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Review

Human health concerns regarding microplastics in the aquatic environment - From marine to food systems

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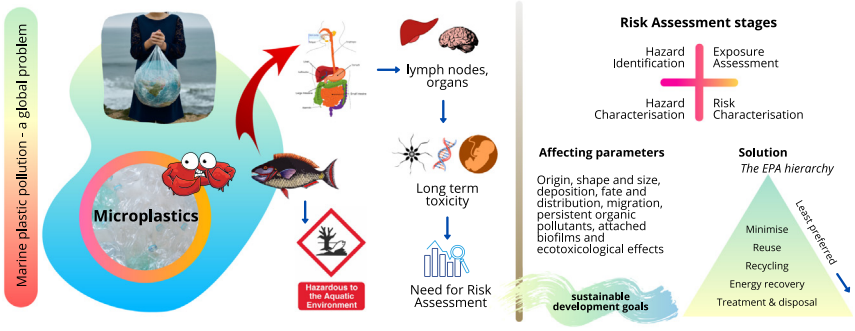
HIGHLIGHTS

- Small plastic particles (<150 μm) can be absorbed by biota tissue, organs, and even cells.
- Microplastics may pose acute and (sub) chronic toxicity, carcinogenicity, and developmental toxicity.
- Similarly, nanoplastics may pose chronic toxicity, genotoxicity, and developmental toxicity.
- PUR, PAN, PVC, Epoxy resin, and ABS are identified as the most toxic polymers.

GRAPHICAL ABSTRACT



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ABSTRACT

Marine plastic waste pollution is one of the most urgent global marine environmental problems worldwide. It has attracted worldwide attention from governments, the public, the scientific community, media and non-governmental organizations and has become a hot issue in current marine ecology and environmental research. This research aimed to conduct a traditional review of the current state of the art regarding microplastics (MPs) definition and characterisation, including an assessment of MPs detected in marine and food systems. The review revealed that plastic waste is not biodegraded and can only be broken down, predominantly by physical processes, into small particles of micron to nanometre size. Particles (<150 μm) can be ingested by living organisms, migrate through the intestinal wall and reach lymph nodes and other body organs. The primary pathway of human exposure to MPs has been identified as gastrointestinal ingestion (mainly seafood for the general population), pulmonary inhalation, and dermal infiltration. MPs may pollute drinking water, accumulate in the food chain, and release toxic chemicals that

Abbreviations: ABS, acrylonitrile-butadiene-styrene; ASEAN, Association of Southeast Asian Nations; ATR, attenuated total reflection; BPA, bisphenol A; CARS, Coherent anti-Stokes Raman scattering; CPR, continuous plankton recorder; DDT, dichlorodiphenyltrichloroethane; DEHP, di-2-ethylhexyl phthalate; DnOP, di-n-octyl phthalate; EDC, endocrine disrupting chemical; EDX, energy dispersive X-ray spectroscopy; EPA, United States Environmental Protection Agency; EDAX Genesis, energy dispersive X-ray microanalyzer; EUSES, European Union System for the Evaluation of Substances; FPA, focal plane array; FTIR, Fourier Transform Interferometer; GHS, Global Harmonized System; GIT, gastrointestinal tract; HDPE, high-density polyethylene; IR, infrared radiation; IUPAC, International Union for Pure and Applied Chemistry; LDPE, low-density polyethylene; MDA, malondialdehyde; MP(s), microplastic(s); MSFD, European Maritime Strategic Framework Directive; NP(s), nanoplastic(s); OPA, organic plastic additive; PA, polyamide; PAH, polycyclic aromatic hydrocarbons; PAN, Polyacrylonitrile; PBDE, polybrominated diphenyl ether; PC, polycarbonate; PCB, polychlorinated biphenyls; PE, polyethylene; PET, polyethylene terephthalate; PICES, North Pacific Ocean Scientific Organization; PLA, polylactic acid; PLI, pollution load index; PMMA, Poly(methyl methacrylate); PMP, plastic microbead; PMPs, primary microplastics; POM, polyoxymethylene; POPs, persistent organic pollutants; PP, Polypropylene; PS, Polystyrene; PUR, Polyurethane; PVC, Polyvinyl chloride; Py-GC/MS, pyrolysis gas chromatography–mass spectrometry; REACH, Registration, Evaluation, Authorization and Restriction of Chemicals; RI, risk index; ROS, reactive oxygen specie; RS, Raman spectroscopy; SAMP, suspended atmospheric microplastic; SAN, styrene acrylonitrile; SEM, scanning electron microscope; SEM/EDX, scanning electron microscope and energy dispersive X-ray spectroscopy; SMPs, secondary microplastics; SOD, superoxide dismutase; SOP, standard operating procedures; TBBPA, tetrabromobisphenol; TED-GC/MS, thermal extraction and desorption and gas chromatography mass spectrometry; TDS-GC/MS, thermal desorption gas chromatography mass spectrometry; TGA-DSC, thermal imaging and differential scanning calorimetry; TGA-SPE, thermogravimetric analysis with solid phase extraction; Triton X-100, octylphenol ethoxylate; UNESCO-IOC/WESTPAC, Western Pacific Sub-Committee of the UNESCO Intergovernmental Oceanographic Commission.

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Human health risk
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Risk assessment

may cause disease, including certain cancers. Micro/nano-plastics may pose acute toxicity, (sub) chronic toxicity, carcinogenicity, genotoxicity, and developmental toxicity. In addition, nanoplastics (NPs) may pose chronic toxicity (cardiovascular toxicity, hepatotoxicity, and neurotoxicity). The toxicity of MPs/NPs primarily depends on the particle size distribution and monomeric composition/characteristics of polymers. Polyurethane (PUR), Polyacrylonitrile (PAN), Polyvinyl chloride (PVC), Epoxy resin, and Acrylonitrile-butadiene-styrene (ABS) are categorised as the most toxic polymers based on monomer toxicity. MP detection methods include combinations of spectroscopic analysis (RS and FTIR) and chromatography (TED-GC/MS). MP/NP toxicological properties and general quantitative and qualitative analysis methods used in MPs Risk Assessment (RA) are summarised. A robust dose-response model for MPs/NPs requires further investigation. This study lays the foundation for the evaluation of MP/NP risk assessment in the marine ecosystem and potential implications for human health.

Contents

1. Introduction

Since the mass production of plastics in the 1950s, output has increased steadily year on year. In 2016, the total global output of plastic products reached 3.35×10^8 t (Novotna et al., 2019), with an average annual increase of about 4% (The Lancet Planetary Health, 2017). As a result, between 4.8×10^6 and 1.27×10^7 t of plastic waste ends up in the ocean every year; among them, nearly 1.15×10^6– 2.41×10^6 t of plastic garbage enters the ocean from rivers (Novotna et al., 2019). Within the EU, between 80 and 85% of marine waste is plastic, with 50% of these being single-use plastic products (Plastics Europe, 2019). In Ireland, it is estimated that 73% of deep-sea fish ingest plastic (Wieczorek et al., 2018), 90% of seabirds are found with plastics in their intestines (Wilcox et al., 2015), and the main material used in nesting for the gannet colony, Little Skellig, on the coastal islands of County Kerry has shifted from natural materials to plastic (Hilliard, 2018). Up until 2019, Europe manufactures approximately 62 million tons of plastics worth €350 billion per year (Plastics Europe, 2019), accounting for approximately 8% of total global production. By 2050, Europe is forecast to produce over 1800 million tons of plastic waste per year (Gallo et al., 2018; Wright and Kelly, 2017). In addition, it is estimated that Europe will release between 75,000 and 300,000 tons of MP particles into the environment annually (European Commission, 2018a).

Land-based plastic waste enters marine environments under the influence of external driving forces (such as hydrodynamic processes) and is then transported long distances by ocean currents into ocean circulation and the deep seabed (Li, 2019). Most polymer plastic products will need decades, or even hundreds of years, to degrade, and about 60% of these plastics have a lower density than seawater, resulting in floating plastics entering ocean circulation aided by ocean currents and winds, forming

the world's five major 'vortex plastic garbage gathering areas' (Pivokonsky et al., 2018). In addition, there is a famous "Great Pacific Garbage Belt" in the eastern part of subtropical circulation in the North Pacific Ocean (Pan et al., 2019). Most of this plastic waste comes from marine fishery activities originating in East Asia (Japan (29.79%) and China (29.27%) (Pan et al., 2019)) and are migrated there by the Kuroshio extension system. According to surveys by Cheung et al. (2018), 'Great Pacific Garbage Belt' covers an area of approximately 1.6×10^6 km^2 with c. 0.45×10^5– 1.29×10^5 t of plastic where MP accounted for 8% of the total mass and 94% of the total quantity by the number of MPs.

MP contamination is common in the marine ecosystem and human food system (Barboza et al., 2018). MPs may carry toxic chemicals to marine organisms, including toxic additives, persistent organic pollutants, and heavy metals enriched from the surrounding environment (Tiwari et al., 2019; Digka et al., 2018). Toxic chemicals may be enriched along the food chain, which may cause a detrimental effect on marine life and human health. While plastic not only threatens the survival of more than 800 species of animals, including large marine mammals (such as whales and dolphins) to various birds, fish, and invertebrates, it also leads to chemical pollution, invasion of exotic species[1] and damage to local tourism and fisheries (Pan et al., 2019). In 2015, marine plastic pollution and global climate change, ozone depletion, and ocean acidification were listed as major global environmental issues (Pivokonsky et al., 2018). Marine plastic pollution has caused the global economy to lose more than $\$ 80 \times 10^8$, including a loss of $\$ 31 \times 10^8$ in aquatic products (Pan et al., 2019).

In light of the seriousness of this emerging issue, this study aims to conduct a traditional review, which is a well established format (Virginia Tech. University Libraries, 2021) providing trends, update, practice and a broader overview on the topic. The primary objectives of this review are to:

- assess the current state of the art regarding MP definition and characterisation,
- review of the detection methods and abundance of MPs in marine and food systems,
- identify risk assessment methodologies employed and toxicological properties of human health concerns.

Search strategy aimed at assembling relevant studies on risk assessment of microplastics and nanoplastics published in international databases such as Scopus, Web of Science, PubMed, EmBase. The following terms were utilised in each database:

- Scopus: ((("Microplastics"[Mesh] OR microplastic* [tw]) OR (nanoplastic* [tw] "nano plastic*" [tw])) AND ("Risk Assessment"[Mesh] OR "Risk assessment*" [tw]));
- Webofscience: (((ALL = microplastic*) OR (ALL = (nanoplastic* OR "nano plastic*")) AND (ALL = "risk assessment*")));
- PubMed: ((("Microplastics"[Mesh] OR microplastic* [tw]) OR (nanoplastic* [tw] "nano plastic*" [tw])) AND ("Risk Assessment"[Mesh] OR "Risk assessment*" [tw]));
- EmBase: ((('microplastic'/exp OR 'microplastic') OR ('nanoplastic'/exp OR 'nanoplastic')) AND ('risk assessment'/exp OR 'risk assessment')).

Next, abstracts of all articles filtered by the above process were read, and the most relevant references were included in this traditional review. A priority was given to articles published in the last ten years.

