

Source-Specific Probabilistic Risk Assessment of Microplastics in Soils applying Quality Criteria and Data Alignment Methods

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Research Paper

Source-specific probabilistic risk assessment of microplastics in soils applying quality criteria and data alignment methods

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Literature search

Exposure assessment

Measured environmenta

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concentrations (MECs)

- • · ·

Effect assessment No observed effect

oncentrations (NOECs)

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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

QA/QC screeni

Sampling reporting

X Clean air conditions

Positive controls Polymer identification

X Chemical purity

X Lab preparation

Quality of doseresponse relationship

Addition of food

M Endpoints

X Sample size

×

M

Study 2

Study 3

Effect thresholds

- First fully aligned ecological risk assessment for microplastics (MP) in soils.
- Alignment of exposure and effect data for particle properties and bioaccessibility.
- Risks assessed probabilistically, accounting for uncertainties in data alignments.
- Quality criteria tools used to evaluate reliability of exposure and effect data.
- Risks found for soils with mulching or sewage use, and with background MP pollution.

ARTICLE INFO

Keywords: Mulching Compost Sewage Species Sensitivity Distributions Quality assurance

ABSTRACT

The risk characterization of microplastics (MP) in soil is challenging due to the non-alignment of existing exposure and effect data. Therefore, we applied data alignment methods to assess the risks of MP in soils subject to different sources of MP pollution. Our findings reveal variations in MP characteristics among sources, emphasizing the need for source-specific alignments. To assess the reliability of the data, we applied Quality Assurance/Quality Control (QA/QC) screening tools. Risk assessment was carried out probabilistically, considering uncertainties in data alignments and effect thresholds. The Hazardous Concentrations for 5% (HC5) of the species were significantly higher compared to earlier studies and ranged between 4.0×10^7 and 2.3×10^8 particles (1–5000 µm)/kg of dry soil for different MP sources and ecologically relevant metrics. The highest risk was calculated for soils with MP entering *via* diffuse and unspecified local sources, *i.e.*, "background pollution".

Probabilistic Risk Assessme

2 3

Risk Characterization Ratio

RISK: SEWAGE > BACKGROUND MF

POLLUTION > MULCHING >> COMPOST

No Risk

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Received 7 November 2023; Received in revised form 24 January 2024; Accepted 4 February 2024 Available online 6 February 2024 0304-3894/© 2024 Elsevier B.V. All rights reserved. However, the source with the highest proportion of high-risk values was sewage, followed by background pollution and mulching. Notably, locations exceeding the risk threshold obtained low scores in the QA/QC assessment. No risks were observed for soils with compost. To improve future risk assessments, we advise to primarily test environmentally relevant MP mixtures and adhere to strict quality criteria.

1. Introduction

Plastic materials are extensively used in agriculture due to their versatility, durability, and low production costs. Mulching films and greenhouses are used to improve crop yield and quality, while drip irrigation pipes ensure efficient crop watering, and polymer-coated fertilizers gradually release nutrients [1-4]. Both agricultural fossil and bio-based plastics can degrade and fragment into smaller plastic items over time, contributing to macro- (>5 mm), micro- (MP, 1 µm-5 mm) and nanoplastic ($<1 \mu m$) accumulation in soils [5]. In addition to plastics intentionally used in agriculture, soils also face unintentional exposure to plastic through organic fertilizers, irrigation water, littering and atmospheric deposition [6,7]. Commonly applied organic fertilizers, such as sewage sludge or compost, can contain large quantities of MP [8–11]. Furthermore, atmospheric deposition significantly contributes to the presence of MP in soils, especially in urban and industrial areas [12,13]. As a result, intentional and unintentional plastic sources have resulted in the widespread presence of MP in soils [7]. Estimates suggest that 14% of the total plastic released into the environment can end up in agricultural soils [14]. Actual concentrations of MPs measured in worldwide soils from farmlands with mulching, compost application, sewage use, and background MP pollution (i.e., MPs entering via diffuse and unspecified local sources) range up to 3.2×10^5 , 2.7×10^3 , $1.7 \times$ 10^5 , and 6.2×10^5 particles/kg dry soil, respectively [15–18]. Additionally, the concentrations and properties of MP (e.g., size, shape, and polymer type) are likely dependent on the source of the plastic [16,19]. Even though agricultural soils have been recognized as major sinks for MP due to the aforementioned practices [20], elevated levels of MP have also been found in soils across different land uses, due to unintentional plastic sources [18,21,22]. In grasslands, forests, and mangroves, MP levels quantified in soils range up to 2.0×10^5 , 6.9×10^5 , and 2.9×10^4 particles/kg dry soil, respectively [18,23].

Given these estimations and the high MP concentrations detected in soils worldwide, there is growing concern about whether MP could pose a threat to soil biota. MP can directly impact soil physical and chemical properties, bacterial communities, soil fauna and plants [24–31], but can also lead to indirect effects on soil properties and biota [26,32,33]. In agricultural soils, MP can be ingested by farm animals, potentially raising concerns for human health [34]. The concentration, size, shape, and polymer type of MP are key factors driving their effects in soils [27, 35,36]. Studies have demonstrated that MP up to 5 μ m can be absorbed by plant roots, leaves, epidermis, and seeds [37,38], and several studies have shown that soil invertebrates can take up MP [31,39,40]. The size and shape of the MP determine their species-specific bioaccessibility, defined as the fraction of the total amount present that is, or can be, taken up from the environment and can cause a biological response [41, 42].

To date, several studies have assessed the risks of MP for soil biota based on available literature data [43–48]. However, the measured concentrations of MP in the soils used are virtually incomparable with each other, as they target different size ranges due to the various sampling techniques and analytical methods used [49]. Similarly, the toxicity data used pertain to MP with different particle characteristics. Hence, the exposure and effect data lack alignment, leading to unreliable risk estimations [50]. Furthermore, these earlier assessments did not explicitly account for the bioaccessibility of MP and the underlying mechanisms behind the observed effects [51–53]. Recently, a method was developed to align MP exposure and effect data, making them comparable by considering differences in their particle properties while factoring in the bioaccessibility of MP [54]. This method has already been applied in risk assessments conducted for MP in sediments and surface waters [50,55–57]. However, a fully aligned risk assessment for soils is not yet available.

Another aspect that was not considered in previous risk assessments for MP in soils is the quality of the input data used. Due to the absence of guidelines and methodological harmonization for quantifying MP in environmental samples and conducting toxicity tests with MP, the quality of available studies and their usability in risk assessment has been questioned [58]. Consequently, Quality Assurance and Quality Control (QA/QC) criteria have been developed to assess the quality of the available literature studies and to serve as a guidance protocol on good practices for future studies measuring MP concentrations in biota [59], water [60], sediment [50], and soil [49,61], as well as effects of MP on biota [62]. OA/OC tools facilitate the identification of data that may not be suitable for environmental risk assessment (ERA), as they could otherwise lead to biased results. Therefore, it is useful to screen the quality of studies reporting input data in the context of risk assessment. As of now, no QA/QC tool has been applied to assess the quality of studies reporting MP toxicity data for soil organisms.

