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On the probability of ecological risks from microplastics in the Laurentian Great lakes☆

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ABSTRACT

The Laurentian Great Lakes represent important and iconic ecosystems. Microplastic pollution has become a major problem among other anthropogenic stressors in these lakes. There is a need for policy development, however, assessing the risks of microplastics is complicated due to the uncertainty and poor quality of the data and incompatibility of exposure and effect data for microplastics with different properties. Here we provide a prospective probabilistic risk assessment for Great Lakes sediments and surface waters that corrects for the misalignment between exposure and effect data, accounts for variability due to sample volume when using trawl samples, for the random spatiotemporal variability of exposure data, for uncertainty in data quality (QA/QC), in the slope of the power law used to rescale the data, and in the HC5 threshold effect concentration obtained from Species Sensitivity Distributions (SSDs). We rank the lakes in order of the increasing likelihood of risks from microplastics, for pelagic and benthic exposures. A lake-wide risk, i.e. where each location exceeds the risk limit, is not found for any of the lakes. However, the probability of a risk from food dilution occurring in parts of the lakes is 13–15% of the benthic exposures in Lakes Erie and Huron, and 8.3–10.3% of the pelagic exposures in Lake Michigan, Lake Huron, Lake Superior, and Lake Erie, and 24% of the pelagic exposures in Lake Ontario. To reduce the identified uncertainties, we recommend that future research focuses on characterizing and quantifying environmentally relevant microplastic (ERMP) over a wider size range (ideally 1–5000 μm) so that probability density functions (PDFs) can be better calibrated for different habitats. Toxicity effect testing should use a similarly wide range of sizes and other ERMP characteristics so that complex data alignments can be minimized and assumptions regarding ecologically relevant dose metrics (ERMs) can be validated.

1. Introduction

The Laurentian Great Lakes are an iconic ecosystem. They support a population of approximately 48 million people and 3500 wildlife species, with large differences across socio-economic and ecological characteristics (Bunell et al., 2014; Hartig and Munawar, 2021). Stressors associated with changes in socio-economic activity within the region, such as population growth, and agricultural and industrial activity, have been estimated and mapped for the period between 1790 and 2010 (Reavie et al., 2018; Pillsbury et al., 2021), and give an indication of the different activities that can lead to nonlinear ecosystem responses to stressors. Ecosystem-wide responses can be negligible until a threshold is reached, at which point a small disturbance can result in the collapse of important system-dependent functions from which recovery is difficult (Scheffer et al., 2015). The timely implementation of local risk management measures is thus important in helping to prevent ecosystem collapse (Pillsbury et al., 2021).

Plastic debris has become a major concern for the Laurentian Great Lakes (reviewed by Earn et al., 2021; Fuschi et al., 2022). In our earlier work, we incorporated data available for the Laurentian Great Lakes into our global non-probabilistic risk assessment for freshwater systems (Koelmans et al., 2020; Redondo-Hasselerharm et al., 2023). The

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assessment used quality assurance and control (QA/QC) screening, alignment, and rescaling of exposure and effect data, and estimated low probability of risk (Koelmans et al., 2020; Redondo-Hasselerharm et al., 2023). Recently, however, it was suggested that concentrations of microplastic in the Great Lakes may now exceed effect thresholds, with the implication that the Great Lakes should be assigned high priority towards managing the risks associated with exposure to microplastic particles (MPs) (Munno et al., 2022; Rochman et al., 2022).

Environmental risk assessment (ERA) is an important pillar in the risk management of environmental stressors and in the development of policies to reduce the negative effects of those stressors. Environmental MPs represent a very complex, heterogeneous mixture of particles of varying size, shape, and polymer type (Gouin et al., 2019; Kooi et al., 2021; Koelmans et al., 2022). This represents a challenge when attempting to extrapolate between lab-based effects data and environmental concentrations, whereby significant concerns have been raised regarding the quality of input data and the reliability of assessing risks (de Ruijter et al., 2020; Koelmans et al., 2020; Mehinto et al., 2022). In previous work, we developed an ERA framework that provides a consistent characterization of ecological risks to MPs by correcting for the heterogeneity of MP characteristics (Koelmans et al., 2017, 2020; 2022; Redondo-Hasselerharm et al., 2023). The main feature of the framework is the use of data alignment methods that allow data comparability between environmental monitoring data, typically obtained using different analytical methods, and which can then be translated into an Environmentally Relevant MP (ERMP) metric. For example, to avoid the arbitrary and limited representativeness of MP samples, exposure, and effect data are rescaled to a standard ERMP size range (1–5000 μm), and effect threshold concentrations for species used in Species Sensitivity Distributions (SSDs) are then aligned to the same ERMP characteristics and corrected for the biologically accessible ERMP fraction (Koelmans et al., 2020). The alignments require that the ecologically relevant dose metrics (ERMs) be known (Koelmans et al., 2017), as this information is needed to translate the data. ERMs must therefore be defined based on curated and confirmed ERMP effect mechanisms (Koelmans et al., 2017). A second feature of the framework is the use of QA/QC evaluation to screen available data for their suitability for ERA. To support this evaluation, we have developed various quantitative QA/QC evaluation tools (Hermsen et al., 2018; Koelmans et al., 2019; de Ruijter et al., 2020; Wright et al., 2021; Redondo-Hasselerharm et al., 2023) and they have been applied to various assessments worldwide (Koelmans et al., 2020; Redondo-Hasselerharm et al., 2023; WHO, 2019, 2021; Mehinto et al., 2022; Coffin et al., 2022).