1.1. Early research on microplastics (MPs) levels

Although the first report on MP pollution in the marine environment was published in the early 1970s, this topic did not regain attention until 2001. A review from Pan et al. (2019) indicates that from 1993 to 2019, global marine MP abundance increased by 2 to 2.5 times, with more than 5 trillion plastic scraps and 250 million tons of plastics eventually floating to the ocean due to industrial emissions and surface runoff. Sediment and surface water are the main environmental matrices measured to evaluate MP abundance (Di and Wang, 2018). The latest studies documented the accumulation of MPs in the North Atlantic ocean over 22 years (from 1986 to 2008) (Ivleva et al., 2017), compared to the North Atlantic coastal surface water levels in 1972 (only 3500 debris km^{-2} PE particles), 62% of more than 64,000 water samples contained MPs in 2008 with a total level between 10,000 and 100,000 debris km^{-2} PE particles. As of 2010, the total weight of global MPs in the surface water is estimated to have reached approximately 4.9×10^5 t with a concentration of between 0.2 and 0.9 particles m^{-3} (Li, 2019). According to recent research reports, MPs are widely distributed in coastal sediments from the UK, India, Singapore, Sweden, Belgium, Italy, China, and Germany, with concentrations from 0.3 to 5000 particles kg^{-1} (Xiong et al., 2018). MPs were also detected in deep-sea sediments from the Atlantic, Mediterranean, Pacific Northwest, and Arctic Oceans (Pivokonsky et al., 2018; Di and Wang, 2018).

1.2. MP definition and characterisation

Due to the different sampling, processing, units, and current identification methods (used for different types, shapes, and sizes), it is difficult to standardise and compare results between different studies. Therefore, unified identification methods and analysis of size ranges have to be developed. From an analytical perspective, MPs typically cover particles in the micrometre range (1 μm to 1 mm). Others, larger than 5 mm in diameter, are considered meso (5 mm to 25 mm) and macro (>25 mm) plastic (Pan et al., 2019). The European Maritime Strategic Framework Directive (MSFD) guidelines (Rist et al., 2018) have defined MP as plastics with a diameter of less than 5 mm (1 mm to 5 mm and <1 mm), and it is used as a standard measure in the analysis of marine and freshwater ecosystems.

There are also spectral identification methods used to characterise size (greater than or equal to 500 μm) and biological methods (<100 μm, <25 μm and <10 μm) (Pan et al., 2019). Depending on size, MPs are typically divided into small MPs (0.001–1 mm) and large MPs (1–5 mm). The size of MPs is considered with 5 mm as the upper limit, and the lower limit is specifically determined by sampling and identification, usually 500 μm (Ivleva et al., 2017). The lower limit of sediment samples is usually between 0.5 and 2 mm, while surface water samples depend on mesh size, often 300 μm, but typically between 53 μm and 3 mm ((Di and Wang, 2018). For bulk samples, the lower limit of filter cut-off size efficiency is as low as 1 μm. Also, analyses must not ignore MPs below 1 μm (100 nm to 1 μm, and nano-plastics (NP) (<100 nm)) (Pan et al., 2019). It must be noted that the International Union for Pure and Applied Chemistry (IUPAC) has defined the lower limit of MP size as 0.1 μm (Revel et al., 2018). However, due to technical limitations, MPs detection at sizes smaller than 1 μm in aquatic environments does not yet have appropriate analytical technology support. So, the minimum recognition size of MPs is considered 1 μm.

According to the MP size classification method developed by the National Oceanic and Atmospheric Administration (Revel et al., 2018), 50% of MPs found in aquatic systems are between 0.5 and 1.0 mm, 29.8% of MPs are between 1 and 2.5 mm and 17.6% of MPs are between 2.5 and 5.0 mm. Since large plastics in the marine environment are broken down into smaller ones by mechanical action (plastic aging process and forced crushing by weather), photo-oxidation, and biodegradation, the abundance of MPs counts increases with decreasing size (Novotna et al., 2019). Simultaneously, concern revolves around the toxicity of MPs associated with its size; the smaller the size, the greater the potential toxicity to marine zooplankton. In vitro tests by Novotna et al. (2019) have shown that activating antioxidant-related enzymes and mitogen-activated protein kinase signaling pathways is sensitive to MP exposure in a size-dependent manner. However, diverse size classification among studies hinders the comparison and collection of information and the differentiation of MPs effects due to different sizes. Therefore, it is important to set a uniform size classification based on MP studies in marine ecosystems.

[1] A plant species or an animal species that is non-native, or alien, nonindigenous, or introduced species (Minchin, 2001), which means a species that has been intentionally or inadvertently brought into a region or area outside their normal distribution (Root et al., 2013).

Table 1
Classes used to describe microplastics (MPs).

Class	Description
Source	Primary microplastics (PMPs) and secondary microplastics (SMPs)
Type	Films, spheres, fibers, foams, and particles
Shape	Film: fragments, crystals, fluff, powder, granules, shavings, round, sub-round, subangular, and angular; Fibers: filaments, microfibers, strands, and threads; Foams: Polystyrene, expanded Polystyrene; Spheres: beads, grains, microbeads, microspheres, cylinder, disc, flat, ovate, pellet, and ellipse; Particles: resin pellets, nurdles, pre-production pellets, nibs; General: irregular, elongated, degraded, rough, and broken edges flakes.
Colour	Transparent, crystalline, white, clear-white-cream, red, orange, blue, opaque, black, grey, brown, green, pink, tan, yellow, and pigmented.
Erosion	Fresh, un-weathered, initial change, degree of cracking, weathering, grooves, surface irregularity, jagged fragments, linear fractures, subparallel ridges, and degradation.

MPs are generally classified according to their origin (Koelmans et al., 2019) (Table 1). Primary microplastics (PMPs) (Novotna et al., 2019) are artificial industrial products with a particle size of less than 5 mm. They may include remnants of toothpaste, hair gel, cleansing milk, particle air fresheners and usually enter the surrounding environment with the discharge of domestic sewage. Secondary microplastics (SMPs) are derived from meso (5 mm–25 mm)/macro (>25 mm) plastic waste through physical, chemical and biological processes (Novotna et al., 2019). Based on the report of Pan et al. (2019) that the main origin of SMPs is the result of abrasion of paint and plastic products, fragments of plastic waste that have not been properly treated, and MP fibers in discarded fishing equipment and textiles. Research indicates that MPs are ubiquitous in environmental compartments of marine ecosystems, including near-shore sediments, seafloor, water columns, and surface layers (Li, 2019).

Due to the multiple sources, MPs appear in various types (films, spheres, fibers, foams, and particles shown in Table 1) (Adam et al., 2019). Among samples obtained from the Pacific Northwest (Xu et al., 2018), 39.7% were spheres, 24.7% were films, and 8.9% were fibers. The shape of MPs is influenced by the following conditions (Pan et al., 2019): (1) initial form of plastic; (2) surface degradation and erosion processes (mechanical wear, photodegradation and biological activity); (3) residence time at sea (indicator: rough surface degree, cracking and brittleness). Previous investigations have shown that the origin and pathway of MPs are inextricably linked to their shape. For example, in the study of the Yellow Sea (Xu et al., 2018), it was found that the main shape of MPs was film and fiber, accounting for 58.1% and 39.1%, respectively. This is since fibers come from offshore sources and the fragmentation of marine floating debris. Films are mainly derived from agricultural production and plastic bags used in daily life. Also, the most popular fishing tools (plastic net and rope) release fiber MPs. According to Xu et al. (2018), MP fibers accounted for 93%, 72%, 75%, 90% and 90.07% in sediment samples from the Chinese Yangtze River estuary, Singapore, Slovenia, South Africa, and Croatia, respectively. Evidence suggests frequent industrial and aquaculture activities discharge tons of wastewater carrying MPs (Pan et al., 2019). Ziajahromi et al. (2017) reported that PET micro-fibers induced stronger toxicity effects than PE micro-beads in *Ceriodaphnia dubia*. However, Zimmermann et al. (2020) reported that shape is not the driving factor for toxicity. So, this may be different when investigating particles with irregular shapes (e.g., beads vs fibers).

The main colours of MPs (Table 1) in surface seawater and sediment samples are transparent and white, while multicolour and black are less prevalent (Pivokonsky et al., 2018). In the Pacific Northwest, the main colour of MPs is white (57.4%), followed by transparent (22.8%), green (6.6%), black (6.4%), blue (2.8%), yellow (2.4%), and purple (1.5%) (Pan et al., 2019). It is in line with 94% of current translucent and light-coloured marine litter in the Sargasso Sea, 82–89% in the South Atlantic, and 72% in the North Pacific (Xu et al., 2018).

The diversity of MP colours indicates that it may have a colour similar to that of natural marine foods. Novotna et al. (2019) report that animals always tend to ingest MPs similar in colour to prey; colour is an important factor affecting the intake of MPs by marine organisms. For example, turtles often die due to the inadvertent eating of transparent and white plastic fragments, and fibers are often eaten by fish (Koelmans et al., 2019). So, MPs can confuse natural prey and predator's behaviour, causing marine biomes to ingest MPs of specific colours (especially white and transparent). Also, colour is a significant index of residence time and weathering degree of plastic on the ocean surface. For example, the degree of yellowing or blackening indicates the carbonyl index's increase, the degree of aging or degradation (Xu et al., 2018). Presently almost all MPs show a faint and faded hue (Rist et al., 2018), which means that MPs are transported to the ocean and undergo long-term weathering, degradation, and aging. Also, there are numerous measures to describe erosion status used in the characterisation of MP, including the freshness, degree of cracking, weathering, irregularity, and degradation.

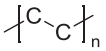
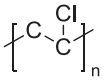
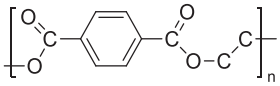
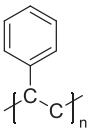
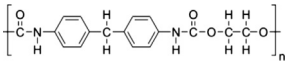
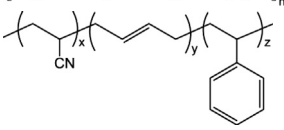
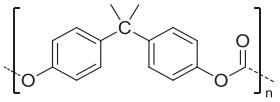
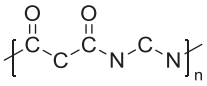
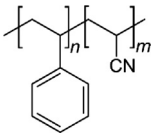
By analysing Raman spectroscopy (RS) characteristic peaks of MPs collected from the Pacific Northwest, seven kinds of polymers commonly found as MPs are identified (as shown in Table 2), including Polyethylene (PE) (58%), Polypropylene (PP) (36%), Polyamide (PA) (3%), Polyvinyl chloride (PVC), Polystyrene (PS), rubber, and polyethylene terephthalate (PET). The polymers found as MPs at the estuary of Yangtze River were PE (82.4%), PP (9.1%), and PVC (6.5%) (Xiong et al., 2018), while remaining polymers account for less than 3%. According to a survey by Pivokonsky et al. (2018), PE is widely used in agricultural films, food packaging films, plastic bottles, and plastic bags, while PP is commonly used in plastic containers, food packaging, carpets, and pipes. In addition, since PE and PP have a lower density than water, they are often transported by ocean currents. In addition to these, other types of MPs that have appeared in reports about marine ecosystems include Polycarbonate (PC), Poly(methyl methacrylate) (PMMA), Polyurethane (PUR), nylon, synthetic (vulcanised) rubber (including tire wear products), and synthetics fiber (Pan et al., 2019). Although MPs have been defined, there is still controversy about whether particles in silicone foam, paints, and coatings contribute to MP levels.