The aim of the present work is to assess the risks of MP in soils using a probabilistic approach. This involves the application of data alignment methods for soils subjected to different sources of MP pollution, along with an evaluation of the existing data through a data quality screening method. An extensive literature search was conducted to obtain measured concentrations of MP in soils with different land uses, as well as toxicity data for soil invertebrates and plants exposed to MP. Study characteristics were extracted and summarized, and the toxicity data relevant to the purpose of this study were selected for use in the risk assessment. The quality of studies reporting exposure and effect data for MP in soils was assessed [50,62], and exposure and effect data were aligned across the entire MP size range following established methods [50,54]. A probabilistic risk characterization was performed, taking into account uncertainties in the data alignment calculations, the diversity of MP, and the effect thresholds derived from fitting Species Sensitivity Distributions (SSDs). Risk Characterization Ratios (RCRs) were calculated by dividing each rescaled Measured Environmental Concentration (MEC) by the Hazardous Concentrations for 5% (HC5) of the species obtained for the food-dilution effect mechanism, with particle volume as the ecologically relevant metric (ERM), as well as by the HC5 value for translocation-mediated effects, with surface area as the ERM [50,51,63, 64]. This analysis was performed in a source-specific manner, which means it was conducted separately for different categories of soils: those with mulching, compost-treated soils, soils treated with sewage in the form of biosolids or sludge, and 'background MP soils'. Background MP soils typically have MP levels attributed to diffuse sources such as atmospheric deposition, transport or run-off from adjacent land areas, and other local anthropogenic sources, such as littering.

2. Methods

2.1. Data collection

An extensive literature search was conducted until April 2023 using the Web of Science (WOS) and ProQuest databases to obtain measured exposure concentrations (MECs) of MP in soils and toxicity data for soil invertebrates and plants exposed to MP through soil. To collect the MECs, the following search strings were used: (concentrations OR levels OR occurrence) AND (microplastic OR plastic particle OR polymer OR bioplastic OR biopolymer OR film OR fragment OR polyethylene) AND (mulching OR greenhouse OR compost OR manure OR waste OR biosolid OR sewage sludge OR wastewater OR dust OR atmospheric) AND (soil OR terrestrial OR land OR farmland OR agricultural OR urban OR industrial OR park OR grassland OR forest OR mangrove). To obtain the toxicity data, the following search strings were used: (effect OR impact OR toxicity OR exposure) AND (microplastic OR plastic particle OR polymer OR bioplastic OR biopolymer OR film OR fragment OR polyethylene) AND (soil OR terrestrial) AND (species OR organism OR animal OR plant OR crop OR invertebrate OR insect OR worm). Studies assessing the effects of MP on microorganisms were not considered because the effect mechanisms that we include in this study do not apply to this group.

Every article found in this search was read to obtain MECs in particles/kg dry soil and toxicity data for soil invertebrates and plants complying with six criteria, which are: 1) use of soil in the tests; 2) reporting of the effect threshold concentrations per mass of soil; 3) chronic exposure duration; 4) evaluation of endpoints that target effects at the individual level (e.g., survival, growth, reproduction); 5) detection of significant concentration-dependent adverse effects (i.e., excluding positive or non-concentration dependant effects) or the highest no observed effect concentration (HNOEC) only when at least 4 concentrations were included in the tests; 6) reporting of the size of the MP used in the tests. Ideally, according to criterion #6 in De Ruijter et al. (2020) and as applied in Redondo-Hasselerharm et al. (2023) [50,62], data from tested MP that have chemicals interfering with particle effects (i.e., related to the volume and surface area of the MP) should be excluded. However, this approach was not implemented in the present study due to the lack of data. We only omitted data when authors demonstrated that the effect was entirely caused by plastic-associated chemicals.

A total of 241 MECs of MP in soils in particles/kg dry soil were obtained from 51 studies. Their characteristics are summarized in the Supporting Information (SI; Table S1), which include land use, potential plastic source(s), country and continent where the sampled soil was located, method used for the identification of MP, depth of the soil layer sampled, number of samples analysed per datapoint, size range of the MP measured, most abundant polymers and shapes found, and minimum, mean and maximum concentrations of MP reported in particles/ kg dry soil. The characteristics of the remaining 10 studies from which the MECs of MP could not be obtained in particles/kg dry soil are listed in Table S2. A total of 50 effect threshold concentrations were recorded from 27 studies complying with the six listed criteria. Their study characteristics are summarized in Table S3, which include species name, taxonomic class, polymer type, shape, and size of the MP tested, exposure duration, number of concentrations used in the tests, ecologically relevant endpoints studied and affected, and concentration descriptor provided by the study. The characteristics of the 41 studies that did not comply with the listed criteria are shown in Table S4.

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

The quality of the selected exposure and effect studies was quantitatively evaluated using previously published Quality Assurance and Quality Control (QA/QC) screening methods [50,62]. To assess the quality of studies reporting MECs of MP in soils, the QA/QC screening tool for sediment samples developed by Redondo-Hasselerharm *et al.* (2023) [50] was revised and deemed applicable for soils. This QA/QC screening tool is composed of ten criteria included in four categories: 1) sampling; 2) contamination mitigation in the laboratory; 3) sample purification and handling; and 4) polymer analysis [50]. After revising the 10 criteria, only criterion #2 "sample size" was modified following the reasoning given in Redondo-Hasselerharm *et al.* (2023) [50] (Table 1).

The quality of the studies reporting effect thresholds for terrestrial species was assessed using the QA/QC screening tool for aquatic species developed by de Ruijter *et al.* (2020) [62], which was also used to assess

the quality of studies reporting toxicity data for freshwater benthic species exposed to MP via the sediment [50]. The same twenty criteria defined in De Ruijter et al. (2020) [62] were deemed applicable to data from soil toxicity tests, which are included in four categories: 1) particle characterization; 2) experimental design; 3) applicability for risk assessment; and 4) ecological relevance. In both QA/QC screening tools, a score of 2 (adequate), 1 (adequate with restrictions), or 0 (inadequate) was given per criterion, and a 'Total Accumulated Score' (TAS) was calculated by adding up the scores for each criterion, with a maximum TAS of 20 and 40 points for studies reporting MECs and toxicity data, respectively. The motivations for the scoring of each criterion within both QA/QC screening tools are given in the SI of Redondo-Hasselerharm et al. (2023) [50].

2.3. Data alignments and construction of species sensitivity distributions for relevant dose metrics

2.3.1. Alignment of microplastic exposure concentrations

From the available datasets we retrieved the mean, minimum and maximum MP number concentrations in soil (particles/kg dry soil). These MECs cannot be compared directly between studies because they targeted different particle size ranges [54]. Across different studies, minimum sizes ranged from 8 to 1000 μ m, while maximum sizes ranged from 500 to 5000 μ m (Table S1). For the same reason, these MECs cannot directly be compared to the reported toxicity data for soil organisms to characterize the environmental risks of MP. To allow for consistent comparisons of exposure concentration data, all MEC data were rescaled to a standard MP size range from 1 to 5000 μ m by multiplying them by a correction factor (CF) [54]:

$$CF = \frac{5000^{1-a} - 1^{1-a}}{x_2^{1-a} - x_1^{1-a}} \tag{1}$$

Here, x_1 and x_2 are the minimum and maximum values of the targeted size range in the soil studies (µm). Alpha (α) is the exponent of the MP power law size distribution $y = bx^{-\alpha}$ (or the power law slope in the log transformed equation log $y = -\alpha \log x + \log b$), in which y and x are the relative abundance and size (*i.e.*, length of the longest axis), respectively.

