

REVIEW

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# Microplastics in food: scoping review on health effects, occurrence, and human exposure

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## Abstract

With most of the plastics ever produced now being waste, slowly degrading and fragmenting in the environment, microplastics (MPs) have become an emerging concern regarding their presence in food and influence on human health. While many studies on marine ecotoxicology and the occurrence of MPs in fish and shellfish exist, research on the occurrence of MPs in other foods and their effect on human health is still in early-stage, but the attention is increasing. This review aimed to provide relevant information on the possible health effect of ingested MPs, the occurrence, and levels of MPs contamination in various foods and estimated exposure to MPs through food. Potential toxic consequences from exposure to MPs through food can arise from MPs themselves, diffused monomers and additives but also from sorbed contaminants or microorganisms that colonise MPs. Recent publications have confirmed widespread contamination of our food with MPs including basic and life-essential constituents such as water and salt providing the basis for chronic exposure. Available exposure assessments indicate that we ingest up to several hundred thousand MPs particles yearly.

**Keywords:** Microplastics, Human health, Occurrence, Food, Human exposure

## Introduction

A little more than a century since the first modern plastic polymer was invented, plastics have travelled from the scientific wonder to one of the greatest global environmental challenges today. Plastic has a great potential to become a marker horizon of human pollution in the Anthropocene era (Corcoran *et al.* 2014), an era of significant human impact on Earth's geology, climate and ecosystems. Global plastics production reached almost 370 million tons in 2019 (Plastics Europe 2019), with over one-third of plastic in both the United States and Europe used in disposable products such as packaging,

eating utensils and trash bags, which are designed to be discarded within 3 years of their production (Gewert *et al.* 2015). Despite efforts towards recycling, a substantial volume of debris has accumulated in the environment and is slowly degraded to micro and nano size by weathering and ageing (Paul *et al.* 2020). It was estimated that over 75% of all plastic ever produced is now a waste (Geyer *et al.* 2017).

Defined as plastic particles with a diameter under 5 mm, microplastics (MPs) became a ubiquitous environmental pollutant present in marine and freshwater systems, soil, air and subsequently the food (Andrady 2017; Gasperi *et al.* 2018; Horton *et al.* 2017; Li *et al.* 2018a; van Raamsdonk *et al.* 2020; Zhou *et al.* 2021). While the immense amount of the scientific evidence on the hazards of the uncontrolled, irreversible, and long-term ecological risks due to MPs do exist for some coastal waters and sediments (SAM 2019), the implications of

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MPs and their potential threats to the other ecosystems and humans are not yet thoroughly investigated and understood.

MPs can be found in food because of environmental pollution but also, there is a growing interest in the possibility of release/leakage of MPs from food packaging materials, such as tea bags, bottles etc. MPs internalised by plants are another potential source of human exposure. The true amount of MP humans may be exposed to via food still cannot be assessed with certainty as many data gaps in MPs research exists (Smith et al. 2018). Also, little is known about the fate of MPs in the digestive tract, absorption uptake kinetics, bioavailability and distribution of MPs in the human body. Exposure to MPs is a complex issue as potential negative health effects can arise from plastics themselves, monomers, additives and various pollutants. One contaminated food might, and mostly do, contain several types of MPs. These particles differ in many ways, including chemical structure, size, shape and biological and chemical load on them. Furthermore, food is not the only source of MPs intake as exposure may occur by inhalation and dermal contact and therefore it could be difficult to distinguish negative health effects caused by intake of MPs by food.

This review aims to provide relevant information on the possible health effect of ingested MPs, the occurrence, and levels of MPs contamination in various food products and estimated exposure to MPs through food.

### Microplastics

Plastics are polymers formed by polymerising monomers into macromolecular chains with the addition of certain additives. Additives catalyze polymerisation reactions or give the plastics functional properties such as elasticity, rigidity, UV stability, flame retardants and colour. On average, 4% of the weight of the plastics predominantly found in MPs are additives (CONTAM 2016), but certain plastics may contain up to 50% of additives by weight. The two main categories of plastics are thermoplastics and thermosetting plastics (thermosets), with the difference being that a thermoset cannot be re-melted, and a thermoplastic can. Thermoplastics include, among others, polyethylene (PE), polypropylene (PP), polyvinyl-chloride (PVC), polyethylene terephthalate (PET), polystyrene (PS), polycarbonate (PC) and polyamide (PA) while main types of thermosets are polyurethane (PU), epoxy resins, vinyl esters and silicones. Five major commodity plastics commonly encountered in MPs are thermoplastics: PE, PP, PVC, PS and PET (Andrady 2017). These five types are also accounted for more of the 70% of European plastics demand (Plastics Europe 2019).

MPs are defined as a heterogeneous mixture of differently shaped materials referred to as fragments, fibres,

spheroids, granules, pellets, flakes or beads, in the range of 0.1–5000  $\mu\text{m}$  (CONTAM 2016). Regarding the size of the particles, further distinction to nano plastics (NPs) can be made. NPs can be defined as a material with any external dimension in the nanoscale or having the internal structure or surface structure in the nanoscale i.e., 0.001–0.1  $\mu\text{m}$  (CONTAM 2016). NPs are considered more damaging than microplastics as they are small enough to permeate through biological membranes (Yee et al. 2021). In terms of this paper, MPs includes NPs, unless otherwise stated.

Regarding origin, MPs fall within two categories: primary and secondary. Primary MPs are plastics that were industrially manufactured to be that size and they are found in textiles, sandblasting media, medicines, and such personal care products as facial and body scrubs (Browne 2015; Cole et al. 2011; Sundt et al. 2014). These particles enter the environment via 'leakage' during manufacture, transportation or use (Andrady 2017). Secondary MPs, more abundant in the environment, mostly originate from the fragmentation of larger plastic litter (macro and meso plastics) but also from usual everyday processes such as laundering of fabrics and use of agricultural mulch plastics (Andrady 2017; Browne et al. 2011; Kyrikou and Briassoulis 2007). Plastics can be fragmented into MPs and subsequently NPs by abiotic and biotic processes. A solitary MPs will break down into billions of NPs particles suggesting that NPs pollution at one point become relevant across the globe (Yee et al. 2021). Generally, abiotic degradation precedes biodegradation and is initiated thermally, hydrolytically, or by UV light in the environment (Andrady 2011; Yee et al. 2021). Environmental bacteria and other microorganisms can biodegrade MPs by the action of either intracellular or extracellular depolymerases (Liu et al. 2010).

