

Dossier – Microplastics

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1 Introduction

For decades, plastic pollution has been recognized as an environmental problem, but its urgency has only been acknowledged more recently. Current discussions address not only the visible, but also the invisible form of plastic pollution, the microplastic particles. As early as the 1970s, small plastic particles were detected and characterized in seawater [1, 2], but the term microplastics was not introduced until the early 2000s [3]. Since 2010, an exponentially increasing number of scientific studies on microplastics has been published. Food packaging is one source of microplastics leading to human and environmental exposure.

The physical hazards of visible macroplastic items are obvious and well-documented, e.g., for marine animals, but the potential hazards of microplastics for humans and the environment are much more difficult to identify. Quantification and characterization of microplastics also present unique challenges, making it difficult to assess their risk for humans and the environment. This dossier summarizes main aspects as well as open questions arising from the links between microplastics, food packaging and human health.

2 Definitions

Microplastics are synthetic or heavily modified natural particles with a high polymer content [4-6]. Microplastics form a highly heterogenous group differing in size, shape, surface characteristics, source, material type, and chemical composition. Shapes of microplastics include fragments, fibers, spheroids, granules, pellets, flakes, and beads.

Microplastic particles are frequently categorized according to their size (Figure 1) [5, 7-9]. However, different size categories have been defined, and their limits are based, e.g., on conventional size units or limit values imposed by sampling or analysis techniques. Whether the

definitions cover all or only the largest dimension of a microplastic particle can also differ.

In 2009, a pragmatic definition described all plastic particles below 5 mm as microplastics without defining a lower size limit [10]. Although the upper size limit has found wide application, it does not represent an internationally recognized consensus and is rather an arbitrary definition [5] with exemptions that may be made, e.g., for fibers [11]. For particles in the size range of 1-1000 nm, a proposal for the term nanoplastics was made that specifies this subgroup in more detail [12].

Throughout this dossier, the term microplastics is used for all plastic particles below 5 mm unless it is explicitly important to differentiate between particles in the nano- and micrometer range.

3 Sources

Depending on their source, microplastics can be grouped into primary and secondary microplastics [9]. Primary microplastics are intentionally manufactured in that specific size and applied, e.g., in cosmetics, as industrial abrasives, and as virgin resin pellets used as feedstock during plastic production. Hence, their use is known at least to the manufacturers. Secondary microplastics are degradation products of larger plastic items that are released either during use or after disposal of bigger plastic parts. The absolute amount of secondary microplastics can only be estimated based on production and modelling data [13, 14]. Important sources of secondary microplastics in the environment include fibers released from synthetic textiles during washing and various degradation products of, e.g., plastic litter (including large proportions of single-use food packaging) and agricultural foils.

