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Quantifying ecological risks of aquatic micro- and nanoplastic

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ABSTRACT

Diverse effects of nano- and microplastic (NMP) have been demonstrated in the laboratory. We provide a broad review of current knowledge on occurrence, measurement, modeling approaches, fate, exposure, effects, and effect thresholds as regard to microplastics in the aquatic environment. Using this information, we perform a 'proof of concept' risk assessment for NMP, accounting for the diversity of the material. New data is included showing how bioturbation affects exposure, and exposure is evaluated based on literature data and model analyses. We review exposure and effect data and provide a worst case risk characterization, by comparing HC_5 effect thresholds from 'all inclusive' Species Sensitivity Distributions (SSDs) with the highest environmental concentrations reported. HC_5 values show wide confidence intervals yet suggest that sensitive aquatic organisms in near-shore surface waters might be at risk.

1. Introduction

Pollution with micro- and nanoplastic is thought to cause risks to aquatic ecosystems (Koelmans, Besseling, et al., 2017; Koelmans, Besseling, & Shim, 2015). Several effect mechanisms of microplastic have been hypothesized or demonstrated, such as alteration of sediment porosity, shading, dilution of food, blockage of the gastrointestinal tract of animals, increased transport of invasive species and increased transfer of chemicals along the food chain (the vector effect) (Bhattacharya, Lin, Turner, & Ke, 2010; Galgani, Hanke, Werner, & De Vrees, 2013; Zarfl et al., 2011). Effects hypothesized or demonstrated for nanoplastic and the smaller size range of microplastic in

particular include translocation from the intestines into other tissues, oxidative stress, immune response, and particle toxicity (Bhattacharya et al., 2010; Browne, Dissanayake, Galloway, Lowe, & Thompson, 2008; Cole, Lindeque, Halsband, & Galloway, 2011; Koehler, Marx, Broeg, Bahns, & Bressling, 2008; Zarfl et al., 2011). However, whether effects will occur in nature depends on the actual exposure. Environmental risk assessment (ERA) methodology generally requires exposure concentrations to exceed effect threshold concentrations in order to conclude that a risk exists (Connors, Dyer, & Belanger, 2017). Here, we provide a review of current knowledge on occurrence, measurement, modeling approaches, fate, exposure, effects and effect thresholds as regard to microplastics in the aquatic environment. A case study for The Netherlands is provided, for the first time reviewing the available concentration data for that country. Subsequentially, we present a provisional quantitative assessment of the risk of micro- and nanoplastic in the aquatic environment. The assessment is based on the reviewed data and was done following six steps: (1) assessing ranges of exposure concentrations that are currently found in the aquatic environment globally, (2) refining the expected exposure by use of exposure models, (3) assessing the nano- and microplastic effect thresholds reported to date, (4) assessing community level effect thresholds using a species sensitivity distribution (SSD) approach, (5) reflecting on the latest insights with respect to the effect of micro- and nano-plastic on chemical transfer and risk, and (6) comparison of exposure and effect levels to characterize risk (Figure 1). This approach recognizes that although many of the tools required to obtain the necessary information for each of these steps differ from those used for soluble chemicals, the risk assessment paradigm applies evenly well to solid polymer particles.

Finally, we reflect on some of the trends in microplastic research and on the possible direction of further research.

2. Exposure assessment

Defining the actual risk of micro- and nanoplastic requires information on exposure concentrations. Below, these exposure concentrations are compared with effect concentrations in the risk characterization of the actual risk assessment (Figure 1). This section provides an overview of measured and modeled ranges of micro- and nanoplastic concentrations.

2.1. Microplastic: Global concentrations from source to sea

Whether aquatic organisms are being exposed to micro- and nanoplastic in the environment, and if so, at what concentrations, are questions that underlie the so-called retrospective exposure assessment. The aquatic environment
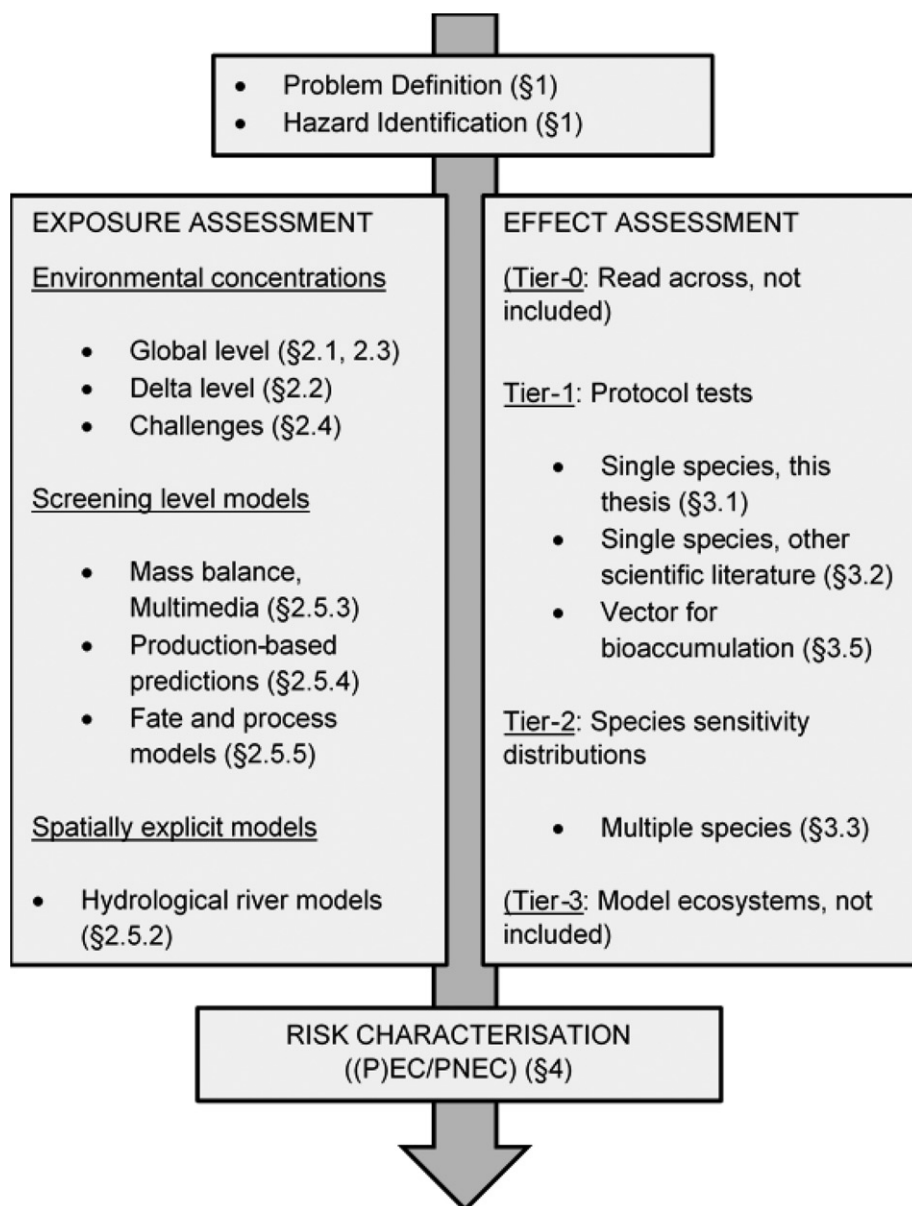
Figure 1. Tools for exposure and effect assessment as part of the general environmental risk assessment framework for micro- and nanoplastic. Based on Koelmans et al. (2017). The symbol § marks the section in which each tool is discussed.

consists of various habitats, and microplastic concentrations have been found to differ among regions. Concentrations of microplastic (plastic with a 0.1 μm–5 mm particle size) found in the environment are summarized in Table 1, arranged from inland water locations towards the ocean: surface freshwater, freshwater sediment, near-shore or estuarine surface water, subtidal sediment, beach sediment, open sea or ocean surface water and seafloor sediment. The concentrations in freshwater and estuarine habitats are shown

Table 1. Highest limits of reported ranges (HLRR) of particle concentrations going from inland toward the ocean, in the following compartments: freshwater surface water, freshwater sediment, near-shore or estuarine surface water, subtidal sediment, beach sediment, open sea or ocean surface water and seafloor sediment, per continent or oceanic region. Because of the orders of magnitude of variation, concentrations are presented on a logarithmic scale. As most studies have reported environmental concentrations in numbers of particles per mass or per volume, these are the units used in this table. The following conversion factors were used to convert to approximate particles per mass or volume where needed: microplastic particle weight 5 μg/particle, trawling depth 0.1 m, sediment density 1.8 kg/L and sediment porewater fraction 20%. These factors are explained in more detail in Section 2.4. Studies up to and including the year 2016 were used.

Continent	Africa	Asia	Europe	North America	South America	Oceania	Ref.
Freshwater surface water (log particles L⁻¹)		−5.36	−0.85	−3.92			(Eerkes-Medrano et al., 2015)
Freshwater sediment (log particles kg⁻¹ DW)			1.81	3.18			(Eerkes-Medrano et al., 2015)
Near-shore surface water (log particles L⁻¹)	−6.14	1.20	2.01	−0.27	−3.59	−6.07	(Amy L. Lusher, 2015)
Subtidal sediment (log particles kg⁻¹ DW)			2.32	2.43	−0.55		(Amy L. Lusher, 2015)
Beach sediment (log particles kg⁻¹ DW)		4.31	3.62	3.00	2.32	1.84	(Amy L. Lusher, 2015)

Oceanic region	North Atlantic	South Atlantic	North Pacific	South Pacific			Ref.
Open ocean surface water (log particles L⁻¹)	−4.40	−4.52	−2.66	−5.27			(Amy L. Lusher, 2015)
Seafloor sediment (log particles kg⁻¹ DW)	−0.55						(Amy L. Lusher, 2015)

for each continent. Open sea or ocean concentrations are provided for each oceanic region for which microplastic concentrations could be found in the scientific literature. Data was gathered from about 120 studies mentioned in the reviews by Eerkes-Medrano et al. (Eerkes-Medrano, Thompson, & Aldridge, 2015) and Lusher (Lusher, 2015) supplemented with studies on the occurrence of microplastic in the aquatic environment published in the years 2015–2016. The concentrations reported until those years vary greatly among compartments and regions but also within studies. Following Aus der Beek et al. (Aus der Beek et al., 2016), here only the highest limits of reported ranges (HLRR) are summarized, because (1) the lower limits of the reported ranges often exclude zero-encounters, which would thus imply bias, and (2) this represents the worst-case information with respect to exposure. Most concentrations in the water compartment rely on surface sampling. A recent study of samples taken from different depths down to 5 m below the ocean surface revealed that microplastic concentrations approach zero at a depth of 5 m, but are considerably higher in the water column above. The data indicated that total buoyant microplastic amounts are underestimated by a factor of 1.04 – 30 when based on surface sampling by instruments like manta trawls (Kooi et al., 2016).

The HLRR of microplastic concentrations in the environment vary by more than ten orders of magnitude (Table 1). Even after correcting for the sediment density (approximated as 1.8 kg/L) concentrations in sediment on a volume basis are generally higher than in surface water, which is explained by the settling of particles. The particles settle either as singular particles, or in aggregated or fouled form, as long as their density is higher than that of the surrounding water (Besseling, Quik, Sun, & Koelmans, 2017). Consequently, although sediment-buried plastic may be less bioavailable, sediment-dwelling organisms can be expected to be exposed to higher concentrations than pelagic organisms. Nevertheless, microplastic in sediment is expected to affect the pelagic foodweb, via transfer through the food chain and, in case of shallow aquatic systems, resuspension. Due to increased water turbulence or defouling, originally settled plastic particles are expected to become resuspended in the water column and lead to exposure of pelagic organisms (Kooi, van Nes, Scheffer, & Koelmans, 2017).