Due to the similarity between the composition of the plastic pollution source and samples collected from the adjacent seas, the concentration of MPs tends to gradually decrease with distance away from industry sources. For example, the content of MPs in samples obtained in Japan's coastal areas is in line with the report on plastic production materials provided by the Japan Plastics Industry Association (Pan et al., 2019). PP and PET are increasingly used in the textile industry for clothing, nonwovens, carpets, and wastewater from the textile and apparel industries may enter the marine environment through rivers. For example, sediments from the estuary of Mexico and Hong Kong have relatively high levels of PP and PET (Cheung et al., 2018). Due to adhesion by microorganisms and other organisms, a proportion of MPs is deposited on the seafloor, including nylon, a synthetic plastic commonly used in clothing and fishing gears, which has a density greater than water (Li, 2019). Sediments from southern Portugal and the Yangtze River estuary are also reported to have high nylon levels (Pivokonsky et al., 2018; Cheung et al., 2018).

1.3. Reporting guidelines and regulations

In September 2017 (Zhu et al., 2018), the Western Pacific Sub-Committee of the UNESCO Intergovernmental Oceanographic Commission (UNESCO-IOC/WESTPAC), the North Pacific Ocean Scientific Organization (PICES), and the Association of Southeast Asian Nations (ASEAN) held international conferences to organise working groups to study marine plastic pollution. In response to the call (Xiong et al., 2018), some member states of the EU, China, US, UK, South Korea, Canada, and Indonesia have enacted relevant laws and taken action to dispose of floating garbage on the coast and prohibited plastic particles in cosmetics (Table 3). In a report released on World Environment Day in June 2015 (Li, 2019), the United Nations Environment Programme proposed that all countries and regions should phase out and ban plastic microbeads for personal care products and

Table 2
Properties of main microplastic polymers found in global aquatic environments.

Polymer type		Characteristics (g cm^{-3}) (Lusher et al., 2017)	Monomer (Lithner et al., 2011)	Chemical structure (Urbanek et al., 2018)	Main use (Lusher et al., 2017)	Approx. global production million tonnes $year^{-1}$ (Lithner et al., 2011)
Polypropylene (PP)		Low density, 0.85–0.94	Propylene		Reusable food containers and packaging, bottle caps, drinking straws, laboratory equipment	45
Polyethylene (PE)	Low-density PE (LDPE)	Low density, non-biodegradable, most common plastics, 0.92	Ethylene		Food wrap film, shopping bags, water pipes	39
	High-density PE (HDPE)	High density, non-biodegradable, 0.96			Toy, milk bottles, pipes, plastic bags, detergent and oil bottles, cable insulation	32
Polyvinyl chloride (PVC)		High density, non-biodegradable, 1.38–1.50	Vinyl chloride		Pipes, floors, window frames, shower curtains, car seat covers, raincoats, bottles, visors, shoe soles, garden hoses, electricity pipes	37
Polyethylene terephthalate (PET)		High density, 1.38–1.41	Terephthalic acid, Ethylene glycol		Plastic Soda beverage bottles and packaging, processed meat packages, peanut butter/jam jars, pillow and sleeping bag filling, textile fibers	33
Polystyrene (PS)		1.04–1.08	Styrene		Fast food container, disposable plastic cups and lids, foam (i.e., "Styrofoam"), CD crystal cases, service ware, packaging materials, laboratory ware, electronic uses	13
Polyurethane (PUR)		Very low density 0.40–0.60	Di/tri-isocyanate, polyol		Upholstery, sports mats, packaging bags.	9
Acrylonitrile-butadiene-styrene (ABS)		1.02–1.08	Acrylonitrile, 1,3-Butadiene, Styrene		Automotive applications, pipes	7[a]
Polycarbonate (PC)		1.20–1.22	Bisphenol A		Construction materials, medical equipment, reusable beverage bottles, CDs, DVDs, street and car lights, sky-lights, baby bottles, roofs of greenhouses, glasses lens, water pipes	2.1
Polyamide (nylon, PA)		High durability and strength, 1.12–1.15	Adipic acid		Textile, carpets, sportswear, small bearings, windshield wipers, water-hose nozzles, helmets, racehorse shoes, inks, rainwear	1.2
Styrene acrylonitrile (SAN)		1.06–1.10	Styrene, Acrylonitrile		Cosmetic containers, ballpoint pens, lighters	0.5[a]

[a] Very varying uncertain data.

cosmetics. The EU and China have implemented this policy (Xiong et al., 2018). Around the world, many governments have developed policies and regulations to ban and tax the sale of lightweight plastic bags to reduce lightweight plastic bag use (Zhu et al., 2018). In 2016, China launched the "Marine Microplastics Monitoring and Ecological Environmental Impact Assessment Technology Research" project to standardise marine MPs monitoring research methods and implement research on the migration diffusion mechanism of offshore MPs and the ecotoxicological effects of MPs (Zhu et al., 2018). In 2018, the EU introduced new regulations in Directive 2019/904 on reducing the environmental impact of common plastic products and promoted fishing waste schemes to encourage voluntary waste collection by trawlers.

Moreover, some non-governmental organizations have carried out a series of actions to treat marine plastic pollution. The existing international marine plastic waste collection and treatment technology is represented by the Ocean Cleanup project in the Netherlands (Li, 2019), which has large international recognition, but its actual effectiveness has yet to be verified. The Seabin sub-project (Li, 2019) has achieved excellent results but on a limited scale by placing bins at fixed points on the dock. To further solve the MPs problem, it is necessary to combine efficient governance actions, extensive media reports, the positive response of large enterprises, and promotion by government agencies.

The first attempt to coordinate sampling, processing and analysis of MPs in marine ecosystems has been made in the European MSFD "Guidelines for Monitoring Marine Garbage in the European Seas" (Li, 2019). As a result, a unified, standardised protocol for MP monitoring methodologies for marine and freshwater ecology has been proposed, which presents the level of pollution and makes results comparable between studies.

According to the United States Environmental Protection Agency's (EPA) Sustainable Materials Management: Non-Hazardous Materials and Waste Management Hierarchy (EPA US, 2020), the reduction or elimination of MP pollution requires a four-tier approach (Fig. 1), including source reduction and reuse, recycling and composting, energy recovery, treatment and disposal. In Ireland, Gdara et al. (2020) collated relevant regulations/policies and the level of human exposure to MPs and NPs, presented evidence on three main potential routes of exposure to MPs and NPs, and reviewed the evidence of the possible health effects of environmental exposure to MPs and NPs on human health based upon in vivo and

Table 3
Guidelines and regulations for MPs in water, food, air, and litter.

Guidelines and regulations	Source	Country	Scope and major aim	Definition of MPs	Reference
Microbeads in toiletries regulations	Freshwater and marine	Canada	Prohibit the manufacture and distribution of toiletries containing MPs in the national territory.	• Plastic microbeads • <5 mm	Government of Canada, 2017
2015 Wisconsin act 43	No mention	USA	Synthetic plastic microbead	• Non–biodegradable • Solid plastic particle • <5 mmUsed to remove or clear skin in rinse-off products	Wisconsin Act 43 (State of Wisconsin, 2015)
2014 Public act 098 – 0638	No mention	USA	Synthetic plastic microbead	• Non–biodegradable • Solid plastic particle • <5 mm • intentionally addedUsed to remove or clear skin in rinse--off products	Public Act 098–0638 (State of Illinois, 2016)
An act to add Chapter 5.9 (commencing with section 42360) to part 3 of Division 30 of the public resources code, relating to waste management	Marine surface water	USA	Prohibits the market supply of personal care products with microbeads additives without toothpaste and prescription drugs.	• <5 mm • Solid plastic particle • Intentionally added • Persistent organic compounds	Assembly Bill No. 888 (State of California, 2015)
Draft ordinance amending the chemicals products (handling, Import and export prohibitions) ordinance (1998:944)	Water	Sweden	Prohibits the market supply of cosmetic products with unnatural MPs additives.	• Solid plastic particle • <5 mm • insoluble in water	European Commission, 2017c
Draft sector agreement to support the replacement of microplastics in consumer products	Surface water	Belgium	(a) Prohibits the market supply of cosmetic products with unnatural MPs additives. (b) Promotes manufacturers to limit MPs in product formulations strictly.	–	European Commission, 2017b
Decree prohibiting the placement on the market of rinse-off cosmetic products for exfoliation or cleaning that contain solid plastic particles, provided for in the third paragraph of point III of Article L541-10-5 of the Environmental Code 6.	No mention	France	Prohibits the market supply of cosmetic products with unnatural MPs additives.	• Solid plastic particle • <5 mm • Thermoforming Process	European Commission, 2016
The Environmental Protection (Microbeads) (England) Regulations 2017; The environmental protection (microbeads) (Northern Ireland) regulations 2018; The environmental protection (microbeads) (Scotland) regulations 2018; The environmental protection (microbeads) (Wales) regulations 2018	Water	UK	Prohibits the market supply of cosmetic products with unnatural MPs additives.	• Solid plastic particle • <5 mm • Water-insoluble	European Commission, 2017a, 2018c,d,e
Draft technical regulation banning the marketing of non-biodegradable and non-compostable cotton buds and exfoliating rinse-off cosmetic products or detergents containing microplastics	Water	Italy	Prohibit the manufacture and distribution of exfoliating or rinse-off cosmetics containing MPs in the national territory	–	European Commission, 2018b
Directive (EU) 2019/904 of the European Parliament and of the council of 5 June 2019 on the reduction of the impact of certain plastic products on the environment	Litter in soil, water, and air	EU	(a) Microplastics are not directly within the scope of this directive. (b) The EU encourages manufacturers to limit MPs in product formulations strictly. © Strictly prohibit single-use plastic product placing on the market and promote sustainable/biodegradable alternatives to minimise the sources of MP pollution.	–	Tajani and Ciamba, 2019
Commission decision (EU) 2017/1219 of 23 June 2017 establishing the EU ecolabel criteria for hand dishwashing detergents, hard surface cleaning products and industrial and institutional laundry detergents	Litter in water	EU	MPs must not be included in product formulations regardless of concentration.	• <5 mm • Based on particles • Insoluble macromolecular plasticBy polymerization processes, chemical modification or microbial fermentation	Vella, 2017
Waste minimisation (microbeads) regulations 2017	No mention	New Zealand	Synthetic, non-biodegradable plastic beads	• Water-insoluble • Plastic particles • <5 mm	Reddy, 2017
An assessment of the sale of microbeads in personal care and cosmetic products currently available within the Australian retail (in-store) market	No mention	Australia	Products, distributor and quantities of products containing microbeads	• Solid manufactured plastic particles • Water-insoluble and non--degradable • <5 mm (typical around 100–300 μm)	O'Farrell and Kate (2018)

in vitro experimental studies. Also, they propose three measures to implement from the plastics strategy for MPs control: 1) implement an effective regulatory and environmental compliance system; 2) provide sound, targeted and timely environmental data, information and assessments; 3) advocate for clean and sustainable production and living environment for all.