Consistent with assessing the risk for other stressors (Halden, 2015), the above framework continues to evolve as new data are made available. Additional components towards the evolution of the framework include the need to strengthen several aspects. First, when communicating the risks of any environmental contaminant, including MPs, it is important to define the associated uncertainties and the sensitivity of those uncertainties with respect to the assessment outcome, as accurately as possible (Wardman et al., 2021). Although conceptually correct, application of the ERA framework has not yet included a quantitative analysis of error propagation when deriving risk characterization ratios (RCR). The ERA framework for MPs relies on rescaling and alignment approaches, which rely on the quality and reliability of input data and parameters, for which there is an inherent level of uncertainty. Therefore, flexibility is needed to enable re-evaluation as new information becomes available, aimed at both quantifying and reducing the relative uncertainty in the RCR derived. It is thus important that the influence of the relative level of uncertainty associated with the input data and assumptions used in model parameterization be transparently communicated. It is for this reason that uncertainty analysis represents a critical part of Good Modelling Practice (GMP) (Buser et al., 2012).

Second, mathematical models used for ERA include various simplifying assumptions used to describe the environment. Consequently, an important challenge is understanding how underlying uncertainty in the assumptions used to realistically account for the ecological relevance of input variables may influence model output. For instance, previous assessments using the ERA framework for MPs are based on a limited number of samples taken at a particular location, with an assumption that the data are representative of the system being assessed (Koelmans et al., 2020; Redondo-Hasselerharm et al., 2023; Rochman et al., 2022; Coffin et al., 2022). Protection goals in ERA, however, require a statistically relevant number of samples to extrapolate exposure concentrations to populations and communities that occupy a much larger spatial scale. Since it is known that concentrations of MPs are highly variable across spatial and temporal scales, data obtained from dynamic water systems, such as lakes or rivers, which represent 'snapshot' samples, may not be representative of the 'true' exposure for the ecosystem under consideration (Mintenig et al., 2020; Talbot and Chang, 2022; Bäuerlein et al., 2023). Thus, risks cannot be reliably defined for a specific location or depth, such as where and when a sample was taken. An alternative approach, aimed at providing a more reliable and relevant estimation of risk, is to model exposure probabilistically based on a system-wide distribution of concentration data that is representative of the habitat or ecosystem being assessed.

Lastly, if tools used in deriving risk based on SSDs are to be used to ensure that data interpretation and communication reflect full transparency, limitations associated with the availability of habitat-specific data for SSDs must be appropriately represented. For instance, the availability of limited effect data, representative of model species for both marine and freshwater systems, results in all available data being combined to enable an SSD to be populated. Although defensible in the absence of data, it can lead to uncertainty if the SSD uses underlying data standardized on ERMP characteristics for environments not representative of the system under investigation. For instance, using data aligned to marine environments, and marine species data (Mehinto et al., 2022) that might be subsequently applied to exposure data for freshwater systems. Application of marine relevant data to exposure data for freshwater systems such as the Great Lakes (e.g., Rochman et al., 2022), would therefore be inappropriate. The subsequent result is an inherent mismatch between the different types of input data, which thus results in the potential for misinterpretation of the potential risk.

The aim of this article is thus threefold. Our primary goal is to comprehensively assess the risk for ERMP in both the sediments and surface waters of the Laurentian Great Lakes. The assessment is supported by our second aim, which is to provide a quantitative update to our framework with new effect thresholds specific to freshwater ecosystems, and which includes new data for the characterization of ERMP and the application of a probabilistic risk characterization model that quantifies and reports the uncertainty propagated through the assessment. Our final aim is to critically review the method used and its results in relation to other published ERA methods, with an emphasis on implications to policy and regulatory decision-makers.

2. Methods

2.1. Data collection and study characteristics

To obtain measured environmental concentrations (MECs) of MPs in sediments and surface waters of the Great Lakes, an extensive literature search (until September 2022) was performed using the Scopus and Web of Science databases. The following string was used: (Microplastic(s) AND Lake AND (Huron OR Erie OR Ontario OR Superior OR Michigan). Furthermore, reference lists were explored and cited reference searches were performed using recent reviews (Earn et al., 2021). From the available datasets, the mean, standard deviation, and/or minimum and maximum MP number concentrations in water (#/L) and sediment (#/kg dw) are extracted (Table S1, Table S2). For effect threshold concentrations, the following strings were used: (effect OR impact OR toxicity) AND (microplastic(s) OR plastic particle(s) OR fiber(s)) AND (freshwater OR aquatic). Studies were used only if a No-Observed Effect

Concentration (NOEC), as well as a Lowest-Observed Effect Concentration (LOEC), was reported, to avoid using "false NOECs". This was not possible for sediments because of the few data available. Furthermore, only organismal endpoints were considered (Table S3). For studies reporting MECs or effect threshold concentrations, study characteristics were summarized (Supporting Information (SI), Table S3).

2.2. Quality assurance/quality control (QA/QC) evaluation

The evaluation of the quality and relevance of data is based on applying transparent criteria aimed at assessing a number of fundamental components. MECs and effects threshold data were screened for their reliability and relevance for ERA, using published QA/QC criteria for freshwater surface water (Koelmans et al., 2019; WHO, 2019), for sediment (Redondo-Hasselerharm et al., 2023), and for laboratory effect studies (de Ruijter et al., 2020). For each criterion, a score of either 2 (adequate), 1 (adequate with restrictions), or 0 (inadequate) was given for each dataset (Tables S4–S31). A 'Total Accumulated Score' (TAS) was calculated by adding the scores for each criterion, with a maximum of 20 and 40 points for MECs and effect threshold concentrations, respectively. As in previous studies, it is the number of non-zero scores that determines the reliability of a study, whereby studies that receive a non-zero score against all QA/QC would be perceived as the most reliable for ERA (Koelmans et al., 2019; de Ruijter et al., 2020; Redondo-Hasselerharm et al., 2023).