The studied soils were classified based on their land use and the potential plastic sources contributing to MP accumulation (Table S1). The land uses considered encompassed farmlands, grasslands, forests, wetlands, mangroves, saltmarshes, and a desert. The MP sources included in the risk characterization were the use of mulching films, the application of compost as organic fertilizer, the application of biosolids or sewage sludge as organic fertilizer, and soils with background MP contamination. We investigated whether the particle properties and the uncertainty parameters that describe these distributions differ among the different soil treatment types. For this, we used the raw data by Huerta-Lwanga et al. (2023) [19] to calibrate the alpha value for MP in mulching- and compost-treated soils, and the raw data by Crossman et al. (2020) for biosolid application [10]. As both studies included data from soils without the evaluated treatments (i.e., control soils), both datasets were also used to obtain the alpha value for background soils. The raw data consisted of two-dimensional length and width data, which were used to estimate particle volume and surface area based on an assumed ellipsoid shape category, and the Simon model [64,65]. The alpha values for particle size (length; mean \pm standard deviation), obtained by fitting the size distribution power law to the data for soils with mulching, compost, and sewage, and background soils were 3.36 \pm 0.07 (number of particles, n = 5617), 3.54 ± 0.22 (n = 603), 3.38 ± 0.50 (n = 729) and 3.05 \pm 0.04 (n = 2498) (Figs. S1-S4; Table S5). Due to the difference in these values, we decided to calculate the alignments using MP source-specific parameters. The rescaling calculations mentioned were performed in a probabilistic manner, considering the uncertainty in the power law slopes. The details of these calculations are further explained

Table 1

Criteria followed in the quantitative evaluation of the quality of papers reporting measured environmental concentrations of microplastics in soils, adapted from Redondo-Hasselerharm *et al.* (2023) [50].

			2	1	0
Sampling	1	Sampling reporting	Detailed reporting of sampling, including date, location (with coordinates or map), method used area and depth of sampling	Date, location, and method are given, but coordinates, map and or info on area and denth are missing	No/insufficient reportage of sampling information.
	2	Sample size ^a	At least 250 g dry weight, if the lower particle size detection limit is $100 \mu\text{m}$ (or smaller). At least 25 g dry weight if the lower particle size detection limit is $20 \mu\text{m}$ (or smaller).	At least 250 g dry weight if the lower particle size detection limit is between 100 and 300 μ m. At least 25 g dry weight if the lower particle size detection limit is 100 μ m (or smaller). At least 5 g dry weight if the lower particle size detection limit is 20 μ m (or	Less than 250 g dry weight is used while the lower particle size detection limit is 300 μ m or higher. Sample size is not reported or is only reported as wet weight.
	3	In-site variability representation	3 or more independent samples collected at the same time at all sites. Samples can be analyzed separately or pooled together.	smaller). 2 independent samples collected at the same time at all sites. Samples can be analyzed separately or pooled together.	1 sample per site or 2 or more samples collected in part of the sites.
	4	Sample processing and storage	Samples are preserved after sampling (<i>i. e.</i> , closed and refrigerated within one day after sampling). Use of non-plastic containers previously rinsed with filtered water or use of aluminum foil. Plastic materials are avoided. Sample processing was done in the laboratory.	Samples are not properly preserved (<i>i.e.</i> , not closed or refrigerated within one day after sampling). Use of plastic containers and/or materials but rinsed with filtered water or covered with aluminum foil. Samples are handled outside but negative controls are placed in the field. Sample storage mentioned but not how samples were preserved.	Sample preservation and storage not mentioned, or information is incomplete (<i>i.e.</i> , material of container not mentioned). Samples handled in the field without controls.
Contamination mitigation in the laboratory	5	Laboratory preparation	Use of cotton lab coat or non-synthetic clothes. Equipment and lab surfaces wiped and rinsed with filtered water or ethanol.	Solely wiping laboratory surfaces and equipment and not wearing a cotton lab coat or wiping laboratory surfaces and equipment with unfiltered distilled water or ethanol IF negative controls were run in parallel and examined for contamination.	No wiping of equipment and lab surfaces and not wearing cotton lab coats and no use of negative controls. Equipment wrapped in plastic.
	6	Clean air conditions	Clean air room or laminar flow cabinet used.	Mitigation of airborne contamination by carefully keeping samples closed as much as possible IF negative samples were run in parallel and examined for occurring contamination	No regard of airborne contamination or use of fume hood without keeping samples closed.
	7	Negative control	Controls (in triplicate, at least) treated and analyzed in parallel to actual samples. Sample concentrations need to be reported accounting for controls per polymer type.	Insufficient form of a control, <i>e.g.</i> , the filtration of air, or the sole examination of petri dishes/soaked papers placed next to the samples OR if less than 3 replicates are used or the number of replicates is not mentioned OR if reported sample concentrations do not account for controls per polymer type.	No negative controls.
Sample purification/ handling	8	Positive control	Controls (in triplicate) with an added amount of a heterogeneous mix of microplastic particles treated alongside the samples, and for which the particle recovery rates are determined.	Insufficient form of a positive control (<i>e.</i> <i>g.</i> , only a part of the protocol is tested, less than 3 replicates are used, or the number of replicates is unknown, or less than 3 size classes are used).	No positive controls.
	9	Sample treatment	Density separation of sample carried out with pre-filtered solutions of ZnCl ₂ , NaBr, NaI, ZnBr ₂ , CaCl ₂ , or similar (with density > 1.4 g/cm ³). Digestion with KOH or H ₂ O ₂ 30% and/or enzymes. If another chemical was used, effects on different polymers should be tested before application. For samples that are clean from organic debris, no digestion is required. Sample treatment is always carried out below 50 °C.	If proof is missing that polymers are not affected by protocol OR in case studies exclusively focus on the bigger microplastics by sieving the samples (mesh size \geq 300 µm) OR in case the authors state no visible debris was found, OR density separation was done with NaCl. Chemicals not filtered. Sample treatment is always carried out below 80 °C.	No digestion of sample unless the sample is clean. Sample treatment is carried out above 80 °C. Density separation not done or done with water.
Polymer analysis	10	Polymer identification	Suspected particles are analyzed with FTIR, Raman or pyrolysis/TED-GC/MS IF numbers of pre-sorted particles are < 100. For particle numbers > 100, 50% should be identified, with a minimum of 100 particles. For filters \geq 25% of the surface area should be analyzed.	Insufficient number of particles identified, OR when a specific size is left out of the polymer identification but is included in the reported concentrations (<i>e.g.</i> , small MP that can't be handled with tweezers) OR when the number of particles in the analyzed subsample is not reported.	No polymer identification with FTIR, Raman or pyrolysis/TED-GC/MS

^a Motivation for the selection of the minimum sample sizes in soil samples provided in the supporting information.

in the risk characterization section (Section 2.4), which provides additional information on how the uncertainty was accounted for in the analysis.