### Identification methods

In general, the analysis of MPs consists of two phases: physical characterization of the displayed fragments, followed by chemical characterization thus confirming the chemical nature of the particles found (Mariano et al. 2021). Several microscopy methods are used for physical characterisation, including stereo and fluorescence microscopy, transmission electron microscopy (TEM), atomic force microscopy and scanning electron microscopy (SEM). Among them, TEM, SEM, and fluorescence microscopy have an analytical potential that allows to identify and determine the chemical and physical properties of many polymers (Mariano et al. 2021). SEM is commonly used for this purpose, especially when coupled with detectors for energy dispersive X-ray analysis (EDX) (Mariano et al. 2021; Oliveri Conti et al. 2020; Shrutti et al. 2020; Zuccarello et al. 2019). Chemical characterization

is mostly performed with Fourier-transform infrared spectroscopy (FTIR), Raman spectroscopy, and thermal techniques like differential scanning calorimetry, thermogravimetry, pyrolysis-gas chromatography-mass spectrometry (py-GC-MS), and combinations of these methods (Mariano et al. 2021). The use of integrated techniques such as Py-GC/MS and thermal extraction desorption gas chromatography-mass spectrometry (TED-GC/MS) are gaining popularity because they are faster and often do not need additional, time-consuming sample preparation steps and they give mass content as a final analysis result (Braun et al. 2021).

Figure 1 shows the ratio of main identification methods for microplastics in food used in research papers presented in this review. In most cases overall analysis has included the initial physical characterization microscopy step, and, in several cases, more methods were used. As shown, FTIR and Raman spectroscopy were most commonly used which is also observed in the review papers by Markic et al. (2020) and Kwon et al. (2020).

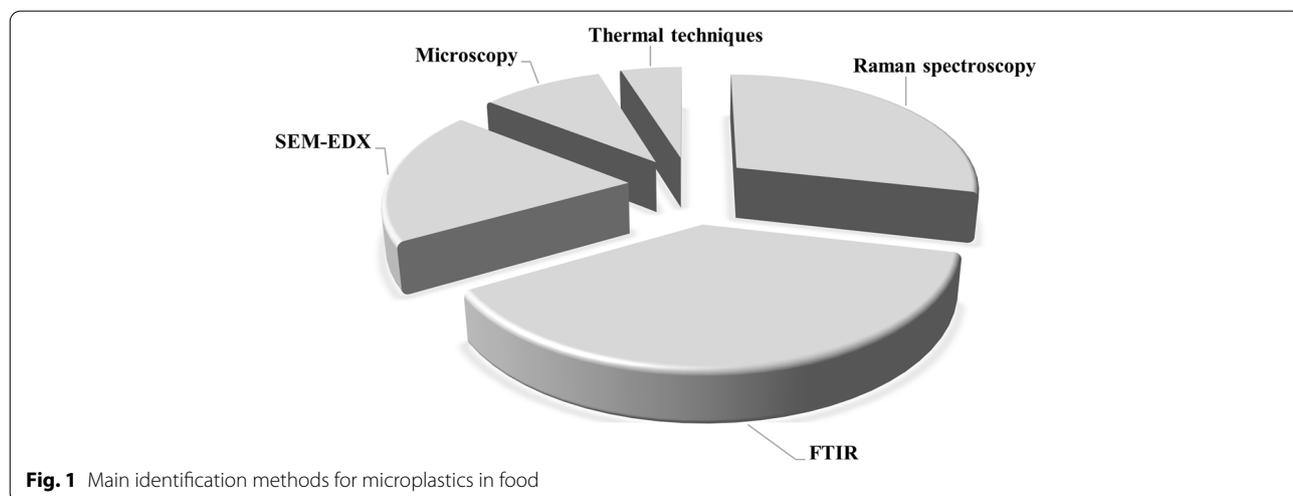
**Microplastics as a hazard**

**Direct effects**

Health effects from exposure to MPs can arise from MPs themselves or diffused monomers and additives used in the production. When discussing MPs impact on human health it is important to distinguish between physical and chemical induction of these effects. Although MPs are not characterised as chemicals, and generally considered chemically inert, they may eventually have similar health outcomes involving the immune system, depending on the amount of the material that gains access to the immune system (CONTAM 2016). An example of a physical effect is the so-called “frustrated phagocytosis”, which is the failure of macrophages to engulf their target

and remove or destroy it, leading to a prolonged inflammatory process and possible tissue damage (van Raamsdonk et al. 2020).

Most of the information on the effects of MPs are obtained from marine wildlife, laboratory animals or from in vitro studies. Ingestion of MPs particles by marine invertebrates has been linked with a wide range of sub-lethal effects including reduced reproduction, reduced growth of individuals and reduced fitness, caused primarily by physical effects of consumed MPs, such as internal damage, gastrointestinal blockage, reduced feeding inflammatory responses and plastic particles replacing digestible food (Horton et al. 2017; Lusher et al. 2013). While some of these effects are highly unlikely to occur in humans, when MPs enter tissues, potential effects might include physical stress and damage, apoptosis, necrosis, inflammation, oxidative stress, and immune responses (van Raamsdonk et al. 2020). Therefore, an important question is whether MPs can be absorbed and enter the systemic circulation and various tissues or stay in the gut lumen following dietary intake. It was estimated that the human excretory system most likely eliminates more than 90% of ingested MPs via faeces (CONTAM 2016; Wright and Kelly 2017). Intestinal absorption is dependent on the size of the particles and it appeared to be very low. Particles on the scale of a few microns or less may be directly taken up by cells phagocytosis and endocytosis mechanisms, particles up to 10µm may be taken up by specialized cells in the Peyer’s patch of the ileum, particles as large as 130µm can enter tissue through paracellular transport in the form of persorption, while particles larger than 150µm are not absorbed, and only local effects on the immune system and inflammation of the gut are to be expected (CONTAM 2016; Powell et al. 2010; Steffens 1995). Not much



**Fig. 1** Main identification methods for microplastics in food

information is available for the absorption rates and bio-availability of MPs. For the particles in the range of 2  $\mu\text{m}$  it was estimated in the range of 0.04–0.3% (Carr et al. 2012), while particles in the nanoscale could reach 0.2 and 1.7% depending on the charge of the particles (Walczak et al. 2015). Once absorbed, MPs can be distributed in the whole organism with only the smallest particles penetrating deep into the tissues and even crossing the blood-brain barrier (CONTAM 2016).

