACT

Quantifying the transport of plastic debris from river to sea is crucial for assessing the risks of plastic debris to human health and the environment. We present a global modelling approach to analyse the composition and quantity of point-source microplastic fluxes from European rivers to the sea. The model accounts for different types and sources of microplastics entering river systems via point sources. We combine information on these sources with information on sewage management and plastic retention during river transport for the largest European rivers. Sources of microplastics include personal care products, laundry, household dust and tyre and road wear particles (TRWP). Most of the modelled microplastics exported by rivers to seas are synthetic polymers from TRWP (42%) and plastic-based textiles abraded during laundry (29%). Smaller sources are synthetic polymers and plastic fibres in household dust (19%) and microbeads in personal care products (10%). Microplastic export differs largely among European rivers, as a result of differences in socio-economic development and technological status of sewage treatment facilities. About two-thirds of the microplastics modelled in this study flow into the Mediterranean and Black Sea. This can be explained by the relatively low microplastic removal efficiency of sewage treatment plants in the river basins draining into these two seas. Sewage treatment is generally more efficient in river basins draining into the North Sea, the Baltic Sea and the Atlantic Ocean. We use our model to explore future trends up to the year 2050. Our scenarios indicate that in the future river export of microplastics may increase in some river basins, but decrease in others. Remarkably, for many basins we calculate a reduction in river export of microplastics from point-sources, mainly due to an anticipated improvement in sewage treatment.

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1. Introduction

Plastic pollution is considered one of today's main environmental problem and pollutants in oceans, rivers and streams (Barnes et al., 2009) and have potential risks to human health (Wright and Kelly, 2017) and the environment. The occurrence of plastic debris in the marine environment, in lakes (Eriksen et al., 2013; Free et al., 2014), at shorelines (Browne et al., 2011) and in rivers (McCormick et al., 2014; Klein et al., 2015; Lechner et al., 2014; Yonkos et al., 2014; Kooi et al., 2016) received increased attention and attempts have been made to quantify microplastic (plastic particles with the dimension of 1 μm up to 5 mm) pollution in the marine environment (Barnes et al., 2009; Cózar et al., 2014; Eriksen et al., 2014; Van Sebille et al., 2015). Recent estimates indicate that rivers transport between 1.15 and 2.41 million tonnes of plastic waste to seas (Lebreton et al., 2017) and this is expected to increase in the coming decades (Jambeck et al., 2015). However, quantitative information about microplastics entering the sea from land is scarce, whereas the relative contributions of the different sources and pathways of plastics are not well documented (Kooi et al., in press). Most studies of marine litter in urban run-off focus on macro-rather than on microplastic debris (Ryan et al., 2009).

Microplastics are known to originate from different sources, which can be divided in two broad categories: primary- and secondary sources (Bergmann et al., 2015). Primary sources are microplastics that are manufactured in microscopic size for domestic and industrial applications, like plastic pellets used as raw material in the plastic industry and/or abrasive microbeads in cosmetics, detergents, other hygiene and personal care products (Arthur et al., 2009; Cole et al., 2011; Fendall and Sewell, 2009).

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Secondary microplastics originate from larger plastic materials and are formed from the breakdown of macroplastics through photo-degradation and mechanical abrasion of marine debris into small plastic particles (Gewert et al., 2015).

Microplastic pollution can originate from point- or diffuse sources. Whereas there is an urgent knowledge gap to understand the characteristics of either one of these sources, this study focuses on microplastic point-source fluxes. The considered pathway of point-source microplastics from land to the sea is effluents of centralized sewage systems. Microplastics in domestic wastewater are being transported to wastewater treatment plants (WWTPs) or are discharged untreated to adjacent water bodies. A potential significant input of microplastics that is not included are fluxes from combined sewer overflows. Rivers connect land to sea and present an important pathway of microplastic waste generated inland to reach the marine environment. Microplastic transport from rivers to seas from such point sources is, therefore, important to better understand the processes underlying the contamination of aquatic ecosystems with plastic debris. Diffuse sources are sources without a specific point of discharge. Examples are plastics entering a body of water through surface run-off, rainfall or wind. Such inputs occur over a wide area and are much more difficult to characterise at present, due to fundamental data gaps with respect to source composition and process rates that govern the transport (e.g. run off) of plastic debris. In areas not connected to centralized sewage systems, decentralized systems such as septic tanks are often used.

Scarcity of quantitative data is one of the biggest constraints encountered in environmental research of microplastic pollution. There are studies available on accumulation of plastic debris in the environment (Barnes et al., 2009), sources of (micro)plastics (Arthur et al., 2009; Cole et al., 2011; Fendall and Sewell, 2009) and consequences of plastic pollution in the marine environment (Kühn et al., 2015). Quantitative assessments of per capita microplastic consumption from different sources are available (Essel et al., 2015; Sundt et al., 2014), as well as information on the microplastics content in incoming wastewater at sewage treatment plants (Brandsma et al., 2013; Magnusson and Norén, 2014; Mintenig et al., 2017; Kalčikova et al., 2017; Talvitie and Heinonen, 2014), and river retentions (Besseling et al., 2017). However, on the continental or global scale, the explicit quantitative analyses of the export of microplastics from land to the sea has not been addressed. Quantities that are released into rivers from sewage treatment plants and subsequently enter the sea on these spatial scales are largely unknown, yet crucial for assessing short- and long-term impacts caused by plastics (GESAMP, 2016).

The aim of this paper is to provide an estimate of microplastic fluxes from land to sea for European rivers. Our approach accounts for point-sources of plastics in river systems and selected types and sources of microplastics. Our analysis builds on earlier applications of a river export model for nutrients that includes information that is relevant for plastic pollution (Kroeze et al., 2016). We apply our model to river basins draining into the North Sea, Baltic Sea, Black Sea, Mediterranean Sea and European river basins draining into the Atlantic Ocean, to calculate microplastic fluxes for the year 2000 and two scenarios for the year 2050.

2. Method

2.1. Model overview

Our modelling approach is inspired by an existing global model for nutrients, the Global *NEWS* (Nutrient Export from WaterSheds) model (Seitzinger et al., 2010; Mayorga et al., 2010). Global *NEWS* calculates point source inputs of nutrients to rivers (from sewage). We take the same approach, and calculated river export of microplastics from point-sources as a function of human activities on land and river retention (Fig. 1). Global NEWS has been applied using input data from the IMAGE model (land use, agricultural and socio-economic parameters) and WBMplus (hydrology). It is thus based on the STN-30p river system (Vörösmarty et al., 2000) at a grid of 0.5 × 0.5° to delineate flow directions and all major basins draining to the coast. Input datasets for point sources (population, sewage treatment) are described in Van Drecht et al. (2009).