2.3.2. Alignment of laboratory effect threshold concentrations to the environmentally relevant and bioaccessible effective concentration

To convert effect concentrations for mono- or polydisperse particles determined in laboratory toxicity tests to effect concentrations for an environmentally relevant mixture of MP particles with a more realistic degree of polydispersity, a correction was made that takes into account the ERM [51]. For a given ERM 'x', effect threshold concentrations of particles with different degrees of polydispersity can be related to each other, as long as the overall size of the ERM remains the same [54]:

$$EC_{poly}^{env,bio} = \frac{EC_{test} \times \mu_{x,test}}{\mu_{x,poly}^{env}}$$
(2)

Here, EC_{poly}^{emvbio} is the effect concentration for bioaccessible environmentally relevant polydisperse MP, EC_{test} is the effect concentration reported in the laboratory toxicity test, $\mu_{x,test}$ is the average value for ERM 'x' for either the mono- or polydisperse MP used in the laboratory toxicity test, and $\mu_{x,poly}^{emv}$ is the average value for ERM 'x' for the polydisperse MP as they occur in nature. Following earlier studies [50, 55–57], the ERMs of particle volume and surface area were selected. However, we now distinguish the effects caused by particle volume into: 1) MP ingestion by invertebrates, which can lead to a feeling of satiation and a food-dilution effect mechanism [29,31,62,66,67]; and 2) physical blockage in plants, as the accumulation of MP in seeds and roots could lead to a reduction in plant growth due to blocking the absorption and/or uptake of nutrients and water [37,68].

The EC_{test} values used in this study are the No Observed Effect Concentrations (NOECs) and Lowest Observed Effect Concentrations (LOECs) shown in Table S3 [50,55,58]. Following previous risk assessments of MP in aquatic systems [55,69], the LOECs were first converted into NOECs using an assessment factor (AF) of 10. For each species, the geometric mean NOEC was calculated per endpoint, and the most sensitive endpoint was selected as effect threshold concentration for that species.

Then, alignments were applied to account for the bioaccessibility of MP [54]. When the suspected mechanism of effect relied on particle ingestion by soil invertebrates, only MP that could be ingested by a particular organism were considered biologically available. Particles wider than the mouth opening of the organism were deemed too large to be ingested and were excluded from further analysis [54,64]. If the MP used in the test were smaller than the maximum size the test organism could ingest, no correction was necessary. The bioaccessible size fractions of MP for each species were determined based on MP ingestion data, food ingestion data, or mouth opening sizes, as specified in Table S6.

For plant species, a bioaccessibility limit of 5 μ m was applied based on the demonstrated accumulation of 4.8 μ m plastic particles in seed capsule pores, root hairs, leaves and epidermis of *Lepidium sativum* [37] and the accumulation of 5.6 μ m plastic particles in the roots of *Hordeum vulgare* [38]. In cases where toxicity depended on tissue translocation, particles that were deemed too large to be displaced through tissue were considered biologically unavailable [54,64]. Following earlier studies [50,55–58], 83 μ m was used as the maximum particle size for translocation. For organisms with mouth openings smaller than 83 μ m, their mouth opening size was used as the upper limit for bioaccessibility. In cases where the maximum size of the tested particles passed the bioaccessibility criterion, the *EC*_{test} values were corrected using Eq. 1. The numerator of the equation represented the bioaccessible size range, while the denominator represented the exposure size range covered. This correction was necessary for 50% of the studies.

When tests used polydisperse particles, for instance prepared by grinding, a power law dependency of the ERMs was assumed. Because the actual power law slopes for these tested particles are unknown, a probabilistic approach was employed to account for the associated uncertainty. Given that power law slopes typically range between 2 and 3, a normal distribution of power law slopes was assumed, characterized by a mean α of 2.5 and a standard deviation of 0.25.

To obtain estimates of power law slopes for particle surface area and volume, we assessed the ratios between power-law slopes for particle dimensions (*i.e.*, length, surface area, and volume) also with literature values. Interestingly, the ratios between power-law slopes for particle dimensions: $\alpha_L / \alpha_A = 1.48 \pm 0.10$, $\alpha_A / \alpha_V = 1.20 \pm 0.07$ and $\alpha_L / \alpha_V = 1.78 \pm 0.22$ (n = 10) (Table S7), have small relative standard deviations ranging from only 6 to 12%. This indicates that these ratios remain relatively constant across different environmental compartments. In other words, the proportions of particle shapes in environmentally realistic MP mixtures are similar across different compartments, including soils, and if power law slopes α for particle length are known, the slopes for surface area and volume can be estimated using these ratios.

For $\mu_{x,poly}^{env}$, with x = particle volume, or x = particle surface area, power law distributions with power law slopes a_x were calibrated to characteristics of individual MP particles, for the soil treatments mulching, compost, and sewage, and for background soils. With known a_x values for these treatments (Tables S5 and S7), $\mu_{x,poly}^{env}$ was calculated as (in case $\alpha \ddagger \{1,2\}$) [64]:

$$\mu_{x,poly}^{env} = \frac{1 - a_x}{2 - a_x} \times \frac{X_{UL}^{2-a_x} - X_{LL}^{2-a_x}}{X_{UL}^{1-a_x} - X_{LL}^{1-a_x}}$$
(3)

Now that all the variables in Eq. 2 are known, the value of $EC_{poly}^{env,bio}$ can be calculated for each species. This value represents the effect number concentration for environmentally relevant MP found in the soils, but only for the species-specific bioaccessible MP fraction. To compare these concentrations with the exposure number concentrations in a risk characterization, a final species-specific rescaling was performed to calculate the effect concentration in terms of particles ranging from 1 to 5000 µm. This rescaling was done using Eq. 4:

$$EC_{env} = EC_{poly}^{env,bio} \times CF_{bio} \tag{4}$$

The correction factor CF_{bio} was calculated using Eq. 1, with the bioaccessible size range covered by the denominator and the probabilistic power law slope for particle size: $a = 2.5 \pm 0.25$, as mentioned earlier. Despite the considerable uncertainty in the power law slope for the tested particles, CF_{bio} ranged from 1.00 to 1.12 only, indicating that the outcome of the assessment is not highly sensitive to the uncertainty in this parameter. The resulting aligned effect concentrations (EC_{env}) expressed in particles per kilogram dry soil, were used to construct SSDs for further analysis and interpretation.

2.3.3. Species sensitivity distributions for ecologically relevant metrics

Eight SSDs were constructed with the selected rescaled NOECs for both volume and surface area as ERMs, and for each of the four soil MP sources (mulching, sewage, and compost, and background soils). Because of the uncertainties in the rescaled NOECs due to the sourcespecific uncertainties in power law slopes for particle length, surface area, and volume, they were calculated probabilistically with 10^4 iterations. Median NOECs were taken from the resulting distributions and were used for the construction of the SSDs.

The SSDs were constructed using the ssdtools package in Rstudio (version 4.3.1.) [70], which uses maximum likelihood estimation to fit 10 different cumulative distribution functions to the effect threshold concentrations for the soil species. The Anderson-Darling, Kolmogor-ov-Smirnov, and Cramer-von Mises tests, and the Akaike's Information Criterion (Aic), Akaike's Information Criterion corrected for sample size (Aicc) and Bayesian Information Criterion (Bic) were used to evaluate the goodness of fit of all 10 distributions. The 5% Hazardous

Concentration (HC5) and its corresponding 95% confidence limits were calculated for the best fitting distribution using parametric bootstrapping (based on 1000 bootstrap iterations). All graphs were made with ggplot2 in Rstudio (version 4.3.1.) [71].