Several recent rodent, zebrafish and in vitro studies have investigated bioaccumulation and the effects of MPs. Most of them used pristine PS particles, thus the main presumption was that showed effect is from MPs themselves. Research by Deng et al. (2017) has shown the accumulation of particles of PS in the liver, kidney, and gut of mice. MPs of 5 and 20  $\mu\text{m}$  were administered through oral gavage at concentrations of 0.01, 0.1, and 0.5 mg per day for 28 days. They reported liver inflammation and the presence of lipid droplets with disturbance of energy and lipid metabolism, oxidative stress, and blood alternation of biomarkers of neurotoxicity. Lu et al. (2018) reported reduced body and liver weight, reduced mucin excretion in the colon, decreased fat metabolism and reduced abundance of microbiota in mice after exposure with high doses of 0.5 and 50  $\mu\text{m}$  pristine PS particles. MPs were introduced through drinking water in a concentration of 100 and 1000 (high)  $\mu\text{g/L}$  and a duration of 5 weeks. Jin et al. (2019) reported somewhat similar findings: decreased colon mucus secretion, altered microbiota composition along with amino acid metabolism and bile acid metabolism disorders. In two related studies, Luo et al. (2019a, b) investigated the effects of 0.5 and 5  $\mu\text{m}$  PS particles on F1 generation of mice through maternal exposure during gestation. Results indicated that maternal exposure during gestation with PS MPs increased risks of metabolic disorder in their offspring, and greater effects were observed in 5  $\mu\text{m}$  MPs-treated groups. Further research has shown that long-term metabolic consequences can be present even in the F2 generation (Luo et al. 2019a). Hou et al. (2021) investigated the effects of 5  $\mu\text{m}$  PS MPs on spermatogenesis in mice. MPs were introduced through drinking water in a concentration of 100, 1000 and 10,000  $\mu\text{g/L}$  and a duration of 5 weeks reaching an estimated average daily dose of 0.6–0.7  $\mu\text{g/day}$ , 6–7  $\mu\text{g/day}$ , and 60–70  $\mu\text{g/day}$ , respectively. After microplastic exposure, the viable epididymis sperm count was significantly reduced with the increased rate of sperm deformity. Atrophy, shedding, and apoptosis of sperm cells at all levels of the testis were observed. Research by Jin et al. (2021) showed that after exposure for 24 h, 4  $\mu\text{m}$  and 10  $\mu\text{m}$  PS MPs accumulated in the testis of mice while sperm quality and testosterone level of mice were declined after exposure to 0.5  $\mu\text{m}$ , 4  $\mu\text{m}$ , and

10  $\mu\text{m}$  PS MPs for 28 days. Zebrafish experiments have shown that PS MPs could induce microbiota dysbiosis and inflammation in the gut (Jin et al. 2018). Furthermore, compared to MPs, NPs may have the potential to induce more serious effects on microbiota dysbiosis and inflammation in the gut of adult zebrafish (Xie et al. 2021). Several recent in vitro studies evaluated the uptake and effects of various polymers particles on various cell lines (Caco-2, HepG2, Caco-2/HT29-MTX mucus model, Caco-2/Raji B M-cell model). They reported variable levels of uptake and in general, cytotoxicity only for high levels of MPs (Abdelkhalik et al. 2018; Agata et al. 1994; Hesler et al. 2019; Lehner et al. 2020; Magri et al. 2018; Stock et al. 2019, 2020; Wu et al. 2019a).

Next to the effect of MPs itself, great attention from the public health point of view is focused on the toxicity of diffusing monomers and additives as there is a vast amount of knowledge on the toxicity of these substances. And while it is estimated that amount of these chemicals from MPs is low, compared to the other sources, chronic exposure is of great concern (Smith et al. 2018). Polymerisation reactions are rarely complete and unreacted residual monomers can be found in the polymeric material (Lithner et al. 2011). Additives, because of their weak non-covalent bond to the polymer backbone, leach rapidly in the environment (OECD 2009). In the environment process of leaching of additives and monomers is further enhanced as fragmentation of MPs is occurring during weathering (Gewert et al. 2015). Little is known on the degradation of MPs in the digestive tract but it has been shown that fragmentation of MPs occurs as part of the feeding and digestion process of earthworms, freshwater amphipods and Antarctic krill (Dawson et al. 2018; Kwak and An 2021; Mateos-Cárdenas et al. 2020).

Lithner et al. (2011) identified 16 out of 55 monomers used in most common polymers as carcinogenic, mutagenic or toxic for reproduction. Among them, bisphenol A (BPA), vinyl chloride, acrylamide and styrene are of greatest concern. BPA constitutes polycarbonate plastics and epoxy resins and is used as an additive in other plastics. BPA is found in food contact materials such as reusable beverage bottles, baby bottles, tableware, storage containers and food cans. Based on these and many other sources BPA represents one of the most abundant chemicals that come in direct contact with human populations worldwide (Welshons et al. 2006). BPA has shown endocrine disrupting effects on humans by interacting with various biological receptors, such as estrogen receptor, androgen receptor, and thyroid hormone receptor resulting in health hazards for the reproductive system, nervous system, metabolic function, immune function, as well as for the growth and development of offspring (Ma et al. 2019). Even though at present-day BPA-free

plastic products are getting more widespread in many cases BPA is simply substituted with one of its analogues that may exhibit similar behaviour to BPA (Pjanic 2017). Vinyl chloride is used primarily in the manufacture of PVC. The largest use of PVC is in the production of plastic piping. Vinyl chloride causes angiosarcoma of the liver, and hepatocellular carcinoma and is classified by International Agency for Research on Cancer (IARC) as Group 1 carcinogen (IARC 2008). Acrylamide is a monomer of polyacrylamide used mostly in water treatment or industry. Acrylamide and its metabolite glycidamide are genotoxic and carcinogenic substances associated with the occurrence of various types of cancers (CONTAM 2015). Worldwide, styrene is one of the most important monomers for polymers and copolymers that are used in a wide range of applications. IARC has classified styrene as probably carcinogenic to humans (Group 2A) due to its association with lymphohematopoietic malignancies (IARC 2018).

Among several hundreds of additives used in plastic production today, the greatest concerns arise from the leaching of phthalates, polybrominated diphenyl ethers, and heavy metals. Phthalates are a group of diesters of phthalic acid and have been widely used by the industry as plasticisers giving flexibility and durability to PVC plastics. There is evidence that phthalates can induce a disruption in oestrogenic activity, reproductive, developmental and liver toxicity both in experimental animals and potentially in humans (Gkrillas et al. 2020). Polybrominated diphenyl ethers are flame-retardant chemicals that were added to plastics and foam products to make them fire-resistant. They have been related to several major aspects of health and behaviour: carcinogenicity, reproductive health, disruption of hormonal signalling, neurotoxicity, neurodevelopmental disorders, behavioural deficits in humans, IQ drop in children and autism spectrum disorders (Poston and Saha 2019). Principal uses of metal additives are inert fillers, pigments for colour, and stabilizers (Janssen et al. 2016; Turner and Filella 2021). Heavy metals such as lead, mercury, chromium, cadmium, and antimony are the most common cause of concern. And while their use is today generally restricted and regulated, hazardous metals have become dispersed amongst contemporary consumer goods through material recycling, the pervasiveness of plastics, poor management and disposal of historical plastics (Turner and Filella 2021).