2.3. Data alignment and construction of species sensitivity distributions (SSDs) for ingested particle volume and area as relevant dose metrics

2.3.1. Alignment of measured environmental concentrations of microplastic

Due to the inherent heterogeneity associated with ERMPs, the MECs can't be directly compared across studies or against effect threshold concentrations (Koelmans et al., 2020). For the concentration data used in the present study, the target minimum particle sizes ranged from 53 to 333 μm (Table S1, Table S2). Therefore, rescaling MECs to be representative of a size range of between 1 and 5000 μm by multiplying by a correction factor based on Probability Density Functions (PDFs) for ERMP is required (Koelmans et al., 2020; Kooi et al., 2021; Alkema et al., 2022). For power-law PDFs, slope values for freshwater and sediment concentration data of $\alpha = 2.64 \pm 0.01$ and $\alpha = 3.25 \pm 0.19$ have been reported, respectively (Kooi et al., 2021). These values are considered to represent appropriate proxies of the slopes used to describe the generic distribution of power laws for MPs reported in aqueous and sediment samples collected from the Laurentian Great Lakes. Details are described in the SI (see equation S1).

2.3.2. Alignment of laboratory effect threshold concentrations

We briefly summarize the alignment methods used here, with more detailed information available in previous studies (Koelmans et al., 2020, 2022; Kooi et al., 2021; Coffin et al., 2022; Redondo-Hasselerharm et al., 2023). The lab-based threshold effect concentration data derived for mono-, and in some instances, polydisperse particles, is extrapolated into an effect concentration representative of an approximation of the true degree of polydispersity of ERMP (Koelmans et al., 2020). The extrapolation can only be performed in instances where sufficient knowledge regarding the unit that the extrapolation is based upon is available, with the unit used referring to the ERM (Koelmans et al., 2017). ERMs are defined based on knowledge regarding the toxicological mechanism of action, which directly relate the physicochemical properties of ERMP to the relevant adverse effect on biota (de Ruijter et al., 2020). Specifically, either a food dilution mechanism of action or a tissue translocation-mediated toxicity mechanism of action, which can result in e.g., inflammation and oxidative stress, represent the two most plausible mechanisms of toxicological action. For these mechanisms, ingested particle volume and particle surface area are used as the quantifiable ERMs, respectively (Koelmans et al., 2020; Kooi et al., 2021; Thornton Hampton et al., 2022).

2.3.3. Determination of HC5 thresholds from SSDs

For sediment exposures to ERMP, we recently determined Hazardous Concentrations for 5% of the species (HC 5, with 95% CI) of 4.9×10^9 (6.6×10^7–1.9×10^{11}) and 1.1×10^{10} (3.2×10^8–4.0×10^{11}) particles/kg sediment d. w., for food dilution and translocation-mediated toxicity, respectively (Redondo-Hasselerharm et al., 2023). Because the most recent data and insights were used to calculate these thresholds, we use these numbers in our ERA framework for benthic communities in the Laurentian Great Lakes. Applying the most recently published data we also derive a new freshwater SSD for both food dilution and translocation-mediated toxicity.

The water-exposure threshold effect concentrations use the NOECs reported as particle number/L, which were selected after screening the compiled data (Table S3). In some cases, the NOEC was only given in mg/L, and the NOEC in particles/L was calculated using established methods (Leusch and Ziajahromi, 2021). Assessment factors 10 and 2 were used to convert acute to chronic toxicity data, and to convert LOEC or EC10 data to NOEC data, respectively (Koelmans et al., 2020; Wigger et al., 2020; Mehinto et al., 2022). For alignment of thresholds in the context of the food dilution mechanism, particles that are too large to be ingested by the organism in question were considered biologically unavailable and thus excluded from further calculations (Koelmans et al., 2020; Kooi et al., 2021; Mehinto et al., 2022; Coffin et al., 2022; Redondo-Hasselerharm et al., 2023). For each of the species, bioaccessible size fractions of ERMPs were obtained from either MP ingestion data, food ingestion data, or mouth opening size (Table S3). For the alignment of thresholds in the context of tissue translocation-mediated toxicity, particles that are too large to be displaced through tissue were considered biologically unavailable. Following recent studies (Mehinto et al., 2022; Coffin et al., 2022; Redondo-Hasselerharm et al., 2023), a length of 83 μm was used as the maximum particle size for translocation. Note that the total surface area of an ERMP mixture with power law dimensions is largely determined by the smallest particles, e.g., those $\ll$ 83 μm. Therefore, the calculations are not sensitive to the cut-off value. In the case of organisms with mouth openings smaller than 83 μm, their mouth opening size was used as the upper limit for bioaccessibility in the context of tissue translocation (Table S3).

The new freshwater SSDs contain 39 data points, representing 15 species, from 11 taxonomic groups. While the artifacts of evaluating ERMP toxicity based on non-representative particles are theoretically corrected by the alignments, we recommend that the set of particles on which the SSDs are calibrated is as diverse as possible. The data on which the new SSDs are based represent the following shape categories: sphere (54%), fragment/irregular (36%), fiber (10%); polymer types: PE (44%), PS (31%), PET (7.7%), PP (2.6%), PVC (2.6%); and tested particle sizes between 1 and 5761 μm, with an average of 70 μm. An analysis of 20,004 individual ERMP particles sampled from freshwater showed a very similar mean and spread of 91 ± 228 μm (Kooi et al., 2021).

Thresholds included NOECs (85% of the data points, AF = 1), LOECs (10% of the data points, AF = 2), and EC50 (5% of the data points, AF = 10), indicating minimal overall use of AFs. For acute toxicity data (46% of the data points) an AF of 10 was used. The derivation of SSDs specific to freshwater species was calculated by normalizing the NOECs based on corrections for both volume and surface area, which were used as the ERMs, and using the ssdtools package in Rstudio (version 4.1.3) (Thorley and Schwarz, 2018). The ssdtools package uses the maximum likelihood estimation to fit 10 cumulative distribution functions to the NOECs of the different species and to evaluate the goodness of fit. Estimates of the 5% Hazardous Concentration (HC5) and the 95% confidence limits using the best-fitting distribution were based on 1000 bootstrap iterations.