Microplastic yield (Yld_{MP}; Eq. (1)) is the amount of microplastic from point-sources that is exported to the river mouth per unit area

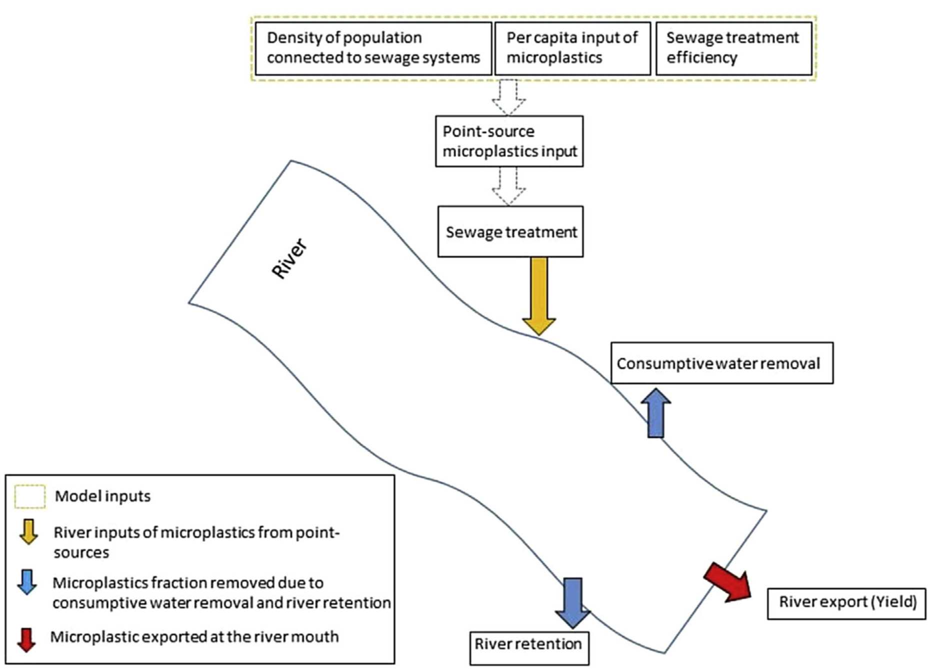

Fig. 1. Schematic overview of microplastic point-source inputs to rivers and export to the river mouth.

Table 1
Microplastic data sources.

Micro plastic point-sources ($WShw_{cap,i}$)	Quantitative estimate	Reference
Personal care products ($WShw_{cap,PCP}$)	0.0071 kg capita^{-1} year^{-1}	Sundt et al., 2014, Essel et al., 2015
Household dust ($WShw_{cap,HD}$)	0.08 kg capita^{-1} year^{-1}	Sundt et al., 2014
Laundry inputs ($WShw_{cap,LD}$)	0.12 kg capita^{-1} year^{-1}	Sundt et al., 2014
Tyre wear ($WShw_{cap,TRWP}$)	0.18 kg capita^{-1} year^{-1}	Sundt et al., 2014, Rijkswaterstaat, 2014, Norén and Naustvoll, 2010, Essel et al., 2015

of the basin (kg km^{-2} year^{-1}). Yields are calculated as a fraction ($FE_{riv,i}$) of the input of microplastics to rivers from point-sources ($RS_{pnt,i}$ kg km^{-2} year^{-1}) (Mayorga et al., 2010):

$$Yld_{MP} = \sum_{i=1}^{n}(FE_{riv,i} \times RS_{pnt,i}) \quad (1)$$

where:

MP = microplastic of type n. We accounted for four sources of microplastics (n = 4): personal care products (PCP), household dust (HD), laundry textiles (LD) and tyre and road wear particles (TRWP);
$FE_{riv,i}$ is the fraction of microplastic inputs to streams that is exported by rivers and aquatic system for microplastic type i; and
$RS_{pnt,i}$ is the microplastics input to rivers from the point-source of microplastic type i (kg km^{-2} year^{-1}).

Micro plastic loads are calculated from yields and the river basin area.

$$L_{MP} = Yield_{MP} \times A \quad (2)$$

where:

L_{MP} = micro plastics load (kg year^{-1});
$Yield_{MP}$ = micro plastics yield, Equation (1) (kg km^{-2} year^{-1}); and
A = basin area (km^2), derived from the existing Global *NEWS* model.

Point source inputs of microplastic to rivers are estimated in the same way as nitrogen point sources in Global *NEWS*, as described in Van Drecht et al. (2009). Point source inputs are calculated per river basin. We account for sewage treatment efficiencies, the extent to which people are connected to sewage treatment systems and the per capita input of microplastics to the river basin:

$$RS_{pnt,i} = (1 - hw_{frem,i}) \times PConDen \times WShw_{cap,i} \quad (3)$$

where:

$hw_{frem,i}$ = fraction of microplastics of type i in sewage influent which is removed via sewage treatment (see SI Appendix);
PConDen = Density of population connected to sewage system (inhabitant km^{-2}); and
$WShw_{cap,i}$ = per capita input of microplastics of type i to the river basin (kg capita^{-1} year^{-1}).

2.2. Per capita inputs of microplastics ($WShw_{cap}$)

Data on microplastics are scarce (Arthur et al., 2009), yet estimates from several studies can be combined (Sundt et al., 2014; Essel et al., 2015) (Table 1). We accounted for microbeads in personal care products, synthetic polymers and plastic fibres in household dust, abraded plastic based textiles during laundry and point-source inputs from synthetic polymers from tyre abrasion mixed with road wear particles.

Personal care products (e.g. facial and body scrubs, toothpaste, shaving cream, peeling products and make-up, etc.) are products containing microbeads (micro sized synthetic polymers, most commonly made of polypropylene, polyethylene, and nylon) (Fendall and Sewell, 2009; Napper et al., 2015; Sundt et al., 2014). Households generally have many plastic materials and products containing synthetic polymers (Sundt et al., 2014). Abrasion and weathering of these materials and products contribute to household dust. The microplastic particles eventually will be cleaned by air conditioner filters and cleaning of floors and dusty surfaces. When cleaned up, they end up in the drain and make their way to sewage treatment plants (Webster et al., 2009).

Plastic polymer based textiles containing microplastics are abraded as microplastic particles during laundry washing. In addition, collected particles will be discharged to the sewer. Abraded synthetic polymers from tyre related wear are another source of microplastic pollution (Norén and Naustvoll, 2010). The car tyre surface is based on synthetic polymers (Styrene Butadiene Rubber, and other additives) and natural rubber that slowly degrade and loose tyre material due to abrasion (Sundt et al., 2014). Car tyre wear particle inputs are mixed with road pavement particles due to friction at the pavement surface interface during rolling of the tyre (Verschoor et al., 2016) resulting in tyre and road wear particles (TRWP). The composition of TRWP is influenced by several factors as described by Verschoor et al. (2016). Their inputs estimates are based on tyre wear fraction and do not include particles released from the road surface. Furthermore, TRWP does not solely occur as a point-source input. TRWP is emitted into the air, soil, surface waters and sewage systems via runoff (Verschoor et al., 2016). Verschoor et al. (2016) estimate that point sources account for 13% of the total microplastic inputs to rivers, Kole et al. (2015) 15%. We use the estimate of 15% in the present study to account for point-sources of TRWP emissions. TRWP emissions is an uncertain parameter. We acknowledge that the share that ends up in WWTPs may be lower in countries where storm water is not entering a centralized sewage system and bypasses a WWTP. For those, we may be overestimating the point source inputs to some extent but there are no data to model this more explicitly.