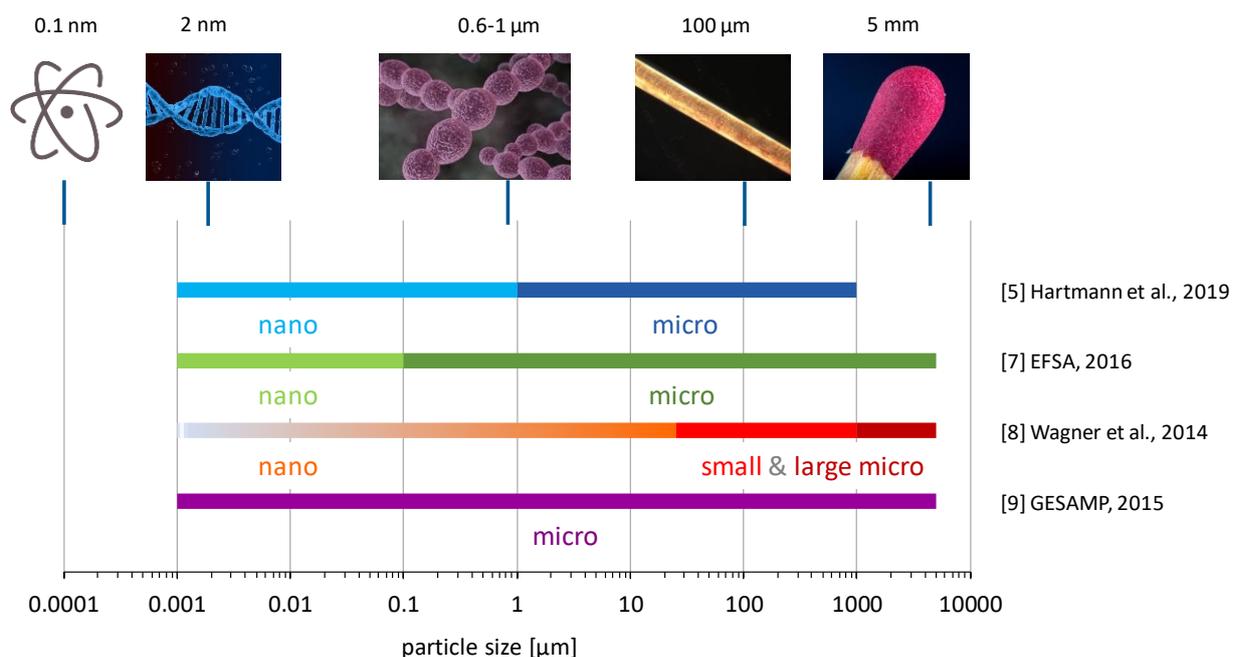


Figure 1: Selected examples of definitions of micro- and nanoplastic particle sizes in scientific literature and institutional reports. For comparison: size of an atom, diameter of a DNA molecule, size of a typical bacterial cell, diameter of a hair, size of a match head.

Abrasion products from car tires and emissions from coatings and paints are also frequently listed among the highly relevant environmental sources of secondary microplastics [15]. However, car tires are mainly made of rubber, and coatings and paints are not exclusively based on plastic polymers. There is no agreement in the scientific community on whether these elastomer particles fall under the definition of *microplastics*.

4 Physico-chemical properties

Plastics can be broadly grouped into thermoplastics and thermosets. Many different polymers and an even greater variety of additives are used to produce the various plastic products that are on the market today [16-18]. However, the properties of microplastics are not only determined by their chemical composition but also by many other factors such as their particle size, particle shape, crystallinity, density, and surface chemistry [19-21].

Depending on the source, size, and sample matrix, it may be very challenging to analyze all these parameters. In addition, the properties of microplastics can change over their life cycle and also affect each other, as the following examples illustrate: (i) physical degradation leads to a higher number of particles with different shapes, thereby generating a larger total surface area and higher particle concentrations; (ii) weathering processes change the surface properties and size of microplastics; and (iii) chemical migration from microplastic particles into the surrounding medium results in altered stability which in turn changes physical degradation processes.

The density of microplastic particles is often considered to be an important physical parameter because it may strongly determine transport in the environment. Together with other inherent properties such as size and shape, density directs whether microplastics would, e.g., settle in sediments or be transported over long distances in water or air. The density of common consumer plastics is in the range of 0.85 to 1.41 g/cm³ which leads to a polymer-type dependent distribution of microplastics between the sediment and the water [22].

5 Methods of sampling and analysis

Methods to measure microplastics in different types of samples, e.g., water, beach sand, or food, typically include the following three steps: (1) sampling, (2) microplastic extraction, i.e., separation and purification of microplastics from the surrounding material, and (3) analysis, i.e., chemical identification, physical characterization, and quantification of microplastic particles [22, 23]. It is important to mention that no internationally standardized sampling procedures or analytical methods are available so far. Therefore, due to the different methods currently used, it is difficult to compare the results of individual studies.

Here we provide a short summary of the analytical approaches currently employed, focusing on the methods that target microplastics in water and biota, since the presence of microplastics in these media may lead to direct human consumption.

5.1 Sampling

Surface water

Microplastics from water samples are most frequently sampled using nets that retain particles above a defined cut-off size [24]. Typical mesh sizes are 300-350 µm, but smaller cut-off sizes were also selected in some studies, demonstrating higher microplastic concentrations and raising a concern over comparability of studies [25]. Further methods include surface microlayer sampling, hand-net collection and bulk water sampling [26].

Animals

Biological tissues are commonly analyzed by dissecting an animal. Depending on the study design and species, either the entire organism (e.g. mussels) or the contents of the stomach and/or the gastrointestinal tract (e.g. birds, turtles) are typically examined [27]. Anything that birds swallow and regurgitate to feed their offspring can also be analyzed. Marine zooplankton can be enzymatically digested as a whole as microplastics remain intact during this procedure [28].

Sediments, soil, and other solids

Plastic-free box corers, iron spoons, or spades are typically used to sample microplastics from solid environmental matrices such as soil, beach sand, and sediment [29]. Especially in these matrices the distribution of microplastics can be highly heterogeneous. Therefore, it is important to take several samples that are distributed over the entire site.