For comparison, we also include a probabilistic ERA using the effect thresholds reported by Mehinto et al. (2022), who derived different thresholds based on a series of increasingly conservative choices regarding the inclusion of endpoints, which is representative of how the proposed ERA framework could be used to inform risk management.

2.4. Probabilistic assessment of risk characterization ratios

The probabilistic ERA of ERMP is performed by quantifying the uncertainties in both the numerator and denominator of the risk characterization ratio (RCR = MEC/NOEC). The ERA performed enables an assessment of the spatiotemporal scale of ecological communities, which is not consistent with the scale of data obtained from a specific sample (i.e. location). Given the inherently dynamic nature of hydrology (e.g. water flow rates), concentration data for MP in water cannot be considered site- or time-specific (Mintenig et al., 2020; Talbot and Chang, 2022). Similar constraints also apply to the dynamics of the sediment-water interface, whereby wind- and flow-induced pressure gradients can cause resuspension and lateral transport when critical shear stress is exceeded (Hawley et al., 2009; Besseling et al., 2017). Concentration data were thus seen as a statistical sample of the true distribution of concentrations at the lake system level.

Gaussian distributions are assumed for exposure concentration data that are reported as mean values with standard deviations; triangular distributions are used in instances where only a mean value is reported accompanied by data of both the minimum and maximum values (Table S1, Table S2). The concentration data from the distributions derived are then rescaled to reflect particle sizes between 1 and 5000 μm as a particle number concentration (Equation S1), with the uncertainty modeled as a power law slope, α, based on the derived standard deviation of the normal distribution. The uncertainty for trawl-based surface water concentrations is determined as the relative standard deviation of 0.48 which is based on a recent review of trawl data (Karlsson et al., 2020; Pasquier et al., 2022). Given the relative extent of uncertainty intervals in reported effect thresholds (e.g., HC5), a log-normal distribution related to the data is assumed. In this instance, Monte Carlo simulations are based on 10^5 iterations, with each iteration representing a random sampling of the aforementioned distributions. The results are reported as log-risk characterization ratio (Log RCR) distributions, which include a quantitative evaluation of the overall uncertainty in the RCR by error propagation. Metrics used to characterize the RCR distributions are mean LogRCR, 5–95% percentile range, and 99% percentile of the distribution. The probability of a risk occurring is calculated as the percentage of the distribution (area under the curve) where the MECs are greater than the PNEC, i.e. where Log RCR >0 (%RCR>1). Risk of ERMP was considered to be absent if the RCR was estimated as < 1 in the 95% percentile of the Log RCR distribution. Calculations were performed with Microsoft Excel (MCSim version May 4, 2013) (Barreto and Howland, 2006).

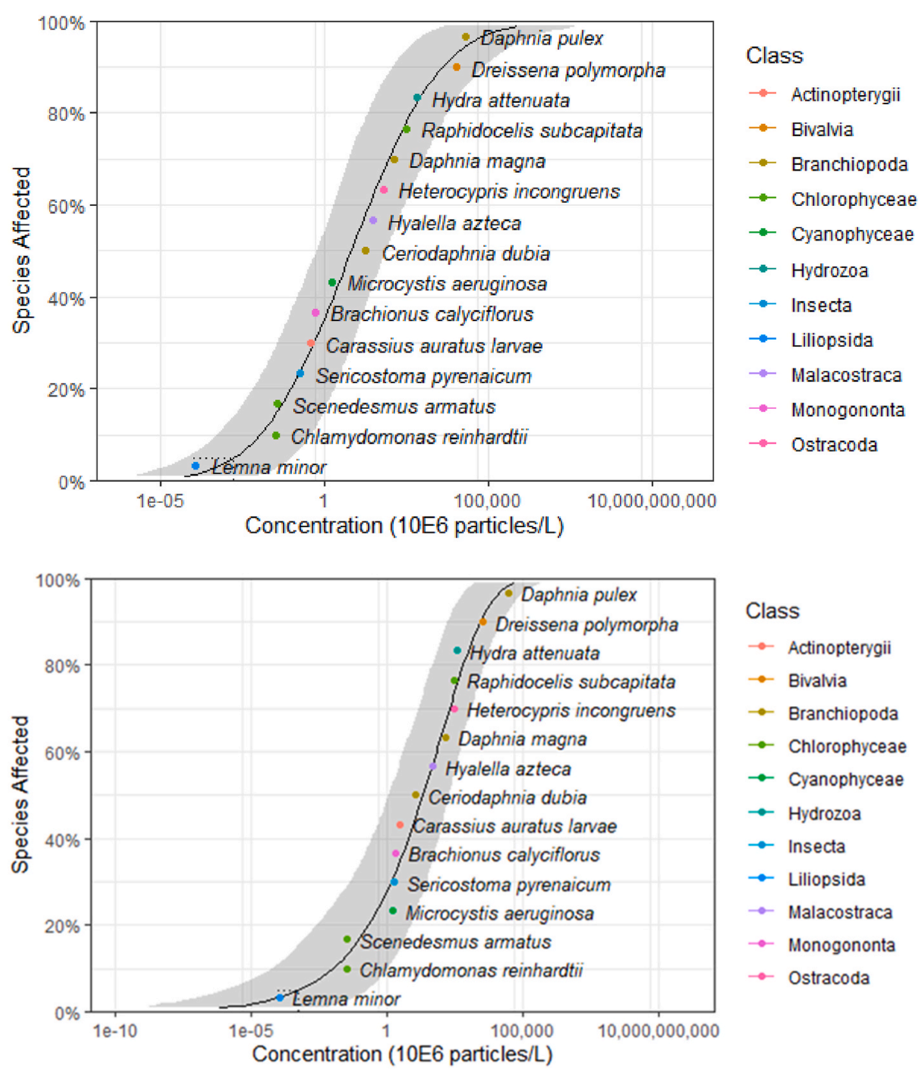

Fig. 1. Microplastic Species Sensitivity Distributions for freshwater species including vertebrates, invertebrates, macrophytes and algae, (A) for the ERM particle volume, to represent a food dilution effect mechanism, and (B) for the ERM particle area to represent a toxicity mechanisms triggered by translocation.