2.3. River retention

Retention of microplastic particles relates to the fraction of the total amount of microplastics retained within the river system, which thus is not exported at the river mouth. This can occur due to net settling which in turn depends on the density and morphology of the particles (Table 2) (Besseling et al., 2017; Kooi et al., in press). We use retention fractions for each microplastics source (Table 2) and account for the length of the rivers (i.e. for basins with shorter

Table 2
Overview of microplastic sources, their particle size (μm), polymer type and density (g/cm^3) (Based on Lassen et al., 2015). Grey rows indicate the four microplastics sources included in this study, white rows provide more details about personal care products.

Micro plastic sources ($_i$)	Size of particles (μm)	Plastic type	Density (g/cm^3)	Retention fraction (Ret_i)[a]	References
Personal care products				Weighted average of 0,2[b]	
Toothpaste	2-5, >10 (white spherical), >100 (blue spherical)	Polyethylene	0.91–0.94	0	Hintersteiner et al., 2015, Polymer Data Handbook 1999.
Facial scrubs	40–800	Polyethylene	0.91–0.94	0	Gregory, 1996, Strand 2014, Polymer Data Handbook 1999.
Hand cleaning	100–1000	Polyethylene	0.91–0.94	0	Gregory, 1996, Polymer Data Handbook 1999.
Shower Gel	>100 (white spherical), >300 (blue elongated)	Polyethylene	0.91–0.94	0	Hintersteiner et al., 2015, Polymer Data Handbook1999.
Shaving Foam	5–15	Polytetrafluorethylene	2.28–2.29	1	Sundt et al., 2014, Polymer Data Handbook 1999.
Household dust	10–100	Polyamide, Polystyrene, Acrylic	1.13–1.15, 1.04–1.09, 1.09–1.20	0.75[c], 0.9[c]	Sundt et al., 2014.
Laundry textiles	10–100	Polyamide, Polystyrene, Acrylic	1.13–1.15, 1.04–1.09, 1.09–1.20	0.75[c], 0.9[c]	Sundt et al., 2014, Polymer Data Handbook 1999.
Tyre and road wear particles (TRWP)	10–400	Styrene Butadiene Rubber as in TRWP	1.2–1.3	0.75[c], 0.9[c]	Sundt et al., 2014, Verschoor et al., 2016.

[a] Retention fractions (Ret,i) for each microplastics source i (i.e. personal care products, household dust, laundry and tyre and road wear particles) were estimated based on spatially explicit hydrodynamic river model simulations as provided by Besseling et al. (2017).
[b] All sub-sources of personal care products are considered to have equal weight as data availability only permitted estimates for personal care products as a sum of sub-sources.
[c] We assumed TWRP would dominate the inputs of car tyre wear, however a non-associated fraction of SBR cannot be excluded, which is why the retention was at 0.75 or 0.90 (depending on the basin scale) rather than at 1.0. A retention fraction of 0.75 is applied to small basins (≤3 cells), and 0.90 to the remaining river basins respectively. Smaller basins have a shorter distance to the sea and it is assumed that microplastic particles are less retained in river systems with shorter distance to the river mouth (Besseling et al., 2017).

distance to the river mouth, a lower retention is defined). The fraction of water removed from rivers for consumptive water use ($FQrem_{,i}$) for each basin was adopted from Global *NEWS* (Van Drecht et al., 2009). The model calculations do not account for remobilisation or burial of microplastic particles, given the lack of knowledge on occurrence and frequency of these processes. The fraction of the microplastics inputs to rivers that is exported to the river mouth ($FE_{riv,i}$) is calculated as follows:

$$FE_{riv,i} = (1 - Ret_{,i}) \times (1 - FQrem_{,i}) \quad (4)$$

where:

$Ret_{,i}$ is the retention fraction of the different microplastic sources i; and
$FQrem_{,i}$ is the fraction of microplastic types i removed through consumptive water use.

Equations (1)–(4) were applied to 623 individual river basins draining into European seas, as provided by the Global *NEWS* model.

2.4. Population connected to sewage systems (PConDen) and microplastic fraction removal by sewage treatment (hw_{frem})

The number of people connected to sewage systems (PConDen) for each river basin is derived from Global *NEWS* (Van Drecht et al., 2009). Existing studies on the removal of microplastics in sewage treatment plants (Brandsma et al., 2013; Magnusson and Norén, 2014; Mintenig et al., 2017; Kalčikova et al., 2017; Talvitie and Heinonen, 2014) indicate a microplastics removal efficiency of 95% or more for sewage treatment plants with at least primary treatment in place. Secondary and tertiary treatment remove somewhat, but not much, more (Carr et al., 2016). We use sewage treatment efficiencies for microplastics that are based on the degree of treatment, i.e. no treatment, primary-secondary or tertiary treatment. These degrees of treatment in river basins are deduced from phosphorous removal efficiencies in sewage treatment from Global *NEWS* (Van Drecht et al., 2009). Different densities and morphologies of microplastic particles that may affect the removal efficiency during treatment are not accounted for. Microplastic parameters are motivated as fair approximations of average microplastic behaviour. More detailed information on the implementation of the removal efficiencies of sewage treatment plants is available in the supplementary material (SI Appendix).

2.5. Scenarios

River export of microplastics to European seas was calculated for the past (2000) and future (2050). For the year 2050 we used two existing scenarios as a starting point. These are the Global Orchestration (GO) and the Adaptive Mosaic (AM) scenario from the Millennium Ecosystem Assessment (Alcamo et al., 2005). These scenarios have been implemented in Global *NEWS* (Seitzinger et al., 2010). The scenarios differ in assumptions on socioeconomic development (globalization or regionalization) and environmental management (proactive or reactive). Earlier Global NEWS studies provide input datasets for these scenarios for land use, hydrology and point sources (Bouwman et al., 2009; Fekete et al., 2010; Van Drecht et al., 2009).

The GO scenario presents a globalized world with a reactive approach towards environmental management, focusing on rapid economic growth and low population growth. In GO sewage connection and sewage treatment efficiencies are generally higher than in AM in 2050, and also higher than in the year 2000. Consumptive water use is generally higher in AM. The AM scenario is characterized by proactive environmental management with relatively simple and economically feasible technologies at regional level. For a more detailed description of the MA scenarios the reader is referred to the supplementary material (SI Appendix, Table S1).