5.2 Extraction

Microplastics need to be separated from any surrounding matrix before they can be analyzed. Solid environmental samples are commonly suspended in a saturated salt solution of a defined density. Microplastic particles with a lower density than the solution will float in the upper water layer while higher-density particles such as clay sink to the bottom. A variety of different salts has been applied for this purpose during the last years [25, 27]. Alternative methods for initial separation are filtration with size fractionation or sieving through size exclusion [30]. After separation, the samples require further purification steps to remove organic and inorganic material that could interfere with analyses. Chemical and enzymatic degradation processes are used for this purpose, e.g., treatment with hydrogen peroxide, acids, bases, or hydrolytic enzymes [25, 28].

5.3 Analysis

The analytical techniques applied to identify, characterize, and quantify microplastics can be broadly grouped into visual, spectroscopic, and chromatographic methods [25, 29, 31-33].

Visual inspection by a human and counting of microplastics under a stereomicroscope is a broadly used approach to describe, e.g., color, size, shape, surface texture, and number of particles. However, this method has several limitations and error sources that can lead to high error rates [32]. The analysis is strongly influenced by the sample matrix, the experience of the individual investigator, and the quality of the microscope. Furthermore, this method does not allow reliable identification of the sample material, because the chemical composition cannot be confirmed.

For a more precise analysis, optical vibrational spectroscopic methods can be applied to differentiate plastics from other particles and to identify the polymer types of microplastics. Fourier transform infrared (FTIR) and Raman spectroscopy are common tools that do not destroy the samples and allow the analysis of particles down to 10-20 µm and 1 µm, respectively [25, 31]. Both methods are based on the energy absorption by the different polymer-types leading to highly specific spectra which can be directly related to a given type of polymer. The advantages and limitations of these two methods have been discussed in more detail by several authors [25, 29, 31, 32].

In addition, chromatographic methods could provide different information not only on the polymer type but also on other properties [25, 29]. For example, pyrolysis gas chromatography/mass spectrometry may be used to measure the polymer type and additives at the same time, whereas high performance liquid chromatography/size exclusion chromatography can determine the molar mass distribution of a polymer. Both methods are standard in analytical chemistry. However, they include thermal treatment and

dissolution steps, respectively, leading to the destruction of the microplastic particles. Furthermore, the particles need to be relatively large in order to allow them to be handled and to produce enough material for the analyses. Other methods that could provide additional information include scanning electron microscopy and flow cytometry [25, 31].

5.4 Quality control

Recently, quality assessment methods were published evaluating studies on microplastics in drinking water, freshwater, and aquatic biota [34, 35]. During these assessments, the following key areas were rated: sampling method, sample size, sample processing and storage, laboratory preparation and clean air conditions, negative controls, positive controls, sample treatment, and polymer identification. Based on the outcomes for “completeness of information” and “reliability,” the authors concluded that stricter quality assurance is needed for studies analyzing microplastics in different types of samples. The application of such quality criteria also becomes very important once analytical methods for nanoplastics have been developed, since the work with particles in the nanometer range is expected to be even more error prone.

5.5 Lower size limit

Attempts to quantify small microplastics (20/25-1000 μm) in environmental samples showed that their concentrations were considerably higher than those of the larger microplastic fractions (1000 - 5000 μm) [36, 37].

The sampling and analysis of plastic particles in the low micro- and nanometer range is technically very challenging and it is currently not possible to detect plastic particles below 1 μm in complex samples. However, laboratory studies have shown that the degradation of plastics can lead to the formation nanoplastics [38, 39]. Therefore, it is a major challenge to reduce the lower detectable size limit in order to also detect such small plastic particles. For human health considerations, the smallest plastic particles may be of high relevance (see 8).

6 Occurrence of microplastics

Microplastics have been found all over the planet. Microplastic particles have been detected not only in the abiotic environment, but also in animals and humans. The presence of microplastics within the food chain as well as in processed human food contributes to human exposure.

6.1 In the environment

Microplastics have been measured in all environmental compartments, such as marine, freshwater, and terrestrial ecosystems, and the atmosphere [6]. First research on microplastics focused on the marine ecosystems, in particular the sea surface and coastlines [40], but increasing information is now available on microplastics in the water column, as well as in sediments [41, 42], the deep sea [43-45], and sea ice [46]. Emerging research suggests that freshwater ecosystems are contaminated to at least a similar extent as the marine environment, with high levels detected in sediments and near urban areas [25, 47, 48]. The few existing studies investigating the atmosphere [49-51] and terrestrial ecosystems [52, 53] demonstrate that microplastics are present there as well.