Meanwhile, the strategy introduces a wide range of legislative and non-legislative measures to address the issue of MPs in a diversified manner. According to the strategy, by 2030, all plastic packaging will be reusable or recyclable in a cost-effective manner through: 1) improved economics and quality of plastic recycling; 2) an end to plastic waste production and littering; 3) investment and promotion of recycling solutions; 4) global action (O'Callaghan-Platt and O'Brien, 2018). In addition, O'Callaghan-Platt and O'Brien (2018) have developed a citizen science (CS) protocol to increase awareness of plastic use levels and promote behavioural change to reduce plastic waste. The protocol captures data on daily plastic use through two formats, the household programme and school programme, these adopt a technology-mediated plastic waste audit tool and use

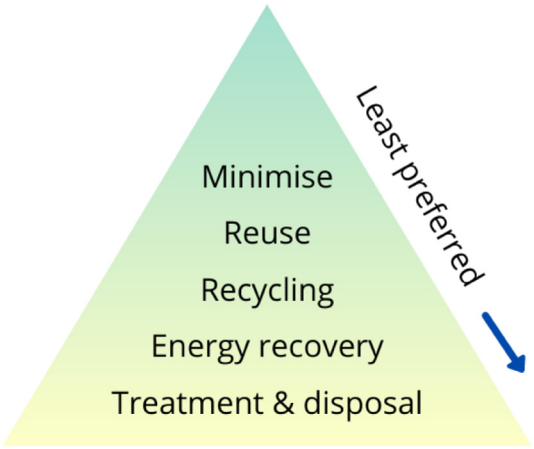


Fig. 1. Non-hazardous materials and waste management hierarchy.

feedback from participants to measure the programme's effectiveness (O'Callaghan-Platt and O'Brien, 2018). Furthermore, with the deepening of worldwide awareness of the MP pollution problem, research and investigation in different sea areas worldwide will make the risk assessment of MPs more viable and comprehensive. In addition, with the enactment of guidelines and regulations (Table 3) and the implementation of new techniques, contamination by MPs should be significantly reduced.

2. Identification methods

2.1. Sampling and separation

2.1.1. Sediment

Due to a lack of uniform guidelines, studies on sampling locations (area along the tidal line and below intertidal zone), sampling depth (0 to 32 cm), number of beaches analysed (1 to 300), sampling tools (tweezers, spoons, core diggers, trowels) and sample weights (0.15 kg to 10 kg) vary widely (Li et al., 2018). MSFD guidelines (Ivleva et al., 2017) recommend a sampling depth of 5 cm, with intervals greater than 5 m with repetition more than 5 times. In general, fractional sampling of between 1 μm–500 μm and 500 μm–5 mm particles are suitable for further analysis (Ivleva et al., 2017).

Quantifying the number of MPs in sediment is usually based on density separation, which extracts all particles from sediment (Di and Wang, 2018). Different sampling methods are used according to the selected matrix. Usually, selective, batch or volume reduction methods are used. Selective sampling is suitable when samples contain a relatively large number of big MPs since a combination of the naked eye and visual recognition is used in direct identification and selection (Li et al., 2018). Batch sampling is suitable for an entire sample volume of sediment (Di and Wang, 2018). Reduced volume sampling (Ivleva et al., 2017) is performed by filtering or sieving to minimise the sample volume (density separation and removal of the organic matrix), and is common in both liquid and sediment analysis.

2.1.2. Surface water

Collecting MPs from surface water usually involves the use of manta trawls or neuston nets, while plankton nets and continuous plankton recorders (CPR) are often applied in standing water bodies (Wang and Wang, 2018). Although mesh processing flow is large, and the flow rate is rapid, the choice of mesh size, omission of smaller MPs, and mesh clogging limit its application (Wang et al., 2018). Meanwhile, the number of MPs increases exponentially as the plastic fragments into MP of decreasing particle size; many particles are lost during sampling of the upper compartment in the ocean. Therefore, MSFD guidance recommends a mesh size of 333 μm, a total net length of 6 m and a fishing time of 30 min (Li et al., 2018). Recently, a Nigerian team (Zhu et al., 2018) released a new inlet water filtration technology that limits mesh size to 50 mm or 10 mm based on content in the sampling area (for high suspended matter, 50 mm aperture is used to prevent clogging). Especially for bulk samples, MPs (density range 0.90–2.30 kg l^{-1}) (Zhu et al., 2018) must be separated from other impurities (sand or stone) (density is about 2.65 kg l^{-1}). Samples can be separated with a certain density liquid (usually saturated salt solution, such as non-toxic and saturated sodium chloride (NaCl) solution with a density of 1.2 kg l^{-1}) (Zhu et al., 2018), and then shaken, stirred and aerated. When the mixture settles, low-density particles float, high-density particles sink to the bottom; MPs are separated by filtering the supernatant (Cheung et al., 2018).

2.2. Pre-treatment

Depending on the type of components, sample preparation with HNO_3 or hydrogen peroxide (H_2O_2) (seafood, honey, and salt) is required to digest organic matter (Ivleva et al., 2017). In addition, MPs in opaque organisms (bivalves and fish) require pre-treatment. In bivalve samples, MPs are extracted using a combination of boiling for 5 h after pre-treatment in HNO_3 overnight. Also, MPs in fish samples are extracted using a combination of rinsing in Sodium hypochlorite (NaClO)/HNO_3 during overnight pre-treatment followed by ultrasonication in methanol (MeOH) (Ivleva et al., 2017). However, all of these chemical methods have the potential to decompose the original MPs.

In laboratory studies, the size and composition of biologically ingested MPs are defined variables. Hence, particles are usually separated from samples and analysed on a filter surface. According to MSFD guidelines (Ivleva et al., 2017), samples are dissected under a microscope with scissors, and all particles of unnatural shape and colour are counted before spectral analysis. Also, large MPs need to be picked and cleaned with tweezers before identification.

2.3. Isolation and digestion

Isolation processes for MPs from sediment samples, consisting entirely of biomass, most commonly include: manual sorting, density separation, chemical digestion and enzymatic digestion (Munno et al., 2018). However, all of these methods have the disadvantage of being expensive and difficult to operate. In practice, reducing the sample size not only reduces the amount of material required for extraction but also reduces the cost, in addition, further improvements to standardised procedures are still required (Nabi et al., 2022). Chemical digestion includes acid digestion, alkaline digestion and wet hydrogen peroxide digestion. Currently, alkaline (potassium hydroxide (KOH) 224 g/L) digestions are often combined with wet hydrogen peroxide digestions for fish tissue processing (Nabi et al., 2022). However, all chemical methods have been shown to affect the recovery rate of MP particles. Enzymatic digestion is the method with the highest digestibility (>97%), however, this method is more sensitive to temperature and needs to be controlled within the room temperature range (Nabi et al., 2022). In addition, the new isolation technique of acetonitrile digestion membranes (with the use of glass fiber filters, nitrocellulose filters or cellulose acetate membranes to filter the post-digestion solution) has been shown in the study by Malafaia et al. (2022) to be a reproducible digestion procedure with good microplastic recovery and also prevents particle loss due to adhesion, resulting in good dispersion of MPs in the resulting analytical solution (Malafaia et al., 2022).

2.4. Purification

To filter out higher-density poisonous polymers, such as polyvinyl chloride (PVC) or polyoxymethylene (POM) (Ivleva et al., 2017), a Sodium polytungstate (SPT, $3Na_2WO_4{\cdot}9WO_3{\cdot}H_2O$) solution with a density of 1.4 kg l^{-1} is typically used, with a low separation efficiency (Mintenig et al., 2019). So, the best choice is to use a mixture of SPT saturated solution with a density of 3.1 kg l^{-1}, calcium chloride ($CaCl_2$) solution with a density of 1.3 kg l^{-1}, sodium iodide (NaI) solution with a density of 1.8 kg l^{-1}, and zinc chloride ($ZnCl_2$) solution with a density of 1.6 kg l^{-1} (cost-

effective but toxic, requiring careful recycling after use) to separate MPs (Ivleva et al., 2017). However, the NaCl solution is still recommended by MSFD guidelines due to its wider range of applications, low cost and disruption (Li et al., 2018).

There are several extraction methods to choose when using NaCl solution, from ordinary stirring (conventional setting) to the combination of fluidization and flotation, with different purification rates. For large MP particles (>1 mm), the purification rate by stirring is between 80% and 100%. However, only 40% of small MP particles (<1 mm) are separated using $ZnCl_2$ solution (Mintenig et al., 2019). The fluidizing samples in NaCl and then flotation in NaI have good purification results (−8–99%) (Mintenig et al., 2019), but the type of polymer plays a decisive role. Recently, $ZnCl_2$ has been used in a newly developed instrument, "Munich Plastic Sediment Separator" (MPSS), with mass purification of between 96% to 100% (large MPs) and 96% (small MPs), which saves time and money (Mintenig et al., 2019).

Samples are generally purified before quantitative analysis to improve and simplify identification. Two purification methods that have been applied so far are chemical degradation and enzymatic degradation of organic substrates. In chemical degradation (Li et al., 2018), samples are treated with different chemicals to purify them. Generally, a 30% H_2O_2 solution or a mixture of H_2O_2 and sulfuric acid (H_2SO_4) is used. Also, Li et al. (2018) have tried to combine deionised water or sodium lauryl sulfate (SDS) solution and an ultrasonic bath. Enzymatic degradation processes use MPs mixed with technical enzymes (lipase, amylase, protease, chitinase, and cellulase) to achieve purification (Li et al., 2018). However, it still needs research to determine whether it is feasible for routine analysis.

2.5. Identification and quantification

There are various methods for particle identification and quantification, as shown in Table 4 and Fig. 2. After visual recognition using microscope pre-sorting of particles, researchers generally use spectroscopic methods, Fourier Transform Interferometer (FTIR) or Raman Spectroscopy (RS), to identify polymers (Li et al., 2018). FTIR and RS are non-destructive technologies. At the same time, Pyrolysis Gas Chromatography Mass Spectrometry (Py-GC/MS) and Thermal Extraction and Desorption coupled with Gas Chromatography Mass Spectrometry (TED-GC/MS) analysis are thermal decomposition technologies. Both techniques identify polymer types and

Table 4
Summary of common detection techniques of MPs.

Method		Methodology	Suitable MPs size	Advantages	Limitations	References
Visual recognition methods	Microscopic counting	Pre-treat samples and count directly.	–m - mm (stereomicroscope)	Suitable for large MPs, quickly and low cost.	High error to distinguish between natural and MPs.	Li et al., 2018; Looder and Gerdts, 2015; Hidalgo-Ruz et al., 2012
Spectroscopic analysis methods	Fourier Transform Interferometer (FTIR)	Plastic polymers have specific interferometer (IR) spectra. Samples are irradiated in the defined range of IR, and the excitable vibrations depend on the composition and molecular structure.	(Attenuated total reflection) ATR-FTIR (>500 μm); (micro) μ-FTIR (>20 μm) couple with microscopy.	Non-destructive, well established, fast and quite reliable; focal plane array, FPA (fast acquisition of several thousand spectra within an area); shorten analysis time after one measurement.	Just suitable for IR active transparent samples (>20 mm); expensive specific instruments; experienced researchers for operation and data processing; difficulty in data interpretation due to effect on the environmental matrix (biofilm); must pre-treat sample to eliminate IR active water.	Song et al., 2014; Besseling et al., 2015; Qiu et al., 2016
	Raman Spectroscopy (RS)	The interaction of the sample molecules with the irradiated laser light; compared to the irradiating laser results, there are differences in the back-scattered light frequency; detection Raman shift leading to substance-specific Raman spectra.	1 to 20 μm; RS couple with microscopy (>1 mm)	High spatial resolution and low sensitivity to water; suitable for opaque and dark particles; fast chemical mapping for fast and automatic data collection and processing.	Great interferences of fluorescence from biological, organic, and inorganic impurities; sample purification before analysis; need appropriate Raman parameters (wavelength, laser power, and photobleaching); time-consuming.	Cole et al., 2013; Collard et al., 2015; Imhof et al., 2016; Qiu et al., 2016; Wiesheu et al., 2016; Zhao et al., 2017
	Scanning Electron Spectroscopy (SEM)	Interaction of electron beams with samples to generate images to measure secondary ions.	>100 nm.	High-resolution image.	Coat samples at high vacuum; no detailed identification information.	Li et al., 2018
Chromatographic methods	Pyrolysis Gas Chromatography Mass Spectrometry (Py-GC/MS)	Thermally treat samples under ambient conditions; trap released gaseous and transfer to a gas chromatography column coupled to a quadrupole mass spectrometry; compare spectra with a common plastic-type database.	>500 μm (handled by tweezers).	Analyse samples with organic plastic additives per run without solvents; avoid background contamination; sensitive and reliable.	One certain weight particle per run; the database is only for PE and PP.	Fabbri et al., 2000; Fabbri, 2001; Fries et al., 2013; Nuelle et al., 2014
	Thermal Extraction and Desorption coupled with Gas Chromatography Mass Spectrometry (TED-GC/MS)	Combine thermogravimetric analysis coupled with solid-phase extraction and thermal desorption gas chromatography mass spectrometry to identify polymers in environmental samples directly.	>500 μm (handled by tweezers).	Robustness, especially regarding impurities and the relatively short analysis time.	Used only for the identification of PE and PP; conclusions only for the total mass fraction of involved polymers; must combine with a concentration method.	Dümichen et al., 2015
	Liquid chromatography	Dissolve samples by selected solvents; measure different molar mass distribution by size exclusion chromatography.	100 μm–1 mm; sufficient sample	High recoveries.	Inability to determine physical properties; restriction on selected polymer types (PS and PET); Small samples per run.	Hintersteiner et al., 2015; Elert et al., 2017;
Other methods	Tagging method	MPs adsorb hydrophobic dye onto their surfaces and fluoresce when exposed to blue light.	>200 μm.	Simple, low cost, and fast screening.	Stain organic debris by the dye; overestimation of MPs.	Shim et al., 2016