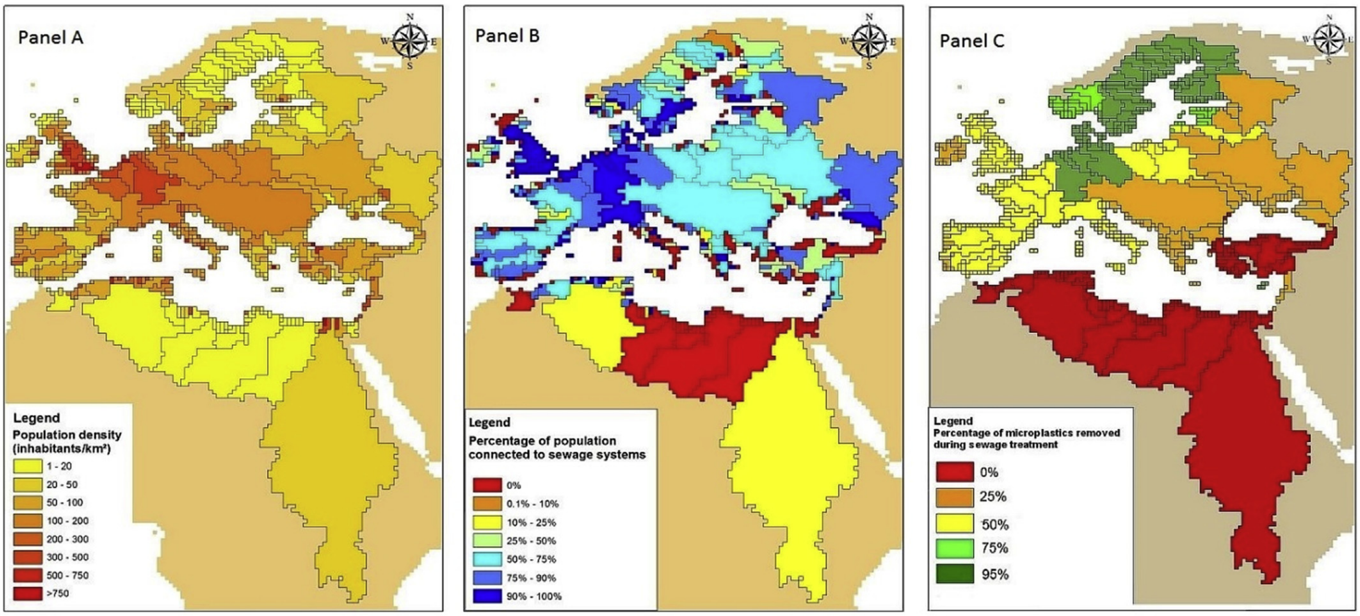

Fig. 2. Panel A: Population densities (inhabitants/km^2) in the study area. **Panel B:** Percentage of inhabitants connected to sewage treatment plants. **Panel C:** sewage treatment efficiencies for microplastics. Source: Deduced from Global *NEWS* input data for the year 2000.

3. Results and discussion (<1500)

3.1. Population densities, sewage connection and sewage treatment

Population densities vary largely among river basins, ranging from 0 to more than 1000 persons per km^2 in 2000 (Fig. 2A). Population densities are high along the coastlines in the northern part of the African continent (mouth of the Nile, Morocco, Algeria and Tunisia), in west Asia (Israel, Lebanon), and in parts of Europe (Greece, Italy, around the Black Sea, the United Kingdom, northern France, Belgium and the Netherlands). Population densities are lower in large parts of Africa, and in Sweden, Norway and Finland.

A crucial factor for point-source river export of microplastics is the connection of the inhabitants of river basins to sewage treatment plants. The more inhabitants are connected to sewage systems, the more microplastics potentially enter the sewage treatments plants in sewage influents or are discharged without sewage treatment. The map visualizes the percentage of inhabitants per km^2 connected to sewage systems, based on the total population density within the river basin (Fig. 2, Panel B).

Generally, in northern African, many river basins have no- or very low connectivity to sewage systems. Only in some small basins close to the Mediterranean Sea the sewage connection rate is higher (60–100%). In western Asia sewage connection is typically 50–100%, especially around Israel and Lebanon. The Black Sea region generally has many rivers with no connection, however in some of the large river basins draining into this sea (Danube, Don), between 50 and 75% of all inhabitants have a connection to sewage installation. Dark red indicates that the population is not connected to sewage systems and, therefore, there are no point-source inputs of microplastics in these rivers.

The efficiencies of sewage treatment plants in removing microplastics from sewage influent differ among river basins. As described earlier, the efficiency of sewage treatment plants to filter out microplastics during sewage treatment is the single most influential factor in preventing microplastics to enter aquatic systems and ultimately reach the marine environment. In the year 2000, there was no sewage treatment in around 20% of all river basins (Fig. 2C, red colour). These river basins are located in the northern part of the African continent and Turkey. In another 20% of the basins, the basin-average treatment efficiency is 25% (Fig. 2C, orange colour); these are primarily located in Ireland, partly Portugal, Greece, Eastern Europe, the northern part of the Black Sea region and western Asian river basins. Around 35% of the river basins have sewage treatment efficiencies of 50% (Fig. 2C, yellow colour). These river basins are mostly concentrated in the western European countries, the United Kingdom, Italy and partly in eastern European countries. Only around 5% of all basins have a sewage treatment efficiency of 75%, which are mostly concentrated in Norway and Estonia. Less than 20% of all river basins have the highest treatment efficiencies (set at a conservative 95%, dark green), most of which are concentrated in Denmark, Sweden, the Netherlands and Finland as well as around Germany and Cyprus.

3.2. River export of microplastics

The calculated river export (yields) of microplastics to coastal seas ranges between 0 and 192 kg km^{-2} river basin year^{-1} for the year 2000 (Fig. 3).

Zero microplastic yield from point sources are calculated for basins in northern Africa, around the Black Sea and in Italy, Greece, northern Scotland and parts of Turkey. This can be explained from the fact that no population is connected to centralized sewage systems (dark red, Fig. 2B). Highest yields are calculated for basins in North Africa (around Morocco, Algeria and Tunisia) at the coastline, the western Asian river basins (Israel and Lebanon and river basins in Turkey) along the coastline and river basins in Greece, Italy. All of these river basins drain into the Mediterranean Sea. Relatively low sewage efficiencies (0%–50%) (Fig. 2C) as well as relatively high percentage of people connected to centralized sewage systems (Fig. 2B) are the main reasons behind the high yields. High yields are also calculated for rivers in southern France, western Portugal and northern and eastern Spain, draining into the Atlantic Ocean, as well as for the United Kingdom (particularly in England with the Thames and other river basins, in Ireland and in Wales). The large ranges in microplastic yields reflect the variation