2.4. Probabilistic risk characterization

The risk characterization was based on the average exposure concentrations reported in the studies used. This is because in cases where maximum values were also reported, these values were still lower than some averages reported in other studies. Consequently, our use of average exposure concentrations did not lead to missing the maximums in the observed exposure concentrations in the overall assessment. The RCR was calculated by dividing each rescaled MEC by the HC5 value obtained for the food-dilution effect mechanism with particle volume as the ERM, and by the HC5 value for translocation-mediated effects, with surface area as the ERM, separately. These effect mechanisms were selected in previous risk assessments for aquatic organisms [50,55–57], and have also been identified for soil organisms [29,31,37,66-68,72]. A calculated RCR for a specific MEC less than 1 indicates that no risk of MP is expected for the species under the specific soil treatment. However, an RCR greater than 1 suggests that the species inhabiting soils under that treatment might be at risk. The RCR were probabilistically modelled using Monte Carlo simulation with 10⁵ iterations. To account for uncertainty in the numerator (i.e., the rescaled MECs), normal distributions were sampled based on measured means and standard deviations of α values (Eq. 1). The HC5 distributions, which were skewed, were sampled using a log-normal distribution. The results are reported as log-risk characterization ratio (logRCR) distributions, which provide a quantitative evaluation of the overall uncertainty in the RCR through error propagation. Metrics used to characterize the RCR distributions include the mean logRCR, the 5-95% percentile range, and the 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 (Predicted No Effect Concentration), i.e., where logRCR> 0 (%RCR>1). Risk of MP was considered absent if the estimated RCR was less than 1 in the 95% percentile of the logRCR distribution. The calculations were performed using Microsoft Excel using a plug-in for Monte Carlo simulations (MCSim version May 4, 2013) [73]. A schematic summary of the methodology followed for the selection and alignment of data included in the risk characterization of MP in soils is shown in Fig. 1.

3. Results and discussion

3.1. Study characteristics and QA/QC evaluation

3.1.1. Characteristics of exposure data

A total of 241 MECs of MP in soils were obtained from 51 studies (Table S1, SI), all of them located in Asia, Europe, and America (Fig. S5). Most of the MECs were reported for soils in China (57%), followed by The Netherlands (16%), Iran (6%) and Denmark (4%) (Fig. S5). Regarding land uses, 52% of the total MECs correspond to soils from farmlands, 36% for grasslands and only 3% for forests (Fig. S6).

When it comes to MP sources associated with agricultural practices in farmlands, 26% of the soils had mulching, while 13% and 9% had sewage and compost applied as fertilizer, respectively (Fig. S6). Among the total number of MECs in farmlands, 37% were associated with fields where no plastic-related agricultural practices had been conducted for a specific duration, i.e., background MP soils, and for which we assume that MP contamination mainly depends on diffuse and unspecified local sources (Fig. S6). Of this 37%, 17% correspond to soils influenced by urban pollution (Fig. S6). In the case of grasslands, only one plasticrelated source was found, namely the use of mulch as dust-proof nets to prevent the release of fine particulate pollution. Therefore, MP exposure data from this field was categorized under "mulching". The remaining MECs of MP in grasslands and other land uses in Table S1 were considered as "background MP soils" in the risk characterization, since the MP concentration in these locations originate from diffuse and local sources.

When considering all MECs compiled for all land uses and plastic sources, fiber was the most often found shape in soils, followed by



Fig. 1. Summary of the methods followed for the selection and alignment of data included in the risk characterization of microplastics (MP) in soils (modified from Redondo-Hasselerharm *et al.*, 2023).

fragment and film (Fig. S7, Table S1). Fibers were the most often detected shape in soils from farmlands with sewage application (Fig. S8) and fragments were the most often found shape in soils with compost (Fig. S9). Fibers and fragments were equally reported in background soils from farmlands (Fig. S10), and fibers, fragments, and films were equally detected in soils from farmlands with mulching (Fig. S11). Polyethylene (PE) and polypropylene (PP) were always the most often detected polymer types in soils from all land uses (Fig. S7) and in soils from farmlands (Figs. S8-S10), except for those with compost application, where PE and polystyrene (PS) were the most commonly found polymer types (Fig. S11).

Scores obtained in the QA/QC evaluation of the 51 studies reporting MECs of MP in soils are shown in Tables S8-S58, in the SI. By displaying the QA/QC scores in this way, we provide transparency regarding the reliability of the exposure data used. As highlighted in earlier QA/QC assessment reports [50,55,59,60,62,74], the scores assigned to each study are not meant to judge the relative merit of a study, as a dataset that has a limited suitability in the context of risk assessment may provide highly valuable information for other purposes. Of the studies evaluated, 78% obtained a Total Accumulate Score (TAS) of 10 or lower, and 22% a TAS above 11 (out of a maximum of 20 points) (Fig. S12). These results are similar to those found by Redondo-Hasselerharm et al. (2023) [50], who assessed the quality of 60 studies reporting MECs of MP in freshwater sediments and found a TAS> 10 in only 20% of the studies, while 80% had a TAS equal or lower to 10 [50]. In contrast to Redondo-Hasselerharm et al. (2023) [50], no study obtained a TAS in the highest range (16 - 20) in the present evaluation (Fig. S12). The highest TAS corresponds to Crossman et al. (2020) (TAS=15, Table S30, SI) [10] and Feng et al. (2020) (TAS=14, Table S21) [75]. From all studies assessed, only Zhang et al. (2022) [16] obtained non-zero values in all criteria (Table S32). The average TAS was 8.4 (range 3 - 15) and the average scores per criterion ranged from 0.31 to 1.82 (Fig. S13). Only for two criteria, "in-site variability representation" (1.82) and "polymer identification" (1.24), the average score was > 1 (Fig. S13). The lowest average scores were found for the criteria "positive controls" (0.37) and "clean air conditions" (0.31) (Fig. S13). These outcomes are in agreement with previous studies reporting MP analysis in drinking water and surface water [60], sediments [50], and soils [61], where the lowest average scores were also found for "clean air conditions" and "positive controls", and where almost no study obtained non-zero scores in all criteria. This shows that the quality limitations in MP analytical methods are independent of the matrix evaluated, and that the conclusions obtained here are consistent among different QA/QC assessments.

3.1.2. Effect data for soil organisms

A total of 50 effect threshold concentrations for 21 soil organisms (9 invertebrates and 12 plants) were obtained from 27 studies complying with the six criteria previously described (Table S3). Fragments, irregular particles, and fibers were the more commonly used shapes in the tests, and PE was the most often used polymer type, followed by PVC (Fig. S14). In four occasions, a combination of polymer types was tested (Fig. S14) [24,27,35]. As previously stated for toxicity tests with aquatic organisms [62], the shapes and polymer types used in effect studies with soil organisms are different from those commonly observed in soils. There is a clear underrepresentation of the shape categories 'fibers' and 'films', and of the polymer type PP in toxicity tests. Moreover, only one study used a polydisperse mixture including multiple shapes and polymer types [76], as the MP used in the tests were extracted from farmlands. Therefore, the use of polydisperse MP in toxicity tests is needed to conduct more robust risk assessments.

Scores obtained in the QA/QC evaluation of the 27 studies reporting toxicity data for soil biota exposed to MP are shown in Tables S59-S76. One third of the studies assessed obtained a TAS higher than 21 out of a maximum of 40, while two third of the studies had a TAS of 20 or lower (Fig. S15). No study obtained a TAS in the lowest range (TAS \leq 10) or in the highest range (TAS \geq 31) (Fig. S15). The highest TAS were obtained

by Rodriguez-Seijó *et al.* (2017) (TAS=24, Table S65) [72], Zhou *et al.* (2020) (TAS=23, Table S62) [77], and Huerta-Lwanga *et al.* (2016) (TAS = 23, Table S60) [29], who reported toxicity data for the earthworms *Eisenia andrei, Eisenia fetida* and *Lumbricus terrestris*, respectively (Table S3). The average TAS was 19.0 (range 15–24) and the average scores per criterion ranged from 0.04 to 2 (Fig. S16).