Variations in HLRR of microplastic concentrations among continents seem to be lower in the open sea and ocean areas compared to that in freshwater systems and near-shore areas. For example, the HLRR of microplastic concentrations in surface water vary by three orders of magnitude in seas and oceans, compared to five and nine orders of magnitude in fresh and near-shore water, respectively. This difference in observed heterogeneity might reflect better mixing or a difference in representativeness of the sampling in the marine environment and less mixing and larger variability

in magnitude of sources and flow conditions for fresh- and coastal waters. Sampling with a trawl over several hundreds of meters in open water might give a more representative sample than pointwise sampling with a bucket or flask from the shore. Furthermore, one could expect less mixing and a greater influence of location-specific hotspot characteristics for sediment compared to water (Besseling et al., 2017). Although this is not yet reflected in a greater variation of microplastic concentrations in sediment compared to water among continents, such a large variation is seen among studies. For example, the HLRR in near-shore sediment in Europe are found at a plastic factory site and are generally a factor 10^2 to 10^8 higher than in the surrounding areas. The absence of observations of large variations in HLRR in sediment concentrations among continents might be due to the fact that sediment sampling has been done in fewer continents than water sampling. Intercontinental comparison is especially hampered for marine sediment, as the number of studies that have included marine sediment is very limited.

The HLRR of microplastic concentrations in near-shore or estuarine areas seem to be higher than concentrations in freshwater systems and open sea or ocean. Accumulation of microplastic in these areas might be high, due to input from rivers and beaches, and from wash-back by marine water currents. Near-shore hydrodynamics, combined with high biological activity, might trap, degrade and foul both microplastic and macroplastic (Gewert, Plassmann, & MacLeod, 2015; Isobe et al., 2014; Van Sebille, England, & Froyland, 2012), which could explain the high microplastic concentrations found here. Yet, the historical development of the research field may also have influenced these differences between areas. After all, whereas awareness of marine microplastic pollution has increased through studies of the marine environment (Andrady, 2011), the freshwater environment has remained relatively underexamined (Eerkes-Medrano et al., 2015; Wagner et al., 2014), Furthermore, logistics make remote open seas or oceans harder to study than near-shore locations (Lusher, 2015). Since the chances of finding a high microplastic concentration are affected not only by the actual environmental concentrations but also by the sampling effort, this factor might partly explain the high HLRR of microplastic concentrations found in near-shore areas.

The HLRR of concentrations in the near-shore regions are higher in sediments than in the overlying water, which also applies to freshwater and the open ocean. Concentrations in beach sediments are even higher than in subtidal sediments. This can most probably be explained by the relatively low density of plastic compared to seawater, causing floating and suspended plastic to be washed ashore (Barnes & Milner, 2005; Kako, Isobe, Kataoka, & Hinata, 2014; Kataoka & Hinata, 2015; Kataoka, Hinata, & Kato, 2013; Katsanevakis et al., 2008; Wilcox et al., 2015), implying beaches may act as a filter for plastic (and other) particles. The high exposure to

sunlight, wind and waves near and on beaches increases the degradation of larger plastic items to microplastic (<5 mm) (Gewert et al., 2015; Isobe et al., 2014; Van Sebille et al., 2012). The chances of detecting these high concentrations are substantial, as beaches have by far the highest sampling effort of all habitats (Lusher, 2015).

Globally, the overall HLRR of microplastic concentrations in freshwater and near-shore surface water are to be found in Europe. Similarly, HLRR in freshwater sediment and subtidal sediment are to be found in North America, those on beaches in Asia, those in open ocean surface water in the North Pacific and those in marine sediment in the North Atlantic. However, there are remarkable data gaps regarding microplastic concentrations in several compartments for the continents of Africa and Oceania in particular, and to a lesser extent for Asia and South America (Table 1). Macroplastic concentrations are known to be high on West coast African beaches, which makes it very likely that high microplastic concentrations occur there too.

2.2. Distribution of microplastic along river deltas: A case study for The Netherlands

Although high microplastic concentrations have been found along European river deltas (Table 1), little is known about their spatial distribution in terms of concentrations, hotspots and characteristics such as shape or size. Here, as a case study for an area with little published concentration data, we summarize such data for microplastic in freshwater, estuarine and marine sediment and surface water locations of the main river delta in The Netherlands (Besseling, 2018; Besseling, Foekema, De Hoon, et al., 2015; Foekema, Hoornsman, Sonneveld, & Arenoe, 2015), as provided by Besseling (2015) and Leslie, Brandsma, Van Velzen, and Vethaak (2017). For sediment, the highest concentration was 56.3 particles/kg dry weight (DW), with highest concentrations in the freshwater part of the delta, moderate in the estuarine part and lowest in de marine part (Besseling, Foekema, De Hoon, et al., 2015). The microplastic concentrations in the sediment were in the same range as those in the German part of the Rhine catchment (up to 64 particles/kg, Wagner et al. (Wagner et al., 2014). A concentration of 20.6 particles/kg DW was found in the sediment of a ditch into which a WWTP discharges (Besseling, Foekema, De Hoon, et al., 2015). For the sake of comparison, the effluent and sludge of this WWTP were analyzed too, and contained on average 2.7 ± 1.9 (SD) particles/m^3 and $7.1 \times 10^2 \pm 7.7 \times 10^2$ particles/kg DW, respectively (Besseling, Foekema, De Hoon, et al., 2015).

Concentrations of 50 μm – 5 mm microplastic in freshwater ranged from <0.1 to 6 particles/m^3. Of these particles, 12% were in the 50-300 μm size

class, 58% were in the 300 μm – 1 mm size class and 30% were in the >1 mm size class. Number concentrations of >0.45 μm microplastic in freshwater ranged from <5 to 40 particles/L. As the latter is at least three orders of magnitude higher than the concentration range for >50 μm particles, this implies that a major fraction of the microplastic in freshwater consists of the smaller size range of microplastic particles. A comparison with the >50 μm particle numbers found in sediment and water also shows a three orders of magnitude difference, with concentrations being much higher in sediment than in water. This indicates that microplastic is removed from the water phase by settling, as already predicted by theoretical modeling (Besseling et al., 2017). The same modeling predicts that this settling causes the smaller size fraction to accumulate in sediments too. This implies that the >0.45 μm particle concentrations of up to 40 particles/L found here in freshwater might also be much higher for freshwater sediments. The microplastic concentrations found in freshwater systems were at the lower end of the 0.3-0.5 particles/m^3 range in freshwater elsewhere in Europe and the US (Eriksen et al., 2013; Faure, Corbaz, Baecher, & Felippe, 2012; Lechner et al., 2014).

Elsewhere in the Netherlands, Leslie et al. (2017) found much higher concentrations of microplastic in freshwater systems, especially in freshwater sediments. Nevertheless, a similar trend could be observed in their data, with highest concentrations in freshwater systems (48 – 187 particles/ L in urban canal water; 1.4 – 4.9 × 10^3 particles/kg DW in riverine suspended matter; <68 – 1.05 × 10^4 particles/kg DW in urban canal sediment) compared to coastal waters (100 – 3.6 × 10^3 particles/kg DW in coastal or offshore sediment). Furthermore, the latter study included analyses of 7 WWTPs, whose influent, effluent and sewage sludge contained 68 – 910 particles/L, 51 – 81 particles/L and 510 – 760 particles/kg WW, respectively. That these concentrations in sediment, surface water and WWTP influent and effluent are higher than the concentrations found elsewhere in the Netherlands and elsewhere in Europe and the US might relate to local differences or to methodological differences. Since some studies include fibre-shaped particles, and extrapolations are made based on relatively small samples, (Leslie et al., 2017) these outcomes might be prone to over- or underestimation. Differences between freshwater concentrations in rural versus urban areas were not large, and similarly no large differences were found between inlets and outlets from and towards international rivers. This either means that there are no large regional or international differences in microplastic pollution, or that water is not the appropriate medium to detect microplastic in, due to high removal rates from the water (Besseling et al., 2017). The large regional variations in shapes of particles that were found point to the latter explanation. The concentrations of

microplastic in WWTP effluents were generally about a factor of 4.4 ± 4.0 (SD) lower than those in WWTP influents (Table 2, factor difference calculated by dividing the effluent by the influent concentration, using the detection limit as concentration when no particles were found), although this varied among the different plants. This corresponds with data by Leslie et al. (2017) on different WWTPs. Concentrations of microplastic in effluent were higher than those in the surrounding freshwater systems. Nevertheless, there was no large difference between upstream and downstream concentrations near WWTPs. This further supports the idea that surface water is not the ideal medium in which to monitor microplastic pollution, due to dilution and settling out of the water column, and due to the fact that the concentrations are greatly influenced by rain, wind and the flow rate of the water on the specific sampling date. The triplicate samples of sewage sludge taken at one WWTP still showed a range of 2×10^2 – 1.6×10^3 particles/kg DW. One year later, no particles were found in the sewage sludge from this WWTP and two others. Detection limits by then were <400 – 500 particles/kg DW, so that concentrations can be assumed to be below this limit. Despite these differences between time points and WWTPs, sewage sludge can be considered a potentially large source of microplastic when released in the environment.

In summary, data suggest there is relatively low spatial variation in microplastic concentrations in water, but a large spatial variation in microplastic concentrations in sediment. This corresponds with global findings, as presented above, suggesting that:

(a) many diffuse sources contribute to microplastic pollution (Kooi, Besseling, Kroeze, Wezel, & Koelmans, 2017);

Table 2. Microplastic concentrations in freshwater in the Dutch delta (Besseling, 2018). For samples in which no particles were found, the $<$ sign indicates a detection limit (DL). The DL is the number of particles that would have been detected in a standard volume in case one particle had been present in the sample. Thus, the DL is lower when a larger sample volume was analyzed (Foekema et al., 2015).

Location type	> 50 μm microplastic particle concentration in sediment (particles/kg DW)	> 50 μm microplastic particle concentration in water (particles/m³)	> 0.45 μm microplastic particle concentration in water (particles/L)
WWTP influent		<10 – <25	20 – 40
WWTP effluent		2.2 – 9.6	<6 – 21
WWTP sewage sludge	<408 – 706		
Upstream WWTP	<2.5 – < 25.9	<0.2 – 6.0	<5 – 10
Downstream WWTP	<2.6 – 21	0.1 – 3.2	<5
Urban area	<2.6 – 56	0.1 – 0.4	<5
Rural area	<4.5 – 17	0.1 – 0.4	<5
Outlet rural area	<2.4 – <30.8	<0.1 – 0.3	<5 – <7
Inlet river	<2.7 – <10.3	<0.1 – 0.1	<5 – 5
Subtidal zone	≤ 9.9	–	–
Off-coast	≤ 9.7	–	–

(b) non-buoyant microplastic settles out of the water column within relatively short distances from their source (Besseling et al., 2017);

(c) model-guided sediment monitoring focusing on hotspots is the best way to reveal the extent of microplastic pollution (Besseling et al., 2017), for example using sediment traps;

(d) sediment-dwelling organisms living at hotspot locations are exposed chronically to microplastic (Redondo-Hasselerharm, Falahudin, Peeters, & Koelmans, 2018; Wagner et al., 2014), whereas acute exposure of pelagic organisms may occur mainly during resuspension events;

(e) differences between sampling locations might not be detectable when selecting microplastic visually, which calls for the use of polymer identification techniques.

