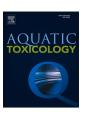
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# Probabilistic environmental risk assessment of microplastics in marine habitats

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#### ABSTRACT

Microplastics are ubiquitous in the environment and given the large number of published hazard and exposure studies, quantitative environmental risks assessments of microplastics become feasible. We present here the first environmental risk assessment for marine waters based only on measured concentrations. The Theyariability and uncertainty of the measured data was accounted for in the exposure assessment, while probabilistic species sensitivity distributions were used for hazard assessment, from which a probability distribution was extracted for the predicted no-effect concentration (PNEC). By dividing the exposure distribution by the PNEC-distribution, we were able to calculate probabilistic risk characterisation ratios for each water body in which measurements were performed. Results show a good coverage of the world's major water bodies by measured exposure concentrations (MECs), while the hazard assessment could be improved by aligning the type of particles tested in hazard studies (size, form, polymer) to those actually found in the oceans. Overall, the mean predicted no-effect concentration (PNEC) is  $3.84 \cdot 10^6$  part m $^{-3}$ , with Oryzias melastigma being the most sensitive species (calculated  $mean \ NOEC \ of \ 3.90 \cdot 10^6 \ part \ m^{-3}). \ Interestingly, the \ only \ type \ of \ dose \ descriptor \ that \ could \ be \ extracted \ from \ the$ literature for particles above  $10-20 \mu m$  was the highest observed no effect concentration (HONEC), which indicates a very low or null toxicity of these larger MPs towards marine organisms. The mean MEC is 1.5 103 part m<sup>-3</sup>, the highest concentrations being measured in the Atlantic and Pacific Ocean. Although there is a very small overlap of the probability distribution associated with the RCR (0.00002 % of the data points), the mean RCR is 4·10<sup>-4</sup> and therefore risks are unlikely given the available data. However, as increasing amounts of plastic reach the environment, RCRs can be expected to increase in the future.

#### 1. Introduction

Plastic is ubiquitous. Due to a variety of desirable characteristics such as malleability and durability, plastics have permeated nearly all economic sectors and everyday lives in society as well. Between 1950 and 2015, the global production has increased from 2 million metric tons (Mt) to 380 Mt and is expected to continue to grow. While the benefits of plastic materials are undeniable, public attention to possible negative impacts on the environment and human health has risen. Attention is paid to the subject by regulatory bodies all over the world, with for example the recent microbead-free waters act in the United States (US Congress, 2015) and the ban of single-use plastic products in the European Union (European Parliament, 2018).

Reports on the occurrence of small synthetic particles floating in marine waters go back as far as the 1970s (Carpenter et al., 1972; Colton Jr. et al., 1974), but the term "microplastic" was only coined about 30

years later. It was first proposed by Arthur et al. (2009) that all synthetic polymer particles smaller than 5 mm in their longest dimension fall under the category of microplastics (MPs). Subsequently, this definition has been widely accepted and used in scientific literature as well as in media and public discourse, and is the one adopted in this work.

MPs are commonly divided into primary and secondary MPs. Primary MPs are intentionally produced for use in industry or consumer products, while secondary MPs result from the breakdown of larger plastic debris, either during use or while in the environment. It was estimated that 15–31 % of all microplastic in the ocean could originate from primary sources (Boucher and Friot, 2017). Fragmentation can take place through photodegradation from UV-light, hydrolysis in the presence of water, (thermal) oxidation, physical abrasion in sediments and soils but also by waves, and biological degradation by organisms (Andrady, 2011; Horton et al., 2017).

A possible source of primary MPs into the environment are spills or

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industrial effluents (de Souza Machado et al., 2018). Primary MPs in personal care products will eventually enter wastewater treatment systems. While removal efficiency in wastewater treatment plants (WWTPs) is high, a certain amount of particles can still occur in the effluents (Auta et al., 2017). Sources of larger plastic items that can break down to secondary microplastics are littering and improper waste management through which macroplastic can reach the terrestrial, freshwater and marine environments (Geyer et al., 2017; Jambeck et al., 2015). The main inputs of MPs to marine environment stem directly from the inland or via freshwater (Andrady, 2011). A more recent study by Lebreton et al. (2018) also highlights the importance of oceanic MPs originating from activities within this same compartment, such as fishing.

With the presence of MPs in the environment and their persistence established, it is likely that they are readily available to living biota. Several studies have shown that organisms take up MPs from their environment (for compilation of these studies, refer to Burns and Boxall, 2018; Phuong et al., 2016; Lusher, 2015). The majority of these studies focused on aquatic species. Due to the diversity of MPs as a material class comprising several different chemicals (Rochman et al., 2019), determining their toxicity is not straightforward. Some studies have found detrimental effects on endpoints like survival, reproduction or sub-lethal endpoints (e.g. gene expression), suggesting that MPs may possess an innate toxicity based on their chemical composition. For some species investigated, no negative impacts were observed at all. Such differing findings clearly illustrate the need to put toxicity studies into context by means of hazard assessment.

Risk assessment (RA) combines an exposure assessment and a hazard assessment to determine whether any given substance is present in the environment at concentrations known to exert negative effects on organisms living in this compartment, thus posing a risk. Burns and Boxall (2018) were the first to systematically assess the possible risk of MPs in water columns and sediments of both freshwater and marine environments. The aggregated data (combined marine and freshwater) suggested that MPs pose no immediate threat to the environment, but no risk assessment specific to one compartment or the other was performed in this study. A risk assessment in freshwater was performed by Adam et al. (2018), who showed that risks in this compartment were unlikely worldwide, although they could not be completely excluded in Asia. Everaert et al. (2018) modelled the environmental concentration in the oceans, integrating global plastic production data and the paths of (micro-)plastic into the environment. They were further able to model the environmental concentration in the future for two different scenarios based on the magnitude of future MP emissions into the ocean. Their findings indicate that there is currently no risk from MPs on a global scale, though in some hotspot regions, environmental concentrations may already reach levels that can cause detrimental effects in biota. Projecting a growing amount of plastic emissions into the ocean, their model predicts that after the year 2050, environmental concentrations will reach high enough levels for significant ecotoxicological impacts to

The risk assessment performed here in the marine environment is the first to look at the marine water column only (as opposed to Burns and Boxall (2018), who included freshwater and sediments) and base the exposure assessment solely on measured environmental concentrations (as opposed to Everaert et al. (2018) who modelled the environmental concentrations). A probabilistic RA approach is employed in these assessments, to account for data variability and uncertainty.