All studies obtained the maximum score of 2 in the criteria "replication", "endpoints" and "presence of natural particles" (Fig. S16). No study scored non-zero in all criteria, and the lowest average scores were found for "verification of background contamination" (0.15), "laboratory preparation" (0.11) and "verification of exposure" (0.04) (Fig. S16). This is consistent with previous quality assessments done for effect studies with MP on benthic [50] and pelagic species [62], where no study received non-zero values in all criteria, and where the criteria "verification of background contamination" and "laboratory preparation" obtained the lowest scores.

3.2. SSDs for volume and surface area as ecologically relevant metrics

From the plants included in the previous selection, only 5 were exposed to MP with a size smaller than the bioaccessibility limit of 5 µm [37,38]. Therefore, the effect threshold concentrations from the remaining 7 plants were excluded from the SSDs. The final SSDs were built with the best-fitting distribution, which was the log gumbel distribution in all cases, to the rescaled effect threshold concentrations from 14 species of 7 taxonomic classes. The invertebrates in the SSDs include four earthworms, L. terrestris, E. andrei, E. fetida, and Aporrectodea rosea, the potworm Enchytraeus crypticus (Class Clitellata), the springtail Folsomia candida (Class Collembola), the oribatid mite Oppia nitens (Class Arachnida), the woodlouse Porcellio scaber (Class Malacostraca) and the snail Achatina fulica (Class Gastropoda). The plants included are ryegrass, Lolium perenne, maize, Zea mays (Class Liliopsida), garden cress, Lepidium sativum, cucumber, Cucumis sativus, and pak choi, Brassica rapa ssp. chinensis (Class Magnoliopsida). Eight SSDs were built, one per treatment (mulching, compost, sewage, and background soils), each of them for the ERMs of particle volume and surface area (Figs. S17-S18).

The HC5 (95% CIs) values obtained for the ERM of volume were: 2.0×10^8 (4.1 \times 10^7 – 3.1×10^9) (mulching), 2.3×10^8 (4.6 \times 10^7 – $3.6\times10^9)$ (compost), 1.5×10^8 $(3.0\times10^7$ – $2.3\times10^9)$ (sewage), and $7.3\times10^7~(1.5\times10^7-1.1\times10^9)$ (background soils) particles/kg dry soil, (Table S77, SI). The HC5 (95% CIs) values obtained for the ERM of particle surface area were: 8.1×10^7 ($1.9 \times 10^7 - 9.5 \times 10^8$) (mulching), 1.1×10^{8} ($2.8 \times 10^{7} - 1.4 \times 10^{9}$) (compost), 8.0×10^{7} (1.9×10^{7} -9.5×10^{8}) (sewage), 4.0×10^{7} (9.4×10^{6} - 4.8×10^{8}) (background soils) particles/kg dry soil (Fig. 2, Table S77). The HC5s obtained from the SSDs for surface area as ERM were consistently lower than those obtained for volume as ERM. Furthermore, within each ERM, the HC5 value for background soils consistently ranked as the lowest (Fig. 2, Table S77). Notably, in all SSDs, the most sensitive species was the snail A. fulica, which exhibited a significant impact on its feeding rate due to chronic exposure to polyethylene terephthalate (PET) fibers [78]. In the case of volume as the ERM, the second most sensitive species was L. sativum for soils with mulching, compost and sewage, while E. fetida was the second most sensitive species in the case of background soils (Fig. S17). For surface area as ERM, the second most sensitive species was always the earthworm E. fetida (Fig. S18).

We compared our HC5 values with those recently reported by Tunali *et al.* (2023) [47] and Jacques and Prosser (2021) [48], which are three and six orders of magnitude lower, respectively (Fig. 2). The reasons for these differences are two-fold. Firstly, the criteria defined to select the effect data included in the SSDs were different. For instance, both previous risk assessments included toxicity data for the nematode *Caenorhabditis elegans*, which was the most sensitive species in both studies [47,48]. Jacques & Prosser (2021) included toxicity data for *C. elegans* where no soil was used in the tests [48,79] (Table S4) and included



Fig. 2. Hazardous Concentrations of microplastics (MP) in particles/kg dry soil for 5% of the species (HC5) obtained in Species Sensitivity Distributions (SSDs) with error bars relating to the 95% confidence intervals (A – H and J), or 25–75 interquartile ranges (I, Tunali *et al.* 2023).

toxicity data for nanoplastics (size < 496 nm) [80]. Although exposures without soil are certainly valid and commonly used for chemicals, they might not be representative of exposure in an actual soil system with the presence of natural particles as well as MP. Tunali et al. (2023) [47] included toxicity data from a test on C. elegans in soil, but they demonstrated that the effects were caused by the additives extracted, while the MP themselves had no significant effects on nematode reproduction [81]. Therefore, it is a chemical rather than an MP effect threshold. This artifact explains the difference between their HC5 and ours, emphasizing the importance of distinguishing between effect data for MP particles and for associated chemicals. Without this distinction, the data cannot be compared, remain fundamentally uninterpretable and therefore unreliable for risk assessment [51,53]. Secondly, the data in the earlier SSDs were not aligned and rescaled to match the characteristics of environmentally realistic MP mixtures to which soil organisms are exposed. These data are mutually incomparable because they pertain to different types of particles. The HC5s reported by Tunali et al. (2023) [47] and Jacques and Prosser (2021) [48] can be considered as values for the average type of particles used in toxicity tests and therefore are not directly applicable to an ERA for environmentally relevant MP mixtures. The HC5 values we present have been adjusted and rescaled to align with environmentally realistic MP mixtures. Nonetheless, the alignment calculations introduce additional uncertainties, as evidenced by the slightly wider confidence intervals of our HC5s compared to those reported by Tunali et al. (2023) [47] and Jacques and Prosser (2021) [48].

With the aim of transparency regarding the reliability of the data used in the current probabilistic risk assessment, the SSDs were also made by showing the TAS obtained for each effect study instead of the taxonomic class in Figs. S19-S20. There is no clear tendency for studies with a lower score to be located in the lower tail or with a higher score to be in the upper tail of the SSD. This suggests that the SSDs are sufficiently reliable for our purpose. However, it should be noted that two criteria that were reported to be crucial in a previous risk assessment for MP in sediment [50], "chemical purity" and "quality of the concentration-response relationship", could not be set here as crucial criteria, as only 3 and 9 studies had non-zero values in these criteria, respectively (Tables S60-S77). This highlights the need for future effects testing with soil organisms to comply with minimum methodological quality standards to improve the quality of SSDs for MP in soils to be used in risk assessment.