2.3. Nanoplastics: Estimated field concentrations

It is commonly assumed that nanoplastic, that is, plastic with a size of ≤100 nm in at least one of the particles' dimensions, is emitted to (Azimi, Zhao, Pouzet, Crain, & Stephens, 2016; Stephens, Azimi, El Orch, & Ramos, 2013) and formed within the environment (Lee, Shim, Kwon, & Kang, 2013). The presence of nanoplastic has been proved under controlled laboratory conditions using nanoparticle tracking analysis (Lambert & Wagner, 2016) and dynamic light scattering (Gigault, Pedrono, Maxit, & Ter Halle, 2016). Recently, the first report of nanosized polymers occurring in ocean surface samples has been provided (Ter Halle et al., 2017). Amongst others, this study shows that pyrolysis GC-MS can be used to identify low concentrations of polymers in an environmental matrix (Besseling, 2018; Fischer & Scholz-Böttcher, 2017; Ter Halle et al., 2017; Mintenig, Bäuerlein, Koelmans, Dekker, & van Wezel, 2018), whereas UV-VIS spectroscopy and field flow fractionation (FFF) are other techniques under development which might be promising in this respect (Koelmans et al., 2015; Mintenig et al., 2018). Since no technique is so far able to detect nanoplastic number concentrations in environmental samples, it is not possible to provide actual measured environmental exposure concentrations for it. Due to the diffuse nature of sources and the lack of information on routes towards and removal mechanisms from the aquatic environment, even estimates of current concentrations are hard to make. The expected trend over time, however, is that environmental concentrations of nanoplastic will increase, because of (1) their increased application in a variety of products (Hernandez, Yousefi, & Tufenkji, 2017), (2) its production as by-product during manufacturing (Stephens et al., 2013; Zhang, Kuo, Gerecke, & Wang, 2012) and (3) the huge potential release by

fragmentation and degradation of macro- and microplastic (Koelmans et al., 2015; Lee et al., 2013; Song et al., 2017). Nanoplastic is used, inter alia, in paints, coatings, medicines, electronics and research (Koelmans et al., 2015). Manufacturing processes that are known to release nanoplastic as a by-product include thermal cutting of polystyrene foam and 3 D printing (Stephens et al., 2013; Zhang et al., 2012).

Under the influence of mechanical, thermal, chemical and biological stressors in the environment, microplastic is expected to degrade into smaller particles (Koelmans et al., 2015; Lee et al., 2013; Song et al., 2017). The highest reported values on aquatic concentrations of microplastic in the Netherlands are, as mentioned above, 10 particles/L in surface water and 56.3 particles/kg DW in sediment until the year 2016, and as published by Leslie et al. (2017) 187 particles/L in surface water and 1.05×10^4 particles/kg DW in sediment. On a global scale, highest reported microplastic concentrations in the aquatic environment are 102 particles/L water and 1529 particles/kg DW sediment, with considerable differences between regions and between freshwater, estuarine and marine habitats. Values for beach sediments, which are not included in these global maxima, are even an order of magnitude higher. Fragmentation of microplastics to nanoparticles has been measured in the laboratory (Gigault et al., 2016; Lambert & Wagner, 2016), and nanoplastics have been detected in the marine environment (Ter Halle et al., 2017). It is unknown how fast and to what extent nanofragmentation will occur in nature. Just based on mass conservation principles, fragmentation of spherical microplastic particles with a size of >0.1 μm – 5 mm into 100 nm nanoplastic particles would lead to particle concentrations that are ultimately $>10^{14}$ times higher than the currently found microplastic particle concentrations. Although non-spherical particles, such as sheets, degrade into smaller numbers of nanoplastic particles, macroplastic can potentially degrade to even larger numbers of nanoplastic particles. Thus, the estimate of 10^{14} times higher nanoplastic particle concentrations than presently measured microplastic particle concentrations is not unthinkable. Validation of this speculation however, is urgently needed. Model-based estimations of the time-scale at which such fragmentation and degradation into nanoplastic would occur are of the order of several hundreds of years (Koelmans et al., 2015).

2.4. Challenges in defining micro- and nanoplastic concentrations

The previous sections reported on assessments of the HLRR of microplastic concentrations in different habitats of different continents (Table 1). Mean concentrations from the same studies or study regions were generally a factor of two to a thousand lower. However, the differences between

methodologies makes generalization unreliable (Connors et al., 2017). Variations in methodology regard the included size range and particle shapes; reported units; calculation of maxima and the extraction and identification of plastic. These factors are briefly discussed below.

Size range and shape. We define particles with a diameter between 0.1 μm and 5 mm as microplastics. However, some studies include only particles in a narrower range as microplastic, often determined by their sampling methodology or the detection limit of devices they used. A commonly used lower limit due to mesh size lies between 300 – 800 μm, while the upper limit is often set between 2.16 and 4.75 mm or up to 5 mm (Lechner et al., 2014). The limits set in these studies result in microplastic numbers being underestimated compared to the aforementioned definition. Furthermore, studies differ as to whether all particle shapes are included, distinguishing between fragments, spheres, sheets, pellets, ropes and fibers. The choice of particle shapes being included is affected by variations in the conditions under which samples in different studies are collected and analyzed. In addition, microplastic literature in recent years has involved some discussion on the effect of contamination of samples during sampling and analyses (Foekema et al., 2013). Nowadays, attempts are made to perform studies under fully clean air and plastic-free conditions, that is, completely eliminating exposure of samples to air, as the latter could possibly contain fibers from the clothes of researchers or other dust, as well as the use of plastic materials for collection, storage and analysis (Hermsen, Pompe, Besseling, & Koelmans, 2017). Consequently, reported microplastic concentrations vary due to:

(a) having fibers excluded due to their high risk of methodological contamination;
(b) no fibers being reported, without mention of exclusion;
(c) all particle shapes being included, irrespective of methodology;
(d) inclusion of fibers because of clean air and plastic-free methods (Lusher, 2015).

Recent work increasingly aims to include the whole 0.1 μm – 5 mm microplastic size range and to avoid methodological contamination and exclude fibers. Nevertheless, the studies listed in Table 1 were included irrespective of whether they gave reasons for including or excluding certain particle shapes. Nor does the present work use any correction for the size ranges included in these studies, because insufficient information is often provided by the individual studies to allow conversions.

Reported unit and unit conversions. Microplastic concentrations are variably reported as mass or as particle numbers per mass, per volume or per

surface area of water or sediment, or even per study site. Some studies, even from recent years, only compare the results of their investigated sites qualitatively. These differences in units and lack of quantification make quantitative comparisons between studies very difficult. Several studies have used conversion factors to be able to make comparisons (Connors et al., 2017). However, this adds another source of uncertainty. Also, sampling depth, time of day and sampling technique were often not described in detail in the studies reviewed. Conversion factors used to calculate the values given in Table 1 are as follows. Concentrations reported as particle mass have been converted to numbers and vice versa (Besseling, Wang, Lürling, & Koelmans, 2014b) by using a mass per particle factor of 5 µg/particle, based on the weight of an average microplastic particle as detected on shores (Van Cauwenberghe, Claessens, Vandegehuchte, Mees, & Janssen, 2013). When outcomes were given per surface area, an assumed manta trawl sampling depth of 0.1 m has been used to convert surface area-based concentrations (particles/km^2) to volume-based concentrations (particles/m^3) (Besseling, Wang, Lürling, & Koelmans, 2014a). Volume concentrations or mass concentrations in wet weight have been transferred to dry weight concentrations by assuming a sediment density of 1.8 kg/L (Besseling, Wegner, Foekema, Van den Heuvel-Greve, & Koelmans, 2013) and a porewater fraction of 20% (Claessens, Van Cauwenberghe, Vandegehuchte, & Janssen, 2013).

Calculation of highest limits of reported ranges of microplastic concentrations. Comparing environmental concentrations of microplastic often requires medians or ranges of reported concentrations per water body, habitat type, ocean or compartment. However, an extra source of uncertainty in defining maximum, or average, concentrations is that the calculations underlying reported environmental concentrations are often unclear and differ between studies (Lusher, 2015). The present literature appears to report highly variable metrics of concentration, such as averages, medians, maximum averages, average maxima and maxima. Even a description like 'average' can imply multiple calculation methods: it can imply that the average of all samples in the study is given, or that averages per subsample or sampling site were first calculated and then the study average. Remarkably, zero-encounters or non-detects are often excluded before calculation of the averages (Eerkes-Medrano et al., 2015; Lusher, 2015), which erroneously leads to higher predicted environmental concentrations (PECs). Furthermore, calculation of a meaningful average concentration for a certain region based on data from different studies should involve a weighted average, because of the varying numbers of sampling sites used in studies. In view of these obvious deficiencies and to provide an impression of exposure under worst-case conditions as mentioned earlier, Table 1 only shows the

HLRR of microplastic concentrations. These may also have been affected by differences in calculations, e.g. because of differences between average maxima and real maxima, and may be highly dependent on the number of sampling sites because of spatial heterogeneity. However, the number of additional calculation steps used to combine studies is smaller than for averages. Furthermore, the high variability of concentrations among sampling sites means that if averages are below effect threshold concentrations, they are not indicative of whether there could be a risk or not. By using the HLRR only, one cannot assess the risk for the entire region, but at least one can assess whether there is a risk for a certain spot. Furthermore, from a risk perspective, evaluating the HLRR of microplastic concentrations would comply with the 'worst case' approach in risk assessment. If, for instance, the HLRR were sufficiently below effect threshold concentrations, all other sites studied in the region can be assumed to be free of risk.

Extraction and identification of plastic. Environmental media that may contain nano- or microplastic will also contain a fraction of water, mineral constituents and/or organic matter. This is the case for abiotic as well as biotic samples. A variety of methods have been applied to isolate nano- or microplastic from the samples for plastic analyses. These include density separations with varying types and concentrations of salts, sieving, removal of organic matter with acids, bases, peroxide and enzymes (or combinations thereof) and drying or decomposing samples at different temperatures (Hermsen et al., 2017; Hidalgo-Ruz, Gutow, Thompson, & Thiel, 2012; Kühn et al., 2017; Lusher, Welden, Sobral, & Cole, 2017; Rocha-Santos & Duarte, 2015; Song et al., 2015; Van Cauwenberghe, Devriese, Galgani, Robbens, & Janssen, 2015). These methods differ in the extent to which particles other than plastic are removed, the likelihood of contamination of the samples with procedural plastic particles from materials used or the work space (Foekema et al., 2013; Torre, Digka, Anastasopoulou, Tsangaris, & Mytilineou, 2016), but also in the extent to which plastic particles that are present in the environmental samples are retained in the samples. Losses of plastic particles occur during these procedures, depending on the number of extraction cycles involved and the aggressiveness of the chemicals used. Several chemicals used for sample preservation or for the digestion of tissue or organic matter, as well as high temperatures, are known to degrade specific polymer types, which thus will affect the analysis results (Dümichen et al., 2015; Kühn et al., 2017; Lenz, Enders, & Gissel, 2016; Lusher et al., 2017). After clean-up of the samples, the subsequent detection of microplastic particles is usually done by visual inspection and increasingly by polymer identification techniques using reference spectra, like FTIR and Raman spectroscopy. The eventual identification, however, is highly dependent on the method used. For instance, of the particles visually

judged as plastic, a percentage varying from 1.4 to 70% was identified as plastic by focal plane array (FPA), micro-Fourier transform infrared (micro-FTIR) spectroscopy or Raman spectro-scopy (Connors et al., 2017; Eriksen et al., 2013; Hermsen et al., 2017; Hidalgo-Ruz et al., 2012; Löder & Gerdts, 2015).

The above challenges illustrate that the analysis of plastic debris in environmental samples is not straightforward. Nevertheless, the number of studies underlying the data summarized in Table 1 is high. If the above restrictions are kept in mind, Table 1 thus gives us at least an impression of the HLRR of environmental microplastic concentrations known to date. Future studies should stick to standardized quality assurance criteria such as those provided by Hermsen et al. (Connors et al., 2017; Hermsen et al., 2017; Twiss, 2016), as there seems to exist a relation between reported particle counts and the degree of quality assurance (Hermsen et al., 2017).