Additionally, as several of the observed toxic effects of MPs are intricately interconnected in such a manner that disturbance of one process may initiate a cascade of other toxicological responses, Kannan and Vimalkumar (2021) hypothesized that MPs and plastic additives could play a role in global obesity pandemic in association to

other obesogens. This role is suggestive as the prevalence of obesity/overweight has increased by three-fold worldwide over the past five decades, which is in congruence with the use of plastics.

#### Microplastics as a vectors

Besides monomers and additives, attention has been drawn to the presence of persistent organic pollutants, such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs), heavy metals and antibiotics in MPs. In marine environments MPs accumulate hydrophobic persistent organic pollutants, resulting in concentrations that can be several orders of magnitude higher than those in the surrounding seawater (Andrady 2011). The concentration of such pollutants in plastics could reach an order of  $10^6$  (Mato et al. 2001), and due to their composition and relatively large surface MPs are especially susceptible to adhering to waterborne organic pollutants and their subsequent leaching (Cole et al. 2011). PCBs suppresses the immune system, they are tumour promoters that enhance the effects of other carcinogenic substances and alter thyroid and reproductive function in both males and females and increase the risk of developing cardiovascular and liver disease and diabetes. PCBs exposure, especially during fetal and early life, reduces IQ and alters behaviour (Carpenter 2006). PAHs are a group of organic pollutants of a critical environmental and public health concern due to their toxic, genotoxic, mutagenic and carcinogenic properties (Ghosal et al. 2016). Toxicological endpoints of exposure to PBDEs are likely to be thyroid homeostasis disruption, neurodevelopmental deficits, reproductive changes, and even cancer (Linares et al. 2015). Classified as emerging contaminants, antibiotics are receiving increased attention as large amounts are released into the environment every year. Sorbing of hydrophilic antibiotics such as trimethoprim, ciprofloxacin hydrochloride, tetracycline, amoxicillin, and sulfadiazine by PE, PS, PP, PA, and PVC was reported (Li et al. 2018b; Shen et al. 2018).

Microorganisms, including plastic decomposing organisms and pathogens, have been shown to colonise MPs (CONTAM 2016). Currently, studies on the presence of microorganisms on MPs in aquatic environments are the only source of information and the broader relevance to food and the consequences to human health are still unknown. MPs are colonised by a wide variety of microorganisms, forming multispecies biofilms characterised by surface-associated microbial cells enclosed in an extracellular polymeric substance matrix (Fabra et al. 2021). Next to the filamentous fungi and algae, several bacterial strains bacteria have been found on the surface of MPs (Mammo et al. 2020). Next to the most common *Vibrio*

spp. other pathogens or opportunistic human pathogens have been confirmed on MPs, including *Escherichia coli*, *Pseudomonas*, *Aeromonas*, *Haemophilus*, *Acinetobacter* and bacteria of the *Pseudomonadaceae*, *Proteobacteria*, and *Campylobacteraceae* families (McCormick et al. 2014; Silva et al. 2019; Viršek et al. 2017; Wu et al. 2019b). Additionally, the presence of biofilms could enhance the accumulation of organic pollutants as they increase the BET surface area of MPs, thus increasing sorption capacity (Wang et al. 2021).

Another issue related to the presence of microorganisms in MPs is increasing antimicrobial resistance. MPs could act as a microcosm for more effective gene exchange between bacteria as many pathogenic bacteria are in close association with other microbes and sorbed contaminants, especially metals which often co select for antimicrobial resistance (Bowley et al. 2021). Furthermore, increased frequency of plasmid transfer was observed in bacteria on MPs (Arias-Andres et al. 2018).

## Sources of microplastics in the human diet

### Microplastics in water, beverages and alcoholic drinks

Water represents a perfect vehicle for chronic exposure to MPs because is consumed on daily basis and it is without a doubt the most important source of MPs in our diet. Furthermore, water is used in considerable amounts during primary production of food, cleaning, and sanitation of food processing plants, as an ingredient or as a component of food ingredients and for various processing operations. MPs may enter drinking-water sources in several ways: from surface run-off (e.g. after a rain event) to wastewater effluent (both treated and untreated), combined sewer overflows, industrial effluent, degraded plastic waste and atmospheric deposition (WHO 2019). According to available data, conventional and optimised wastewater treatment can effectively remove more than 90% of MPs from wastewater (WHO 2019). Plastic bottles and caps that are used in bottled water may also be sources of MPs in drinking water. To date, several studies investigated the presence of MPs in water. As shown in Table 1 contamination of water with MPs is a global issue with comparatively higher amounts of MPs present in bottled water. Similar to water, MPs contamination was recorded in several types of other beverages and alcoholic drinks (Table 1). While we can speculate that the source of contamination for some products, such as beer or soft drinks, is water, for other contamination could occur through the environment, other constitute components, production processes or packaging materials.

### Microplastics in fish and shellfish

Production of fishery and aquaculture products has outpaced human population growth during the last five

decades (Lusher et al. 2017). These products represent around 17% of animal protein intake by the world's population and in 2015, production reached a record high of around 170 million tonnes of animal products, without considering the addition of approximately 29 million tonnes from farmed aquatic plants (Lusher et al. 2017). Plastics enters aquatic systems from both land-based and sea/freshwater activities. However, it is estimated that 80% of plastic waste in the marine environment is from land-based sources (Jambeck et al. 2015) e.g. from trash, industrial discharge, through inland waterways, wastewater outflows, and transport by winds or tides. Depending on the particle size and the physiological and behavioural traits of the organism, there is an opportunity for the ingestion of these particles by invertebrates and vertebrates and such consumption has been widely observed in many marine species (Horton et al. 2017). Levels of MPs in fish and shellfish are good indicators of MPs contamination of the aquatic environments. The risk of intake of MPs by humans is generally considered lower for larger fish species as the gastrointestinal tract is removed before consumption. On the other hand, small fish species, such as sardines, anchovies, spats and many shellfish species are consumed whole which increases the risk of MPs exposure. Also, there is a growing concern for the possible trophic transfer of MPs in aquatic, benthic and pelagic food webs as predatory organisms may indirectly accumulate MPs during the ingestion of MPs contaminated prey (Lusher et al. 2017). Similarly, predators and detritivores may ingest MPs while scavenging detrital matter containing MPs. Fish and shellfish are the most investigated and understood sources of MPs in the human diet as many studies have investigated their occurrence and abundance. In the last decade, researchers have identified the presence of MPs in fish and shellfish captured in the wild and obtained from aquaculture farms or markets (Kwon et al. 2020). In this period more than 400 original research papers were published reporting MPs in fish, with a majority (62%) focused on marine species and a minority (38%) specifically focused on freshwater species (Galafassi et al. 2021).