3.3. Risk characterization

The logRCR distributions were plotted for each source of MP (mulching, compost, sewage, and background). To characterize the risks of MP in soils with compost, we combined the MECs for soils with compost, manure, and domestic or municipal waste (Table S1). For sewage, we combined the MECs in soils with biosolids and sludge (Table S1). In the case of compost and sewage, only soils from farmlands contained these sources. In contrast, data for mulching and background MP soils were composed by other land uses (Table S1). Farmlands with MP linked to other agricultural practices besides mulching, compost, and sewage (*e.g.*, irrigation with wastewater, use of shed films, *etc.*) (Table S1), were excluded from the analysis due to the lack of data available for these sources.

Risk was quantified probabilistically as the logRCR distribution, with minimum, maximum, mean, standard deviation, 5%, 95%, and 99% CIs, and %RCR>1 as metrics to characterize the distribution (Table S78, Fig. 3, Fig. 4, Figs. S21-S24). The cumulative frequency distributions of logRCR and their 95% CIs, show that for some sites with high exposure concentrations, and both ERMs, a fraction of the RCR distribution exceeded the logRCR= 0 risk threshold (vertical purple line in Fig. 3, Fig. 4, Figs. S21-S24) for soils with mulching, sewage, and background MP pollution. Only for soils with compost use (Fig. 3b, Fig. 4b, Fig. S22) no exceedances of the risk thresholds are found, even if the lower limit of the confidence interval is taken into account. These results indicate that for three out of the four investigated sources of plastic contamination, "hotspot" locations exist where risks are likely to occur. The mean of only one location (3%, n = 34) for soils with mulching exceeded the logRCR= 0 threshold for particle volume and surface area as ERMs, while 21% and 26% of their 95 CIs exceeded the threshold for volume (Fig. 3a, Fig. S21) and surface area (Fig. 4a, Fig. S21), respectively. In the case of soils with sewage application, the mean RCR from two locations (13%, n = 16) in the case of volume (Fig. 3c, Fig. S23) and from three locations (19%, n = 16) in the case of surface area (Fig. 4c, Fig. S23) exceeded the threshold, and for both ERMs 31% of the 95 CIs exceeded the threshold. For soils with background MP pollution, the mean RCR from eleven locations (7%, n = 161) for volume (Fig. 3d, Fig. S24) and fifteen locations (9%, n = 161) for surface area (Fig. 4d, Fig. S24) exceeded the threshold, and the 95% CI of 15% and 19% of the locations exceeded the logRCR=0 for volume and surface area, respectively.

The mean RCRs are plotted for all treatments showing the 'exposure-TAS' *i.e.*, obtained by the study reporting the concentration of MP at



Fig. 3. Risk Characterization Ratios (RCR) for microplastics (MP) in soils, based on particle *volume* as the physical blocking ecologically relevant metric. The four panels show cumulative frequency distributions of the average logRCR calculated for locations for which MP measured concentrations data in soils were available (with 95% confidence intervals, CI), for the sources of contamination: a) mulching, b) compost, or c) sewage use, and d) background. The vertical purple line (-) represents the value logRCR= 0, which separates the part of the distribution where risk would apply (RCR>1) from the part where risk would not apply (RCR<1). Different land uses (farmlands, forests, grasslands, mangroves, and sand) are presented with different colors.

each location, for particle volume (Fig. S25) and surface area (Fig. S26) as ERM. The concentrations of MP from all locations with mulching and sewage use exceeding the threshold were always reported by studies with the lowest TAS (\leq 5 points out of 20) (Figs. S25-S26). Thus, those farmlands with mulching and sewage sludge or biosolid application that are presumed to be at risk based on our probabilistic risk assessment, might come from studies where no fully adequate quality measures were considered. For background MP soils, about 20% of the locations exceeding the threshold obtained the lowest TAS (\leq 5 points out of 20), while the rest obtained the second lowest TAS (\leq 5 points out of 20). This indicates that, together with the need for high quality toxicity data, also high-quality exposure data are required to obtain consistent and reliable outcomes in risk assessment.

The location that exceeded the risk threshold for soils with mulching corresponds to a cotton field in China with over 30 years of mulch use, with a reported concentration of $3.2 \pm 0.41 \times 10^5$ particles/kg of soil [1]. In soils with 5, 10, and 20 years of mulch use, the RCRs were still below the risk threshold, which demonstrates how progressive contamination with MP over time caused the exceedance of risk thresholds [1]. As for soils with sewage, the two locations that exceeded the threshold were in Denmark, with concentrations of 1.65×10^5 and 1.43×10^5 particles/kg of dry soil [17]. Interestingly, all farmland soils with background MP pollution that exceeded the threshold were reported in the same Danish study, where twice as many MP were found in soils with background MP pollution compared to soils with sewage sludge [17]. The authors speculated that these uncertainties were

caused by the sampling and analytical methods used [17], a point supported by the low score obtained in the QA/QC evaluation (5 out of 20) (Figs. S25-S26). The rest of the background MP soils that exceeded the thresholds for both particle volume and surface area as ERMs were mangroves in Brazil, with reported average concentrations ranging from 2.9×10^3 to 1.7×10^4 particles/kg dry soil (size range: $150 - 5000 \ \mu$ m) and wetlands in the United States, with mean concentrations ranging from 8.3×10^2 to 2.0×10^3 particles/kg dry soil (size range $250 - 5000 \ \mu$ m) [22]. For particle surface area as the ERM, one roadside grassland [82] and one suburban forest [18] in China also exceeded the threshold, with mean MP concentrations of 1.2×10^5 and 3.9×10^5 particles/kg dry soil, respectively.

In our assessment, we considered four sources of contamination relevant to soils of different land uses. Although the distinction was not initially designed, it became necessary as alignment parameters appeared to differ among these sources. It should be noted that the concentration data and MP characteristic data for each of these sources cannot be claimed to be representative of the global distribution of these characteristics. This limitation arises because only limited data were available, and data for each MP source predominantly came from different studies, regions, and locations. Nonetheless, it is still relevant to provide a direct, albeit provisional, comparison between the four categories of contaminated soils (Fig. 5). According to our probabilistic assessment, the most accurate metric for comparing the occurrence of risks is to provide and compare the exact percentage of the RCR distribution (*i.e.*, % area under the curve) that exceeds logRCR= 0 (*i.e.*,



Fig. 4. Risk Characterization Ratios (RCR) for microplastics (MP) in soils, based on particle *surface area* as the ecologically relevant metric for translocation-mediated toxicity. Four panels show cumulative frequency distributions of the average logRCR (with 95% confidence intervals, CI) for the sources of contamination: a) mulching, b) compost, or c) sewage use, and d) background (*i.e.*, untreated). The vertical purple line (–) represents the value logRCR= 0, which separates the part of the distribution where risk would apply (RCR>1) from the part where risk would not apply (RCR<1). Different land uses (farmlands, forests, grasslands, mangroves, and sand) are presented with different colors.

RCR=1), for each of the MP sources. Subsequently, these percentages (% logRCR>0; Table S77) for all locations within a source category can be plotted as cumulative frequency distributions (Fig. 5). For curves positioned higher on the graph, a given likelihood of exceeding the risk threshold (%logRCR>0; x-axis) applies to a smaller proportion of the investigated sites (y-axis). Therefore, the proportion of investigated soils at high risk that received sewage application is greater than the proportion of investigated soils at high risk with MP from diffuse and local sources (i.e., background MP pollution), and for soils with mulching. The vertical array of green data points for composted soils shows that the risk threshold exceedance of zero applies to 100% of the locations (Fig. 5). For the other applications, %logRCR> 0 ranges from zero to 60-99%, depending on the source of MP, ERM, and location (Fig. 5). We observe that for none of the soils, the risk probability is 100%, and that the highest risk is calculated for soils with background MP levels (Fig. 5). For the location with the highest concentration of MP in background soils, almost the entire proportion of the logRCR distribution is higher than the threshold (96% for volume and 99% for surface area).