2.5. Modeling the fate of and exposure to microplastic

Introduction. Models can be used to assess the exposure to micro- and nanoplastic in the context of risk assessment (Figure 1). For microplastic, models can be used for spatiotemporal interpolation based on occurrence data, and can guide monitoring campaigns (Aalderink, Zoeteman, & Jovin, 1996; Besseling et al., 2017). As the current techniques are not yet capable of detecting nanoplastic in environmental samples, prospective exposure assessment for nanoplastic is completely dependent on modeling (e.g., Everaert et al., 2018). Based on future emission scenarios, models can be used to forecast future microplastic concentrations in environmental media (Koelmans, Kooi, Law, & Van Sebille, 2017), and can be used to predict when critical effect thresholds are exceeded (Everaert et al., 2018). Since transport and fate models have been used before for other pollutants and other particle types like algae, micro-organisms, sediment and nanomaterials, models for micro- and nanoplastic can build upon this existing knowledge (Aalderink et al., 1996; Besseling et al., 2017). Spatiotemporally explicit models are most helpful when it comes to transport and retention patterns within freshwater and marine systems. For freshwater systems, this type of model has been developed for micro- and nanoplastic at different scales, ranging from single river catchments (Besseling et al., 2017; Nizzetto, Bussi, Futter, Butterfield, & Whitehead, 2016) to the global scale (Kooi et al., 2017; Lebreton et al., 2017; Schmidt, Krauth, & Wagner, 2017; Siegfried, Koelmans, Besseling, & Kroeze, 2017). So far, these models have been theoretical or empirical, and though validated with other particle types, they have not been fully validated for plastic particles yet. For marine systems, particle behavior and ocean circulation models are used to model

the fate of microplastic and to find optimal clean-up strategies (Kooi, van Nes, et al., 2017; Sherman & van Sebille, 2016). Below, currently available microplastic models are summarized going from their source towards and inside the oceans.

River transport modeling. Spatiotemporally explicit models have addressed catchment hydrology, soil erosion, sediment budgets (Nizzetto et al., 2016), advective transport, homo- and heteroaggregation, sedimentation-resuspension, polymer degradation, presence of biofilm and sediment burial processes (Besseling et al., 2017). Besseling et al. (2017) showed that for particles in the 100 nm to 10 μm size range, concentrations in the water of a 40 km river stretch dropped from an input concentration of 1 ng/L to steady-state concentrations between 0.5 and 0.8 ng/L at the end of the stretch (Besseling et al., 2017). Another model study indicated that leaving the aggregation of small plastic particles with other suspended solids out of consideration results in higher percentages being predicted to remain in the water column (Nizzetto et al., 2016). However, several empirical studies have shown that fouling followed by aggregation and settling is relevant for particle fate in aquatic systems (Besseling et al., 2017; De Klein, Quik, Bauerlein, & Koelmans, 2016; Long et al., 2015). Therefore, leaving aggregation out of consideration might lead to overestimation of aquatic plastic particle concentrations. Both of these river catchment modeling studies agree that concentrations of particles in the upper part of the microplastic size range become reduced to 0 to 20% of the input concentration in the water column (Besseling et al., 2017; Nizzetto et al., 2016). High flow periods might, however, cause resuspension and remobilise this pool (Nizzetto et al., 2016). The locations of hotspot concentrations in riverine sediment largely depend on the particle size of the plastic and on river morphology. In our study, the highest steady-state concentration in riverine sediment was predicted to be 5 mg/kg for >1 mm particles at <1 km from the 1 ng/L input source (Besseling et al., 2017). For most other sizes and locations, the steady-state concentrations were a factor 2 to 15 lower. The calculated steady-state concentrations within 1 km from the source are in line with concentrations of microplastic found by Wagner et al. (2014) in the sediment of freshwater systems in Europe (Besseling et al., 2017). Apart from relatively close to the source, hotspot concentrations of microplastic are also expected in sedimentation areas characterized by a reduced flow velocity. Hence, monitoring campaigns and impact assessments should focus on these regions (Besseling et al., 2017; Nizzetto et al., 2016).

Modeled effects of polymer density (Besseling et al., 2017; Nizzetto et al., 2016) and formation of biofilms (Besseling et al., 2017) on the fate of plastic particles were not large, although this formation of biofilms and its effects on fate processes has been empirically confirmed, and is receiving

increasing attention in the recent literature (Artham et al., 2009; Carson, Nerheim, Carroll, & Eriksen, 2013; Kooi, Nes, Scheffer, & Koelmans, 2017; Lobelle & Cunliffe, 2011; Long et al., 2015). As the modeled plastic particles were spherical, particles of different shape might behave differently (Besseling et al., 2017; Kooi, Besseling, et al., 2017). A global modeling study incorporating microplastic of different shapes and point sources predicted that the majority of riverine transport of microplastic to sea would consist of synthetic polymers from tyre abrasion (>40%) (Siegfried et al., 2017). The other fractions are made up by plastic-based textiles abraded during laundry (29%), synthetic polymers and plastic fibers in household dust (19.4%) and microbeads in personal care products (9.8%). Yearly, 1.15 to 2.41 million tonnes of plastic waste are estimated to enter the oceans from rivers, 67% of which is made up by the top 20 most polluting rivers (Lebreton et al., 2017).

Emission-based mass flow modeling and multi-media modeling. Emission-based mass flow modeling and multi-media modeling have been used before to estimate fluxes of pollutants and particles between compartments like the atmosphere, terrestrial soil, surface water and water-bottom sediment (Koelmans et al., 2015; Kooi, Besseling, Kroeze, van Wezel, et al., 2017; Meesters, Koelmans, Quik, Hendriks, & van de Meent, 2014). For microplastic, the first provisional calculations of mass flows from WWTPs to water bodies predicted a retention for microplastic of 40 to 96% in WWTPs, depending on the type of plant (Van Wezel, Caris, & Kools, 2016). In comparison, the new data provided in section 2.2 indicate a retention of 53% ± 46 (SD) in three WWTPs, while Leslie et al. (2017) reported 72 ± 61% (SD). Multi-media models like SimpleBox4Nano (SB4N) calculate average background concentrations in different compartments (Kooi, Besseling, Kroeze, van Wezel, et al., 2017). Assuming a yearly emission of 20 kilotons (Sherrington, Darrah, Hann, Cordle, & Cole, 2016), of which 50% to the water and 50% to the soil, resulted in the average background predicted environmental steady-state concentrations (PECs) in the Rhine catchment (Europe) shown for each compartment in Table 3 (Kooi, Besseling, Kroeze, van Wezel, et al., 2017). Note that these PECs relate to mass concentration and thus cannot be compared directly with measured number concentrations. Furthermore, these PECs per size class are based

Table 3. Distribution of plastic particles of different sizes over the soil, water and aquatic sediment compartments, as predicted by the multi-media model SB4N. PECs are based on a yearly emission of 20 kt (Kooi et al., 2017).

Particle size	0.1 μm	1 μm	10 μm	100 μm	1 mm
Soil (log μg/m^3)	6.43	6.38	6.17	4.57	2.62
Water (log μg/m^3)	5.45	5.44	5.39	4.89	3.08
Sediment (log μg/m^3)	1.52	2.41	4.42	6.07	6.26

on the yearly emission of 20 kt, such that PECs based on new estimated yearly predictions can be derived from the ratios between the 20 kt used here and the respective emissions. Although the resulting output concentrations are highly dependent on the assumed yearly emissions in a catchment, SB4N clearly predicted different fates for microplastic particles of different sizes (Kooi, Besseling, Kroeze, van Wezel, et al., 2017). Whereas PECs of nanometer and micrometre sized plastic particles are highest in the soil and water compartments, PECs of particles bigger than 0.1 mm are expected to be highest in aquatic sediments (Table 3). This largely corresponds with the distribution of micrometre and millimeter sized plastic particles over water and sediment calculated with the aforementioned spatiotemporally explicit models (Besseling et al., 2017; Nizzetto et al., 2016). However, compared to the outcomes of SB4N, spatiotemporally explicit modeling predicted a higher retention of nanoplastic in sediments due to settling of aggregated nanoplastic. Another emission-based mass flow model was used by Siegfried et al. (2017) to calculated riverine transport of microplastic in Europe. The model revealed large spatial differences in transported plastic mass, largely as the result of differences in the technological status of WWTPs. For the transport of plastic from production and consumption sites towards the coastal and marine environment, Siegfried et al. (2017) used the output from modeling simulations provided by Besseling et al. (2017). An extrapolation towards the year 2050 showed that riverine transport of plastic will increase in some rivers while decreasing in others, the latter decreases being mainly explained by the expected improved wastewater treatment (Siegfried et al., 2017).

Coastal and marine microplastic concentration estimates based on plastic production. Recently, Van Cauwenberghe predicted microplastic concentrations in the coastal and marine environment by using plastic production data (Van Cauwenberghe, 2016). Van Cauwenberghe calculated the cumulative emission of plastic based on the world plastic production, assuming that 1.7 to 4.7% of this annual plastic production reaches the oceans (Van Cauwenberghe, 2016). Using this percentage, current coastal and open ocean microplastic abundance as well as extrapolations of future concentrations were calculated (Table 4) (Van Cauwenberghe, 2016). Two scenarios to forecast a range of future environmental plastic concentrations were used: a business-as-usual continuation of the current growth of the annual plastic production of 4.5% per year, and an immediate stop to plastic production (Table 4). (Van Cauwenberghe, 2016) The production-based estimates obtained this way appeared to exceed current measured concentrations of total floating plastic debris particles by only one order of magnitude. Given the uncertainties in data and calculation, this overestimation was considered acceptable (Van Cauwenberghe, 2016), also because

Table 4. Coastal and open-ocean microplastic concentrations in surface water and sediment, estimated by VanCauwenberghe (2016).

Particle size	2015	2100	
		Immediate production stop	Business-as-usual
Coastal surface water (particles/L)[a]	$4.7 \times 10^{-4} - 2.1$	$2.7 \times 10^{-3} - 11.9$	$0.03 - 129.4$
Coastal sediment (particles/L)	$9.5 - 3.5 \times 10^3$	$55.1 - 2.1 \times 10^4$	$597 - 2.2 \times 10^5$
Open ocean surface water (particles/L)	$1.3 \times 10^{-4} - 0.3$	$7.5 \times 10^{-4} - 2.0$	$0.01 - 21.1$
Seafloor sediment (particles/L)	$0.7 - 15.7$	$3.7 - 91.1$	$40.5 - 987.2$

[a]Based on an assumed average polymer density of 1100 kg/m^3 and a particle size distribution of 10% 1 – 5 mm, 25% 0.3 – 1 mm and 65% 1 – 300 μm sized particles.

the calculation did not take into account the fact that part of the emitted plastic is missed during sampling, settles in deeper layers (Koelmans, Kooi, Lavender-Law, & Van Sebille, 2017) and/or resides at beaches.