3. Results and discussion

3.1. SSDs for effects of ERMP on freshwater species based on food dilution and translocation-mediated toxicity mechanisms

SSDs developed are based on data alignment between effect data and the complete ERMP size range (1–5000 μm). Fig. 1 summarizes the SSDs for both a particle volume-based 'food dilution' effect mechanism (Fig. 1a) and a particle area-based toxicity mechanism, triggered by translocation (Fig. 1b). The HC5, derived from the assumption that food dilution is responsible for the observed adverse effect in laboratory tests, was 547 (95% CI; 2.24–1.72 $\times$ 10^5) particles/L based on a Weibull best fitting distribution. This is approximately seven times higher than a previously reported freshwater HC5 threshold of 75.6 particles/L (Koelmans et al., 2020). The HC5 derived for translocation-mediated toxicity was 1688 (95% CI; 54.5–1.07 $\times$ 10^5) particles/L, assuming a log-normal distribution. The observation that the HC5 is lower for a food dilution than for a translocation based toxicity mechanism is consistent with results reported in previous studies (Mehinto et al., 2022; Redondo-Hasselerharm et al., 2023). The observed consistency is related to underlying assumptions in the calculation. Specifically, the volume (ERM for food dilution) uses the third power of the particle size (l^3), as opposed to surface area (ERM for toxicity triggered by translocation) which uses the second power (l^2).

The most sensitive species influencing the SSD and the HC5 are observed to be *Lemna minor* and *Chlamydomonas reinhardtii* (Fig. 1). We note that the ERM for these species is agnostic with regard to the relevant effect mechanisms, consequently caution is warranted when interpreting the relative mechanism of action. Specifically, both an ingestion-based and a translocation-based mechanism are either not applicable or cannot yet be adequately parameterized. Due to this observation, we therefore also developed SSDs that omit algae and macrophyte species, using only aquatic heterotrophic species capable of ingesting particles (Fig. S1). In this case, the $HC5^i$ (with 'i' for ingestion only) for a food dilution mechanism (log-Gumbel distribution) was 1.2 $\times$ 10^6 (2.9 $\times$ 10^5–1.3 $\times$ 10^7), and for a translocation-mediated toxicity (log-normal distribution) it was 7.3 $\times$ 10^4 (3.5 $\times$ 10^3–3.7 $\times$ 10^6) particles/L. Without the aforementioned algae and macrophyte species, the effect thresholds are observed to be significantly greater, i.e. 2000 and 43 times higher for a food dilution and a translocation-mediated mechanism, respectively.

3.2. ERA of ERMP in Great Lakes sediments

Our risk characterizations for the Laurentian Great Lakes benthic environment are based on realigned exposure data and an HC5 effect threshold developed in the same way as those described above (Redondo-Hasselerharm et al., 2023). The analysis yields an ERA representative of the ecosystem scale, which suggests negligible risks for benthic organisms across the Laurentian Great Lakes (Fig. 2). In this instance the mean RCR are <1 for the sediments in all systems studied (Fig. 2, Table S32). The mean RCR represents the most probable value from the distribution and implies that the mean is most likely. However, system-level heterogeneity in ERMP concentrations due to hydrological dynamics has been considered. This means that the exposure can be arbitrarily higher or lower both locally or transiently, than the most probable value. The variability is illustrated by the distributions (Fig. 2). Additionally, each of the calculations is inherently uncertain due to uncertainties in the input parameters, further broadening the distribution. Thus, while the mean RCR represents the most likely outcome at the ecosystem level, the distribution represents the probability of risk occurring at an unspecified random location in the system.

For the systems studied here, the distribution of RCR values has a range of seven to eight orders of magnitude (Table S32: Min – Max range). Tails of the distributions of RCR at the 95% percentile are greater than 1 for Lake Erie and Lake Huron at 13.7 and 14.6% (Table S32). This means that there is a probability that risks are likely to occur for some parts of the lake sediment benthic community, even if the mean RCR is < 1. Whereas, for the majority of the lake sediment areas, we also note negligible risk.