In the review paper on plastic ingestion by marine fish in the wild, Markic et al. (2020) systematically reviewed 93 papers published between 1972 and 2019. Results of the review study revealed that plastic ingestion was recorded in 323 species (65.4%) out of a total of 494 examined marine fish species. In the review of papers published from March 2019 to March 2020 Sequeira et al. (2020) revealed that a median of 60% of fish, belonging to 198 species captured in 24 countries, contained MPs in their organs. Environments included marine (52%), freshwater (31%) or mangrove (7%), estuarine and marine (5%), and only estuarine (5%). Minimum and maximum

**Table 1** Occurrence and characteristics of microplastics in water, beverages, and alcoholic drinks

Type of product	Type of polymer	N/Np	Size range (µm)	Analytical method	Microplastic content range (particles/L) <sup>a</sup>	Sample origin	Reference
Ground and drinking water	PE, PVC, PA, polyester, epoxy resin	24/10	50-150	FTIR microscope	0-7 <sup>b</sup>	Germany	(Mintenig et al. 2019)
Water from three treatment plants	PET, PE, PP, PS, PVC, polyacrylamide, polybutylacrylate	3/3	1-100	SEM + FTIR + Raman spectroscopy	338 ± 76-628 ± 28 (treated water)	Czech Republic	(Pivokonsky et al. 2018)
Tap water	nr	110/86	50-4830	Stereo microscope	0-8.605	Hong Kong	(Lam et al. 2020)
Tap water	Anthropogenic debris <sup>c</sup>	159/126	100-5000	Stereo microscope	0-61	Global	(Kosuth et al. 2018)
Bottled water (plastic and glass), beverages, tap water	PE, PP, PS, PET, PA, polymethyl methacrylate	15/15	nr	TED-GC/MS	< 0.01 µg/L – 2 µg/L (mass content)		(Braun et al. 2021)
Bottled mineral water (PET bottles)	–	10/10	1.28-4.2	SEM-EDX	3.16 × 10 <sup>7</sup> -1.1 × 10 <sup>8</sup>	Italy	(Zuccarello et al. 2019)
Bottled mineral water	PP, PE, PET, PS, PA, polyester	38/38	5-100	Micro-Raman spectroscopy	2-44 (single use) 28-241 (returnable) 5-20 (cartons) 4-156 (glass)	Germany	(Schymanski et al. 2018)
Bottled mineral water	PET, PE, PP styrene-butadiene	32/32	1-> 10	Micro-Raman spectroscopy	4889 ± 5432 (reusable) 2649 ± 2857 (single use) 3074 ± 2531 (glass)	Germany	(Oßmann et al. 2018)
Bottled water	PP, PE, PA	259/242	6.5-5000	Optical microscope+FTIR	0-10,000	Global	(Mason et al. 2018)
Beer	Anthropogenic debris <sup>c</sup>	12/12	100-5000	Stereo microscope	0 <sup>d</sup> -14.3	Laurentian Great Lakes	(Kosuth et al. 2018)
Beer	nr	24/24	nr	Stereo microscope	2-79 (fibres) 12-109 (fragments) 2-66 (granules)	Germany	(Liebezeit and Liebezeit 2014)
Beer and soft drinks	PP, PE, polyacrylamide	29/29	3,5–2224.25	Inverted microscope+FTIR	8-117	Ecuador	(Diaz-Basantes et al. 2020)
Soft and energy drinks, beer, cold tea	PET, PA, polyester, acrylonitrile-butadiene-styrene	57/48	100-3000	Epifluorescence microscope+SEM-EDX	0-7 (soft and energy drinks) 0-28 (beer) 1-6 (cold tea)	Mexico	(Shruti et al. 2020)
White wine	PE	26/24	7–475	Micro-Raman spectroscopy	2563-5857	Italy	(Prata et al. 2020)

N/Np Number of samples/positive samples, nr Not reported

<sup>a</sup> Presented as Range, Mean ± SD or Range of Mean ± SD

<sup>b</sup> Per cubic meter

<sup>c</sup> Not stained by the Rose Bengal dye

<sup>d</sup> The three-trial average was less than the number found in the blank

concentrations of MPs in fish were in the range of 0 to 5 and 4 to 56 particles, respectively. Carnivores species contained more MPs than omnivores and the most common polymer types were PE, PP, PET and PA. Galafassi et al. (2021) reviewed papers on the occurrence of MPs in freshwater fish species. Environments included rivers, lakes, estuarine environments, aquaculture ponds,

wetlands or mangrove forests and drinking water reservoirs. Results of the review revealed that plastic ingestion was recorded in 257 species from over 32 different countries. The occurrence of MPs in tested samples reached 90% in some cases and MPs abundance ranged from values of 0 to 4 particles/fish to maximum observations of ~6 to 30 particles/fish. The most common polymer

types identified were PE, PS, PP, rayon, PA, cellophane and acrylonitrile. Only a few studies have investigated the presence of MPs in biological matrices different from the digestive system, gills and skin. In his review paper, Kwon et al. (2020) reviewed over 30 papers reporting the presence of MPs in various types of shellfish. The most investigated species was blue mussel and the majority of reported MPs concentrations were less than one particle/gram. Concentrations were slightly higher in other types of shellfish with an exception of a high reported mean concentration of 297.74 particles/gram found in Atlantic mud crab.

### Microplastics in salts and sugars

Salts and sugars represent another perfect vehicle for chronic exposure to MPs as they are mostly consumed on daily basis, both solely and as part of various food products. Salts and sugars are also commonly used in the cosmetic and pharmaceutical industry as additives, stabilizers, and thickeners. According to the origin, salts are

classified as sea salt and lake salt produced by evaporation, rock salt produced by mining, and river or well salt produced from wells in non-coastal zones (Iñiguez et al. 2017). Several studies have investigated MPs presence in salts while only one early study has investigated the presence of MPs in sugars (Table 2). While researchers in this study did not use spectroscopic identification methods for MPs particulates it is a clear indication of the possible presence of high levels of MPs in sugars.