Fate processes and models for the estuarine and marine environment. Overview of current models. As plastic particles reach the river mouth, the relative importance of the different processes that affect their fate in rivers is assumed to change (Figure 2). Here, the higher salinity causes a wider variety of polymer types to drift within or float upon the water column. The change in salinity can also destabilize dispersed submicron particles, causing aggregation of particles (Koelmans et al., 2015) that might become non-buoyant, and may result in settling. Particles with intermediate polymer density would settle in freshwater but float in marine water. Polymer types that are on their way to settle, might change direction when they reach the estuarine environment. Their fate depends on the type of estuarine system (Lima, Costa, & Barletta, 2014; Williams, Hindell, Swearer, & Jenkins, 2012). In a salt-wedge estuary, these polymer types will most probably drift above the halocline within the relatively fresh upper part of the water column, whereas in well-mixed estuaries they are likely to be pushed even further upward towards the water surface. The presence of a halocline (Obbard et al., 2014) as well as the strength of the tides (Ballent, Pando, Purser, Juliano, & Thomsen, 2013; Mathalon & Hill, 2014) will affect the time during which plastic particles remain within the water column. In the marine environment too, the vertical distribution might consist of a buoyant, a settling and an in-between fraction. The in-between fraction, at intermediate depth, is thought to remain there due to the opposing mechanisms of fouling and buoyancy (Kooi et al., 2017). Whereas many previous studies predicted that the majority of microplastic in the marine environment would be present in the surface layer of the upper few meters (Kooi et al., 2016), recent modeling scenario studies indicate that the fraction at a lower, intermediate, depth may well be larger (Kooi, van Nes, et al., 2017). Aggregation of plastic particles with materials of higher density will accelerate settling (Besseling et al., 2017; Kooi, van Nes, et al., 2017), but the same process will slow down the settling rate of other mineral or organic
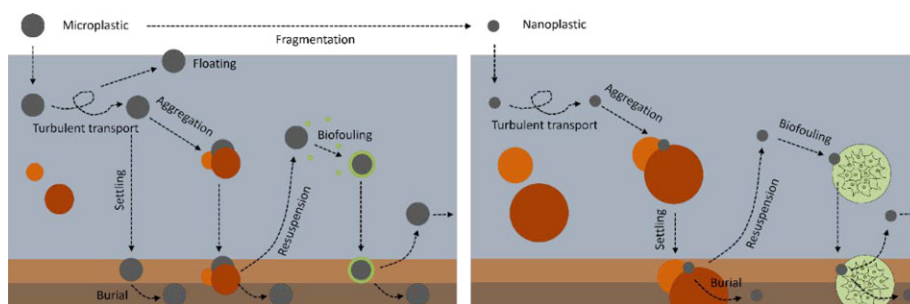
Figure 2. Processes that affect the fate of plastic particles in the aquatic environment, adapted from Kooi et al. (2017).

particles, as inclusion of plastic in the aggregate reduces their overall density (Long et al., 2015). Diurnal and seasonal fluctuations in photosynthesis, and hence in biofilm growth, might induce an oscillating movement of micrometre sized plastic over a depth range down to about 75 m below the ocean surface (Kooi, van Nes, et al., 2017; Van Sebille et al., 2012).

The global spatial distribution of plastic is affected by several oceanographic processes. Global ocean circulation, tides and Ekman transport cause accumulation to occur in five to six main regions (Maximenko, Hafner, & Niiler, 2012; Van Sebille et al., 2012). Using ocean surface current models, combined with debris concentrations or spatial macroplastic beach clean-up data, resulted in surface concentration estimates for macroplastic which could be compared with spatial distributions or species-specific habitats of sea turtles and birds (Schuyler, Hardesty, Wilcox, & Townsend, 2014; Wilcox et al., 2013; Wilcox, Van Sebille, & Hardesty, 2015). These modeling efforts revealed where turtles and sea birds have the highest encounter rates with plastic and suggested where cleaning efforts might be most cost-effective. Interestingly, this is not so much in the main subtropical oceanic plastic accumulation regions, but instead in parts of the Southern Ocean where plastic concentrations are not extremely high but the number of seabird species is (Wilcox et al., 2015), namely off the coast of China and in the Indonesian archipelago near large sources of debris from land (Schuyler et al., 2014; Sherman & van Sebille, 2016) and near the typical entry point where debris enters the Gulf of Carpentaria (Wilcox et al., 2013).

Some examples of processes currently missed by transport models. So far, plastic particle fate models have mainly included abiotic processes only, the exception being the influence of biofilm formation. However, it is not only the abiotic processes of beaching and incorporation into marine snow, but also more biotic processes, like ingestion and bioturbation, which are likely to affect the fate of plastic particles. Besseling et al. (2015) studied the feedback between microplastic and bioturbation at the sediment-water interface (Besseling, Foekema, Ren, Van den Heuvel-Greve, & Koelmans, 2015).

They used parallel cosms with and without lugworms (*Arenicola marina*). Cosms with lugworms inhabiting polyethylene-contaminated sediment contained 200 individuals/m^2, which is within the range of environmentally realistic population densities (Beukema & De Vlas, 1979). Microplastic concentrations in water were measured by microplate reader with Microplate Data Collection & Analysis Software (BioTek, USA). Bioturbation by *A. marina* appeared to increase the concentration of microplastic in the overlying water (Figure 3). The production of feces heaps and the movement of lugworms along the sediment-water interface facilitated the release of polyethylene particles from the sediment. The data thus reveal that microplastic can not only have effects on organisms (Besseling et al., 2013; Besseling et al, 2017), but organisms can also affect the distribution of microplastic in their surrounding compartments. This suggests a direct feedback between microplastic fate and effects. Bioturbating organisms can be hypothesized to affect the relative importance of sediments as a sink and the temporal bioavailability of microplastic (Besseling, Foekema, Ren, Heuvel-Greve, & Koelmans, 2015). Other species of ecosystem engineers, including bivalves (Paredes, Koelmans, & Besseling, 2015; Wegner, Besseling, Foekema,
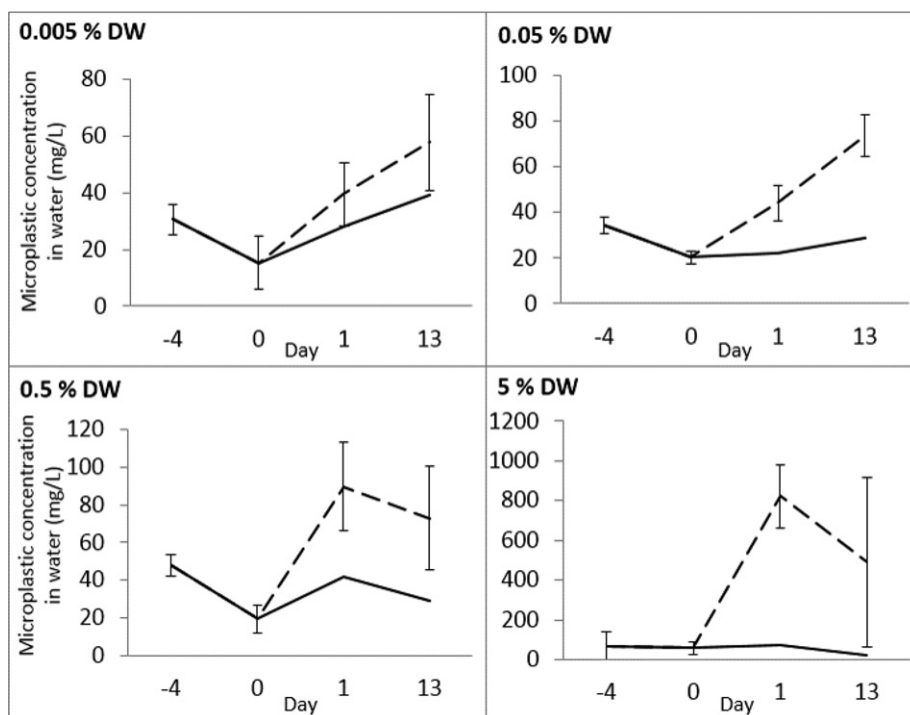
Figure 3. Microplastic concentrations (g/L ± SD) in overlying water as a result of bioturbation in sediment, with initial microplastic concentrations of (top - bottom) 0.005, 0.05, 0.5 and 5% dry weight (DW). Lugworms were added to the cosms on day 0 (dotted lines). Solid lines represent cosms without lugworms (Besseling et al., 2015).

Kamermans, & Koelmans, 2012), can also be expected to alter micro- and nanoplastic concentrations in sediment and water compartments.

Apart from the environmental concentrations, internal exposure to plastic particles in the aquatic environment depends on the actual ingestion by organisms. Plastic uptake can be modeled as a mass balance of ingestion and loss processes, such as that calculated previously for a humpback whale (Besseling, Foekema, Van Franeker, et al., 2015). This biodynamic modeling approach has also been used to model the ingestion of nano-, micro- and macroplastic by worms, fish and birds (Besseling et al., 2015; Herzke et al., 2016; Koelmans, Besseling, & Foekema, 2014; Koelmans, Besseling, Wegner, & Foekema, 2013). Most of these studies have modeled ingestion to determine bioaccumulation of hydrophobic contaminants (Herzke et al., 2016; Koelmans et al., 2014; Koelmans et al., 2013) and rarely solely in the interest of ingestion itself (Besseling et al., 2015). This calculation of steady-state microplastic concentration may apply more universally to other organisms too and can be used to model microplastic transfer for entire food webs (Diepens & Koelmans, 2018). Tissue or organ concentrations of nanoplastic particles that are possibly transferred beyond the gut system might be modeled in a similar way.

Since plastic concentrations can vary along the vertical gradient of the water column, one should take care to use the appropriate average aqueous concentration over an appropriate depth interval to calculate steady-state concentrations in organisms. The presence or absence of a halocline in estuaries, as well as oscillatory movements in oceans (Kooi, van Nes, et al., 2017), affect the concentrations of plastic particles and to some extent also those of organisms. Consequently, the use of average aquatic concentrations cannot directly be used to predict actual exposure concentrations.

Furthermore, unless microplastic concentrations are too low to be detected, characteristics of different filter feeding species probably affect whether microplastic ingestion is solely a matter of chance of encounters. Several copepoda and bivalves are known to be able to discriminate between edible and non-edible particles, leading to post-ingestive food selection or pausing of feeding when edible to non-edible particle ratios are insufficient (Ayukai, 1987; Brillant & MacDonald, 2000; Donaghay & Small, 1979; Huntley, Barthel, & Star, 1983; Vroom, Koelmans, Besseling, & Halsband, 2017; Wegner et al., 2012). Other species are known to adjust ingestion rates depending on the nutritional value or the size of the food (Koelmans et al., 2013; Wilson, 1973). Thus, species-specific characteristics affect whether lower or higher ingestion rates than determined by chance encounter can be anticipated. For species with different feeding types, such as scavengers and active predators, ingestion rates are differently related to the prevailing environmental concentrations. One way to model internal

concentrations in these organisms could be by defining a plastic encounter rate, using their beak width, beak open-to-close ratio, beak open-to-feeding ratio, swimming distance and the aqueous plastic concentration. Large differences in stomach concentrations between individuals of the same species (De Stephanis, Giménez, Carpinelli, Gutierrez-Exposito, & Cañadas, 2013; Foekema et al., 2013; González Carman et al., 2014; Van Franeker et al., 2011) are partly explained by the regions they inhabit. However, when large differences between individuals of one species are found within the same region, it might suggest that individual food selection preferences greatly affect ingestion. Such large differences in individual food selection strategies are seen in many species and mean that plastic encounter rates can mainly be useful to predict average ingestion rates by a species in a region, not aiming to represent the possibly large individual differences.

3. Effect assessment

To define the actual risks of micro- and nanoplastic, an assessment of exposure needs to be combined with an assessment of effect thresholds. Eventually, these can be combined with one another in a risk characterization (Figure 1). The sections below present an overview of effect thresholds for physiological effects of micro- and nanoplastic and effects on bioaccumulation of chemicals in organisms.

3.1. Review of the literature on effect thresholds

There is considerable data available on the occurrence of macro- and microplastic in biota, mainly for the higher trophic levels, but also for invertebrates (De Stephanis et al., 2013; Foekema et al., 2013; Hermsen et al., 2017; Jabeen et al., 2017; Murray & Cowie, 2011; Santos, Andrades, Boldrini, & Martins, 2015; Secchi & Zarzur, 1999; Taylor, Gwinnett, Robinson, & Woodall, 2016; Wright, Thompson, & Galloway, 2013). Effects of plastic on organisms have been hypothesized or demonstrated to relate to entanglement, blocking of intestines, reduced nutritional value of food, increased exposure to plastic-associated chemicals and particle toxicity. Some of these effects have mostly been observed for macroplastic, such as entanglement and blocking of intestines of organisms in the wild (Kühn, Bravo Rebolledo, & Van Franeker, 2015). Other effects are more likely to be caused by smaller particles such as micro- and nanoplastic.

The scientific literature was searched for data on effect thresholds in order to ascertain how adverse effects of micro- and nanoplastic are distributed among species, ecosystems, exposure media and plastic particles with varying characteristics. The overview presented here is based on 168

published effect thresholds from 66 different studies, as well as effect thresholds summarized by Lusher (2015) and Connors et al. (2017) and supplemented with effect thresholds published until July 2017. These threshold data are summarized in Table 5 for each exposure medium, size category, ecosystem and threshold value.