6.2 In wildlife

For wildlife, likely the most important route of exposure to microplastics is ingestion of plastic debris or ingestion of food and water contaminated with microplastics, both during filter-feeding and

foraging. Microplastic particles have been measured in aquatic invertebrates [54], fish [55-57], seabirds [58], and marine mammals [59, 60]. In contrast, studies reporting and quantifying the ingestion of microplastics by terrestrial animals in their natural habitat are scarce [61]. Since some of the reported aquatic animals are part of the human diet, the following section will focus on the occurrence of microplastics in these animals.

6.3 In the human diet

The scientific evidence demonstrating the presence of microplastics in several foods and beverages has been reviewed in detail [33, 62]. Since the reported studies applied different analytical methods and quality assurance tools, the results are generally not comparable with each other. For this reason, we do not provide an in-depth summary of the quantitative results here but refer to these two review papers and the original studies cited therein [33, 62].

Fish and seafood

The presence of microplastics in marine animals can generally be explained by direct ingestion and trophic transfer. In the case of bigger fish species that are part of the human diet, the gut is usually removed before consumption. This step reduces the potential human exposure to microplastic. In contrast, bivalves, crustaceans, and small fish are consumed as a whole and can thus be a direct source of microplastics. Studies on the presence of microplastics in fish and seafood that are consumed by humans were summarized in several reviews and reports, e.g. [6, 7, 33, 55, 62, 63]. When expressing results as number of microplastic items per g, typical values in blue mussels (*Mytilus edulis*) varied between 0.2 and 2.9 items per g [64]. The measurement of microplastic abundance in *M. edulis* collected from the North Sea and the Canadian Atlantic coast resulted in ranges of 5-19 and 34-126 particles per individual, respectively [65, 66].

Water, beer, and milk

In 2019, the World Health Organisation (WHO) published a report summarizing nine studies that reported microplastics in bottled and unbottled drinking water [22]. The data quality of these studies was further assessed and rated by Koelmans and colleagues [22, 34]. Concentrations were in the range of 0 to more than 10^4 microplastic particles per liter, and mean concentrations varied between 10^{-3} and 10^3 particles per liter. Smaller cut-off sizes of the sampling filters often resulted in higher particle counts. The polymer types of the particles were analyzed in six of nine studies, with polypropylene (PP), polyethylene (PE), polystyrene (PS), polyvinyl chloride (PVC), and PET being the most frequently detected [16]. The application of rigorous quality control measures during drinking water analyses was mentioned as one reason why microplastic was not detected in some studies [67, 68].

Microplastic particles were also found in all 12 beer samples tested from the US, with all breweries drawing water from the Laurentian Great Lakes and using either glass bottles or aluminum cans as packaging [69]. Taking blank samples into account, a range of 0 to 14.3 particles per liter beer was determined. The analysis revealed that almost exclusively fibers and very few fragments were present. Another study of 24 German beer brands reported ranges of 2 to 79 fibers, 6 to 109 fragments and 2 to 66 granules per liter beer [70]. In both studies, microplastics were not further investigated spectroscopically, and therefore no information on the polymer types is available.

In 2020, microplastic particles were detected in all of the analyzed 23 milk samples from the Mexican market [71]. In the samples, 3-11 particles per liter were found, almost three quarters of them were blue and 97.5% of the particles had the form of fibers.

Honey, sugar, and salt

Apart from seafood and beverages, microplastic particles have also been measured in salt, honey, and sugar. In 2019, Lee et al. reviewed the presence of microplastic particles in table salts and concluded that almost all salt products (at least 94%) contained some microplastics [72]. Several studies found a correlation between microplastic concentrations in different types of salt and their origin (e.g. [73, 74]). Highest concentrations were measured in sea salts, followed by lake salts and then rock salts.

By applying negative staining techniques and microscopic examination, microplastic fibers and fragments were investigated in honey and sugar samples [75, 76]. A more recent study also employed spectroscopic methods to characterize microplastic particles in honey [77].

Food processing and packaging

Whether or not food packaging leads to the formation of microplastics during handling and storage, which in turn end up in food, has not been well investigated so far. It is conceivable that different processing steps increase the levels of microplastics in food and beverages, but such information is currently scarcely available. In 2016, the European Food Safety Authority (EFSA) therefore requested more data on the impact of food processing on the formation of microplastics [7].