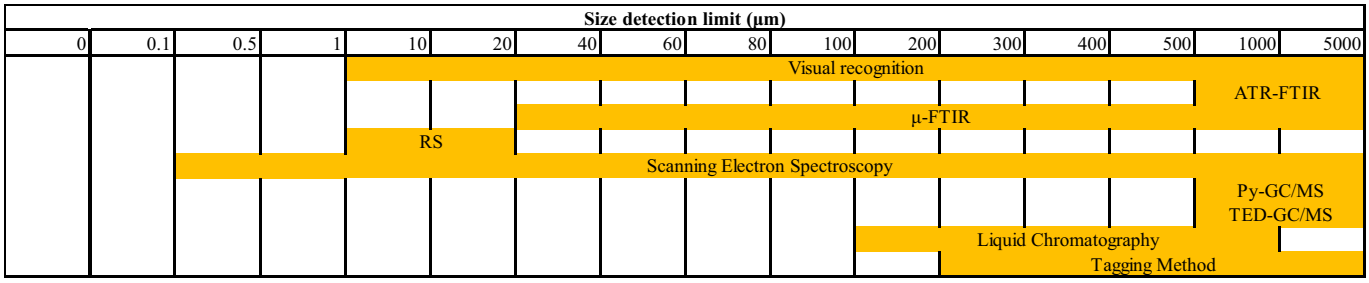


Fig. 2. Size limit of different detection techniques of MPs.

additives. RS and FTIR coupled with an optical microscope are commonly used to provide information on particle size and number, while TED-GC/MS is used to derive the total mass fraction of polymer involved (Mintenig et al., 2019). In the future, thermal analysis will be used to screen samples and analyse contamination levels after determining the number and size of particles using spectroscopic analysis. With continuous development and improvement of various technologies, analytical techniques will be faster, more sensitive and easier to operate in the future.

In addition to these methods, thermal imaging and differential scanning calorimetry (TGA-DSC) (Ivleva et al., 2017), fluorescence microscopy imaging, coherent anti-Stokes Raman scattering (CARS) (particle positioning of known MPs), and 3D Raman imaging (identification of unknown MPs) are also common identification methods (Li et al., 2018).

2.5.1. Visual recognition

There is a need to distinguish synthetic polymer particles from natural substances before identification and quantification. Most experiments distinguish MPs using the naked eye (large MPs) or dissecting microscope (small MPs) (Li et al., 2018). However, these methods rely heavily on the observer's judgment, and even experienced researchers find it hard to distinguish between MPs and natural organic matter, including quartz particles, plant fragments or animal tissues (Li et al., 2018). Visual recognition can lead to errors, especially for smaller particles, so optical identification is necessary. Also, the identification of fibers often uses Bengal Red (4,5,6,7-tetrachloro-2,4,5,7-tetraiodofluorescein) to dye natural organic particles (cellulose fibers) to differentiate from unstained fibers (Ivleva et al., 2017). However, it still requires spectral analysis (FTIR or RS) to identify undyed fibers' synthetic properties. Also, the polymer type of MPs is very hard to identify using visual recognition methods.

After the standard morphological identification of MP samples, spectroscopic analysis is frequently used to characterise the polymer types in MPs. In studying the Atlantic sea surface (Zhu et al., 2018), visual identification was combined with RS for the first time. The overall recognition success rate of particle MPs was 68%, 63% for small MPs, and 83% for large MPs (Zhu et al., 2018). The overall success rate for the identification of fiber MPs was 75%. In addition, coloured MPs have a better recognition rate, with blue being 86%, while only 41% and 42% recognition rate is observed for white and black MPs, respectively. The study of Laurentian Great Lakes (Ivleva et al., 2017) showed a 20% error rate in determining MPs by energy-dispersive X-ray spectroscopy (EDX) analysis when MPs were less than 1 mm. Ivleva et al. (2017) analysed MP content in bivalves using FTIR spectroscopy; many spherical particles were identified as aluminum silicate with an error of up to 70%. Visual identification and spectral methods (infrared (IR), RS, and EDX) are always combined to obtain more accurate results.

2.5.2. Spectroscopic analysis

2.5.2.1. Fourier Transform Interferometer (FTIR). FTIR generates a spectrum by recording a stepwise shift of wavelengths that simultaneously collect all wavelengths and use Fourier transformation to process the data (Ivleva et al., 2017). FTIR set three operating modes, transmission, reflection, and attenuated total reflection (ATR) (Ivleva et al., 2017). Usually, large MPs are separated with tweezers and analysed by ATR-FTIR (Ivleva et al., 2017). Samples placed on an ATR crystal are irradiated with an attenuation wave on the surface. This method quickly and accurately identifies and analyses large MPs. Since single particle analysis is difficult for small MPs, a combination of FTIR and optical microscopy is used with three possible modes (transmission, reflective and attenuated total reflection mode). MPs are usually collected on a filter for analysis. In transmission mode, interferometer-transparent filters (aluminum oxide or special silicon filters) (Ivleva et al., 2017) are used, and effective MP thickness is detected depending on optical characteristics of MPs. Transparent PE particles (Ivleva et al., 2017) are easier to analyse than coloured or dark particles in this mode. When in reflection mode (Ivleva et al., 2017), the signal is often disturbed by light scattering errors, and analysis depends on the morphology of MPs. However, these disadvantages can be avoided by using micro-ATR-FTIR (Ivleva et al., 2017), which directly works on the filter and reduces errors. Nevertheless, this technique makes it necessary for the surface of each particle to interact with the crystal, which is time-consuming.

Recently, grid mode detection technology based on the focal plane array (FPA) (Mintenig et al., 2019) was used to improve FTIR imaging, allowing the instrument to quickly capture and measure thousands of spectra on an area to analyse an entire sample. However, this technique is greatly affected by the environmental matrix, and samples must be kept dry while the analysis is slow (11 μm diameter filters take 10.75 h) and is not suitable for processing large amounts of data (Ivleva et al., 2017). Furthermore, although FTIR effectively and accurately analyses samples and distinguishes various polymer types, the interferometer resolution is limited by diffraction (Zhu et al., 2018). So smaller particles (size less than 20 μm or thickness less than 5 μm, as shown in Fig. 2) cannot be analysed. Therefore, FTIR has only been applied in a few studies.

2.5.2.2. Raman Spectroscopy (RS). When a monochromatic Raman light source (laser) illuminates the sample, some photons are inelastically scattered (Oßmann et al., 2018). The resulting spectrums are compared with a reference database to receive information about the molecular vibrations of samples. RS is often coupled to a standard optical microscope as the light source used in RS is usually in the visible range (Oßmann et al., 2018). Although RS has been used in MP studies of marine and freshwater ecosystems, most studies have only used it to analyse large particles or small subsamples (the smallest size is about 1 μm, as shown in Fig. 2) (Ivleva et al., 2017). Compared with FTIR, RS has the advantages of high spatial resolution (down to 1 μm) and insensitivity to water, enabling more accurate chemical mapping. Even in transmission mode, RS also can analyse dark opaque particles (Oßmann et al., 2018). Small MPs (1 μm to 20 μm) have the greatest impact on aquatic life; the only suitable way to identify them is by using RS. However, if the particle size is less than 1 μm, it is quite time-consuming. Furthermore, the latest autofocus systems do not meet requirements for focusing lasers on MPs for RS mapping (Ivleva et al., 2017).

Large MPs are processed directly with tweezers, and small MPs are handled using filters when using RS for MP identification. Filter analysis is time-consuming and labour-intensive; thus, statistical information for the entire filter is often provided after examining some filter areas (Ivleva et al., 2017). Measurement time depends on the sample matrix, the overall contamination level, and the researcher's expertise. However, RS is

extremely susceptible to fluorescence interference from microbial, organic (humus), and inorganic (clay mineral) pollutants, hindering MP identification. Therefore, samples must be purified before RS. Meanwhile, the selection of acquisition parameters (Ivleva et al., 2017) (laser wavelength, laser power, photobleaching, measurement time, objective magnification, confocal mode) is critical to solving the issue of the strong fluorescent background.

2.5.2.3. Scanning Electron Spectroscopy (SEM). Chemical and morphological characterisation of MPs using a scanning electron microscope (SEM) equipped with an energy dispersive X-ray microanalyzer (EDAX Genesis) requires the transfer of samples onto a conductive and adhesive carbon tape mounted on an aluminum sample holder (Fries et al., 2013). The detection method in low vacuum mode effectively avoids electrical charging effects and any need for sample coating preparation (Fries et al., 2013).

2.5.3. Chromatography

2.5.3.1. Pyrolysis Gas Chromatography Mass Spectrometry (Py-GC/MS). In 2001, Py-GC/MS (Ivleva et al., 2017) was first applied to identify MPs samples. Then, gaseous compounds produced by individual MPs heat-treated in a test tube (Zhao et al., 2018) were captured in a cold injection system, and compounds were then transferred to GC (gas chromatography) column connected to quadrupole MS (mass spectrometry) after thermal desorption was completed. Finally, comparing the obtained pyrolysis product spectrum with a common polymers database accurately identified the MPs while providing information on organic plastic additives (OPA) (Zhao et al., 2018). However, because Py-GC/MS only analyses single particles per time, it is unsuitable for routine analysis (Zhao et al., 2018). Furthermore, since it requires tweezers to place particles in a test tube, the smallest particle analysed is about 100 μm (actual treatable particle size is about 500 μm, as shown in Fig. 2) (Rist et al., 2018). In addition, due to its strong matrix effect and sensitivity to impurities, it is generally not used to analyse entire environmental samples (Rist et al., 2018).