## 2. Materials and method

## 2.1. Scope and general methodology

In this work, the risk assessments were conducted on a global scale, with some geographical differentiation when possible. Baseline assumptions had to be made: Firstly, the hazardous effects of additives

included in polymer matrices or sorbed pollutants need to be clearly distinguished from those that the plastic compound itself possibly exerts. Since the objective here is to assess the risk posed directly by MPs, data on the effect of plastic additives are not included. Studies targeting the effects caused by MPs acting as vectors of other pollutants are also not included. Secondly, the working definition by Arthur et al. (2009) is still the most used to describe MPs in scientific literature. Therefore, for these risk assessments, particles with sizes smaller than 5 mm in their longest dimension are included as MPs. Nanoplastics, so particles below 100 nm, were excluded from the analysis.

The general procedure followed in this work is the same as in Adam et al. (2018). After data collection, probability distributions were calculated for both hazard and exposure assessments, and compared for risk assessments. All calculations were performed in R (R Core Team, 2019). For interpretation of results, data were analysed regarding specific variables described in the following sections. All statistical analyses were conducted with IBM SPSS Statistics for Windows, version 26 (IBM Corporation, 2019).

## 2.2. Hazard assessment

#### 2.2.1. Data collection and harmonisation

For collection of hazard data, the existing risk assessments were used as initial datasets to be reviewed (Burns and Boxall, 2018; Everaert et al., 2018). Additional online literature searches were conducted with the terms "microplastic", "marine", "ecotoxicity", "toxicity" and combinations thereof. All data published until the 31st of July 2019 were taken into account.

Reported toxicities were provided either as particle- or mass-based concentrations. For comparability with the exposure assessment, all data were harmonized by converting to particle-based number concentrations. This was achieved using the reported average size, shape and polymer type of the tested MPs along with the density for the polymers, taken from Polymerdatabase (2019) and ScientificPolymer, Inc (2013).

In total, 152 studies were reviewed for marine toxicities. The preferred endpoints were population growth, survival and reproduction. One further endpoint included is negative impacts on embryo development, as it is relevant on a population basis. Exclusion criteria were used to ensure data quality, detailed in Table S1.

## 2.2.2. Calculation of predicted no-effect concentrations

The species sensitivity distribution (SSD) is an accepted approach for assessing the hazardous effects of a substance on an ecosystem (Monti et al., 2018). It aims to include sensitivities of multiple species from the same habitat and towards a specific stressor. To account for the complexity that an ecosystem presents, the SSD represents the entire set of sensitivities in cumulative order, resulting in a curve described by a distribution - for example a logistic or triangular distribution (Posthuma et al., 2002). The toxicity assessment in the present work was performed on the basis of the probabilistic SSD (pSSD) developed by Gottschalk and Nowack (2013) and modified by Wigger et al. (2019) (pSSD<sup>+</sup>). This method allows to account for the inherent inter-laboratory variation associated with toxicity testing, as well as the inclusion of different dose descriptors and acute toxicity data.

Toxicity data is commonly reported in a variety of different dose descriptors. The effect or inhibition concentrations (E/ICx) or lethal concentrations (LCx) describe the concentrations at which a certain percentage (x) of a population are negatively affected by the substance in question. Other dose descriptors include the lowest observed effect concentration (LOEC), the highest observed no-effect concentration (HONEC) and the no-observed effect concentration (NOEC). For (p)SSD building, no-observed effect concentration (NOEC) is the preferred dose descriptor, and chronic data are preferred over acute data (ECHA, 2008). However, the latter are far more prevalent in the literature. To account for this, the pSSD<sup>+</sup> method makes use of uncertainty factors (UFs) 1) to convert any dose descriptor into a NOEC with the UF<sub>Descriptor</sub>

 $(UF_D)$ ; and 2) to extrapolate acute data to chronic data with the  $UF_{Time}$   $(UF_T)$ .  $UF_D$  values used for the conversion into NOECs and associated references are shown in Table S2. Uncertainty factors for the acute to chronic ratio for particulate pollutants have been reported for nanoparticles and have been considered here when deriving  $UF_{Time}$  (Wigger et al., 2019).

Values for UF $_{\rm T}$  depend on the lifetime and duration of lifecycle stages of each species (Table S3). There exist only few testing guidelines for marine species, which define the durations of toxicity tests, allowing to categorise data as chronic or acute. Therefore, for those species where no specific duration has been defined, the values for UF $_{\rm T}$  were derived from other species within the same order or phylum. Furthermore, duration times for UF $_{\rm T}$  were always chosen in a precautionary manner. For example, for the phyla Myzozoa, Ochrophyta and Proteobacteria, no specific guideline exists. These species were tested for 23 days, between 3 and 22 days, and 6 h, respectively. The cut-off value of 3 days recommended for algae for applying UF $_{\rm T}$  was therefore considered prudent to not underestimate the respective toxicity values.