What has to be considered an effect of plastic on an organism is still being debated in the field of plastic debris research. In some studies, for instance, plastic ingestion or trophic transfer, that is, secondary ingestion via a plastic-containing lower trophic organism (Au, Lee, Weinstein, van den Hurk, & Klaine, 2017; Farrell & Nelson, 2013; Setälä, Fleming-Lehtinen, & Lehtiniemi, 2014; Tosetto, Williamson, & Brown, 2017) has been framed as an adverse effect in itself, whereas the effect actually arises from the physiological consequences of the ingestion, such as gut obstruction and the consequent growth reduction, or an adverse effect may in fact even be absent. For this reason, plastic ingestion as an endpoint of effect is excluded here and only the following endpoints are considered: survival, feeding, growth, weight loss, reproduction, molting, malformation, behavior, photosynthesis, oxidative stress, enzyme activity, inflammation, gene expression and nutrient cycling. These endpoints can all be assumed to affect population size, given time, eventually leading to a change of community composition and possibly of ecological functioning. Only those studies were included that did not report the inclusion of associated chemicals at relevant effect concentrations. Effects of plastic on bioaccumulation of chemical substances are discussed separately further on in the present review. This is in line with the suggestion by Koelmans et al. (2017) to deal with the additional hazard of bioaccumulation of chemical substances separately following existing risk assessment methods.

The effect thresholds derived from the literature were partly EC_{50} (Effect concentration at which 50% of the exposed organisms is affected) values, partly LOEC (Lowest observed effect concentration) values and partly NOEC (No observed effect concentration) values for organisms that were exposed to micro- or nanoplastic via water, food or sediment. As threshold concentrations are provided in varying units in the literature, the following conversions were used to express all data on the basis of weight per liter of water or kg of DW sediment or food: particle numbers were converted into mass data for spherical particles and fibers using the formulas for sphere and cylinder volume, respectively (Connors et al., 2017). For same-diameter but irregular particles, half the volume of a sphere was assumed. If a range of particle sizes was used, the average radius was used in the equations. If no polymer density was provided, the polymer densities given by Andrady (Andrady, 2011) were used, and when exposure included a mixture of polymer types, the average polymer density was calculated. A sediment density

Table 5. Summary of published effect threshold data of micro- and nanoplastic on organisms. Thresholds: LC_{50}: lethal dose 50%, EC_{50}: effect concentration 50%, LOEC: lowest observed effect concentration, NOEC: no observed effect concentration.

Exposure medium	Size category	Ecosystem	LC_{50}	EC_{50}	LOEC	NOEC
Water (mg/L)	Micro	Fresh	0.4 – 57	5 – 172	$6.9 \times 10^{-9} - 2 \times 10^{5}$	0.02 – 400
		Brackish	23.5	0.04 – 0.1	$6.9 \times 10^{-9} - 1.8 \times 10^{4}$	0.4 – 313
		Marine	–	–	$9.1 \times 10^{-3} - 2.5 \times 10^{3}$	$2 \times 10^{-3} - 510$
	Nano	Fresh	4 – 36	0.5 - 1.6	$4.5 - 1 \times 10^{3}$	0.5 – 1
		Brackish	0.2 – 2.2	–	–	1 – 313
		Marine	0.8 – 3.9	13	0.1 – 250	10 – 100
Sediment/food (g/kg DW)	Micro	Fresh	–	–	–	700
		Brackish	–	–	–	–
		Marine	–	–	0.1 - 100	0.3 - 100
	Nano	Fresh	–	–	1	–
		Brackish	–	–	–	–
		Marine	–	–	–	–

of 1.8 kg/L (Besseling et al., 2013) and a porewater fraction of 20% were used where needed (Claessens et al., 2013).

3.2. Species sensitivity distributions (SSDs) as a tool to explore microplastic effect data

Effects of chemical stressors are often reported for individual species. This, however, does not offer insights into the consequences of the respective stressors at community level. To increase the relevance of the effect data for this community level, an approach has been developed that combines effect data for individual species in a species sensitivity distribution (SSD). SSDs are log-linear regressions through measures of effect to determine the affected fraction of species at a given concentration (Posthuma, Suter, & Traas, 2002). SSDs are used in the environmental risk assessment of substances, but also have been used for particles (Connors et al., 2017). SSDs can be used to estimate the concentration at which 5% of the species in a community is affected, which is referred to as the 'Hazardous Concentration for 5% of the Species' (HC_5). Using the literature data, provisional SSDs for the stressors micro- and nanoplastic were generated with the SSD generator from the US-EPA (EPA (United States Environmental Protection Agency), 2016a).

These SSDs are presented as provisional, since they involve several uncertainties. Ideally, SSDs use the effect threshold values of one single endpoint (one type of harm) for ≥ 10 different species, with environmental variables kept constant (Diepens et al., 2016). Consequently, the observed SSD only expresses the variability of the species sensitivities and the experimental variability. Such data is not yet available for plastic as a stressor. Plastic as a stressor has unique features, which means that an SSD for microplastic is fundamentally different from single substance-single endpoint SSDs. First, microplastic or "plastic debris" is a mixture of different sizes and types of particles, which implies that the observed distribution of the stress response reflects this variability. For instance, some studies may have used monodisperse particles that all are ingested, whereas others may have used broader size distributions with large particles blocking the ingestion of smaller ones. Furthermore, the variability covered by the various studies included in the SSD may still not fully represent the variability with respect to microplastic occurring in nature. Second, the different types and sizes of particles trigger responses through different modes of action (different types of harm), which implies that the observed distribution of the stress response reflects this variability too. Previous studies have combined data from different endpoints to overcome the lack of available data (Everaert et al., 2018; Hermsen, 2015; Van Cauwenberghe, 2016). Here, a

pragmatic criterion for combining different endpoints was used, by combining those endpoints that all imply harm at the population level of a species. Hence, from the large set of thresholds summarized in the previous section, we here combine the endpoints of survival, reproduction and growth, the latter including any reduction in weight, length or hampering of molting (while a subsequent effect of these endpoints on reproduction can be expected too) to construct SSDs, as they are all population-threatening. Both acute and chronic LC_{50}, EC_{50} and LOEC values were used, with exposure durations varying from minutes to months. The comparability of these data was improved by using extrapolation factors from Diepens et al. (2016) to infer chronic LOEC values for these different effect thresholds. The ranges of extrapolation factors used for $\leq$21-day LC_{50}, EC_{50} and LOECs values were 10-30, 5-15 and 3-10, respectively (Diepens et al., 2016). When exposure duration was $\leq$5 d, the higher ends of these ranges were used (extrapolation factors of 30, 15 and 10), while for exposure durations >5 d but <15 d, the intermediate values of these ranges of extrapolation factors were used (20, 10, 6.5), and when exposure duration was $\geq$15 d but <21d, the lower ends of these ranges were used (10, 5, 3). For 21-day LC_{50} and EC_{50} values, an extrapolation factor of 5 was used to derive the chronic LOEC (Diepens et al., 2016). Since data included those for several organisms that inhabit a salinity range from fresh to brackish, and since no mechanism is known or expected for an effect of salinity on the physical adverse outcome pathways related to microplastic, effect thresholds for marine, estuarine and freshwater species were combined. This combination of taxa from different habitats and ecosystem types is strictly for calculation purposes, and does not imply that they are supposed to share the same habitat. A similar approach of combining data for freshwater and marine invertebrate species has been used for pesticide risk assessment (Diepens et al., 2016; Jeong et al., 2016). In conclusion, the tentative SSDs for plastic debris presented here reflect the combined variability of species sensitivity, properties of the stressor and effect mechanisms, as a function of the dosage and thus can be referred to as "all-inclusive" SSDs. The extrapolated chronic effect thresholds used to construct the SSDs can be found in the Appendix Tables A1 and A2.

The separately constructed SSDs for organisms exposed to micro- and nanoplastic via the water phase (expressed as plastic mass per volume) are shown in Figure 4. Of the species studied, the one that seems the most sensitive to exposure to microplastic via the water phase is the Japanese oyster *Crassostrea gigas* (2 – 6 µm spherical PS particles), and the least sensitive the amphipod *Gammarus fossarum* (32 – 250 µm irregular PMMA and PHB particles) (Table A1, Supporting Information). For nanoplastic, the most sensitive is the copepod *Tigriopus japonicus* (50 nm spherical PS
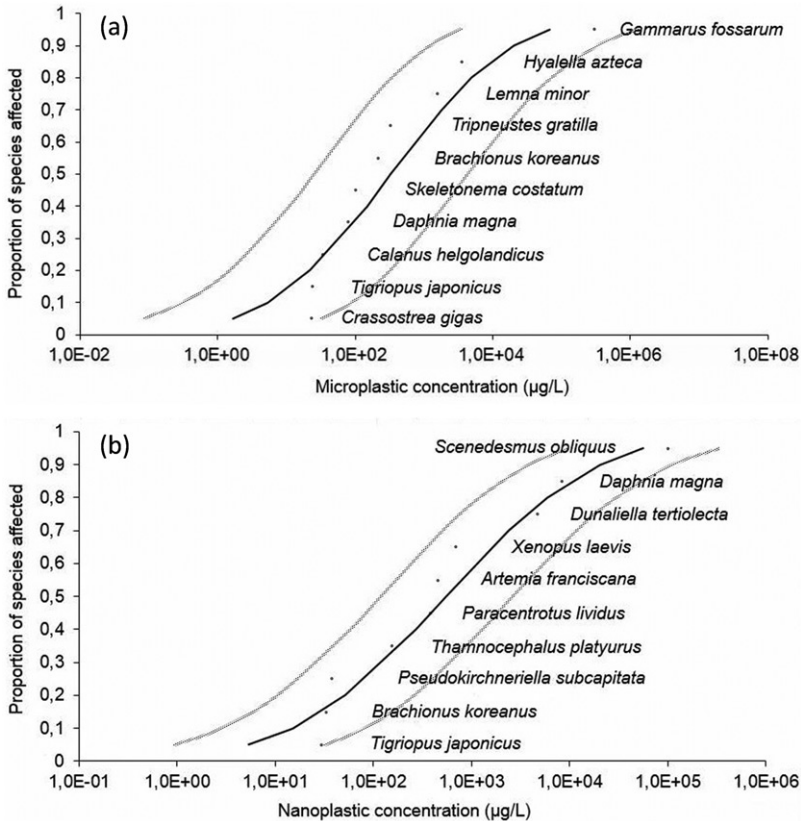
Figure 4. Species sensitivity distributions of organisms from marine, estuarine and freshwater environments exposed to microplastic (Panel A) or nanoplastic (Panel B) via the water phase. Effect thresholds represent chronic LOECs. Grey curves represent the 95% confidence intervals.

particles) and the least sensitive the algae *Scenedesmus obliquus* (70 nm spherical PS particles) (Besseling et al., 2014b) (Table A2, Supporting Information). Relatively high sensitivities of the juveniles and of reproduction endpoints were observed for microplastic (Table A1, Supporting Information) compared to the growth and survival endpoints. Effects on growth might be due to an overall decreased nutritional value of the food as it becomes diluted with plastic (Besseling, Foekema, Van den Heuvel-Greve, & Koelmans, 2017; Besseling et al., 2014b, Besseling et al., 2013; Wright, Rowe, Thompson, & Galloway, 2013). Several studies suggest that such an effect, however, would not necessarily be unique for plastic but could similarly be caused by natural (e.g., mineral) particles (Vroom et al., 2017). On the other hand, the study exposing *G. fossarum* showed that effects of particles were not found when natural silica particles instead of plastic particles were used (Blarer & Burkhardt-Holm, 2016). The mechanism of decreased nutritional value due to dilution of food would not apply to organisms that do not ingest microplastic, like algae. For phytoplankton

however, growth inhibition could occur due to adhesion, pollutant transfer or attenuation of light caused by presence of microplastics (Yokota et al., 2017). From these SSDs, a hazardous concentration (HC$_5$) of 1.67 µg/L (R^2: 0.85, 95% confidence interval (95%CI):   0.086 – 32.6 µg/L) was derived for microplastic. The CI for the SSD of microplastic spans a factor of 380, illustrating the large variability emerging from combining different end-points, particle types and study approaches in one SSD.