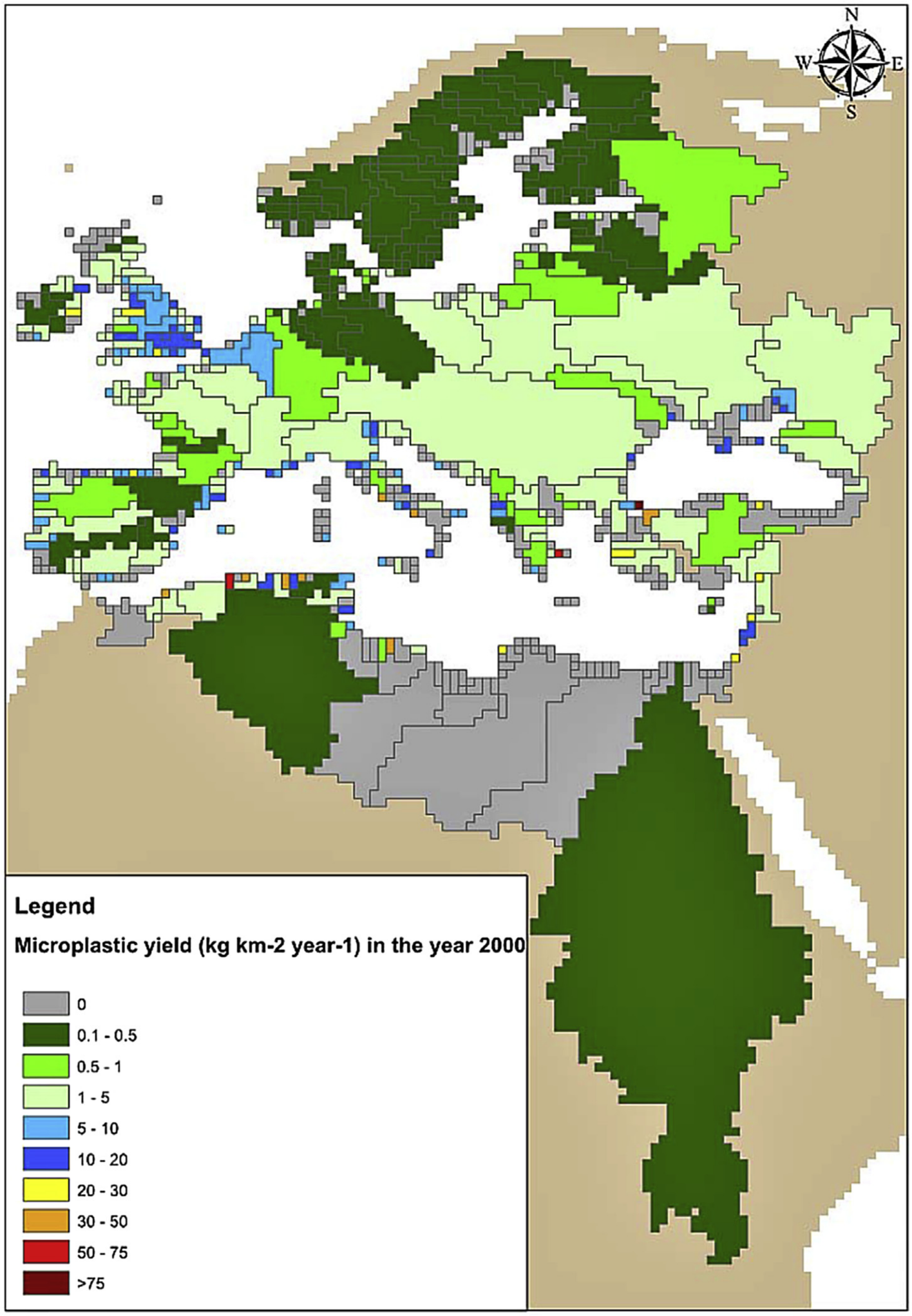

Fig. 3. River export of microplastics (yields in kg km^{-2} year^{-1}) as calculated for the year 2000.

in socio-economic development, and technologies applied in sewage treatment.

We calculate that in total 14.4 kilotonnes of microplastics were exported from point-sources to the North Sea, Baltic Sea, Black Sea, Mediterranean Sea and the European river basins draining into the Atlantic Ocean in 2000 (Loads presented in Fig. 4). The total loads differ by sea. Microplastic export (load) to the Mediterranean Sea was 5.6 kilotonnes, to the Black Sea 4.1 kilotonnes, to the European part of the Atlantic Ocean 2.7 kilotonnes, to the North Sea 1.1 kilotonnes, and to the Baltic Sea 0.9 kilotonnes microplastics. The high load for the Mediterranean Sea is in line with some other studies (Eriksen et al., 2014; Van Sebille et al., 2015).

3.3. *Sources of microplastics in rivers*

Tyre and road wear particles are calculated to be the largest sources of microplastic in the European rivers, and account for 42% of the total exported microplastic load (Fig. 4). Rivers exported 1.6 kilotonnes of TRWP to the Black Sea in the year 2000, and 2.3 kilotonnes to the Mediterranean Sea. River export to the Atlantic Ocean, North Sea and the Baltic Sea amounts to 1.2 kilotonnes, 0.5 kilotonnes and 0.4 kilotonnes, respectively. This is in line with Verschoor et al. (2016), who indicate that tyre and road wear particles are undoubtedly the largest land-based source of microplastic inputs to surface waters. The second largest source of microplastic

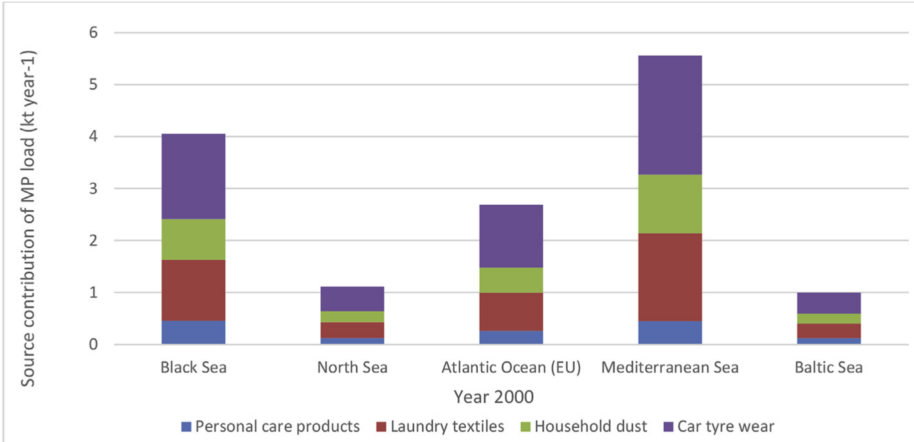

Fig. 4. River export of microplastics (in kilotons per year) to the European Seas by relative point-source contribution as calculated for the year 2000.

inputs to rivers are plastic polymer based textiles, accounting for 29% of the inputs to rivers: rivers exported 1.7 kilotonnes of microplastic from textiles to the Mediterranean Sea, 1.2 kilotonnes to the Black Sea in the year 2000, 0.7 kilotonnes to the Atlantic Ocean, 0.3 kilotonnes to the North Sea and 0.3 kilotonnes to the Baltic Sea. Synthetic polymers in household dust contribute 19% to the total river export of microplastics, and personal care products 10%.