The pSSD<sup>+</sup> method first builds single probability distributions including all selected data and associated uncertainties for a single species. From these species-specific distributions, multiple pSSDs are then calculated using a Monte-Carlo routine (Wigger et al., 2019). The predicted no-effect concentration (PNEC), i.e. the concentration at which no effect is expected to occur in the ecosystem, needs to be derived from the (p)SSD. The recommendation by the European Chemicals Agency (ECHA, 2008) is to extract the PNEC as the fifth percentile of the SSD, therefore protecting 95 % of species. In the case of probabilistic hazard assessment, where multiple pSSDs are calculated based on Monte-Carlo analysis, the result is a probabilistic PNEC distribution instead of a single value.

HONECs are reported in the literature when no toxicity can be measured. However, toxicity could occur at higher doses than those tested. Therefore, this dose descriptor was not considered reliable. pSSDs were built based on both mass and number concentrations. However, in the pSSD based on particle-number concentrations, the sensitivity of two of the species that ranked at the lowest end of the distribution were defined based on HONECs. Since this low part of the pSSD is also the most important to calculate the PNEC, these two species were removed from the calculation. Moreover, pSSDs based on both mass and particle number concentrations were calculated again, excluding all HONECs from the datasets. These more stringent datasets were then used for the final risk assessment, but all pSSDs were kept in this study to show the reader all data available.

## 2.2.3. Data analyses

The definition of MPs as a class of contaminants with different chemical structures, shapes and sizes might also result in differences in their toxicity. Therefore, the dataset for hazard assessment was analysed as to whether or not the different shapes, sizes and polymer types used in toxicity studies resulted in different effects on organisms. Differences in NOECs for different polymer types and shapes were analysed with Mann-Whitney tests by grouping the data into one polymer type or shape against all other remaining data points.

## 2.3. Exposure assessment

## 2.3.1. Data collection and harmonization

Data were obtained from existing reviews or risk assessments, as well as by literature search. Searches were conducted on Web of Science and Google Scholar with the terms "microplastic" and "marine". Data from estuarine samples were included. While being a potential relevant sink for MPs, concentrations from beaches or benthic sediments are not included. All data published until the 31 st of July 2019 were taken into account.

Concentrations measured in marine waters are commonly reported based on particle number either per volume or per surface area sampled. A preference is for the former because of its compatibility with the hazard data, which are reported on a volume basis. For this reason, concentrations per surface area were converted by dividing by the reported sampling depth. If this information was not provided, it was assumed that the sampling devices were submerged half-way (Fossi et al., 2012; Gajst et al., 2016), therefore taking half of the opening height as sampling depth.

In cases where measured concentrations were null, half of the limit of detection (LoD) reported in the dataset was taken. When LoDs were not reported in the field studies, they were calculated based on the size of the net mouth, the speed of the boat and the duration of the sampling as in equation 1:

$$LoD = \frac{1}{volume \ sampled}$$

$$= \frac{1}{surface \ sampled \ \times \ boat \ speed \times \ sampling \ duration}$$

Two papers did not give the size of the net mouth they used. In these cases, the mean of the other LoDs was used.

Criteria used to exclude studies considered of low quality are described in Table S4. When rates of particle misidentification were given and clearly not considered in the reported results, microplastic concentrations were recalculated accordingly. This was the case for three studies. All data points were categorized as "coastal" or "open ocean" samplings based on descriptions of the sampling locations available in the studies. We categorized as "open ocean" those samples taken during ocean transects or clearly in the middle of the ocean. All other samples were considered as "coastal", including estuarine samples. The coastal subset also included the Baltic Sea and Black Sea, which are (semi-)enclosed water bodies."

## 2.3.2. Consideration of uncertainty and variability

To include uncertainties and variabilities of measured environmental concentrations, each data point was assigned a distribution in the following manner. If a study reported a MEC as a mean concentration and standard deviation, it was assigned a normal distribution. When minimum, mean and maximum concentrations were reported, a triangular distribution was created with these values. If only a minimum or mean and a maximum value were given, the assigned distribution was uniform within the range of these two provided values. Lastly, if only single measurements were provided, no variability could be associated. From all data points and their associated probability distributions, several cumulative exposure functions were calculated based on a Monte-Carlo routine.

## 2.3.3. Data analyses

The dataset used here was analysed for possible effects of various variables on the overall outcome of the exposure assessment. To study the potential human influence on measured concentrations, datasets for coastal waters and open waters were compared.

The data used for the analysis were not obtained using a standardized method for MP sampling. However, the minimum size of the particles sampled could have a strong influence on the measured concentrations: wider mesh or filter pore sizes could lead to lower concentrations. The influence of mesh and filter pore sizes on the reported concentrations was investigated using linear regression analysis, after the model was deemed applicable to the datasets by residual error and normal Q-Q-plot analysis.

## 2.4. Risk assessment

To characterize the environmental risks that MP might pose to the marine environment, exposure and hazard probability distributions were compared in two ways. First, the percentage of overlap between both these distributions is given. A risk is expected to occur if the distributions do overlap. Second, risk characterisation ratios (RCRs) were calculated by dividing the MEC probability distribution by that of the PNEC. If the RCR is lower than 1, no risk is expected to occur given the current state of knowledge. Otherwise, a risk can be expected to occur for species of the habitat in question. For the risk assessment only the particle-number based PNEC were used because the exposure data are only available in particle-number metrics.