The here derived HC$_5$ for nanoplastic is 5.4 µg/L (R^2: 0.93, 95% CI 0.93 – 31 mg/L), which, within error limits, is identical to that of microplastic (1.67 µg/L,   with CI 0.086 – 32.6 µg/L). Confidence intervals, however, are less wide but still considerable, again illustrating the uncertainty. When using an SSD approach to derive HC$_5$ values, an assessment factor (AF) of 5 has been applied by Van Cauwenberghe to obtain a predicted no effect concentration (PNEC) (Van Cauwenberghe, 2016). In this section an example is provided of how such a concentration could be estimated, and referred to as a preliminary safe standard (PSS). This results in PSS concentrations of 0.33 µg microplastic/L and 1.1 µg nanoplastic/L water (Table 6).

For exposure via food or sediment, insufficient chronic LOEC data was available to construct SSDs. For the effects of microplastic on survival, growth or reproduction, two LOEC values had been derived before. A LOEC of 12 g/kg food for survival of fish was published by Mazurais et al. (2015) and we provided a LOEC of 74 g/kg DW in sediment for the growth of lugworms in earlier work (Besseling et al., 2013). When only a single or a few effect thresholds are available, an AF of 1000 has been used before to derive PNEC values based on data for the most sensitive endpoint available (Van Cauwenberghe, 2016). For microplastic, in addition to effect thresholds for survival, growth and reproduction, effect thresholds could also be obtained from the literature for a variety of other, partly more sensitive, endpoints (Table A4, Supporting Information). The most sensitive endpoint that could be found in the literature, is translocation to tissue beyond the gastrointestinal tract (Farrell & Nelson, 2013), though it can be debated whether this has to be considered an effect on biological functions. Another aspect to keep in mind while interpreting translocation to tissues beyond the gastrointestinal tract when working with high concentrations (Farrell & Nelson, 2013) is that apparent translocation might be due to cutting during dissection of tissue. For these reasons, we chose not to use this endpoint here and work with the most sensitive endpoint given in Table A4, Supporting Information, which is oxidative stress and liver damage in fish at 0.1 g/kg DW food. The resulting PSS calculated from this effect threshold is 0.1 mg/kg food (Table 6). For nanoplastic, the LOEC of 1 g/kg food for the growth of fish (Table A3, Supporting Information) provided by Cedervall, Hansson, Lard, Frohm, and Linse (2012) is the only available

Table 6. Preliminary safe standard (PSS) values for exposure to micro- and nanoplastic via different compartments in different ecosystems. HC$_5$: hazardous concentration for 5% of the species, LOEC: lowest observed effect concentration, AF: assessment factor, 95% CI: 95% confidence interval.

Size category	Ecosystem	Exposure medium	HC$_5$	LOEC	AF	PSS
Micro plastic	Aquatic environment	Water	2.0 ng/L		5	0.4 ng/L (95% CI: 3.6×10^{-4} – 4.5×10^2)
		Food/Sediment		0.1 g/kg DW	1000	0.1 mg/kg DW
Nano plastic	Aquatic environment	Water	5.4 µg/L		5	1.1 µg/L (95% CI: 0.19 – 6.2)
		Food		1 g/kg DW	1000	1 mg/kg DW

effect threshold for exposure via media other than water, so this value was used with an AF of 1000 to derive a preliminary PSS of 1 mg/kg DW food.

Previous calculations by Van Cauwenberghe for microplastic in the marine environment resulted in a PNEC of 640 particles/L for exposure via water and a PNEC of 540 particles/kg WW for exposure via sediment (Van Cauwenberghe, 2016). Use of the aforementioned conversion factors for particle weight (5 μg/particle) and porewater fraction (20%) converts these to PNECs of 3.2 mg/L and 3.4 mg/kg DW, respectively. The PSSs derived in the present study are a factor 10^4 lower than those for exposure via water, whereas for exposure via sediment or food, the suggested safe concentrations are reasonably within the same range (here a factor 34 lower than the PNEC reported by Van Cauwenberghe et al. (2016). That the PSS values derived here are lower than the PNECs reported by Van Cauwenberghe is because we use more recently available effect thresholds and because all thresholds were scaled to chronic LOEC values using extrapolation factors, leading to relatively conservative estimates. Following Van Cauwenberghe et al. (2016), it should be emphasized that because of the limited availability of suitable threshold data and large confidence intervals, the derived safe values (PSS) are very preliminary and thus should be used with caution.

3.3. Effect thresholds of plastic particles with varying characteristics

In addition to the variability in species sensitivity and effect mechanisms, the data underlying the SSDs reflect that plastic as a stressor is present in a variety of sizes, shapes and types. This is why the SSD was used here as a tool to explore whether a relation could be found between the effect level and the varying characteristics of different plastic particles (SSDs not shown). However, these SSD analyses revealed no relation between effect level and polymer type, and within each size class (i.e., >100 nm for microplastic and <100 nm for nanoplastic) no relation with size could be found. This can mean either that no such dependence exists, or that the large variation in tested species, endpoints and scientific quality of the data used made that no dependence could be detected. Also, effects just can be too small to detect them with statistical rigor, for instance in case a physical effects would primarily depend on particle size, rendering effects of polymer type to be marginal. While nanoplastic is considered the potentially most harmful size class (Koelmans et al., 2015), the EC_{50} values of the smallest nanoplastic particles tested (diameter around 50 nm) lie within the upper right half of the curve. Imhof and Laforsch did not find effects of a mixture of polymer types on mud snails, but suggest that a link between effect levels and either polymer type or size might exist, based on a comparison with effects found in studies using ≤20 μm polystyrene beads

(Imhof & Laforsch, 2016). The constructed SSDs showed no clear relation with nanoparticle size or charge either. Several studies have reported effects on growth, survival and embryo toxicity for positively charged nanoplastic, whereas these effects are lacking, or only occur at higher concentrations, when the same particles are used with negatively charged surface groups (Bergami et al., 2017; Della Torre et al., 2014; Nasser & Lynch, 2016). When combining data from different studies in one SSD, these differences in effects within studies were obscured by the differences between studies. The effect levels for microplastic included here are largely based on particles with a spherical shape (16 studies); only two studies used fibers and 8 studies used irregularly shaped particles. From that, no difference in effect levels between microplastic particles with different shapes could be distinguished. One might expect a more difficult egestion or severe effects from fibre-shaped particles based on the known effects of asbestos and different phagocytic reactions to particles with this shape (Sharma et al., 2010). Indeed, one study found effects on the assimilation efficiency for fibers, and not for spherical particles (Blarer & Burkhardt-Holm, 2016). Yet, the effect data for the fibers of both studies were in the upper right part of the SSD curve, which presents the data for the least sensitive cases. This might be due to the micrometer size and might thus be different for nanoplastic. However, all effect levels reported for nanoplastic thus far relate to spherical particles (Table A2, Supporting Information).

Another particle characteristic that varies among studies is whether pristine particles are used or particles that had weathered in an environmental or laboratory setting. The latter is done for different purposes, to actively promote biofilm growth on the particles, give the particles a natural flavoring or make them more environmentally relevant in a general sense (Besseling et al., 2014b; DeMott, 1988; Vroom et al., 2017). In this SSD analysis, no distinction was made between these different ways of preparing the particles, because there was a large variation in the extent to which these preparation methods were described in the original articles in the first place. However, the inclusion of a biofilm has been found to affect the ingestion and egestion efficiency in organisms (Nasser & Lynch, 2016; Vroom et al., 2017). Where egestion became reduced due to ingestion of plastic particles with biofilms, a reduced feeding rate was also found (Nasser & Lynch, 2016). This implies that although biofilm formation is fast and might (even when not specifically quantified) have been present in studies that claimed to use pristine particles, outcomes of effect studies with pristine particles might be more unfavorable for organisms under environmentally realistic conditions.

The final variable particle characteristic that needs to be mentioned here is the concentration of chemicals within the plastic particles used to derive

effect thresholds. The effect thresholds found might apply to the effects of plastic particles themselves or to chemicals transported by these particles, or to the combined effects of multiple stressors, as many studies assessing effects of plastic particles do not exclude that the particles they used contained additives or other chemicals.

3.4. Role of microplastic in bioaccumulation of chemicals

Previous studies have shown that the effect of microplastic ingestion on the bioaccumulation of omnipresent hydrophobic chemicals, also referred to as persistent organic pollutants (POPs), is restricted roughly to a twofold increase or decrease in tissue of lugworms (Bakir, O'Connor, Rowland, Hendriks, & Thompson, 2016; Besseling, Foekema, et al., 2017; Besseling et al., 2013). Whether an actual increase or decrease of bioaccumulation is found depends on the polymer type and chemical characteristics, as well as on complex counteracting mechanisms of contribution to chemical uptake through plastic versus food, and whether or not there is chemical equilibrium (Besseling, Foekema, et al., 2017; Koelmans, 2015; Koelmans et al., 2013). This has been demonstrated by the fact that the POP concentrations in the surrounding water (or porewater for sediment-dwelling organisms) fully explained the observed bioaccumulation in bioassays (Besseling et al., 2017).

Although the role of microplastic ingestion in the bioaccumulation of POPs by organisms has been suggested to be minor for most aquatic habitats (Besseling, Foekema, Van Den Heuvel-Greve, & Koelmans, 2017; Endo & Koelmans, 2016; Herzke et al., 2016; Koelmans, Bakir, Burton, & Janssen, 2016; Koelmans et al., 2013; Lohmann, 2017; Ziccardi, Edgington, Hentz, Kulacki, & Kane Driscoll, 2016), there has been considerable debate on this. The hypothesis that microplastic affects bioaccumulation has dominated a large part of the microplastic research during the past decade, and both those scientists who do and those who do not think that microplastic increases the uptake of POPs find proof in experimental data (Besseling, Foekema, et al., 2017; Besseling et al., 2013; Chua, Shimeta, Nugegoda, Morrison, & Clarke, 2014; Hartmann et al., 2017; Koelmans et al., 2016; Rochman, Hoh, Kurobe, & Teh, 2013; Wardrop et al., 2016). The contrasting views can be explained by taking a closer look at the precise hypotheses that underlie the different studies. Studies with varying types of polymers and POPs found that the more amorphous, low-density polymer types (i.e. PE) have the highest affinity for POPs and, just as in other absorbing pools such as lipids and organic matter, the most hydrophobic POPs will be most attracted to plastic. Apart from differences in polymer types and chemicals, it is the use of different species and in particular different exposure scenarios in different studies which results in different conclusions (Au

et al., 2017; Koelmans et al., 2016; Rochman et al., 2013; Wardrop et al., 2016) Microplastic ingestion is likely to increase bioaccumulation in organisms when these organisms are relatively clean at the start of exposure and are fed with high doses of microplastic loaded with POPs. Microplastic is less likely to increase bioaccumulation in organisms fed with microplastic when the POP concentrations in the organisms are already closer to – or at chemical equilibrium with – the surrounding environmental media like water and diet components. In hotspot locations, where plastic makes up a relatively large fraction of the diet and the POP concentration gradient allows chemical transfer to the organism, plastic might however significantly increase exposure to chemicals (Chen et al., 2018; Jang et al., 2016).