3.4. Comparison with measured data

Our study is a first attempt to model microplastic loads in rivers. There are many uncertainties associated with our model setup. Validating the model is not easy, as data on microplastics in rivers are scarce. Moreover, also current methodologies to assess concentrations in rivers are surrounded with considerable uncertainty (Kooi et al., in press). This implies that a detailed validation of the model is not yet possible. Still we can compare some of the model results with the available data and carefully reflect on the observed level of consistency. Our model results for two individual rivers can be compared with previously published estimates of microplastics export for these rivers (Faure et al., 2015; Lechner et al., 2014; Van der Wal et al., 2015). By applying the above mentioned Equations (1)–(4) to the Rhone, our model calculated a microplastic export of 163 tonnes microplastics per year by the Rhone into the Mediterranean Sea. Faure et al. (2015) estimated an export rate of 208 tonnes per year for the Rhone, based on upscaling concentration samples with the downstream population increase. For the Danube, our model calculated an export rate of 1503 tonnes per year to the Black Sea, which compares well to the 1534 tonnes per year as estimated by Lechner et al. (2014). Van der Wal et al. (2015) extrapolated their results and estimates a transport of 500 tonnes per year for the Danube. Furthermore, van der Wal estimated an export of 120 tonnes per year for the Po (our model calculated an export of 399 tonnes per year) and 20–30 tonnes per year for the Rhine (105 tonnes per year calculated by our model). Evidently more research is needed for unambiguous comparison and validation purposes, but we show a reasonable comparison to the few measurements available with the use of reliable information as a basis for inputs and model parameters.

3.5. Scenarios for the year 2050

The model results for the GO and AM scenarios for 2050 indicate that the total calculated microplastic river export by many European rivers from point-sources in both scenarios is lower than in the year 2000 (up to 50% lower; Fig. 5). Exceptions are African rivers draining into the Mediterranean Sea and scattered basins throughout Europe, where more microplastic is exported in both scenarios. The generally lower export is remarkable because usually it is assumed that microplastic river export to seas is likely to increase in the future (e.g., Jambeck et al., 2015). However, this expectation is based on proportionality with increasing plastic production and population size (Jambeck et al., 2015) and may not apply to socio-economic and technical developments in Europe.

The scenario analysis indicates that the total point-source river export of microplastics to European seas (load) may decrease by 1% (AM) to 18% (GO) between 2000 and 2050. The reduction is larger for the GO scenario that assumes a globalized world focusing on rapid economic growth and low population growth. For the AM scenario, where simple and economically feasible technologies are implemented, a substantially lower reduction is calculated. There are socio-demographic differences among the two scenarios but the most important factor in this respect is the efficiency to remove microplastic from sewage influent during treatment. Sewage treatment is improving less fast in the AM scenario than in the GO scenario. The future scenarios do not account for potential changes in per capita inputs of microplastics. Such changes may be driven by changes in consumer behaviour due to increased awareness or potential new regulations, like stricter regulations for WWTP to increase the efficiency of filtering out microplastics during treatment, or regulations to address microplastic particle emissions. This would likely result in a further decrease in predicted export from European rivers.

4. Conclusions

We provided the first quantitative assessment of microplastic export by rivers on a continental scale. We do not provide a validated model that is able to predict river export of microplastics with certainty, because this is not possible at the current level of information. However, we provide a highly needed methodology to assess microplastics transported by rivers to seas, which we consider an important first step to obtain a better understanding of the spatial patterns of plastic export to marine ecosystems.

Our study highlights the importance of improving sewage systems and sewage treatment efficiencies. This holds especially true for the Mediterranean Sea, for which we calculate the largest microplastic loads. The Mediterranean Sea is the only sea for which we calculate increasing microplastic loads in the future.

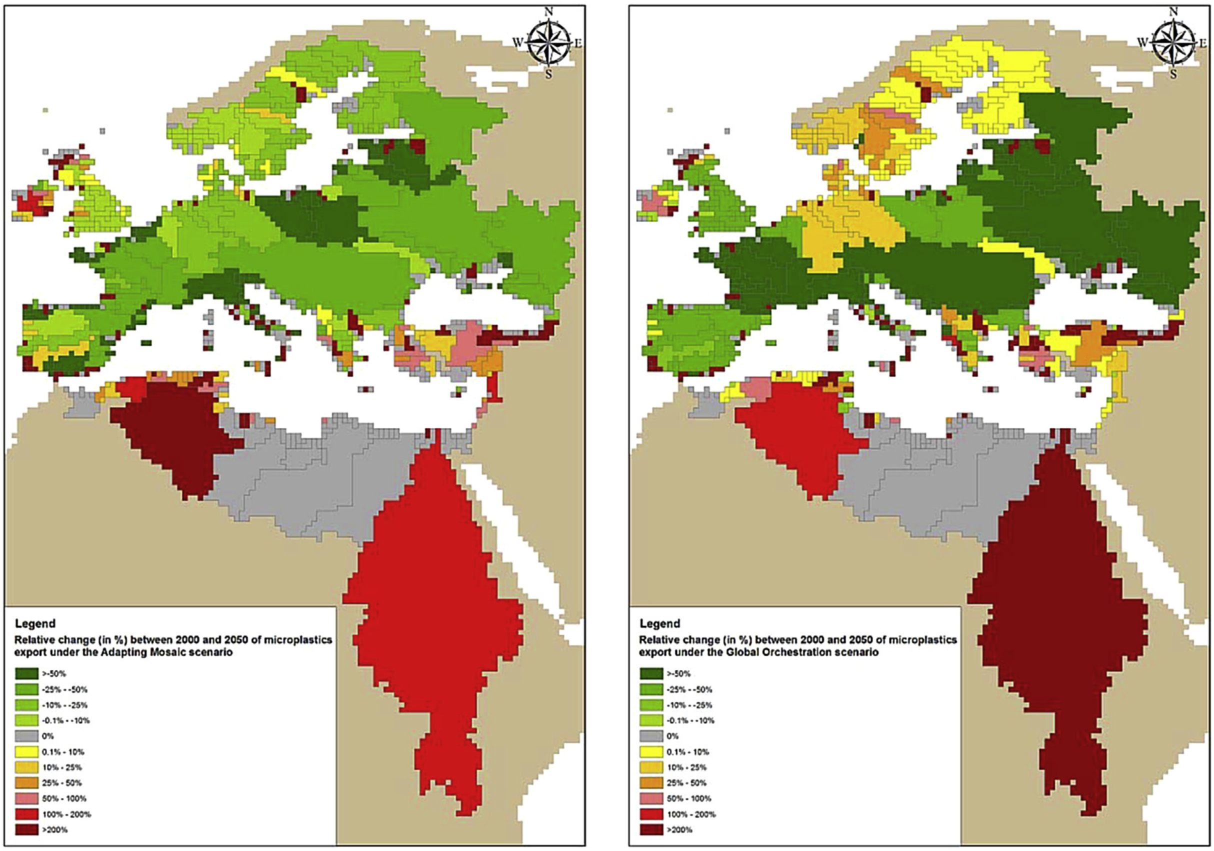

Fig. 5. relative change (in %) between 2000 and 2050 in microplastics river export (load) for two scenarios, Adapting Mosaic (left) and the Global Orchestration (right). Green colours indicate that river export of microplastics in 2050 is lower than in 2000, while yellow to red colours indicate an increase. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Our study also identifies the sources and types of microplastics in European rivers, and their shares in the total loads. This may help to identify effective management strategies to reduce microplastic pollution and potential associated risks to human health and the environment. Regardless of model uncertainty, we show that synthetic polymers from tyre and road wear probably comprise the largest source of microplastic pollution, followed by abraded plastic based textiles containing microplastics and synthetic polymers and plastic fibres in household dust. Microbeads in personal care products constitute only a small part of the total microplastic river export. This information may help to prioritize measures to reduce pollution. It indicates that reducing microplastic inputs from car tyre and road wear is most effective. A focus on the largest sources may help to effectively reduce the microplastic inputs to the European seas. Our estimates do not include all point sources of microplastic in rivers and therefore may be considered minimum estimates. Our model is based on a provisional, heuristic inventory of microplastic data, and would be vastly improved with more and better input data availability. Moreover, it does not include diffuse sources, which need to be assessed in future studies when better data are available.