### Microplastics in processed foods and honey

Contamination of processed foods with MPs generally can be related to the environmental sources, contamination of raw materials and contamination from packaging materials. In some cases, the presence of MPs can be caused by materials used during the manufacturing process such as filtration of beer and milk (Diaz-Basantes et al. 2020; Kutralam-Muniasamy et al. 2020). While the presence of MPs in fish and shellfish could be to a certain extent mitigated by cleaning it can be expected that

**Table 2** Occurrence and characteristics of microplastics in salts and sugar

Type of product	Type of polymer	N/Np	Size range (µm)	Analytical method	Microplastic content (particles/kg) <sup>a</sup>	Sample origin	Reference
Commercial salts, rock salt, lab-grade, and raw salt	PE, PP, PU, PET, PVC, and others, totaling 23 polymer types	24/24	65-2500	Stereo microscope+FTIR	11-193 (commercial) 64 (rock)	Shi Lanka	(Kapukotuwa et al. 2022)
Sea salts	PP, PE, and polyvinyl acetate	23/23	3.3-4460	Optical microscope +SEM-EDX	0.67 ± 1.15- 3.42 ± 4.94	Africa	(Fadare et al. 2021)
Sea salts	PET, PVC, PE, PS, PA, PP	11/11	10-150	Microscopy+Micro FTIR	170-320 (Italy) 70-200 (Croatia)	Italy and Croatia	(Renzi et al. 2019)
Sea and rock salts	Anthropogenic debris <sup>b</sup>	12/12	100-5000	Stereo microscope	46.7-806	Global	(Kosuth et al. 2018)
Sea, lake, and rock salts	PE, PP, PET, PU, PVC, PA	16/16	20-5000	Micro-Raman spectroscopy	16-84 (sea salt) 8-102 (lake salt) 9-16 (rock salt)	Turkey	(Gündoğdu 2018)
Sea salts	PE, PS, PET, PA, polyester	8/8	< 500-5000	Digital microscope+FTIR	56 ± 49-103 ± 39	India	(Seth and Shrivastav 2018)
Sea, lake, and rock salts	PE, PP, PET	39/nr	100-5000	Stereo microscope +FTIR	0-1674 (sea salts) <sup>c</sup> 0-148 (rock salts) 28-462 (lake salts)	Global	(Kim et al. 2018)
Table salts	PE, PP, PET, PS, polyacrylonitrile, PA	17/15	160-980	Stereo microscope +Raman spectroscopy	0-10	Global	(Karami et al. 2017)
Sea and well salts	PET, PE, PP	21/21	30-3500	Stereo microscope +FTIR	115-185 (well salts) 50-280 (sea salts)	Spain	(Iñiguez et al. 2017)
Refined and powdered sugar, unrefined cane sugar	Coloured fibres and fragments	6/6	10-9000	Stereo microscope	217 ± 123 (fibres) 32 ± 7 (fragments) 1100 (total fibres and fragments for cane sugar sample)	Germany	(Liebezeit and Liebezeit 2013)

N/Np Number of samples/number of positive samples, nr Not reported

<sup>a</sup> Presented as Range, Mean ± SD or Range of Mean ± SD

<sup>b</sup> Not stained by the Rose Bengal dye

<sup>c</sup> Excluding one outlier of 13,629 particles/kg

all MPs present in processed food will be consumed. Sources of honey contamination have been identified as environmental, that is particles having been transported by the bees into the hive, or having been introduced during honey processing or both (Liebezeit and Liebezeit 2013). Table 3 shows the current knowledge on the presence of MPs in diverse types of processed foods and honey.

### Microplastics in plants

Terrestrial plants are directly exposed to plastic pollution, deriving from many sources, such as the application of sewage sludge and organic fertilizers, agricultural plastic film or atmospheric deposition (Tympha et al. 2021). While contamination of plants surfaces, and subsequent human exposure, could be reduced by washing or cleaning, it is a prominent issue does MPs can contaminate edible tissues of plants. In the review on the effects of MPs on terrestrial plants and aquatic macrophytes, Mateos-Cárdenas et al. (2021) identified 24 studies that report the ability of plants or macrophytes to adsorb and/or internalise MPs or NPs to a certain degree. The study by Li et al. (2020a, b; 2020a) revealed intake of 0.2 µm and 2.0 µm PS beads in wheat and lettuce, their presence in xylem sap and thus their transport from roots to shoots via the transpiration stream. The internalisation and the migration of MPs to edible plant tissues has also been demonstrated by other authors (Li et al. 2019, 2020b).

In the first study about the presence of MPs in vegetables and fruits, Oliveri Conti et al. (2020) analysed carrots, lettuces, broccoli, potato apples and pears using SEM-EDX method. The higher median level of MPs in fruit and vegetable samples was 223,000 and 97,800 particles/g, respectively. Apples were the most contaminated fruit samples, while carrot was the most contaminated vegetable. The lower median level was 52,050 particles/g observed in lettuce samples. The reported size of the MPs was in the range of 1.36–2.52 µm. Authors hypothesized that the fruits contained more MPs not only because of the very high vascularization of the fruit pulp but also due to the greater size and complexity of the root system and age of the tree compared to the vegetables.

### Microplastics from food contact materials

Food contact materials are materials and articles intended to come into contact with food at any level of the food chain including processing, preparation, storage, serving, etc. As such, they can be a source of various physical, chemical, and biological hazards. The food contact materials are regulated by general safety principles included in EU regulations (EC) No 1935/2004 and (EC) No 2023/2006 (European Commission 2004, 2006).

Besides the general legislation, specific European Union measures exist for some food contact materials such as plastic materials (recycled), ceramics, regenerated cellulose films, active and intelligent materials as well as for some substances including BPA, epoxy derivatives and nitrosamines (EFSA 2020). Despite that, the presence of MPs poses emerging and challenging food safety hazards.

The presence of MPs as a consequence of contact materials in some products such as bottled water is well known (Mason et al. 2018; Schymanski et al. 2018). Research by Schymanski et al. (2018) identified most of the particles in water from returnable plastic bottles as PET (84%) and PP (7%), materials that bottles and caps are made from. For the beverage cartons and glass bottles, they identified PE and biofilms, materials commonly used as coating materials and lubricants. Infant feeding bottles made from PP could shed as much as 16,200,000 particles/L (Li et al. 2020a). Kedzierski et al. (2020) identified PS food tray as a source of MPs contamination of meat. Results of their study revealed PS contamination of packaged meat in the range of 4 to 18.7 particles/kg. The study by Du et al. (2020) showed that MPs are present in take-out containers in the range of 3 to 29 particles/container. The highest abundance occurred in PS containers with a rough surface. The proposed source of MPs was atmospheric fallout and shedding from the container's inner surfaces. Furthermore, treating the containers with hot water did not influence microplastic abundance.

Microplastics could also be generated in everyday activities, such as when we open a plastic package to eat chocolate, cut or tear sealing tape to open a package, twist or open a bottle to drink water, beer, and such everyday simple tasks can generate about 0.46–250 MPs particles/cm (Sobhani et al. 2020). Even steeping a single plastic teabag at brewing temperature can release vast amounts of MPs particles (Hernandez et al. 2019).