As for nanoplastic particles, there are two reasons why they might deliver a more substantial contribution to bioaccumulation of POPs in organisms than microplastic particles. The first reason is a much higher (1-2 orders of magnitude) affinity of POPs to these particles, compared to microplastic (Liu, Fokkink, & Koelmans, 2016; Velzeboer, Kwadijk, & Koelmans, 2014). The second reason is that nanoplastic might be able to reach other tissues than microplastic (Koelmans et al., 2015). The small size of nanoparticles allows them to enter cells via endocytosis, penetrate tissues, move directly from the digestive tract to the circulatory system, and cross the blood-brain barrier (Browne et al., 2008; Kashiwada, 2006; Koziara, Lockman, Allen, & Mumper, 2003). Biofilm formation has been found to decrease sorption to nanoplastic, whereas sorption was enhanced in a marine as opposed to a freshwater setting (Velzeboer et al., 2014). This suggests that the presence of nanoplastic may potentially increase the uptake of POPs from the environment in organisms, the extent of which depends on the specific environment.

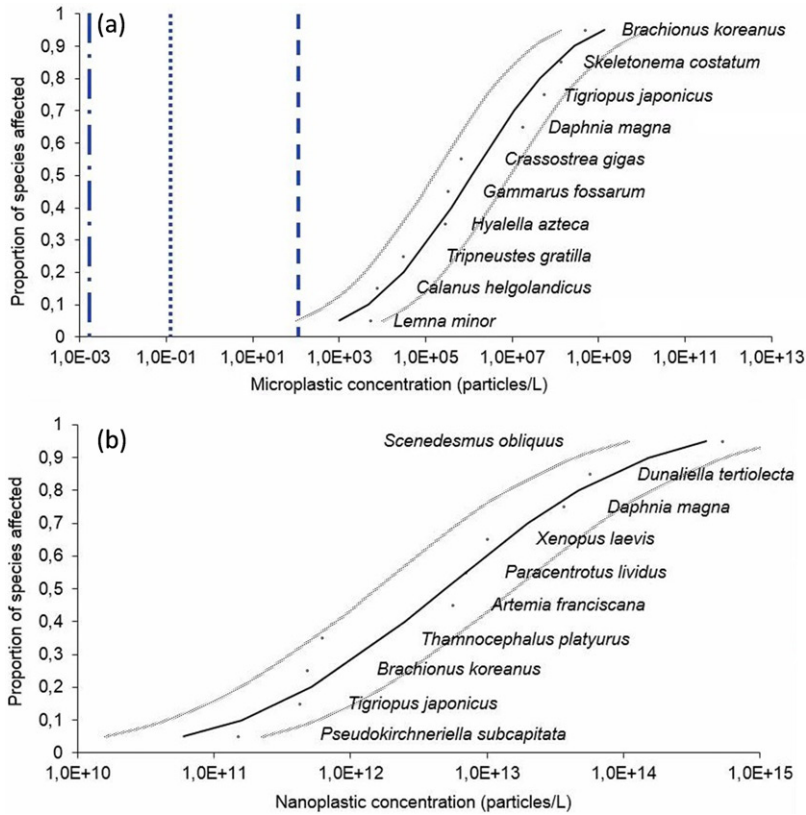## 4. Risk characterization for nano- and microplastic particle effects

A comparison of plastic particle effect thresholds with exposure concentrations reveals to what extent organisms may be at risk (Figure 5). As exposure concentrations of micro- and nanoplastic were given in particles per volume in Table 1, the comparison with chronic LOEC values is made here using the unit of particles/L. It might be preferable to work with particle mass per medium volume or mass, as in Figure 4. However, environmental fate studies (section 2.1) often do not provide detailed information on particle characteristics from which a conversion factor from mass to particle numbers or vice versa could be derived (a general particle mass of 5 μg/particle could be used instead, but this is a rough estimate), whereas the effect threshold studies from the literature (section 8.3.2) often do include this information. Therefore, less uncertainty is introduced when the effect threshold dataset is converted to particle concentrations, rather than

converting the environmental concentrations dataset to mass concentrations. Hence, exposure and effect levels are compared here as particle concentrations. A further advantage is that, additional to the SSDs using mass concentrations (Figure 4), the same data is here presented as SSDs using particle concentrations (Figure 5).

Risks of microplastic in water. To be able to align the metric used for exposure (particles per liter) with those for effects, SSDs are also provided on a particle number per liter basis (Figure 5). For organisms exposed to microplastic in water, the HC_5 derived from the SSD is 1015 particles/L (R^2: 0.95, 95% CI 101 – 10223 particles/L) (Figure 5), which is of the same order of magnitude as the value of 640 particles/L given by Van Cauwenberghe. This HC_5 for microplastic has a high R^2, relatively narrow CI, and is of the same order as the worldwide HLRR of microplastic concentrations in-near shore surface water (Figure 5A). Worldwide HLRR in freshwater up to and including the year 2016 are three orders of magnitude lower, and those in open ocean surface water almost five orders of magnitude lower than this HC_5. Taking into account that amounts of microplastic are underestimated by up to a factor of 30 when based on surface sampling (Kooi et al., 2016), microplastic concentrations might present a risk to the most sensitive species at hotspot locations in near-shore regions.

Risks of nanoplastic in water. For organisms exposed to nanoplastic in water, the HC_5 is 5.97×10^{10} particles/L (R^2: 0.96, 95% CI $(1.6 – 22) \times 10^{10}$ particles/L). Effect threshold concentrations for nanoplastic expressed in particles/L are generally seven orders of magnitude higher than those for microplastic. Since measured concentrations of nanoplastic in the environment are lacking, no direct comparison with environmental nanoplastic concentrations can be made. Ter Halle et al. (2017) demonstrated the polymer composition of colloidal particles in the ocean, but did not provide particle numbers. However, environmental number concentrations of nanoplastic may potentially become 17 orders of magnitude higher than those of microplastic in the future, due to fragmentation of larger plastic particles (calculation based on 5 mm spheres fragmenting into 100 nm spheres). This would mean that environmental nanoplastic concentrations could exceed the effect thresholds over time. Within a timeframe of several hundreds of years (Van Cauwenberghe, 2016), nanoplastic concentrations would then be within the range of effect thresholds included in Figure 5B.

Risks of microplastic in sediment or food. For aquatic organisms exposed to microplastic via the media food and sediment, only two chronic LOEC values could be derived when limiting the selection of data to the endpoints survival, growth and reproduction (2.2×10^5 and 4.1×10^9 particles/kg). Effect threshold values for other endpoints are available in the literature (Table A4, Supporting Information), these endpoints are not by definition threatening populations,

Figure 5. Risk characterization for microplastic and nanoplastic. Separate panels are provided for exposure to microplastic via water (Panel A), and to nanoplastic via water (Panel B). Solid black curves represent SSDs with plastic concentrations expressed in particles per volume. Grey curves represent the 95% confidence intervals. SSDs are based on (Panel A); chronic LOEC concentration data (particles/L) for microplastic and the endpoints survival, growth and reproduction (Table A1, Supporting Information, as in Figure 4A) and on (Panel B); LOEC concentration data (particles/L) for nanoplastic and the endpoints survival, growth and reproduction (Table A2, Supporting Information, as in Figure 4B). Vertical lines indicate measured environmental concentrations as worldwide highest limits of reported ranges (HLRR) (Table 1) for exposure via water (Panel A); microplastic concentrations measured in freshwater surface water (blue dotted line, near-shore surface water (blue dashed line) and open ocean surface water (blue dot-dashed line). For nanoplastic (Panel B) no vertical lines are drawn because no HLRR data are available yet.

although they might lead to community changes. The worldwide HLRR of microplastic concentrations in freshwater sediment and beach sediment are within the range of effect thresholds for these not by definition population threatening endpoints (Table A4, Supporting Information) but microplastic concentrations in freshwater, subtidal, beach and seafloor sediment are below the two chronic LOEC values for population threatening effects.

In conclusion, based on the preliminary SSDs and worst case worldwide HLRR exposure estimates, the exceedance of hazardous microplastic concentrations for the most sensitive species may occur currently in hotspot

locations of near-shore surface waters. Van Cauwenberghe (2016) predicted that only sediment-dwelling organisms would be at risk of exposure to microplastic concentrations exceeding the effect thresholds (Van Cauwenberghe, 2016). The difference between her prediction and the findings in our present study is due to the (recent) availability of more effect thresholds and the use of extrapolation factors to scale these thresholds consistently to chronic LOECs. The present risk assessment is based on comparing effect threshold values from separate bioassays and HLRR values. However, the results should be taken as an illustration of the approach rather than as a basis for strong conclusions. Furthermore, it is advisable to address and quantify the present uncertainties using probabilistic risk assessment methods.

5. Outlook

Hotspots, underestimated environmental concentrations (Hartmann et al., 2017; Kooi et al., 2016) and ongoing degradation of macro- and microplastic into nanoplastic result in approaching of predicted effect thresholds, causing risks for organisms being exposed to plastic particles at specific locations. As regards microplastic, current scientific methods are fairly well able to assess their occurrence, effects and hazards. Previous detection of microplastic and other plastic debris was often based on visual appearance, but this is increasingly being replaced by techniques like FTIR and Raman spectroscopy. These techniques are accessible to more and more research groups, so that confirmation of the occurrence in different media and different organisms with these techniques will continue for several more years. The level of public interest in the subject of microplastic pollution is expected to peak around 2022, as has been forecast based on the history of attention focused on other contaminants of emerging concern (CECs, i.e., harmful environmental agents whose identities, occurrences, effects and hazards are not yet sufficiently understood) (Halden, 2015). Some of the currently assumed effects of microplastic can be considered less harmful than anticipated before. An example is the often limited effect of microplastic on bioaccumulation of other contaminants in organisms. This view appears to receive growing recognition (Bakir et al., 2016; Beckingham & Ghosh, 2017; Besseling, Foekema, et al., 2017; Besseling et al., 2013; Devriese, De Witte, Vethaak, Hostens, & Leslie, 2017; EPA (United States Environmental Protection Agency), 2016b; Gassel, Harwani, Park, & Jahn, 2013; GESAMP, 2015; Herzke et al., 2016; Koelmans et al., 2016; Koelmans et al., 2013), which might constitute a paradigm shift. On the other hand, our presented risk assessment provisionally shows that particle effect thresholds inferred from scientific literature data can be exceeded, in

particular in near-shore surface water hotspot locations and, although very limited population threatening effect threshold data are available, possibly also in freshwater sediment and beach sediment.

The replaceability of microplastic by more natural materials, the public interest in microplastic pollution, the improvement of WWTPs (Siegfried et al., 2017) and bans on the use of microplastic in several countries make it unlikely that the development, marketing, disposal and consumption of products containing microplastic will increase. However, the degradation of macroplastic that is already present in the environment and the ongoing disposal of new macroplastic by our consumer society might lead to new, higher, levels of microplastic pollution, with consequences for species and possible renewed political and scientific attention. Further development of SSDs (Koelmans, Besseling, et al., 2017) based on additional and higher quality effect threshold data and standardized testing methods (Besseling, 2018; Redondo Hasselerharm et al., 2018) will further improve insight in the risk of exposure to plastic. Separating the risk of particle effects from that of chemical transfer effects, and considering the latter within a more complete environmental setting including all relevant chemical transfer pathways, will help to assess the overall risk of exposure to plastic (EPA, 2016b; Koelmans, Besseling, et al., 2017).

The generation of knowledge about the occurrence and effects of nano-plastic has been different from that for microplastic. The level of interest in nanomaterials in general was expected to peak in 2016 (Halden, 2015), but nanoplastic is a specific case. Although these particles are often bracketed together with microplastic, our current technical ability to determine their occurrence, fate and effects is far less highly developed. Breakthroughs in detection techniques for nanoplastic in environmental media, as well as methods to assess their fate and effects at the level of tissues or organisms, will lead to enhanced knowledge about nanoplastic, but this will come later than for microplastic. For other contaminants of emerging concern, attention by policy makers and the development of new regulations have been shown to peak a few years after the peak in scientific attention (Halden, 2015). Therefore, within about a decade, policy regulations might be introduced for microplastic and could be under development for nanoplastic too. Further assessment of the risk of micro- and nanoplastic and further introduction of regulations on the use of plastic will diminish the risk of exposure and will help frame the public concern and the scientific debate relative to those regarding environmental concerns other than plastic.

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