Our model is the first to assess export of microplastics from land to sea, and the first to quantify the relative contributions of sources to this export. We consider it a first quantitative comparison and a crucial first step for future research. Additional measures e.g., environmental policies and management are arguably required to reduce plastic pollution in European seas. Furthermore, the model output indicates that microplastics loads largely differ among rivers as a result of different socio-economic and technological characteristics (i.e. number of inhabitants connected to sewage installations and efficiency to filter out microplastics). Global modelling of microplastic as introduced with this study provides a tool to identify these characteristics and provides a basis for scenario analysis of microplastic fluxes from European wastewater and rivers.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.watres.2017.10.011.

References

Alcamo, J., Van Vuuren, D., Cramer, W., Alder, J., Bennett, E., Carpenter, S., Christensen, V., Foley, J., Maerker, M., Masui, T., et al., 2005. Changes in ecosystem services and their drivers across the scenarios. In: Ecosystems and Human Well-being, vol. 2. Island Press, Washington, D. C, pp. 297–373.

Arthur, C., Baker, J., Bamford, H., 2009. In: Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Mircroplastic Marine Debris. Department of Commerce, National Oceanic and Atmospheric Administration. Technical Memorandum NOS-OR&R-30.

Barnes, D.K., Galgani, F., Thompson, R.C., Barlaz, M., 2009. Accumulation and fragmentation of plastic debris in global environments. Philos. Trans. R. Soc. B Biol. Sci. 364, 1985–1998.

Bergmann, M., Gutow, L., Klages, M., 2015. Marine Anthropogenic Litter (Pp. 245–307). Springer International Publishing, Cham.

Besseling, E., Quik, J.T., Sun, M., Koelmans, A.A., 2017. Fate of nano- and microplastic in freshwater systems: a modeling study. Environ. Pollut. 220, 540–548. https://doi.org/10.1016/j.envpol.2016.10.001.

Bouwman, A., Beusen, A., Billen, G., 2009. Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050. Glob. Biogeochem. Cycles 23.

Brandsma, S., Nijssen, P., Van Velzen, M., Leslie, H., 2013. Microplastics in River