2.5.3.2. Thermal Extraction and Desorption coupled with Gas Chromatography Mass Spectrometry (TED-GC/MS). The latest thermal analysis (Ivleva et al., 2017) method, TED-GC/MS, combines thermogravimetric analysis coupled to solid-phase extraction (TGA-SPE) and thermal desorption gas chromatography mass spectrometry (TDS-GC/MS) to identify polymers directly in environmental samples. Under inert conditions (Zhao et al., 2018), 20 mg of environmental samples (including organic components and PE MPs) are firstly placed in a TGA (thermogravimetric analysis) crucible and heated, and then adsorbed on the extraction matrix (sample with polydimethylsiloxane layer) to then be thermally desorbed into gas chromatography–mass spectrometry. This method calibrates and determines PE MPs' weight percentage in different environmental matrices (soil, bivalve and suspended solids). However, further research is needed to determine a method to distinguish different polymers in the mixture. Although this method has to be pre-concentrated to reach the minimal detectable concentration, the robustness of the technique to impurities and the shorter analysis time make it a technique with great potential.

2.5.3.3. Liquid chromatography. Liquid chromatography utilises the different solubilities of various polymers in specific solvents to quantify the level of MPs, including tetrahydrofuran and hexafluoroisopropanol, to dissolve PS and PET, respectively (Li et al., 2018). Samples are concentrated to obtain the polymer extraction and analysed by comparing the data obtained from the analysis results with the data from the size exclusion system (Li et al., 2018). This method does provide a high recovery rate and an accurate quantification of the level of MPs, although the size of MPs cannot be determined (Li et al., 2018). However, this method is still at the experimental stage and has not been applied to real environmental water samples. Therefore, further research is needed to verify its capabilities.

2.5.4. Tagging method

Tagging methods are commonly used for visualization and counting in quantitative studies of MPs. By adsorbing a hydrophobic dye onto the surface of samples, MPs fluoresce when irradiated by blue light (Shim et al., 2016). This screening method has the advantage of being simple, fast, and low-cost. However, the levels of MPs may be overestimated as they may also stain additional organic debris (Shim et al., 2016).

3. MP in marine ecosystems

3.1. Levels and distribution of MPs in the marine environment (sediment and water)

MPs have been found on the coastal sediment and surface water of all five continents. MPs were found at 18 marine monitoring points in the Northwest Pacific ranging from 6.4×10^2 to 4.2×10^4 items km^{-2} in surface water, where MP abundance in surface water in the South Pacific Ocean and the Northwest Atlantic Ocean is about an order of magnitude lower than that in the Pacific Northwest Ocean (Cai et al., 2018). It appears that MP pollution is widespread in the entire Northwest Pacific, especially in East Asian waters around Japan where MPs concentration is 1.7×10^6 particles km^{-2} in surface water and −2–144 particles kg^{-1} in intertidal sediments, which is about 20 times higher than in deep-sea sediments (1–5 to 6.7 particles kg^{-1}) (Wang et al., 2018), due to the surrounding area with highly industrialised cities, densely populated coastlines, subtropical cyclones, and Kuroshio currents (Xu et al., 2018).

Although MPs in the surface water are still below 6650 particles m^{-3} on a global average, some areas are experiencing severe pollution, potentially threatening ecosystem health, and this includes coastal waters (>100,000 particles m^{-3}) around the Arabian Gulf, the Mediterranean sea (Xu et al., 2018) and narrow straits (9200 particles m^{-3}) around Queen Charlotte Fjord (Pan et al., 2019) due to limited exchanges with oceans as a result of being semi-closed marine ecosystems.

It is reported, by 2100, the number of floating MPs globally will increase to between 2.5×10^7 to 1.3×10^8 t with a concentration of between 9.6 and 48.8 particles m^{-3}, which means that the number of MPs could grow 50 times in nearly a century (Wang et al., 2018; Cheung et al., 2018). According to the forecast of Di and Wang et al. (2018), the worldwide average deep-sea sediment concentrations will be up to between 73 and 373 particles kg^{-1} by 2100. Also, about 5% of marine MPs are washed ashore per year, which will eventually cause environmental concentrations to approach, or even exceed, safe concentrations in the mid-21st century (Pan et al., 2019).

The distribution of MPs is non-homogeneous in the North Yellow Sea, Bohai Sea, Yangtze River estuary, South China Sea, Northeast Atlantic Ocean, Emerald Bay, Northeast Pacific Ocean, Mediterranean Sea, Tyrrhenian Sea, Adriatic Sea, and Baltic Sea (Xiong et al., 2018; Cai et al., 2018; Pivokonsky et al., 2018; Sun et al., 2018; Xu et al., 2018), indicating marine MPs content and distribution is influenced by ocean currents and meteorological conditions, which leads to an uneven distribution of MPs in different sea areas from a diversity of sources (land and other seas).

As Pan et al. (2019) studies have shown, there is a positive correlation between MP contamination levels and population density or river input. The MP abundance in Chinese densely populated areas such as the Bohai Sea and Yellow River Delta (Pan et al., 2019; Sun et al., 2018) is generally high. Up to 80% of MPs in the marine environment originate from land, including rivers, ports and coastal tourism, where wastewater treatment plants and urban runoff are the main sources of MPs (The Lancet Planetary Health, 2017). The other 20% of MPs are reported to originate from fishing (recreation or commercial) and marine industries (oil rigs and aquaculture) (The Lancet Planetary Health, 2017). According to Daly (2018), the west coast of Ireland is sparsely populated, and its offshore Atlantic Ocean is among the few seas worldwide where MPs have not been detected. On the east coast of Ireland, MP concentrations in the surface water of the Irish Sea were as high as 2.46 ± 2.43 particles m^{-3} (Lusher et al.,

2014) due to the impact of a dense human population and developed industry.

3.2. Marine ecosystem concerns

Although the amount of MPs in the ocean is far below critical values, it requires urgent attention due to the exponential growth trend. Because MPs are detected in the food chain from plankton to invertebrate (crustacean, mollusc) to vertebrate (fish), aquatic organisms generally may be exposed to MP contamination (Rist et al., 2018) and varying degrees carry MPs. Many experiments have analysed MPs' effects on marine invertebrates (sea earthworms, sea cucumbers, lobsters, squid, crabs and zooplankton), and a few have analysed effects on vertebrates (fish, seabirds, whales and seal). For instance, for earthworms (Revel et al., 2018), after exposure to MPs (62–1000 mg kg^{-1} of polyethylene), fibrosis, congestion, and inflammatory infiltration were observed. Another experiment by Peters and Bratton (2016) exposed earthworms to polyethylene MPs associated with zinc which showed zinc accumulation and higher zinc desorption (40–60%) in synthetic earthworm guts than in soil (2–15%).

The toxicological risks of MPs ingestion cover three main aspects: particles, polymer additives, and adsorbed pollutants. Laboratory toxicity experiments (Li, 2019) have shown that marine organisms feed on MP particles, resulting in numerous health issues (as shown in Table 5), including organ blockage (digestive tract), bio-storage energy consumption, physical damage (internal abrasions and intestinal adhesions), metabolic disturbances, and even death when the abundance exceeds a critical threshold (Uhrin and Schellinger, 2011; Lusher et al., 2013). Even smaller MPs (<1.5 mm) are more likely to penetrate deep into organs and block them quickly. For example, moderate to severe histopathological changes happen in the intestines of sea bass (*Dicentrarchus labrax*) exposed to PVC MPs for 30 to 90 days through ingestion and intestinal function is completely impaired after exposure for 90 days (Revel et al., 2018). In addition, when European bass was exposed to different concentrations of PS MPs (90 μm, 10,000 to 80,000 particles m^{-3}), it inhibited egg hatching (Lusher et al., 2013).

Meanwhile, their growth rate was reduced, and feeding and innate habits changed. Although the concentration of MPs used in tests is much higher than that found in real environments, indoor toxicology tests show that MPs produce multiple toxic effects on organisms (Koelmans et al., 2019), which cannot be ignored. In particular, given that early life stages of marine life (eggs, embryos, and larvae) are highly vulnerable to waterborne pollutants, MP pollution threatens their survival (embryotoxicity), which may lead to a reduction of fish species (Uhrin and Schellinger, 2011). Fish act as the main pathway to shift MPs to higher nutrient levels, potentially threatening the entire ecosystem.

Furthermore, polymer additives used in production may be leached out after oral ingestion. For instance, heavy metals are often leached from pigment products (Lusher et al., 2013). In addition, MPs have strong adsorption to pollutants in the marine environment, including organic pollutants and heavy metals (Uhrin and Schellinger, 2011). At present, polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH), and dichlorodiphenyltrichloroethane (DDT) have been detected in samples obtained from beaches of the North Yellow Sea (Li, 2019).

According to the in vitro experiments by Piccardo et al. (2021), *Hydropsyche pellucidula* larvae were exposed to PP MPs due to the adsorption of surfactants (superoxide dismutase (SOD), octylphenol ethoxylate (Triton X-100) and malondialdehyde (MDA)) during MP production. This process accelerated the release of superoxide anions (O_2^-) and hydrocarbon metabolism and triggered oxidative stress, raising the concentration of reactive oxygen species (ROS) in larval cells. The oxidative stress to the loss of larval cell biofilm fluidity and increased ion permeability, ultimately disrupting membrane integrity. In addition, the biological activity of proteins and peptides was also affected by surfactants, causing the folding of peptide chains and changes in the surface charge of macromolecules. Even the size, dosage and interaction of MPs with other contaminants can lead to lipid peroxidation and induce oxidative stress (Piccardo et al., 2021).

Table 5
The effect of MPs originating from inhalation and ingestion at various levels of biological organization.

Level of biological organization	Polymer type and size (examples) (Lusher et al., 2017)	Potential impact (Lusher et al., 2017)
Macromolecule	PE 100 nm–30 μm, PS 50 nm–4.7 μm, MMA 1 μm–2 μm, PC 1 μm −55 μm.	DNA damage, genotoxicity, altered gene expression, and protein transcription.
Organelle	PMMA 10 μm	More micronuclei.
Cell	PS 20 nm–4.7 μm, PE 300 nm–10 μm, PS 60 nm–200 nm, PMMA 2 μm–35 μm, PS 20 nm–200 nm.	Cell coagulation, necrosis, apoptosis, altered cellular division, stress response, loss of cell viability, oxidative stress reaction, altered fatty acid metabolism, and increased calcium ions.
Tissue	PE 600 nm–21 μm, PMMA 1 μm–35 μm.	Inflammation, fibrosis, and bone osteolysis.
Organ	PMMA 1 m–10 μm	Lesions and carcinogenesis.
Individual	PS 60 nm–200 nm, LDPE 300 m–10 μm	Changed feeding, increased metabolic demand, and redistribution of energy reserves
Population	PS 20 nm–200 nm	Slower growth, reduced fertility, reduced offspring survival, and reduced population.

4. MP in food systems

4.1. Levels of MPs in food systems

4.1.1. MPs occurrence in mollusks

Barboza et al. (2018) reported fluorescently labelled PS particles (3.0 to 9.6 μm) were used to track MPs, as humans may consume the entire soft tissue portion of bivalves, which is a microplastic-rich part (average concentration: 0.36 particles g^{-1}). After 12 days of MP addition, MPs in the haemolymph peaked, and particle size greatly affected transport efficiency. The survey of MP abundance by Doyle et al. (2019) in the gastropod *Littorina littorea* from Galway Bay, Western Ireland, showed that 60.4% of *Littorina littorea* samples contained MPs with an average MP level of 2.14 particles g^{-1} of wet soft body mass. MPs are detected in bivalves from aquaculture farms in Germany (0.36 ± 0.07 particles g^{-1}), France, Belgium, and Netherlands (0.2 ± 0.3 particles g^{-1}) (Barboza et al., 2018). By comparison, MPs in Canadian bivalves were found to be 500 times higher (Covernton and Cox, 2019). With the global average consumption of bivalve being 2.4 kg $year^{-1}$ $person^{-1}$ (Revel et al., 2018), each individual may eat up to 864 MPs per year by eating mussels. In Europe (Revel et al., 2018), this number could be as high as 1550 to 9474 MPs per year. It is predicted that by 2100 (Rist et al., 2018), the number of MPs consumed by humans from bivalves will increase to 6.6×10^4 particles $year^{-1}$ (minors) and 4.4×10^5 particles $year^{-1}$ (adults). Moreover, for edible shellfish cultured in the North Sea, after digesting mussels directly with concentrated Nitric acid (HNO_3) and purifying them with purified water for three days, only MPs particles (<25 μm) remained (Barboza et al., 2018). Further Raman microscopy (RM) analysis indicates that although bivalves also absorbed larger MP particles, only smaller ones were transferred to tissue (Rist et al., 2018).