#### 3. Results

#### 3.1. Hazard assessment

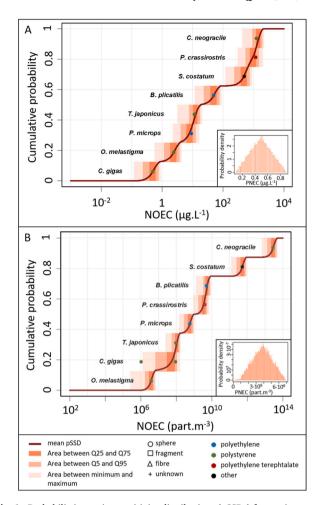
The final dataset for marine hazard assessment included 46 values of ecotoxicity from 23 species. The entire dataset used for the pSSD calculation is compiled in Table S5. It is constituted by 70 % data points derived from acute studies. The REACH guidelines recommend to include algae, invertebrates and fish in PNEC derivation for marine waters (ECHA, 2008). Since these types of species are also included in the guidelines for the derivation of PNEC in freshwater, which include more phyla and families (ECHA, 2008), it was considered prudent to apply the guidelines for the freshwater environment to the marine environment. As shown in Table S6, the overall compliance of the dataset is high, as it includes all required types of organisms except an insect species and a higher plant. It covers more than ten species. In most cases, the ecotoxicological dose descriptor was a HONEC (n = 37), followed by LOEC (n = 8) and NOEC (n = 1). The majority of studies employed spherical particles (26 data points), followed by fragments (n = 19). Only one study used fibres (Table S3). An undetermined shape was treated as a sphere for conversion of concentrations. The most commonly used polymer was PE (n = 21), followed by PS (n = 16) and PET (n = 5). The remaining 9 data points consisted of PET, PP, PVC and tire wear particles.

The particle-based pSSD included several species at the lower end of the curve for which only HONECs were reported (Figure S1). Because of the low reliability of this dose descriptor, the pSSDs based on were calculated excluding HONECs. The datasets then included 9 data points from 8 species. The exclusion of a number of values from the dataset should increase the quality of the derived PNEC distribution, yet some species were dropped entirely for these new pSSD calculations. A strictly brackish species (*O. melastigma*) had to be included to comply with REACH recommendations on as high a level as possible for the dataset including all available data (Table S6).

pSSDs were built in particle- and mass-based units (Figures S1A and S1B, respectively). The figures show a summary of 10000 pSSD simulations. In each run, one value was taken from each species-specific sensitivity (NOEC) distribution. Subsequently, these values were ordered in ascending fashion and plotted as a cumulative curve, resulting in a mean NOECs and associated distributions around it. The deterministic NOECs calculated from the dose descriptors reported in toxicity studies are depicted as points, where differing colours and shapes represent the polymer types and particle shapes. For the mass-based pSSD, one value could not be transformed from particle-based units due to lacking information on the size of the fibre tested, resulting in 45 data points from 23 species.

Removing all HONECs from the datasets (Fig. 1) resulted in O. melastigma showing as the most sensitive species (NOEC of  $3.9\cdot10^6$  part. m $^{-3}$ ). The overall lowest value was reported for C. gigas but due to differences in size, one higher value reported for this species resulted in the higher mean NOEC of  $9.77\cdot10^6$  part. m $^{-3}$ . When using mass-based concentrations, C. gigas was found to be the most sensitive species, with a NOEC of  $0.5~\mu g.L^{-1}$ . On average, C. neogracile and P. crassirostris are the least sensitive species (NOECs of  $1.3~and~1.2~mg.L^{-1}$ , respectively).

The PNEC distributions for marine species as both mass and particle concentrations were derived as the fifth percentiles of the pSSDs for the dataset containing all values (Table 1). The mean values are  $3.84\cdot10^6$ 



**Fig. 1.** Probabilistic species sensitivity distributions (pSSDs) for marine species exposed to microplastics in mass- and particle-based concentrations (A and B, respectively), excluding HONECs from the dataset. The mean pSSD curves are shown by the red lines, surrounding bars indicate the range of uncertainty. Points indicate single NOEC values.

Table 1
Statistical analyses of PNEC probability distributions in marine waters.

Unit	5 <sup>th</sup> quantile	25 <sup>th</sup> quantile	Mean	75 <sup>th</sup> quantile	95 <sup>th</sup> quantile
part. m <sup>-3</sup>	$1.74 \cdot 10^6$	$2.95 \cdot 10^6$	$3.84 \cdot 10^6$	4.74·10 <sup>6</sup>	5.89·10 <sup>6</sup>
μg.L <sup>-1</sup>	0.2	0.4	0.5	0.6	0.8

part m<sup>-3</sup> and 0.5  $\mu$ g L<sup>-1</sup>.

## 3.2. Exposure assessment

The final dataset for the probabilistic exposure assessment included 1056 measurements in total (Table S7). The worldwide coverage of water bodies is wide-ranging, as samplings were conducted even in remote areas such as Antarctica and Arctic waters. Both photic and aphotic environments were sampled (93 % and 7 %, respectively). To provide a general overview of the data, measurements were categorized according to their geographical location into groups of major water bodies. The Mediterranean Sea, the Pacific Ocean and the Atlantic Ocean are best covered by this dataset, with 35 %, 33 %, and 13 % of concentrations reported, respectively. Further concentrations were reported for the Arctic Ocean (10 % of data). Six percent of data consist of measurements from the Indian Ocean and Arabian Sea, with the

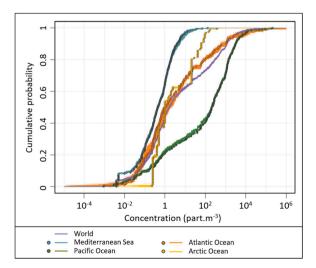
remaining 3% stemming from measurements of the Black Sea and Southern Ocean. Only groups with at least 100 data points were chosen to be depicted in Fig. 2. Concentrations in other water bodies are illustrated in Figure S2. On these figures, measured concentrations and their associated uncertainties and variabilities are represented in a cumulative order.

Key figures from MEC distributions in all water bodies are listed in Table 2. Worldwide, 89 % of reported concentrations are between  $10^{-2}$  and  $10^4$  part m<sup>-3</sup>. Higher concentrations were detected only in a few cases in the Pacific, Atlantic and Indian oceans. The widest ranges of concentrations were also found in these three major water bodies.