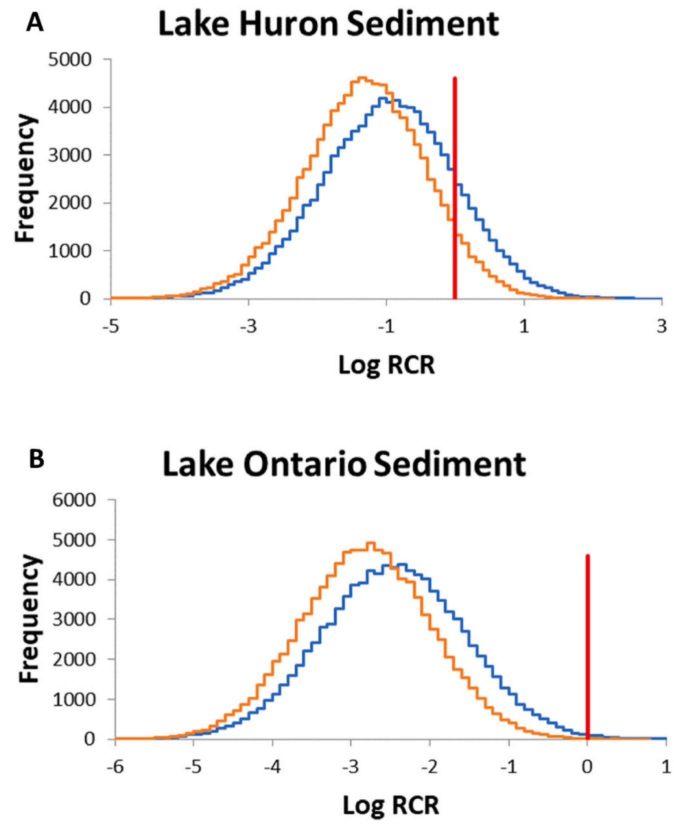

Fig. 2. Log RCR distribution for microplastic exposure from Lake Huron sediment (A) and Lako Ontario sediment (B), based on particle volume as the food dilution effect metric (blue curve)− and on particle area as the translocation-mediated effect metric (orange curve)−. The vertical red line (−)represents the value log RCR = 0, or RCR = 1, which separates the part of the distribution where risk would apply (RCR>1) from the part where risk would not apply (RCR<1). Note that the vertical axis (relative probability) is arbitrary. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

While the mean RCR represents an estimate of the likelihood of risk occurring across the system, guidance towards applying a precautionary approach can be established by considering the 95%, 99%, or the most exact measure: e.g. the %RCR>1 (Table S32). The differences in the RCR metrics can thus be used following a tiered-approach that introduces increasingly more protective measures aimed at helping to inform a risk management strategy.

3.3. ERA of ERMP in Great Lakes surface waters

When considering the RCR distributions calculated for the surface waters of the Laurentian Great Lakes based on the new SSDs, a propagated variability and uncertainty spanning up to 14 orders of magnitude is observed for risks due to food dilution and 8 orders of magnitude for translocation-based toxicity (Fig. 3, Table S33: Min – Max ranges). The distributions are wider as compared to the RCRs estimated for sediment (Table S32), largely influenced by higher variability in ERMP concentrations (i.e. MECs), higher uncertainty in the HC5 values, and the influence of the additional uncertainty in trawling depth for surface water data obtained from trawl samples.

As for sediments, the probability of risk is estimated based on whether risks occur at the ecosystem scale using the mean RCR.

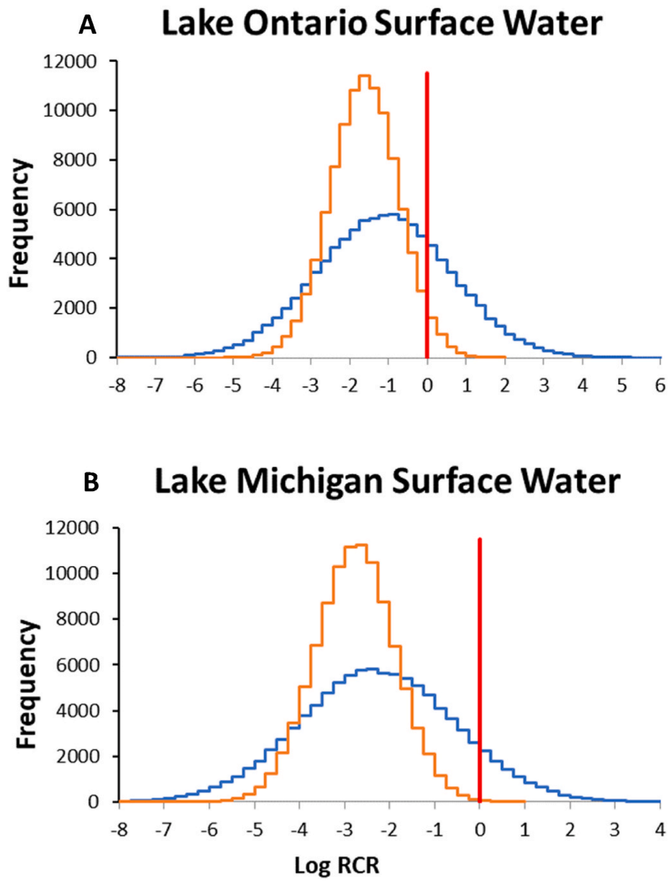

Fig. 3. Log RCR distribution for microplastic exposure from Lake Ontario surface water (A) and Lake Michigan surface water (B), based on particle volume as the food dilution effect metric (blue curve)and on particle area as the translocation-mediated effect metric (orange curve)–. The vertical red line (–) represents the value log RCR = 0, or RCR = 1, which separates the part of the distribution where risk would apply (RCR>1) from the part where risk would not apply (RCR<1). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Consistent with our conclusion for benthic systems across the ecosystem, we suggest the risk to pelagic organisms to be negligible when considering the entire Laurentian Great Lakes system scale. This is because the mean RCR values for the surface waters in all the systems studied are ≪ 1 (Table S33). If we look at the 95% and 99% percentile tails of the RCR distributions derived from SSDs for ingesting species only, then the risks to pelagic species for specific locations of the lake areas are also observed to be negligible. This is because at these percentiles the RCR values are also ≪ 1 (log RCR < zero) and/or from the fact that the percentages of the distributions with RCR>1 (%RCR>1 values) are zero, for all lakes (see FD^i and TM^i values, Table S33). However, for the scenario where all aquatic species data are used for the ERA (FD and TM values, Table S33), a limited partial risk is observed, based on a food dilution mechanism with RCR values of 1.72 (Lake Michigan) to 2.91 (Lake Ontario) at the 99% percentile of the distribution. Percentages of the distribution exceeding the risk threshold (%RCR>1) are very similar for Lake Michigan, Lake Huron, Lake Superior and Lake Erie (8.3%–10.3%). Only for Lake Ontario, is this percentage two times higher (23.6%). For risk based on translocation-mediated effects, risks are negligible (99% percentile values < 0; %RCR>1 values between 0.04 and 0.14), except for Lake Ontario where a limited partial risk is indicated (99% percentile = 0.41, %RCR>1 is 2.49%).