4.1.2. MPs occurrence in crustaceans

When the zooplankton crustacean *Daphnia magna* were exposed to PET MP fibers (62–1400 μm) for 48 h, the majority of ingested fibers were ≈300 μm were found in the gut (Jemec et al., 2016). Also, transgastrointestinal translocation (Rist et al., 2018) of MPs was confirmed in experiments on crabs. In the experiments of Au et al. (2015), the amphipod *Hyalella azteca* was exposed to weathered PP MP fibers (particle size: 20–75 μm) and laboratory-made PE fragments (particle size: 10–27 μm). Gastrointestinal toxicity was recorded during the 10-day acute exposure and reproductive toxicity was demonstrated during the 42-day chronic

exposure. Also, the mortality of daphnids was increased when exposed to PET MP for 48 h (Jemec et al., 2016). In laboratory and field studies by Dawson et al. (2018) and Cau et al. (2020), it has been shown that larger MPs are triturated and fibrillated, and reduced to smaller MPs that are small enough to potentially cross physical barriers or be expelled as a mixture of triturated particles that are more easily uptaken by other marine biotas. Furthermore, the hepatopancreas of crustaceans is usually viewed as the eatable part of the animal. According to G. Wang et al. (2021) and T. Wang et al. (2021), the hepatopancreas is a vital organ for defense mechanisms to alleviate the effects of MPs in crabs. Its consumption is not suggested for humans due to its potential contamination levels (Barrento et al., 2008).

4.1.3. MPs occurrence in fish

Beer et al. (2018) conducted a survey of wild fish, which showed that plastic particles in the intestine were in 35% of the fish analysed and were larger than 5 mm. RM analysis (Beer et al., 2018) showed that only 11 of the 35 particles and fibers present in the digestive tract of fish are plastic, and the rest are cellulose. Also, MP fouling is always caused by the use of fragile Polystyrene plastic containers to store and transport fish. According to a survey by Beer et al. (2018), 1822 microparticles were extracted from the stomach and intestine of 1337 fish samples along the Mediterranean coast of Turkey, most of which were MPs (70%). In the analysis of different fish populations in 21 Chinese coastal seas (Zhao et al., 2018), 26 fish species carry MPs, which account for between 55.9% and 92.3% of their populations. In the study of Revel et al., (2018) of MP particles in intestines of *Gadus morhua* from the North and Baltic Seas, plastic particles were found in 5.5% of test samples, of which 74% were MPs. In comparison, demersal marine fish were found to have a higher content of MPs than pelagic marine fish, which is higher again than that found in freshwater fish (Zhao et al., 2018). In summary, the results obtained vary greatly due to different sampling locations, regional differences in pollution levels, or differences in MP extraction methods.

4.1.4. MPs occurrence in other sea products

In addition to personal care products (toothpaste, scrub), humans may also ingest MPs through beer, honey, sugar, salt (7 to 681 particles kg^{-1}), tap water, and bottled water (Barboza et al., 2018). More than 80% of urban tap water worldwide is reported to be contaminated with MPs (Eerkes-Medrano et al., 2019). According to Barboza et al. (2018) rough estimates in the EU, the maximum annual consumption per person is 4000 plastic particles in tap water and 37 to 1000 plastic particles in sea salt. Other tests have shown that 24 German beer brands contain MP fibers, and 17 sea salt brands in 8 countries contain PP and PE MPs (Barboza et al., 2018).

4.2. Food safety concerns

Since the intake of MPs may result in the transfer of harmful substances to human tissues, it is critical to test for MPs in marine fish of commercial significance. In recent years, reports of plastics in digestive fish systems have become very common, and fish (such as grass carp) have been reported to ingest MPs both along the coast and in deep waters (Lithner et al., 2011). In the report of Esposito et al. (2022), MPs were detected in the gastrointestinal tracts of eight different deep-sea fish in which the main polymer is PE including fibers, films and fragments. These fish are the main food and energy resource for different marine predators. Thus, they are important ecological and economic species and may profoundly impact the entire food chain.

When aquatic organisms consume MPs along with different nutrients, they may enter the food chain and accumulate to higher trophic levels. The negative impact of MPs on living organisms is related to the strong mechanical injury potential of MPs ingested in the digestive tract of the organism and leaching of monomers and additives, including plasticizers, stabilizers, pigments or colourants, antioxidants, and fillers or flame retardants, which are toxic, carcinogenic or are endocrine disruptors (Eerkes-Medrano et al., 2019). Even if low concentrations (ng l^{-1}–mg l^{-1} range) of additives are released from plastic into the environment, they can also negatively affect organisms. Since MPs have a large surface area to volume ratio and surface hydrophobicity, MPs are susceptible to contamination by persistent organic pollutants (POPs), polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB), and dichlorodiphenyltrichloroethane (DDT) (ng l^{-1}–μg l^{-1}) in the water environment (Eerkes-Medrano et al., 2019). Different types of MPs accumulate certain levels of waterborne POPs (PE (HDPE and LDPE), and PP accumulates organic pollutants PCB, PAH more so than PET and PVC) (Eerkes-Medrano et al., 2019) or toxic metals from the marine environment. Also, MPs are often used as carriers of invasive pathogenic microorganisms (Eerkes-Medrano et al., 2019).

4.2.1. Mechanisms of MPs ingestion and translocation

Potential MP exposure routes in the human body include gastrointestinal tract (GIT) ingestion or pulmonary inhalation (Revel et al., 2018). A common mechanism of entry by both pathways is endocytosis. GIT ingestion is recognized as the main exposure route. In this process, MPs enter the GIT circulation system through gaps in the villus tip epithelium to achieve transport. Translocation to secondary target organs is affected by many factors, including particle size, hydrophobicity, surface charge, surface functionalization, and related protein corona. Current research (Revel et al., 2018) indicates that MPs (<2 μm) exist in human blood and organs, though it is unclear whether larger MPs are transported to the same extent. It is worth noting that studies (Revel et al., 2018) on PE MPs show that MPs may cross the GIT layer; even MPs (50 μm) are reported to be transported to lymph nodes in the liver and spleen (Lithner et al., 2011). These particles cause an inflammatory response in surrounding tissues, promoting macrophages' immune activation and cytokine production. In addition, MPs (<2.5 μm) may cross the respiratory barrier and remain in the lungs, which means air pollution caused by MPs is closely related to respiratory and cardiovascular toxicities (Liao et al., 2011).

5. MPs toxicity potential

5.1. Toxicity of chemicals in plastic products

According to the United Nations Global Harmonized System (GHS) (Xu et al., 2018), plastics on the market with special functions are made by arranging monomer or oligomeric structural units of polymer chains through different chemical reactions techniques. The resulting product contains most polymer chains and traces of residual monomers, used catalysts, additives, and by-products. In plastics, various additives (plasticizers, stabilizers, pigments, fillers, and flame retardants) are usually toxic, carcinogenic, or endocrine-active substances. In addition, thousands of chemicals are used in plastic products, giving rise to numerous potential hazards. Lithner et al. (2011) evaluated and ranked the potential hazards of monomers (used in the production of polymers) to organisms and humans. Lithner et al. (2011) also quantified the potential hazards of plastic polymers based on monomers toxicity; PUR, Polyacrylonitrile (PAN), PVC, Epoxy resin, and Acrylonitrile-butadiene-styrene (ABS) were categorised as the most toxic polymers.

It has been shown that the monomers, oligomers, and chemicals contained in many plastic products may pose a threat to human health and often manifest in the form of reproductive toxicity. Common plastics on the market are made of biologically inert polymers, such as PE and PP (Rist et al., 2018). However, certain monomers and oligomers are harmful to human health, which may be leached and transferred into the human body during use. The most common are bisphenol A (BPA) (a key monomer in the production of polycarbonate and epoxy resins) and styrene (the production of foamed PS) (Xu et al., 2018), which are endocrine-disrupting chemicals (EDCs). For example, BPA seriously affects the reproduction of aquatic organisms (Rist et al., 2018). BPA has repeatedly appeared in reports about harmful agents in urine, blood, breast milk, and tissue samples (Xu et al., 2018). It indicates that the main human exposure routes are respiratory inhalation, skin contact, and ingestion (Rist et al., 2018).

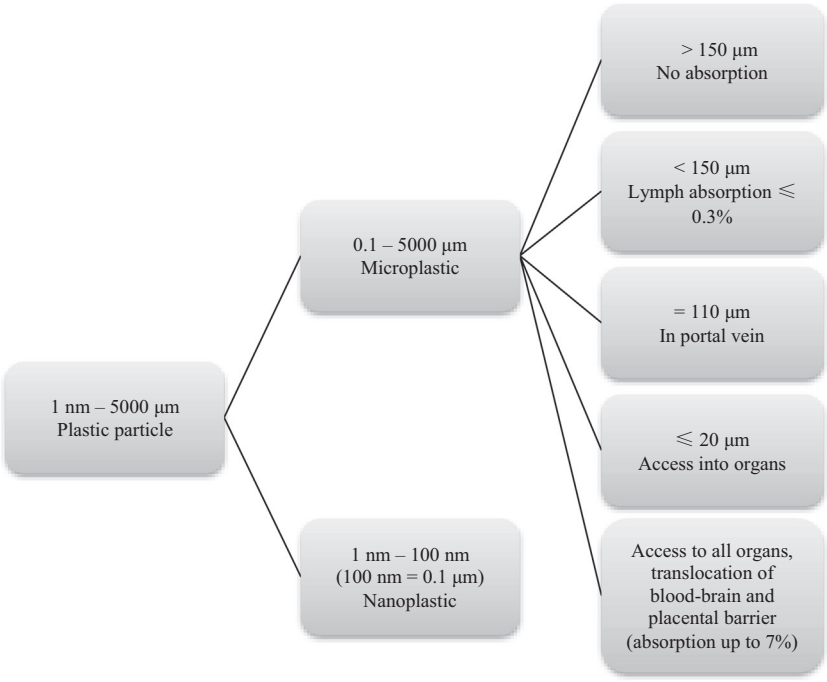


Fig. 3. Relationship between the fate of microplastics and nanoplastics in mammals and particle size.

Furthermore, phthalates (di-n-octyl phthalate (DnOP) and di-2-ethylhexyl phthalate (DEHP)) (Xu et al., 2018) are commonly found in human urine and blood that causes abnormal development of the human body and affect adolescent development, male and female reproductive health, pregnancy outcomes, and respiratory health (Liao et al., 2011). In addition, polybrominated diphenyl ether (PBDE) and tetrabromobisphenol A (TBBPA), which are used as flame retardants, have a role in destroying the homeostasis of thyroid hormones (Xu et al., 2018). Also, PBDE has an antiandrogenic effect. Moreover, there is the release of metal-based catalysts used in plastic water bottles production during use. For example, catalyst antimony (Sb) is released at 60–85 °C, potentially causing nausea, vomiting and diarrhoea (Xu et al., 2018).