The high majority of data (87 %) stems from single station measurements with no reported variability, and was consequently not attributed any probability distribution. When variability was reported, it was mainly from samples taken at different locations or among replicates taken from stations at the same time, sometimes temporal variability was also reported.

Measurements in the Mediterranean Sea provide a good coverage of the entire water body. Sampling campaigns have covered regions from the Strait of Gibraltar to the Turkish and Israeli coasts of the Mediterranean Sea and areas of the Ligurian, Tyrrhenian and Adriatic Sea, as well as the Sea of Marmara. Concentrations commonly ranged between  $10^{-2}$  and 10 part.  $\rm m^{-3}$ , which constitutes 84 % of data, and a mean concentration of 2.4 part.  $\rm m^{-3}$ . The highest concentration reported (129 part  $\rm m^{-3}$ ) was measured after a flooding event at the Turkish coast. Such weather events are thought to increase MP concentrations in oceans due to increased input from rivers (Gündogdu et al., 2018). Approximately 13 % of samplings contained no MP at all. In this case, the reported or calculated LoD were used. Overall, 65 % of data reported concentrations of 1 part  $\rm m^{-3}$  or less.

The concentration data within the Pacific Ocean group cover a wide geographical area. Some sampling efforts in the Pacific Ocean set off the West Coast of Canada and the United States, from Vancouver Island to California. Some of the data stems from Mexico as well as the South Pacific subtropical gyre, (Eriksen et al., 2013). A large number of data from the Western Pacific were also included in this group, which includes the Chinese Sea, Yellow Sea and Japan Sea. The concentrations measured in the Pacific Ocean range between 10<sup>-3</sup> and 10<sup>5</sup> part. m<sup>-3</sup> with some non-detects included. Based on this data, 74 % of samples taken in the Pacific Ocean contained more than 1 part m<sup>-3</sup>, constituting the water body most polluted by MPs. It is noteworthy that of the 199 concentrations above 100 part. m<sup>-3</sup>, which represent approximately 55 % of the data, 192 were reported from studies that sampled the Western



**Fig. 2.** Cumulative probability curves of microplastic concentrations in major water bodies. Points represent reported concentrations, i.e. single measurements or reported means.

Pacific, mainly from coastal and estuarine areas.

The data categorized into the Atlantic Ocean group consists of samples from many different areas, including European, African, Northand South-American waters, as well as in the North Atlantic subtropical gyre. The majority of sampling efforts found concentrations in the range between  $10^{-2}$  and  $10^4$  part m<sup>-3</sup> with a mean concentration of  $3.6 \cdot 10^3$  part m<sup>-3</sup>. In 50 % of cases the MECs were 1 part m<sup>-3</sup> or less. Approximately 76 % of the data were found to be below 40 part. m<sup>-3</sup>. The remaining quarter of data are concentrations between 60 to more than  $1.52 \cdot 10^5$  part m<sup>-3</sup>. This is visible by a small jump and subsequent tailing off towards the higher end of the curve. The majority of data points in this group are from the North and Baltic Sea (30 % and 27 %, respectively), which are (semi-)enclosed water bodies.

Many samplings in the Arctic Ocean found no MP at all, which is visible by data points at the beginning of the curve. Most concentrations (79 %) are between 0.23 and 100 part.  $m^{-3}$ , resulting in the large jump in the cumulative curve. The mean concentration is 21 part.  $m^{-3}$ . Of the data points for the Arctic Ocean, 54 % of concentrations are of 1 part.  $m^{-3}$  or lower. Only four measurements resulted in concentrations above 100 part.  $m^{-3}$  and the highest reported value was 375 part.  $m^{-3}$ . These high concentrations are suggested to be a result of melting sea ice or point sources such as wastewater dumping by ships (Kanhai et al., 2018).

## 3.3. Risk assessment

The risk that MPs might pose to marine ecosystems was characterised in two ways, excluding HONECs from the hazard dataset. First, minima and maxima of MEC and PNEC probability distributions were compared (Fig. 3). Second, the whole probability distributions were used to calculate RCRs, shown in Fig. S3 and Table 3. An overlap does occur on 12 % of the total range of values included in both MEC and PNEC distributions (Fig. 3). This results in a global RCR with a mean of  $4\cdot10^{-4}$  and  $2.10^{-5}$  % of its values above 1, showing a very unlikely but possible risk.

#### 4. Discussion

This research shows the first results on the risk assessment of microplastics that is both specific to the marine environment and based on measured exposure concentrations. To account for the variability and the uncertainty associated with the recent experimental data used for this assessment, a probabilistic approach was used. The following sections discuss the strengths, weaknesses and reliability of the present assessment.

#### 4.1. Hazard assessment

The peer-reviewed literature contains contradictory data regarding the potential effect of particle size on marine toxicity. For example, an increase in particle size lead to a decrease in toxicity towards copepods and rotifers in Jeong et al. (2017,2016), Beiras et al. (2018) and Lee et al. (2013), while no influence of particle size was observed towards bacteria, diatoms, mussels and oysters by Gambardella et al. (2019); Yi et al. (2019) and Beiras et al. (2018). Fig. 4 shows the relation between particle size and toxicity in the whole dataset used in this assessment. While the overall MP toxicity seems to be lower with decreasing size when using particle-based concentrations (Fig. 4A), this could be an artefact, since larger sizes automatically lead to lower particle-based concentrations. The relationship between particle size and mass-based toxic concentrations seems non-existent (Fig. 4B). However, for particles above 12 µm, only HONECs were found, indicating no observed toxicity of these larger particles at the tested concentrations. Some of the animals tested with these particle sizes were planktonic species, for which such particles might not be recognised as food. Nevertheless, several organisms including an urchin, a mussel, a shrimp and two

**Table 2**Key figures of the probability distributions associated with measured environmental concentrations (part. m<sup>-3</sup>) reported from all water bodies. Q5, Q25, Q75, Q95: 5th, 25th, 75th, 95th quantiles, respectively.