4. General discussion

4.1. General merits of the study

A conceptually novel element of our study is that risk is assessed at spatial scales relevant to aquatic communities, rather than at the scale of individual samples. While probabilistic techniques have been used previously for other stressors, including MPs (Koelmans et al., 2014; Mehinto et al., 2022; Coffin et al., 2022), we are not aware of previous ERMP assessments modelling the fully aligned RCR probabilistically. An advantage of the probabilistic approach over discrete modelling is that the distribution of uncertainty is quantified, which meets the call for more transparent and explicit communication of uncertainties to risk managers and policymakers (SAPEA, 2019). Whether or not risks occur is not a binary question, and thus requires approaches that can quantitatively express the probability of risk. Given the diversity of MPs as an environmental stressor, and the variability and uncertainty with regard to how environmental processes influence the environmental fate and behaviour of MPs, we argue that probabilistic approaches have so far been underutilized towards assessing the risks of MPs.

4.2. Findings for the Great Lakes

Application of the ERA framework indicates that lake-wide risks from ERMP are not expected for the Laurentian Great Lakes, neither in the water column nor in benthic habitats. However, exposure concentrations are highly variable in space and time and we show that these exposure concentrations can exceed effect thresholds, both in the water column and in the sediments, although it is not possible to indicate exactly when and where. After all, as mentioned earlier, the available data are limited to snapshots in space and time. In order to improve the overall assessment of MP concentrations across the Laurentian Great Lakes will require either more spatial and iterative data in space and time, a key requirement to enable a statistically significant evaluation of temporal and spatial trends in exposure (Talbot and Chang, 2022), or validated hydrodynamic models to explicitly interpolate spatiotemporal trends (Hawley et al., 2009; Daily and Hoffman, 2020; 2022). However, the fraction of exposures exceeding the effect thresholds has now been quantified and that fraction is expected to be about 10–20% for exposure to water and about 0–20% for exposure to sediment, based on the most conservative criteria used here. This means that there is a high likelihood that ERMP will result in negative impacts on aquatic communities in the Laurentian Great Lakes if innovative measures aimed at reducing the release and formation of MPs are not implemented (SAPEA, 2019). Based on the information presented in this study, the rank of risks in the water column are in the following order: Lake Ontario > Lake Erie > Lake Superior > Lake Huron > Lake Michigan, which differs from the order for sediments, where Lake Huron had the highest and Lake Ontario had one of the lowest risk profiles. We note that there is no reason to necessarily find a correlation between the water and the sediment, largely due to the influence of the low number of datapoints, complex sedimentation behaviour of the particles due to biofouling, and differences in hydrodynamics between each of the lakes (Daily and Hoffman, 2020; 2022).

4.3. Comparison with an assessment based on the Mehinto thresholds

Effect thresholds have recently been developed based on SSDs for marine habitats (Mehinto et al., 2022). In their SSD, the majority of datapoints used by Mehinto et al. (2022) was marine, and the most sensitive species that influenced derivation of the HC5 were marine species (marine bivalve *Pinctada margaritifera* and estuarine fish *Oryzias melastigma*). Mehinto et al. (2022) performed a sensitivity analysis concerning the influence of these species data on the HC5, which resulted in a shift of the HC5 value of up to 300% when the sensitive species were left out. Overall the alignments used were based on parameters for a marine environment, consequently, the SSD developed represents a marine SSD, which would not be fit for purpose when attempting to inform risk management and policy for the Laurentian Great Lakes area. Unless better data is not available, we would not recommend that management decisions for freshwater lakes such as the Laurentian Great Lakes be based on impact thresholds largely derived from marine species data. Intuitively, if there is an obvious mismatch in the parameters used in deriving an SSD, such as differences between marine and freshwater systems, the exposure data alignments applied as part of this and other studies would no longer be consistent with those for enabling a fit-for-purpose ERA.

For illustrative purposes, however, we include here an example of the challenges and limitations associated with an assessment that might be based on the thresholds reported by Mehinto et al. (2022), (i.e. a marine SSD) and applied to the Laurentian Great Lakes. For comparison and for users who may wish to apply the thresholds derived by Mehinto et al. please refer to Table S34. Using the data reported in Table S34, Threshold 1 (Investigative monitoring) would be exceeded across the Laurentian Great Lakes, based on a food dilution effect mechanism, while for a translocation-based effect mechanism exceedances are observed only in the instance of Lake Ontario, with a partial spatio-temporal exceedance of 28.6%. Threshold 2 (Discharge monitoring) would also be exceeded based on a food dilution effect mechanism with average RCRs for all lakes and for 47–95% of the distributions. Again, for a translocation-based effect mechanism this is only the case for Lake Ontario, but only for 5% of the distribution. Exceedance of Threshold 3 (Management planning) for food dilution is only observed for Lake Ontario (average RCR = 8.5), however, partial spatiotemporal exceedances of 36–88% are possible among the lakes. For a translocation-based effect mechanism, a marginal risk is observed for Lake Ontario, at the 99th percentile of the distribution. Threshold 4 would also only be marginally exceeded in Lake Ontario for a food dilution mechanism (average RCR = 1.3) with partial spatiotemporal exceedances of 5–53% among lakes. In summary, applying the Mehinto et al. (2022) risk management framework would indicate Investigative and/or Discharge monitoring for all lakes, and management planning for Lake Ontario, whereas the results for the other lakes are only applicable to specific locations of the lake areas requiring management action.