Suspended Particulate Matter and Sewage Treatment Plants. Amsterdam. IVM Report R14/02. 20 pp.
Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines woldwide: sources and sinks. Environ. Sci. Technol. 45, 9175–9179.
Carr, S.A., Liu, J., Tesoro, A.G., 2016. Transport and fate of microplastic particles in wastewater treatment plants. Water Res. 91, 174–182.
Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the marine environment: a review. Mar. Pollut. Bull. 62, 2588–2597.
Cózar, A., Echevarría, F., González-Gordillo, J.I., Irigoien, X., Úbeda, B., Hernández-León, S., Palma, A., Navarro, S., García-de-Lomas, J., Ruiz, A., Fernández-de-Puelles, M.L., Duarte, C.M., 2014. Plastic debris in the open ocean. Proc. Natl. Acad. Sci. 111 (28), 10239–10244.
Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S., 2013. Microplastic pollution in the surface waters of the Laurentian Great lakes. Mar. Pollut. Bull. 77, 177–182.
Eriksen, M., Lebreton, L.C., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., Galgani, F., Ryan, P.G., Reisser, J., 2014. Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PloS one 9 (12), e111913. https://doi.org/10.1371/journal.pone.0111913.
Essel, R., Engel, L., Carus, M., Ahrens, R., 2015. Sources of microplastics relevant to marine protection in Germany. Texte 64, 2015.
Faure, F., Demars, C., Wieser, O., Kunz, M., De Alencastro, L.F., 2015. Plastic pollution in Swiss surface waters: nature and concentrations, interaction with pollutants. Environ. Chem. 12 (5), 582–591.
Fekete, B.M., Wisser, D., Kroeze, C., Mayorga, E., Bouwman, L., Wollheim, W.M., Vörösmarty, C., 2010. Millennium ecosystem assessment scenario drivers (1970–2050): climate and hydrological alterations. Glob. Biogeochem. Cycles 24.
Fendall, L.S., Sewell, M.A., 2009. Contributing to marine pollution by washing your face: microplastics in facial cleansers. Mar. Pollut. Bull. 58, 1225–1228.
Free, C.M., Jensen, O.P., Mason, S.A., Eriksen, M., Williamson, N.J., Boldgiv, B., 2014. High-levels of microplastic pollution in a large, remote, mountain lake. Mar. Pollut. Bull. 85, 156–163.
GESAMP, 2016. Sources, fate and effects of microplastics in the marine environment: part two of a global assessment. In: Kershaw, P.J., Rochman, C.M. (Eds.), IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/UNEP/UNDP Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection). Rep. Stud. GESAMP No. 93, p. 220.
Gewert, B., Plassmann, M.M., MacLeod, M., 2015. Pathways for degradation of plastic polymers floating in the marine environment. Environ. Sci. Process. Impacts 17 (9), 1513–1521.
Gregory, M.R., 1996. Plastic 'scrubbers' in hand cleansers: a further (and minor) source for marine pollution identified. Marine Pollut. Bull. 32 (12), 867–871.
Hintersteiner, I., Himmelsbach, M., Buchberger, W.W., 2015. Characterization and quantitation of polyolefin microplastics in personal-care products using high-temperature gel-permeation chromatography. Anal. Bioanal. Chem. 407, 1253–1259.
Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347, 768–771.
Kalčíková, G., Alič, B., Skalar, T., Bundschuh, M., Gotvajn, A.Ž., 2017. Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater. Chemosphere 188, 25. https://doi.org/10.1016/j.chemosphere.2017.08.131.
Klein, S., Worch, E., Knepper, T.P., 2015. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. Environ. Sci. Technol. 49 (10), 6070–6076.
Kole, P.J., Löhr, A., Ragas, A., 2015. Autobandenslijtstof: een verwaarloosde bron van microplastics? Milieu: Opinieblad van de Vereniging van Milieuprofessionals (VVM), vol. 21, pp. 39–41 issue 5, (2015). http://hdl.handle.net/2066/151029.
Kooi, M., Besseling, E., Kroeze, C., van Wezel, A.P., Koelmans, A.A., 2017. Modelling the fate and transport of plastic debris in fresh waters. Review and guidance. In: Wagner, M., Lambert, S. (Eds.), Freshwater microplastics. Emerging environmental contaminants? Springer. https://doi.org/10.1007/978-3-319-61615-5. Hdb Env Chem 58.
Kooi, M., Reisser, J., Slat, B., Ferrari, F.F., Schmid, M.S., Cunsolo, S., Brambini, R., Noble, K., Sirks, L.A., Linders, T.E.W., Schoeneich-Argent, R.I., Koelmans, A.A., 2016. The effect of particle properties on the depth profile of buoyant plastics in the ocean. Sci. Rep. 6, 33882. https://doi.org/10.1038/srep33882.
Kroeze, C., Gabbert, S., Hofstra, N., Koelmans, A.A., Li, A., Löhr, A., Ludwig, F., Strokal, M., Verburg, C., Vermeulen, L., de Vries, W., Wang, M., van Vliet, M.T., 2016. Global modelling of surface water quality: a multi-pollutant approach. Curr. Opin. Environ. Sustain. 23, 35–45.
Kühn, S., Rebolledo, E.L.B., van Franeker, J.A., 2015. Deleterious effects of litter on marine life. In: Marine anthropogenic Litter. Springer International Publishing, pp. 75–116.
Lassen, C., Hansen, S.F., Magnusson, K., Hartmann, N.B., Rehne Jensen, P., Nielsen, T.G., Brinch, A., 2015. Microplastics: Occurrence, Effects and Sources of Releases to the Environment in Denmark. Danish Environmental Protection Agency, Copenhagen K.
Lebreton, L.C., van der Zwet, J., Damsteeg, J.W., Slat, B., Andrady, A., Reisser, J., 2017. River plastic emissions to the world's oceans. Nat. Commun. 8, 15611. https://doi.org/10.1038/ncomms15611.
Lechner, A., Keckeis, H., Lumesberger-Loisl, F., Zens, B., Krusch, R., Tritthart, M., Glas, M., Schludermann, E., 2014. The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river. Environ. Pollut. 188, 177–181. https://doi.org/10.1016/j.envpol.2014.02.006.
Magnusson, K., Norén, F., 2014. Screening of Microplastic Particles in and Down-stream a Wastewater Treatment Plant. IVL Swedish Environmental Research Institute, Sweden (2014).
Mayorga, E., Seitzinger, S.P., Harrison, J.A., Dumont, E., Beusen, A.H., Bouwman, A., Fekete, B.M., Kroeze, C., Van Drecht, G., 2010. Global nutrient export from WaterSheds 2 (NEWS 2): model development and implementation. Environ. Model. Softw. 25, 837–853.
McCormick, A., Hoellein, T.J., Mason, S.A., Schluep, J., Kelly, J.J., 2014. Microplastic is an abundant and distinct microbial habitat in an urban river. Environ. Sci. Technol. 48, 11863–11871.
Mintenig, S.M., Int-Veen, I., Löder, M.G., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res. 108, 365–372.
Napper, I.E., Bakir, A., Rowland, S.J., Thompson, R.C., 2015. Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. Mar. Pollut. Bull. 99 (1), 178–185.
Norén, F., Naustvoll, F., 2010. Survey of Microscopic Anthropogenic Particles in Skagerrak (Report commissioned by Klima-og Forurensningsdirektoratet, Norway).
Rijkswaterstaat, 2014. Emissieschattingen Diffuse Bronnen. Emissieregistratie. Bandenslijtage wegverkeer. Rijkswaterstaat-WVL, 22.
Ryan, P.G., Moore, C.J., van Franeker, J.A., Moloney, C.L., 2009. Monitoring the abundance of plastic debris in the marine environment. Philosophical Trans. R. Soc. B Biol. Sci. 364, 1999–2012.
Seitzinger, S., Mayorga, E., Bouwman, A., Kroeze, C., Beusen, A., Billen, G., Van Drecht, G., Dumont, E., Fekete, B., Garnier, J., 2010. Global river nutrient export: a scenario analysis of past and future trends. Glob. Biogeochem. Cycles 24.
Strand, J., 2014. Contents of polyethylene microplastic in some selected personal care products in Denmark. In: International Conference on Plastics in Marine Environments..
Sundt, P., Schulze, P., Syversen, F., 2014. Sources of Microplastics-pollution to the Marine Environment. Mepex for the Norwegian Environment Agency. Report no M-321/2015. Asker: Mepex Consult.
Talvitie, J., Heinonen, M., 2014. BASE Project 2012-2014: Preliminary Study on Synthetic Microfibers and Particles at a Municipal Waste Water Treatment Plant. Baltic Marine Environment Protection Commission HELCOM, Helsinki.
Van der Wal, M., Van der Meulen, M., Tweehuijsen, G., Peterlin, M., Palatinus, A., Viršek, M.K., Coscia, L., Kršan, A., 2015. SFRA0025: Identification and Assessment of Riverine Input of (Marine) Litter. Final Report for the European Commission DG Environment under Framework Contract No ENV. D (Vol. 25). 2/FRA/2012.
Van Drecht, G., Bouwman, A., Harrison, J., Knoop, J., 2009. Global nitrogen and phosphate in urban wastewater for the period 1970 to 2050. Glob. Biogeochem. Cycles 23.
Van Sebille, E., Wilcox, C., Lebreton, L., Maximenko, N., Hardesty, B.D., Van Franeker, J.A., Eriksen, M., Siegel, D., Galgani, F., Law, K.L., 2015. A global inventory of small floating plastic debris. Environ. Res. Lett. 10 (12), 124006.
Verschoor, A., de Poorter, L., Dröge, R., Kuenen, J., de Valk, E., 2016. Emission of Microplastics and Potential Mitigation Measures: Abrasive Cleaning Agents, Paints and Tyre Wear. RIVM rapport 2016-0026.
Vörösmarty, C.J., Fekete, B.M., Meybeck, M., Lammers, R.B., 2000. Global system of rivers: its role in organizing continental land mass and defining land-to-ocean linkages. Glob. Biogeochem. Cycles 14 (2), 599–621.
Webster, T.F., Harrad, S., Millette, J.R., Holbrook, R.D., Davis, J.M., Stapleton, H.M., Allen, J.G., McClean, M.D., Ibarra, C., Abdallah, M.A.-E., 2009. Identifying transfer mechanisms and sources of decabromodiphenyl ether (BDE 209) in indoor environments using environmental forensic microscopy. Environ. Sci. Technol. 43, 3067–3072.
Wright, S.L., Kelly, F.J., 2017. Plastic and human health: a micro issue? Environ. Sci. Technol. https://doi.org/10.1021/acs.est.7b00423.
Yonkos, L.T., Friedel, E.A., Perez-Reyes, A.C., Ghosal, S., Arthur, C.D., 2014. Microplastics in four estuarine rivers in the Chesapeake Bay, USA. Environ. Sci. Technol. 48, 14195–14202.