Water body	Q5	Q25	Mean	Median	Q75	Q95
Worldwide	$1.3 \cdot 10^{-2}$	$2.6 \cdot 10^{-1}$	1.5·10 <sup>3</sup>	1.6	$1.5 \cdot 10^2$	$2.7 \cdot 10^3$
Atlantic Ocean	$2.9 \cdot 10^{-2}$	$2.4 \cdot 10^{-1}$	$3.6 \cdot 10^3$	1.3	$5.3 \cdot 10^{1}$	$4.7 \cdot 10^3$
Arabian Sea	$3.5 \cdot 10^{-2}$	$1.2 \cdot 10^{-1}$	$4.3 \cdot 10^{-1}$	$2.8 \cdot 10^{-1}$	$7.4 \cdot 10^{-1}$	1.1
Arctic Ocean	$2.5 \cdot 10^{-1}$	$4.0 \cdot 10^{-1}$	$2.1 \cdot 10^{1}$	1.0	2.1	95
Black Sea	$2 \cdot 10^{-1}$	$2.1 \cdot 10^{2}$	$8.9 \cdot 10^{2}$	$5.4 \cdot 10^2$	$1.1 \cdot 10^{3}$	$2.7 \cdot 10^3$
Indian Ocean	$1.5 \cdot 10^{-1}$	$1.5 \cdot 10^{1}$	$1.5 \cdot 10^3$	$1.8 \cdot 10^{2}$	$5.2 \cdot 10^2$	$1.2 \cdot 10^4$
Mediterranean Sea	$5.10^{-3}$	$1.0 \cdot 10^{-1}$	2.4	$4.5 \cdot 10^{-1}$	1.6	9.0
Pacific Ocean	$3.10^{-2}$	2.2	$2.8 \cdot 10^3$	$2.0 \cdot 10^{2}$	$1.2 \cdot 10^3$	$6.5 \cdot 10^3$
Southern Ocean	$3.5 \cdot 10^{-3}$	$4.0 \cdot 10^{-3}$	$8.3 \cdot 10^{-2}$	$4.8 \cdot 10^{-3}$	$9.9 \cdot 10^{-2}$	$4.5 \cdot 10^{-1}$

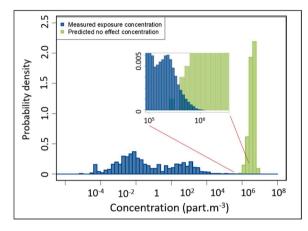


Fig. 3. Worldwide measured environmental concentration (MEC) and predicted no-effect concentration (PNEC) distributions in part  $\rm m^{-3}$  for marine habitats.

**Table 3**Key figures of the risk characterization ratio (RCR) distribution in marine waters.

RCR	RCR
5 <sup>th</sup> quantile	$4.10^{-9}$
25 <sup>th</sup> quantile	$7.10^{-8}$
Mean	$4.10^{-4}$
Median	$5.10^{-7}$
Q75	$4.10^{-5}$
Q95	$8 \cdot 10^{-4}$

fishes, were also tested with particles larger than 12  $\mu m$  and up to a few hundred  $\mu m$ , which are within the size range of their usual food. Moreover, 80 % of the available data points are HONECs. This shows that the overall toxicity of microplastics is low, as actual toxic doses could be observed in only a fifth of the results. The distribution of the different trophic levels is quite even and all levels are tested with small and large particle sizes. A recent hazard assessment for nanoplastics in marine resulted in a PNEC value of 72  $\mu g/l$  for the best available dataset (Yang and Nowack, 2020), so within the range of NOEC values visible in Fig. 4b. These authors also stated that nanoplastics seem to be less toxic than microplastics and engineered nanomaterials. The often mentioned increase in toxicity with smaller particle size is therefore not visible in the currently available datasets on nanoplastic and microplastic.

Two additional points regarding the toxicity assays themselves are worth highlighting. First, it should be remembered that MP tend to aggregate in seawater, so they can be seen as larger particles to organisms and, if dense enough, settle down the water column of the bioassay (Davarpanah and Guilhermino, 2015; Prata et al., 2018). This lowers the bioavailability of the MPs and should be considered in ecotoxicity assessment, but is not always the case. Therefore, the doses reported in

this study could be higher than the actual doses to which the organisms were exposed, and the hazard thereby underestimated. The nominal sizes reported here should be seen as the minimum size of the particles occurring in the test media. Second, although behavioural endpoints were not considered in this assessment, they were shown to be several orders of magnitude more sensitive than mortality (Gambardella et al., 2017). Moreover, almost 7 out of the 9 the data points used here for hazard assessment are based on acute toxicity. Therefore, risk assessment based on more chronic studies and more sensitive endpoints could result in a risk towards marine organisms. This was out of the scope of this study, which focused on following regulatory guidelines, but should be kept in mind.

#### 4.2. Exposure assessment

The exposure assessment showed that concentrations of MP could be as high as 1.3·10<sup>6</sup> part. m<sup>-3</sup> in the water column. Most samplings were conducted near or in coastal areas (72 %) rather than in the open ocean. Only 18 % of the data come from waters that were clearly distinguished as estuarine, all of them being in close proximity to the coast. The coastal subset also included the Baltic Sea and Black Sea, which are (semi-) enclosed water bodies. Worldwide, the mean value of coastal concentrations is about one order of magnitude higher than in open waters  $(1.6\cdot10^3 \text{ and } 4.7\cdot10^2 \text{ part. m}^{-3}, \text{ respectively, Table 4})$ . This is mainly driven by concentrations measured in the Atlantic Ocean, were differences between coastal and open waters are of 2 orders of magnitude. However, not much difference can be seen between both types of waters in the Pacific Ocean and in the Mediterranean Sea. In the Mediterranean Sea, open waters are slightly more polluted than coastal waters (mean concentrations of 3.1 and 1,8 part. m<sup>-3</sup>, respectively). Two main sources of MPs can be distinguished: MPs coming from land, where most plastic is used, would explain high concentrations in coastal areas, while the degradation of larger plastic objects in marine waters could partly explain high concentrations in open waters. Both sources of MPs are moved within seas and oceans by currents, which accumulate plastics in patches localised in open waters.