### Exposure assessments

With the increase in research data on the occurrence of MPs in food, there is an increased effort on assessing MPs amount humans are exposed to. Exposure assessments are an initial step in evaluating the degree of potential health risk that MPs represent. The amount of the microplastics ingested by an individual will depend on a combination of highly variable parameters, not only of the characteristics of the microplastics but also on each individual's age, size, demographics, cultural heritage, geographic location, nature of the development of surrounding environment and lifestyle options (Senathirajah et al. 2021). To date, only two comprehensive studies are assessing the intake of MPs through multiple food and other sources both based on literature data on MPs occurrence. Cox et al. (2019) in the

**Table 3** Occurrence and characteristics of microplastics in processed foods and honey

Type of product	Type of polymer	N/Np	Size range (µm)	Analytical method	Microplastic content (particles/kg(L)) <sup>a</sup>	Sample origin	Reference
Milk	PP, PE, polyacrylamide	10/10	2.48-6742.48	Inverted microscope+FTIR	16-53	Ecuador	(Diaz-Basantes et al. 2020)
Honey (industrial and craft)	PP, PE, polyacrylamide	14/14	5.15-5174.01	Inverted microscope+FTIR	22-114	Ecuador	(Diaz-Basantes et al. 2020)
Honey	Synthetic fibres and fragments	47/47	40-3100	Stereo microscope	10-336 (fibres) 2-82 (fragments)	Germany	(Liebezeit and Liebezeit 2015)
Honey	Coloured fibres and fragments	19/19	10-9000	Stereo microscope	166 ± 147 (fibres) 9 ± 9 (fragments)	Germany, France, Italy, Spain Mexico	(Liebezeit and Liebezeit 2013)
Milk	PE, PP, PS, PA, PU, polysulfone, polyvinyl alcohol	8/8	≥ 5	Micro-Raman spectroscopy	204-1004 (particles/100 mL)	Switzerland	(Da Costa Filho et al. 2021)
Milk	Polyethersulfone, polysulfone	23/23	100-5000	Epifluorescence microscope+SEM-EDS + Micro-Raman spectroscopy	3 ± 2-11 ± 3.54	Mexico (including international brands)	(Kutralam-Muniasamy et al. 2020)
Dried marine fish	PE, PET, PS, PP, PVC	14/12	195-4780	Micro-Raman spectroscopy	0.56 ± 0.03 (particles/g)	Asia	(Piyawardhana et al. 2022)
Canned fish	PET, PS, PP, PS-PP, PS-PET	50/40	10-8000	Optical & fluorescence microscope+Micro-Raman + SEM-EDX	0.05 ± 0.01-0.22 ± 0.02 (particles/g)	Iran	(Akhbarizadeh et al. 2020)
Canned sardines and sprats	PP, PET, PVC	20/4	190-3800	Stereo microscope+Micro-Raman + SEM-EDX	1-3 (per brand/can)	Global	(Karami et al. 2018)
Rice	PE, PET, PP	52/52	nr	Py-GC/MS	52 ± 5.0-283 ± 50 (µg/g dw) (mass content)	Australia	(Dessi et al. 2021)
Vinegar	PE, butylated hydroxytoluene, Irganox, Erucamide	9/9	1-5000	Stereo & fluorescence microscope+FTIR	16.64 ± 6.34 (optically) 51.35 ± 20.73 (fluorescence)	Iran	(Makhdoumi et al. 2021)

N/Np Number of samples/positive samples; nr Not reported

<sup>a</sup> Presented as Range, Mean ± SD or Range of Mean ± SD

research based on the American diet estimated that annual microplastics consumption ranges from 39,000 to 52,000 particles depending on age and sex. These estimates increase to 74,000 and 121,000 when inhalation is considered. Additionally, individuals who meet their recommended water intake through only bottled sources may be ingesting an additional 90,000 microplastics annually. Authors additionally concluded that these values are most likely underestimated. Senathirajah et al. (2021) estimated that global average ingestion of MPs in the range of 0.1 to 5 g weekly, with tap and bottled water being the greatest contributor. European Food Safety Authority (EFSA) estimated in a conservative approach that exposure to microplastic after consumption of a portion of mussels (225 g) would be 900 particles which could be compared to 7 g of plastics (CONTAM 2016). Next to these assessments, there are several exposure assessments based on the intake of individual foods (Table 4).

The true measure of human exposure can be estimated by body fluids analysis. Investigations of the MPs present in the human body are still scarce and future research is necessary to better understand the interaction of MPs and the human body. The presence of MPs was confirmed in human stool samples (Schwabl et al. 2019). All eight tested stool samples tested positive for MPs. A median of 20 MPs (50 to 500  $\mu\text{m}$  in size) per 10 g of human stool was identified. Overall, nine plastic types were detected, with PP and PET being the most abundant. PC, PA and PP MPs were detected in 11 colectomy samples with an average of 331 particles/individual sample suggesting that MPs are ubiquitously present in the human digestive tract (Ibrahim et al. 2021). More concerning, MPs presence was also confirmed in the human placenta (Ragusa et al. 2021). In total, 12 particles, ranging from 5 to 10  $\mu\text{m}$  in size, were found in four out of six human placentas tested. Three particles were identified as stained PP, while for the other nine it was possible to identify only

**Table 4** Exposure assessments to microplastics through various individual sources

Source of exposure	Estimated consumption	Exposure level	Type of polymer	Reference
Water and beverages	2.2-3 L/day	4400-5800 particles/ person/ year	Anthropogenic debris <sup>a</sup>	(Kosuth et al. 2018)
Bottled mineral water	2 L/day (adults) 1 L/day (children)	1,531,524 particles/kg/ body weight/day (adults) 3,350,208 particles/kg/ body weight/day (children)	PET	(Zuccarello et al. 2019)
Salts	14.8-18.01 g/day	Up to 302 particles/person/year	PE, PP, PET, PU, PVC, PA	(Gündoğdu 2018)
Salts	3.95 g/day	Maximum of 37 particles/person/year	PE, PP, PET, PS, polyacrylonitrile, PA	(Karami et al. 2017)
Salts	5 g/day	131.4–372.3 particles/person/year (Croatia) 306.6–580.35 particles/person/year (Italy)	PET, PVC, PE, PS, PA, PP	(Renzi et al. 2019)
Fruit and vegetables	High intake for apples and pears of 165.3 and 115.7 g/day for adults and children, respectively Low intake for carrots of 20.3 and 18.0 g/day for adults and children, respectively	$1.15 \times 10^5$ - $1.41 \times 10^6$ particles/kg body weight /day (children) $2.96 \times 10^4$ - $4.62 \times 10^5$ particles/kg body weigh /day (adults)	nr	(Oliveri Conti et al. 2020)
Seafood	9.6 -57 kg/year	518 – 3078 particles/person/ year	PE, polyester, semisynthetic cellulose	(Barboza et al. 2020)
Mussels	0.082-3.08 kg/year	123-4620 particles/person/year	PET, PU	(Catarino et al. 2018)
Vinegar	3.1 L/year	Up to 3.68 particles/kg/body weight/year (adults) Up to 16.08 particles/kg/body weight/year (children)	PE, butylated hydroxytoluene, Irganox, Erucamide	(Makhdoumi et al. 2021)
Food contact materials	4-7 takeout's weekly	12–203 particles/person/weekly	PP, PS, PE, PET	Du et al. 2020
Infant feeding bottles	–	14,600–4,550,000 particles/person/day	PP	(Li et al. 2020)
Household dust fallout	Evening meals	13,731-68,415 particles/person/ year	na	(Catarino et al. 2018)