However, only a few studies investigating this topic have been published since then. In 2018, Li et al. reported higher microplastic concentrations in processed than in unprocessed mussels [64]. In the same year, Karami et al. measured micro- and mesoplastic particles (up to 10 mm) in canned sardines and sprats [78]. Improper gutting, the canning process, or translocation of small microplastic particles into edible tissue were mentioned as possible reasons for the contamination of the fish.

Plastic bottle caps made of high-density polyethylene (HDPE) were further identified as source of microplastics after being opened/closed 100 times [79]. In contrast, the same study showed that multiple squeezing of polyethylene terephthalate (PET) bottles did not lead to the formation of microplastics. In autumn 2019, the release of high concentrations of micro- and nanoplastic particles from plastic teabags into water was reported [80]. Under realistic use conditions, it was estimated that one cup of tea would contain approximately 2.3×10^6 and 14.7×10^9 particles that are larger and smaller than $1 \mu\text{m}$, respectively.

The potential sources of microplastics were discussed in two studies that analyzed microplastic particles in drinking waters packaged in plastic bottles, glass bottles, and beverage cartons [81, 82]. The results did not identify significant correlations between the packaging type and the microplastic levels. Nevertheless, water packaged in reusable plastic and glass bottles tended to be more contaminated than water in single use plastic bottles and beverage cartons. The authors propose that plastic packaging items themselves, i.e., the plastic bottles, coated cartons, and caps, can release microplastics through mechanical abrasion or ageing. Reusable plastic and glass bottles can also be contaminated during cleaning and refilling processes. This would explain the presence of particles having another polymer type than the packaging.

In 2020, microplastic fibers consisting of sulfone polymers were found in milk samples [71]. Since polyethersulfone and polysulfone are commonly used in ultra- and microfiltration membranes in the food and dairy industry, the authors hypothesized that the fibers could have originated from the filtration units.

7 Human exposure

Human exposure to microplastics occurs primarily through ingestion of food and beverages [33], but oral uptake via household dust [83] and inhalation of airborne microplastics [84] were shown to be additional exposure sources. Human exposure has been confirmed by measuring microplastic particles in human stool samples [85] and the lungs [84]. However, the data on the occurrence of microplastics in food, beverages, and air are still too sparse to allow the quantification of human exposure [6].

8 Health hazards

Current evidence on possible effects of microplastics on human health is limited, but different toxicity pathways were proposed, mainly based on information from animal testing. In the organism, microplastic particles can potentially translocate into the circulatory system and different tissues, cause oxidative stress, cytotoxicity, inflammation, or immune reactions, serve as carriers for harmful chemicals and microorganisms, cause lesions in the respiratory system, and disturb the gut barrier and the microbiome [84, 86-88]. Plastic particles in the nanometer range are of particular concern as they could penetrate cells and the gut epithelium more easily than particles in the micrometer range [89].

However, the applied test systems may not sufficiently simulate real exposure scenarios, as microplastic particles found in the environment usually consist of (weathered) mixtures of materials, shapes, and sizes that cannot be easily reproduced in the laboratory. Additionally, most studies focus on oral exposure, but effects that may occur through inhalation of microplastics are less studied. Therefore, such results should be considered as important indications of the potential adverse effects of microplastics, but they may not be directly applicable for risk assessment [6].

9 Risk assessment

Risk assessments of chemicals are based on exposure and toxicity data and form the basis of regulatory decisions. The results of such assessments strongly depend on the quality of the underlying data and usually address only the risks of individual chemicals. This framework can also be applied to the risk assessment of microplastics, but there are unique technical challenges that may lead to higher uncertainties: The quantitative analysis of a soluble chemical is usually straightforward and results in an unequivocal concentration value. In contrast, the quantitative and qualitative properties of samples containing microplastics can be described in many ways, because particles may differ, e.g., with respect to their polymer composition, size distribution, and surface characteristics. Since sample characterization is fundamental for further toxicity and exposure analyses (and thus for risk assessment), the development of quality criteria and international standards was recommended to harmonize the set of available methods and improve comparability [6].

9.1 Human health

Reports published by EFSA [7], the Food and Agriculture Organization of the United Nations (FAO) [55], Science Advice for Policy by European Academies (SAPEA) [90], and the Norwegian Scientific Committee for Food and Environment (VKM) [6] concluded that neither currently available exposure nor toxicity data provided a sufficient basis to assess the risk of microplastics to human health.