The sampling method has a clear influence on the reported microplastic concentrations. In our dataset, 66 % of concentrations were obtained with manta trawl or neuston net mesh sizes between 200 and 505  $\mu m$ . The lowest mesh size for a net was 20  $\mu m$  (Ter Halle et al., 2017). In some cases, bulk sampling was conducted via a pumping device, the intake system of a ship or Niskin bottles. The bulk water samples were then subsequently filtered with filter pores of 0.22–333  $\mu m$ .

A slight negative correlation ( $R^2=0.4601,\ p$ -value  $<2.2\cdot10^{-16},$  Pearson's coefficient =-0.6787) can be observed between measured concentrations and mesh or filter pore sizes (Fig. 5), indicating that smaller filtration sizes result in higher MECs. Most notably, the slope is steeper for mesh sizes below 200  $\mu$ m, resulting in higher detected concentrations. In the Mediterranean Sea, samples were exclusively taken with mesh sizes larger than 200  $\mu$ m, whereas the employed mesh sizes for samples in the Atlantic and Pacific Ocean ranged from 0.45–500  $\mu$ m. This might partly explain why measured concentrations in the

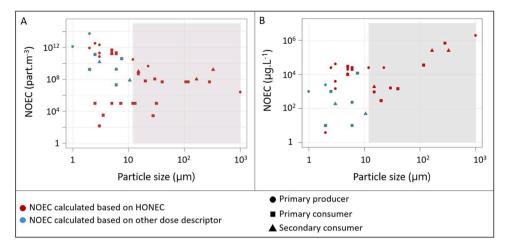
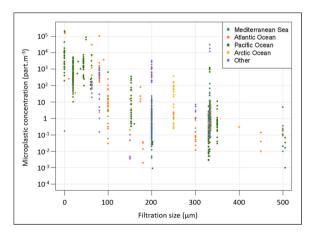


Fig. 4. Relation between particle sizes used in toxicity studies and resulting no-effect concentrations. Dose descriptors other than HONEC include NOECs, LOECs and EC50s. The grey shaded area denotes the particle size larger than  $12 \, \mu m$  where only HONECS were available. The shape of the data points refers to the trophic level of the tested species.

**Table 4**Key figures of probability distributions associated with measured environmental concentrations reported from coastal and open waters (part m<sup>-3</sup>). Q5, Q25, Q75, Q95: 5th, 25th, 75th, 95th quantiles, respectively.

Water body	Q5	Q25	Mean	Median	Q75	Q95
Worldwide	1.3·10-2	2.6·10-1	1.5.103	1.6	1.5.102	2.7·103
Coastal waters	$1.7 \cdot 10 - 2$	$2.4 \cdot 10 - 1$	1.6.103	1.9	2.4.102	2.6.103
Open waters	$1.0 \cdot 10 - 2$	4.010 - 1	4.7.102	1.2	2.1.101	2.9.103
Atlantic Ocean	$2.9 \cdot 10 - 2$	$2.4 \cdot 10 - 1$	3.6.103	1.3	5.3.101	4.7.103
Coastal waters	$3.0 \cdot 10 - 2$	$2.4 \cdot 10 - 1$	2.5.103	$8.8 \cdot 10 - 1$	2.4.101	2.5.103
Open waters	$2.5 \cdot 10 - 1$	$8.0 \cdot 10 - 1$	4.9.101	1.7	2.7.101	2.0.102
Arctic Ocean	$2.5 \cdot 10 - 1$	$4.0 \cdot 10 - 1$	2.1.101	1.0	2.1.101	9.5.101
Coastal waters	NA	NA	NA	NA	NA	NA
Open waters	$2.5 \cdot 10 - 1$	$4.0 \cdot 10 - 1$	2.1.101	1.0	2.1.101	9.5.101
Mediterranean Sea	$5.0 \cdot 10 - 3$	$1.0 \cdot 10 - 1$	2.4	$4.5 \cdot 10 - 1$	1.6	9.0
Coastal waters	$4.8 \cdot 10 - 3$	$7.2 \cdot 10 - 2$	1.8	$2.8 \cdot 10 - 1$	1.1	5.4
Open waters	$3.0 \cdot 10 - 1$	$9.5 \cdot 10 - 1$	3.1	1.7	3.2	1.2.101
Pacific Ocean	$3.0 \cdot 10 - 2$	2.2	2.8.103	2.0.102	1.2.103	6.5.103
Coastal waters	$3.3 \cdot 10 - 1$	1.2.101	3.1.103	2.2.102	1.1.103	4.6.103
Open waters	$3.0 \cdot 10 - 3$	$3.0 \cdot 10 - 2$	1.8.103	1.8	2.2.103	9.1.103



**Fig. 5.** Differences in filter or mesh size used during sampling and the effect on measured concentrations.

Mediterranean Sea were not as high as in the oceans. Recently an approach has been published to rescale measured microplastic concentrations within a certain size range to any other size range based on the power law that relates particle abundance and size (Koelmans et al., 2020). Such an approach might be used in future exposure and risk

assessments.