nr Not reported, na Not applicable

<sup>a</sup> Not stained by the Rose Bengal dye

the pigments, which were all used for man-made coatings, paints, adhesives, plasters, finger paints, polymers, cosmetics and personal care products.

The exposure metrics used to express MP estimates are mostly given in quantities not making a difference in type of polymers, shape, or size of microparticles. These parameters are of importance for the exposure and risk assessment. Exposure to MPs (and NPs) is also related to the exposure to contaminants used as additives in the production or those adhering on the surface. Phthalates with their endocrine disruptive activity, are one of the examples for this. Therefore, plastics pose a number of potential human health and environmental risks. The research shows that MPs act as a hub for potentially pathogenic bacteria (Beloe et al. 2022). Once in the water or food, the MPs can further spread these bacteria, assist in their survival and potentially influence their fitness and virulence.

The size definition in exposure assessment is reflected in the effects they may have in a human body. It is generally believed that large plastic polymers are inert and are not absorbed by the intestinal system (due to their size), or they do not cross brain-blood barrier. This would imply that they are excreted un-metabolized (Kannan and Vimalkumar 2021). However, upon entering the environment and/or in biological systems, plastics break down into small particles resulting in smaller plastic particles in the environment, including MPs. These subsequently formed smaller plastic particles are massively present and moreover may have different toxicokinetic profiles. Based on the results of the previous studies on metal and metal-oxide nanoparticles, such as gold (Au) and titanium dioxide (TiO<sub>2</sub>) nanoparticles, which can reach the brain to exert a range of neurotoxic effects, and the similarities to micro and nano plastic, it may be envisaged that plastic particles may have similar effect (Prüst et al. 2020). Prüst et al. (2020) reviewed such neurotoxic effects. They also reviewed 20 studies regarding the general toxicity of MPs and NPs from animal models. The studies point towards alterations in gene expression, inflammation of gut, gills, liver, kidney and/or muscle, particle accumulation in tissues of gills, intestine, liver, kidneys, gallbladder and/or gonads, (lipid) oxidative damage in body/organs, disturbed metabolism, alterations in motility and behaviour, alterations in intestinal barrier function and gut microbiome, reduction of overall fitness and increased mortality.

Exposure assessment studies refer to exposure to microplastic as an expression of lifetime exposure to MPs for humans. Most often it is expressed on an annual or daily basis per capita (person). The latter is more common for dietary exposures, while annual basis is more relevant for a correlation of the exposure to annual production of plastics. The exposure to MPs may be

modelled by using a mass balance of intake (i.e., dietary and inhalation) and loss processes in the human body (via the GI tract). For this plastic model the source of MPs in GI tract is threefold (dietary intake, inhalation via air and back transformation of MPs in GI) (Mohamed Nor et al. 2021). The authors aimed to estimate the MPs exposure in children and adults via eight food types and inhalation, and to assess the chemical contribution of MPs in relation to total chemical intake. Interestingly they have performed three corrections to account for the particle sizes (realignment following power-law size distribution), false positive occurrence data and correction for the translation of the data from inedible (fish gut) to edible (fish muscle) part. Authors have shown that rescaling of particles in exposure assessment may influence the estimates by a factor of ten. They also pointed out the weakness of one point estimate of MPs intake since this is not representative of the human exposure to MPs.

As the number of human exposure studies is increasing it is relevant to consider the complexity of MPs mixtures and discrepancies in the size ranges. Therefore, general terms like 'microplastics' need to be specified in detail. The exposure estimates will not directly be sufficient to characterise the risk to humans due to the lack of knowledge on possible effects which can be caused by the oral consumption of MPs particles. Nevertheless, the harmonisation and use of comparable exposure metrics as it is done for other nanoparticles is of the utmost importance.

## Conclusion

There is undeniable evidence that MPs are present in our food. As a methodology for the detection of MPs have some limitations in the detection of particles in the nanoscale range presented occurrence and levels could be underestimated. Having in mind that almost all plastic ever produced is still in the environment and as it degrades it is obvious that the contamination of food with MPs will increase in future years and even decades. There is a myriad of possible adverse health effects caused by MPs themselves or adjoined hazards. The interaction of the toxicity of polymers and their chemical cargo, as well as the pathogenic potential of associated microorganisms and their toxins, as well as the role of MPs on the spread of antibiotic resistance in one health paradigm requires further research to bring the new weight of evidence and quantitative data. Moreover, future research must focus on revealing realistic scenarios of exposure to MPs through food and other sources and on the better understanding of MPs fate in the human body and the real consequences of MPs consumed.

## Abbreviations

MPs: Microplastics; PE: Polyethylene; PP: Polypropylene; PVC: Polyvinylchloride; PET: Polyethylene terephthalate; PS: Polystyrene; PC: Polycarbonate; PA: Polyamide; PU: Polyurethane; NPs: Nano plastics; BPA: Bisphenol A; IARC: International Agency for Research on Cancer; PCBs: Polychlorinated biphenyls; PAHs: Polycyclic aromatic hydrocarbons; PBDEs: Polybrominated diphenyl ethers.

## Authors' contributions

Conceptualization: B.U., T.C.-V. and A.R.; data curation: B.U.; writing—original draft preparation: B.U.; writing—review and editing: T.C.-V., M.A. and A.R.; visualization: B.U. and A.R.; supervision: A.R. and T.C.-V.; funding acquisition: A.R., M.A. and T.C.-V. All authors have read and agreed to the published version of the manuscript.

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## Availability of data and materials

All data used for the study are contained within the manuscript.

## Declarations

### Competing interests

The authors declare no conflict of interest.

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