3.4. General discussion, study limitations and prospect

We presented the first fully aligned ecological risk assessment specifically parameterized for MP in global soils. It is considered a global assessment because we incorporated all available MP exposure data for soils into the assessment. Yet many countries could not be accounted for due to a lack of data. This implies that more spatially representative assessments should be conducted in the future when additional data become available.

We found that MP characteristics vary among different sources. Consequently, we employed alignment parameters specific to the sources of plastic contamination. Similarly, compartment-specific alignment parameters have been previously reported and applied in the context of ecological risk assessments for aquatic ecosystems, such as surface waters [55-57] and sediments [50], and components of the human diet, including assessments of human exposure to MP [83]. In MP research, careful alignment of data and the use of consistent units are crucial to avoid 'apple-to-orange' comparisons [53]. Although we chose to use the most specific parameters available for the soil type studies, the HC5 values obtained exhibit minimal differences (as shown in Fig. 2). This suggests that a generic HC5 value may be justifiable for users who do not wish to consider specific sources of MP in their soil risk assessment. In line with the precautionary principle, we recommend utilizing the lowest calculated HC5 value (i.e., 4.0×10^7 '1–5000 µm' particles/kg of dry soil), which should, in principle, provide sufficient protection for all plastic sources examined. This HC5 value can also be considered generic due to the diversity in soil properties represented in our exposure and effect data set.

For none of the soils, the risk probability was 100% (Fig. 5), although the demonstrated high probabilities of risks in the soils exposed to mulching, sewage or contaminated by diffuse and local sources may warrant risk management actions. This is particularly relevant because exposure concentrations and, consequently, the curves in Figs. 4 and 5



Fig. 5. Cumulative frequency distributions of the areas of the Risk Characterization Ratios (RCR) distributions that have logRCR> 0 (%(logRCR)> 0; X-axis) for the different microplastic sources (blue: background, green = compost, black: mulching, red = sewage) for particle volume (upper panel) and surface area (lower panel) as environmentally relevant metric (ERM).

will shift to the right if policies remain unchanged, owing to continued exposure and fragmentation of larger plastic items. We emphasize that the risk profiles provided here are conditional on the fragmentary data available, so that conclusions regarding relative risks for the different soils may change as more representative data becomes available.

Ecological risk assessment is inherently uncertain, and it is advisable to transparently communicate these uncertainties to policymakers and the public, striving for completeness and accuracy [84,85]. Our probabilistic assessment offers a transparent quantification of uncertainties associated with the predicted (absence of) risks. We have considered uncertainties in the data alignment parameters and HC5 values, two factors known for their sensitivity in calculating risks. However, this does not imply the absence of other potential sources of error or uncertainty that should be addressed in future work. Therefore, it is recommended to investigate uncertainties related to soil sampling depth and volume, the homogeneity of (exposure) concentrations in samples, the ingestion of particles by invertebrates or the blockage of seeds and roots in plants, and translocation parameters for both invertebrate and plant species. Moreover, any potential, currently unknown, or difficult-to-quantify effect mechanisms, including, for example, indirect effects due to changes in soil properties, can be considered in the future [7,24,32,33,86].

We aimed to provide complete transparency regarding the QA/QC measures applied in the studies from which we sourced data for both our exposure and effects assessments. A portion of the data was inherently unreliable, introducing a type of uncertainty distinct from the uncertainties addressed through the aforementioned probabilistic approaches. While the cumulative quantity of data may partially mitigate the variabilities and noise resulting from inadequate QA/QC, it is evident that there is significant room for improvement in analytical detection methods and testing strategies for assessing the effects of MP

in soils. However, this undertaking represents a time-consuming research endeavour, critical and urgent though it may be, which will span several years. As a result, during this interim period, risk assessors may need to engage in critical reflection and seek additional advice regarding the credibility of the available methods through expert judgement [51,58].

When addressing the risks associated with MP in soil, several research priorities become evident. Firstly, the detection and quantification of the entire continuum of MP sizes, ranging from 1 to 5000 μ m, in soil pose a substantial challenge. This challenge, while already complex in matrices like surface or drinking water, becomes even more intricate due to the presence of background mineral and organic particles in soil. Isolating MP from these matrices necessitates labour-intensive processes, diminishing the accuracy of MP detection in soil. Furthermore, it is essential that results from effect tests do not depend on substantial differences between the particles being tested. When such differences exist, they require alignment through various corrections and standardizations. A more effective approach is to conduct tests using environmentally relevant mixtures of particles from the outset [62]. This approach results in effect data that are directly comparable and can be efficiently employed in risk assessments.

4. Conclusion

We conducted a comprehensive probabilistic ecological risk assessment of MP in global soils. Our findings indicate that the characteristics of MP vary among different sources of contamination with MP. Consequently, parameters were specifically aligned to these sources. In our assessment, the HC5 values obtained were significantly higher compared to earlier studies and ranged between 4.0×10^7 and 2.3×10^8 particles (1–5000 $\mu m)/kg$ of dry soil for different MP sources and ecologically relevant metrics. The probability of risk for soils was never 100%, but soils exposed to certain sources, such as mulching, sewage, and diffuse or local sources, exhibited high risk probabilities, indicating a need for risk management measures. The proportion of soils at high risk was greater for those subjected to sewage application than for those receiving MP from diffuse and local sources, as well as from mulching. However, the highest risk was calculated for soils with MP entering via diffuse and unspecified local sources. Locations that exceeded the risk threshold scored low in the QA/QC assessment. No risks were associated with soils containing compost. We conclude that, without changes in current policies, the risks for soil organisms are likely to increase due to the continued exposure and fragmentation of larger plastic items.

Environmental Implication

With the detection of high microplastic concentrations in soils globally, there is a growing concern about their potential threat to soil biota. However, no risk assessments have been conducted yet for microplastics in soils accounting for the non-alignment and the quality of the data. We characterize the probabilistic risks of microplastics using plastic source-specific data alignment methods and applying strict quality screening tools. We show that risks from microplastics cannot be excluded in soils with mulching or sewage, nor in soils with microplastics entering via diffuse and local sources. Our risk analysis provides important information for environmental management and policy.

CRediT authorship contribution statement

van Gestel Cornelis A.M.: Writing – review & editing, Validation, Methodology. Koelmans Albert: Writing – original draft, Supervision, Methodology, Investigation, Formal analysis, Conceptualization. Rico Andreu: Writing – review & editing, Validation, Methodology. Huerta Lwanga Esperanza: Writing – review & editing, Validation, Data curation. Redondo Hasselerharm Paula Elisa: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

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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Statement

With the detection of high microplastic concentrations in soils globally, there is a growing concern about their potential threat to soil biota. However, no risk assessments have been conducted yet for microplastics in soils accounting for the non-alignment and the quality of the data. We characterize the probabilistic risks of microplastics using plastic source-specific data alignment methods and applying strict quality screening tools. We show that risks from microplastics cannot be excluded in soils with mulching or sewage, nor in soils with microplastics entering via diffuse and local sources. Our risk analysis provides important information for environmental management and policy.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.133732.

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