## 4.3. Risk assessment

Globally, the mean RCR is  $4.10^{-4}$  but a very small part of the probability distribution is beyond 1. Therefore, although risks are highly unlikely in the present, they cannot be excluded. They might rise in the future, since it is mostly non-biodegradable, more plastic will be discarded even if production stagnates, and will reach the environment (Everaert et al., 2018). However, in order for the mean RCR to reach one, the exposure concentration would need to a factor of 2500 higher and is therefore unlikely to be reached in the near future even if the current plastic leakage persists. This should be taken into consideration by decision makers aiming to protect our environment. However, these conclusions could be better supported if the discrepancies appearing between the dataset used for hazard assessment and that used for exposure assessment were smaller. The RCR for microplastic in the ocean is similar to the one calculated for freshwaters with modes of  $1.3 \cdot 10^{-6}$  in North America,  $3.3 \cdot 10^{-6}$  in Europe and  $4.6 \cdot 10^{-3}$  in Asia (Adam et al., 2018).

PS was used most in toxicity studies (56 % of data points), but was on average only 4% of the MPs found in environmental samples (Fig. 6A). The polymer type information was not available from all studies, so the numbers are from a more limited dataset. The most abundant polymer type in marine waters were PE and PP, with 36 % and 27 % respectively,

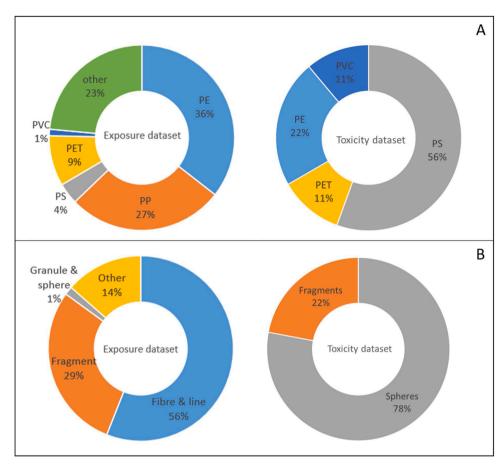


Fig. 6. A: Comparison of polymers found in the environment (exposure dataset) and polymers used in ecotoxicity assays (toxicity dataset). B: Comparison of MP shapes found in environmental samples (exposure dataset) and used in ecotoxicity assays (ecotoxicity dataset). PE: polyethylene, PP: polypropylene, PS: polystyrene, PET: polyethylene terephthalate, PVC: polyvinyl chloride.

of the average concentrations in samples. However, only 22 % of hazard data points were based on PE testing and none on PP. Shapes of MPs tested in bioassays are mainly spheres (78 % of data points), while fibres and fragments were found in the environment as the main MP shapes, counting on average for 56 % and 29 % of particles counted, respectively (Fig. 6B).

Another large discrepancy between environmental concentrations and MPs in toxicity studies concerns their size. Fig. 7 clearly illustrates that particles used for toxicity tests are smaller than most of those found in seawater, as their average diameters are all between 1 and 10.5  $\mu m$ . The disagreement between the polymer types, forms and sizes of particles detected in the ocean and used in the hazard studies, seriously

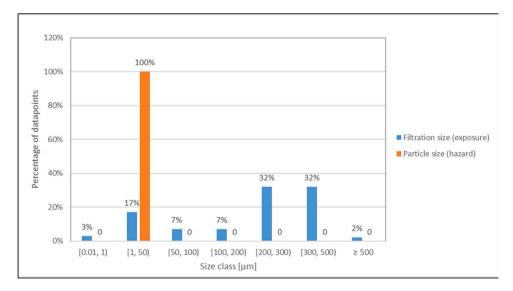


Fig. 7. Percentages of data points in different size classes. Filtration size represents the lower size limit used to sample microplastics from water and particle size refers to those used in toxicity studies.

V. Adam et al. Aquatic Toxicology 230 (2021) 105689

compromises the usefulness of the hazard data set for risk assessment. For a realistic RA, it is urgently necessary that hazard studies are performed using actual particles sampled from the ocean or having the same composition, size and form. However, as sizes larger than  $12~\mu m$  were never shown to have any observable effect on the used test species, using larger sized MP would likely increase the PNEC values and thus decrease the RCR.

Koelmans et al. (2020) recently suggested a method to not only rescale the measured exposure concentrations as discussed above but also apply a correction to SSDs that makes data obtained with different microplastic types used in the laboratory compatible with those observed in nature and applied the method to a freshwater dataset. This is one possible approach to solve the issue of misalignment of exposure and hazard data. However, such a novel approach needs to be backed up with experimental data and therefore toxicological studies under relevant exposure conditions and with realistic microplastic types and sizes need to be performed in order to expand the data basis needed for risk assessment and to understand better the influence of size and particle type on effects.

#### 5. Conclusions

The results of this study show that currently risks of MPs towards marine organisms are unlikely, although they cannot be completely excluded since parts of the MEC and PNEC distributions overlap. This assessment strongly relies on the experimental data available and the analysis would be more accurate if the datasets available for exposure and hazard would rely on more similar types of particles. This means that ecotoxicity assays should be performed more often on the types and sizes of particles that are most frequently observed in the environment. Nevertheless, the hazard dataset shows a good coverage of species required by regulation, and the exposure dataset relies on measurements performed all over the globe. Therefore, the conclusions drawn here remain valid. The strong reliance of this risk assessment on available data also means that the present results could change as more become available or as concentrations of MP in marine waters increase.

## CRediT authorship contribution statement

Véronique Adam: Methodology, Software, Validation, Resources, Writing - original draft, Visualization, Supervision. Alex von Wyl: Formal analysis, Data curation, Writing - original draft, Writing - review & editing, Visualization. Bernd Nowack: Conceptualization, Validation, Investigation, Writing - review & editing, Supervision, Project administration, Funding acquisition.

## **Declaration of Competing Interest**

The authors have no competing interest to declare.

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## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.aquatox.2020.105689.

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