Threshold 3, as reported by Mehinto et al. (2022) was based on an SSD with organism- and population-level endpoints and a median data collapse method, and was set at the median HC5. The implication is that this is the threshold that, in terms of the method by which it is derived, best matches our new thresholds specifically designed for the Laurentian Great Lakes. According to Mehinto et al. (2022), Threshold 3 for food dilution was derived as 5 particles/L, which is 100 times lower than the new HC5 value we obtained for freshwater species at 547 particles/L, and more than 5 orders of magnitude lower than the $HC5^i$ that we obtained by considering only the species that can ingest particles ($HC5^i = 1.19 \times 10^6$). Similarly, these numbers for translocation-mediated toxicity are 890, 1690, and 7.26×10^4 particles/L, also implying that our current novel ecological impact thresholds are much higher than those of Mehinto et al. (2022). In contrast, if the same logic of the Mehinto et al. (2022) management framework would be applied to the Laurentian Great Lakes, while the exposure data and the SSDs specifically applicable to freshwater would be used, 'management planning' would not be indicated in any of the lakes.

The considerable differences in these numbers help to illustrate the sensitivity of the input data used to derive the threshold values. The ERA framework based on the use of QA/QC screening and data alignment to obtain consistent risk characterizations can, as such, be considered robust, as was also assessed in a recent expert elicitation procedure (Mehinto et al., 2022). However, in the Mehinto et al. (2022) risk management framework, crucial QA/QC criteria were omitted, because otherwise an insufficient number of data would be available to derive the thresholds. The inclusion of potentially unreliable data used by Mehinto et al. (2022)was thus meant to be illustrative towards how threshold values might be obtained, and should thus not be used as discrete values to be applied to other systems without full consideration of the associated implications. Recognizing the issues associated with the reliability of the data also helps to explain the relatively low level of confidence in the actual thresholds derived through the elicitation procedure reported by Mehinto et al. (2022), while also emphasizing that the current thresholds should be viewed as snapshots representing the available knowledge and intended purpose at the time of their derivation. New data is continuously being generated, and which is increasingly observed to be compliant with recommended QA/QC standards and experimental protocols. Impact thresholds used for management decisions must therefore consider the implications of using outdated and/or sub-optimal data, which can result in erroneous conclusions. The illustrative exercise presented here aims at demonstrating how this can occur while also stressing the importance of using the best-available data and approaches. Ultimately, the decision regarding the application, interpretation, and acceptability of data generated from an ERA framework based on the development of SSDs is determined by risk managers or policymakers. The purpose of this exercise thus aims to better inform the decision-making process.

4.4. Limitations and perspectives

Several limitations exist regarding QA/QC, which are important when weighing the certainty of the results. While it does not render the data useless, of the 28 datasets considered in this study (Table S4-Table S31), only two received a non-zero score for all criteria (Table S9, Table S25). Furthermore, propagation of uncertainty associated with environmental sampling, analytical and alignment methods, the temporary nature of input data sets, and habitat compatibility can be identified and are well understood. There are, however, a number of important points for discussion. For instance, the alignments used here assume a log-linear extrapolation down to 1 μm, while the actual measurements often go no lower than 20, 100 or often even 300 μm. In empirical data, we often see a deflection at small particle sizes, as a result of which linear extrapolation leads to an overestimation of particle numbers. While this can affect the accuracy of exposure assessment, for example, it is good news that the RCR is not sensitive to this artifact, because the bias in extrapolating the RCR numerator and denominator effectively cancel each other. Nevertheless, better size distributions, that extend to the submicron scale, would greatly increase the quality of the ERA framework. Another issue is that the hypothesized effect mechanisms, such as food dilution and translocation, are based on generally accepted empirical data, but have not been specifically validated for the effect data used in the SSDs. Only the original study using SSDs for food dilution included a criterion that any effect study must have observed ingestion and name food dilution as the most plausible effect mechanism (Koelmans et al., 2020). It is therefore possible that the effects on which the SSDs are based, are actually based on different mechanisms of action than those for which the effect metrics were calculated. The SSDs derived are thus conditional, implying that the results of the assessment are tentative with respect to the assumed effect mechanisms.

In an effort to help reduce the uncertainties identified, we recommend that concrete steps to improve the ERA framework require the characterization and quantification of a wider ERMP size range (ideally 1–5000 μm, and as yet understudied factors such as biofilms, see Amariei et al., 2022) to validate PDFs, to use a similarly wide range of sizes and other ERMP characteristics in toxicity effect tests, aimed at minimizing the need for performing alignments (Redondo-Hasselerharm et al., 2023), and to further validate assumptions with respect to ecologically relevant dose metrics. Finally, we emphasize the need to develop the knowledge and technology to determine which of the concepts used here are useful to estimate exposure, effect, and risk characterization for nanoplastic particles (SAPEA, 2019).

5. Conclusion

In this study, we take the next step in the development of our ERA framework for ERMP in aquatic systems, by probabilistically modelling different sources of uncertainty, variability and diversity and applying the improved framework to the Laurentian Great Lakes. Lake-wide risks from ERMP are not expected in the water column or benthic habitats. However, the fraction of exposures exceeding effect thresholds is expected to be about 10–20% for water exposure and about 0–20% for sediment exposure. The implication is that ERMPs are likely to represent unacceptable risks on aquatic communities where concentration thresholds are exceeded for impacted regions in the Laurentian Great Lakes if innovative measures are not taken to reduce the release and formation of MPs.

Authorship statement

Albert Koelmans: conceptualization, methodology, software, formal analysis, investigation, writing-original draft, visualization, supervision. **Paula Redondo-Hasselerharm**: formal analysis, investigation, visualization, writing-review & editing. **Nur Hazimah Mohamed Nor**: investigation, writing-review & editing. **Todd Gouin**: investigation, writing-review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.121445.

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