The WHO addressed the potential human health impacts of exposure to microplastics through drinking water and concluded that there is (i) no reliable information suggesting that toxicity related to the physical hazard of microplastic particles is a concern through drinking water exposure, (ii) low health concern for exposure to chemicals in

microplastics through ingestion of drinking water, and (iii) no evidence to suggest a human health risk from microplastic-associated biofilms in drinking water [22].

There is no clear consensus in the scientific community as to whether risk assessments based on the available exposure and toxicity data are currently possible [91] or not [34]. However, the authors of both studies called for a better data basis and appropriate and validated methods and reference materials to support further risk assessments.

9.2 Environment

Very few environmental risk assessments for microplastics have been published so far [90]. In 2018, Everaert et al. assessed the risk of microplastics in the oceans by modelling past, present and future microplastic concentrations based on data about global plastic production [92]. A meta-analysis of literature data reporting on the effects of microplastics allowed to estimate safe levels of free-floating microplastics. After comparing predicted and safe concentrations, the authors did not expect any effects of buoyant microplastics up to the year 2100 but excluded heavily polluted sites from this estimate. For the marine benthic compartment, they expected adverse ecological effects from the second half of the 21st century onwards.

In 2019, Adam et al. performed a preliminary probabilistic risk assessment of microplastics in freshwater [93]. Following the analysis of all available peer-reviewed data on exposure and ecotoxicity of microplastics in freshwater, the authors concluded that there is no immediate risk to the environment, but they could not exclude a small risk in some Asian coastal areas.

10 Regulations

Within the last few years, the use of certain primary microplastics in specified personal care products such as rinse-off cosmetics, soap, and toothpaste was banned in many countries all over the world (e.g., the U.S., Canada, New Zealand, Taiwan, South Korea, France [94]). In January 2019, the European Chemicals Agency (ECHA) proposed to restrict intentionally added microplastics in many consumer and professional products, such as cosmetics, detergents and maintenance products, paints and coatings, construction materials and medicinal products, as well as various products used in agriculture and horticulture and in the oil and gas sectors [11]. Many of these provisions set out further definitions and exemptions which need to be considered.

While these restrictions only have a direct impact on primary microplastics, other regulatory and legal frameworks may also indirectly affect the abundance of secondary microplastics. In 2019, two reports on microplastics were delivered to the European Commission by independent scientific advisors [90, 95]. In both reports, implemented European legislation as well as ongoing policy measures were reviewed that could help to prevent and attenuate microplastics in air, soil, and water. The authors identified the water

framework directive, waste legislation, and directives applicable to urban waste-water treatment, the application of sewage sludge as fertilizer, and air quality as relevant areas of action. In addition, the EU plastics strategy and the European action plan for the Circular Economy both address plastic pollution, although they do not specifically mention microplastics. This diverse set of various policy measures shows the many regulatory options through which microplastic pollution can be tackled, but it also illustrates the complexity of the issue.

11 Conclusions

Microplastic pollution is ubiquitous and persistent, and it is likely to increase in the future as plastic production is predicted to grow and the formation of microplastic particles occurs with a time lag [92]. However, knowledge about the effects of microplastics on human health and the environment is limited, and standardized methods to address the occurrence and effects of microplastics are missing. Therefore, intense discussions are currently taking place about whether the risks of microplastics should be managed by applying a strictly evidence-based or precautionary approach [96]. In addition, it might be helpful to consider the persistence of microplastics during risk assessment as it has been already proposed for persistent chemicals [97].

Depending on the approach chosen, future work will focus either on filling existing knowledge gaps on toxicity and exposure of microplastics or on the immediate development and implementation of risk management measures that make use of current knowledge on the ubiquity, persistence, and increasing emissions of microplastics. In any case, there is a widespread consensus that specific measures should be taken to limit the future increase of microplastic particles in the environment and biota, for example, through addressing the use of plastic and preventing environmental plastic pollution.

Abbreviations

ECHA	European Chemicals Agency
EFSA	European Food Safety Authority
FAO	Food and Agriculture Organization of the United Nations
FTIR	Fourier transform infrared
PE	polyethylene
PET	polyethylene terephthalate
PP	polypropylene
PS	polystyrene
PVC	polyvinyl chloride
SAPEA	Science Advice for Policy by European Academies
VKM	Norwegian Scientific Committee for Food and Environment
WHO	World Health Organization

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