5.2. Particle toxicity of MPs

As shown in Fig. 3, lymph nodes are capable of absorbing up to 0.3% of plastic fragments smaller than 150 μm, while MPs smaller than 110 μm are accessible in the portal vein and into organs even when MPs are smaller than 20 μm in size (Lusher et al., 2017). Moreover, 7% of NPs may transfer across epithelial cells and can enter all organs such as the liver and spleen (hepatotoxicity), heart (cardiovascular toxicity), thymus, reproductive organs (reproductive toxicity) and brain (neurotoxicity) (Lusher et al., 2017) (detailed in Table 6). In addition, NPs can cross the blood-brain barrier and the placental barrier for translocation (Lusher et al., 2017).

Dominant pathways of uptake for MPs are via phagocytosis and endocytosis. Macrophages in the small intestine epithelium absorb particles larger than 0.5 μm through phagocytosis, while honeycomb cells internalize 5 μm of particles through endocytosis (Revel et al., 2018). Lusher et al. (2017) confirmed that MPs (particle size: between 0.1 and 150 μm) could be transported into the lymphatic system through the mammalian circulatory system (Lusher et al., 2017). Recently, a study of mice on the distribution and accumulation of PS MPs in tissues and resulting health risks to specific tissues showed MPs accumulated in the liver, kidney (nephrotoxicity), and intestines (gastrointestinal toxicity) (Revel et al., 2018) (Table 6). Also, the particle size of MPs was found to be closely related to tissue accumulation kinetics and distribution pattern (Rist et al., 2018). By analysing biochemical biomarkers and metabolomic profiles in mouse livers, it is noted that MPs affect oxidative stress, energy and lipid metabolism, and neurotoxicity (Rist et al., 2018).

The main reported mechanism of toxicity of MPs is via oxidative stress and its accompanying inflammation. The chemical composition and the particle size of MPs decisively affect the degree of adverse reactions, with larger nano-sized particles producing more ROS (Eerkes-Medrano et al., 2019) and can be more easily translocated. Schirinzi et al. (2017) showed

Table 6
Classification of MP and NP potential toxicity to marine animals and humans.

Plastic-type	Toxicity	Classification	Reference
Microplastics	Acute toxicity	Respiratory toxicity	Zhang et al., 2021
	Acute and subchronic toxicity	Cytotoxicity	Liang et al., 2021
	Acute and subchronic toxicity	Gastrointestinal toxicity	Jin et al., 2019; Qiao et al., 2019
	Chronic and subchronic toxicity	Immunotoxicity	Jin et al., 2019; H. Sun et al., 2021; M. Sun et al., 2021; T. Sun et al., 2021
	Acute toxicity, chronic toxicity and genotoxicity	Reproductive toxicity	Sobhani et al., 2021
	Carcinogenicity	–	Martin et al., 2017
	Developmental toxicity	Embryotoxicity	Uhrin and Schellinger, 2011
Nanoplastics	Chronic and subchronic toxicity	Nephrotoxicity	Gherkhbolagh et al., 2018
	Chronic toxicity	Cardiovascular toxicity	H. Sun et al., 2021; M. Sun et al., 2021; T. Sun et al., 2021
	Acute toxicity, chronic toxicity, developmental toxicity, and genotoxicity	Reproductive toxicity	An et al., 2021
	Acute toxicity and chronic toxicity	Hepatotoxicity	Lusher et al., 2017
	Acute toxicity	Neurotoxicity	Gambardella et al., 2018
	Genotoxicity	–	Lusher et al., 2017

that MPs (particle main size: 10 μm PS) exerted oxidative stress on the human brain and epithelial cells in vitro experiments. Other potential main biological reactions include cytotoxicity (apoptosis and necrosis, and even tissue damage and fibrosis) and carcinogenicity (Lusher et al., 2017) (Table 6).

Also, MPs may induce intestinal obstruction or tissue abrasion. MPs have a large surface area that may activate the intestinal immune system to trigger local inflammation (Rist et al., 2018), thereby further accelerating intestinal uptake of MPs. Animal studies have shown that mucosal colon tissue absorbs 0.2% of 3 μm polylactide-*co*-glycolide particles (Revel et al., 2018). Mucosal colon tissue of patients with inflammatory bowel disease reportedly increases transport volume to 0.45% due to increased intestinal permeability (Rist et al., 2018).

According to H. Sun et al. (2021), M. Sun et al. (2021) and T. Sun et al. (2021), NPs induce significant genotoxicity in aquatic organisms when aquatic organisms are exposed to environmentally relevant concentrations of NPs (≤ 1 mg/L, with mean toxicity increased by 24%), with the higher concentration of NPs with the smaller the particle size inducing greater genotoxic damage (Table 6). The level of genotoxic effects of NPs is independent of the composition, morphology, exposure concentration, and duration of exposure to NPs, although it is closely related to particle size, habitat, and species (H. Sun et al., 2021; M. Sun et al., 2021; T. Sun et al., 2021). In addition, freshwater biota are more susceptible to the effects of NPs (H. Sun et al., 2021; M. Sun et al., 2021; T. Sun et al., 2021).

5.3. Indirect effects of MPs

Adverse effects of MP on human health include, in addition to direct toxicological effects, its ability to act as a carrier for chemical additives and pathogens. MPs have a large surface area to volume ratio and surface activity; they can adsorb various pollutants leached from plastic, resulting in elevated levels, including heavy metals, PAH, PCB, and DDT (Revel et al., 2018). Since MPs often enter the food chain, the possibility of biomagnification in high-level predators (including humans) is increased. By exposing juvenile fish (*Pomatoschistus microps*) to PE MPs carrying pyrene for 96 h (Revel et al., 2018), found that fish aerobic capacity was reduced. In addition, the accumulation of polychlorinated biphenyls in tail tissues of lobsters (*Nephrops norvegicus*) was restricted after crustacean daphnia (*Daphnia magna*) were loaded with polychlorinated biphenyls (PE MPs). Concurrently, research from Revel et al., (2018) on mussels have also detected that PE MPs contaminated with pyrene affected muscle cells immune response (immunotoxicity), including changes in lysosomal compartments peroxisome reproduction and antioxidant systems. Experiments from Lithner et al. (2011) have shown that organisms exposed to MPs contained 2.3 times PAHs, 6 times PCBs, and 2.8 times PBDEs compared to the negative control.

Pathogenic microorganisms tend to form biofilms (Vibrio species) on the surface of MPs. For example, many PE and PP particles were found in the Baltic Sea to carry potentially pathogenic *Vibrio parahaemolyticus*. According to scanning electron microscope (SEM) images (Revel et al., 2018), some bacterial actively break down hydrocarbons in polymers to degrade and reduce plastic particle size. Microbial biofilm communities attached to MP are diverse, with structure and diversity strongly affected by geographical location, season, and plastic type.

6. MP risk assessment

Risk assessment of MPs is a daunting task because even though the materials that form MPs are available for industrial use at the macroscopic scale, which is regulated and approved, this does not mean that the MPs have the same risk potential and same exposure pathways. Fig. 4 shows a potential framework for risk assessment of MPs and the different factors to consider. At the microscopic scale, structure determines behaviour. Therefore, MPs cannot be treated as chemicals in RA. According to the RA methodology issued by Codex Alimentarius in 2011 (FAO and WHO, 2021), the steps in risk assessment include hazard identification, exposure assessment, hazard characterisation, and risk characterisation. The hazard identification of MPs highlights that risk is related to three main factors (Fig. 4): (a) Exposure pathways, including seawater (surface water and water column), sediment (coastal or benthic), and air, (b) the exposure

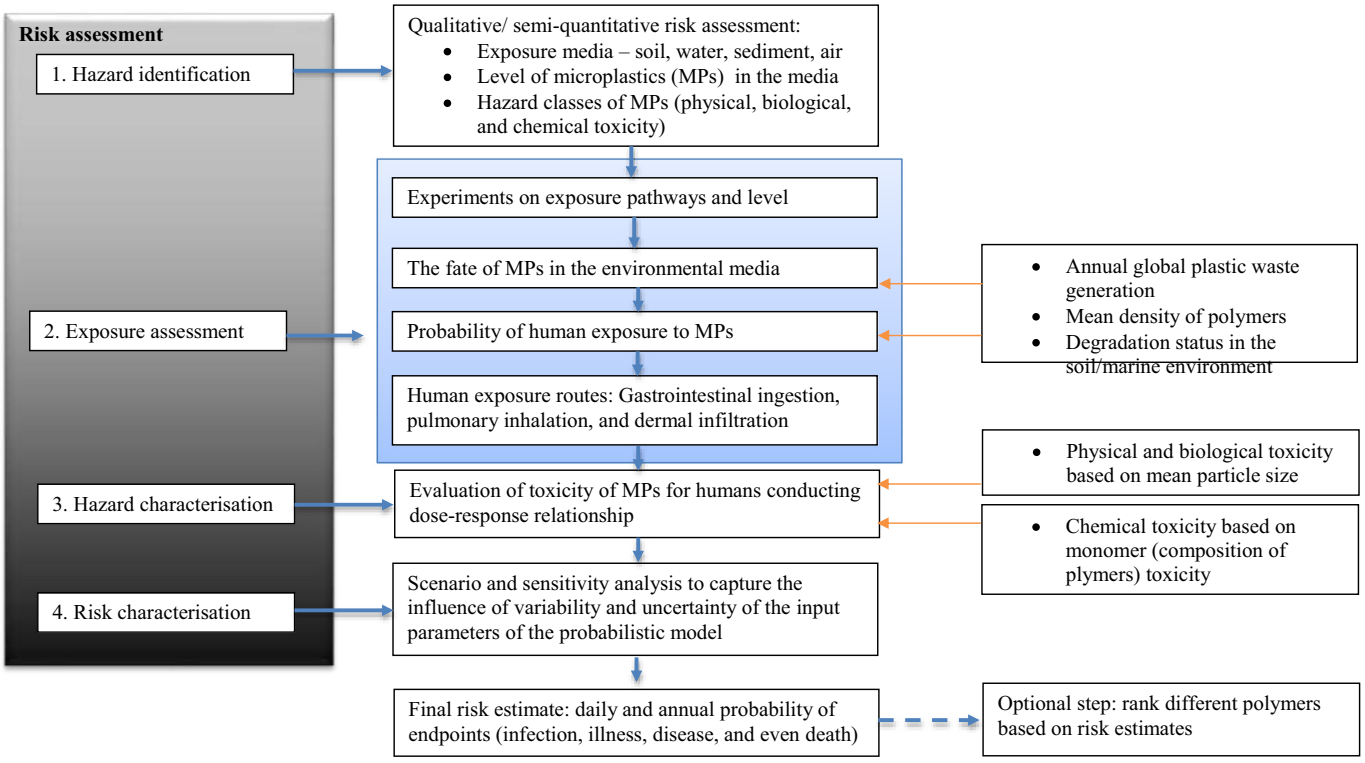


Fig. 4. Framework for Risk Assessment of MPs in the marine environment.

levels of MPs, and (c) the potential hazard of MPs (mechanical, biological, and chemical toxicity) (EPA US, 2016; Yuan et al., 2022).

The hazard identification stage (Figs. 4 and 5) shows the main pathway for humans to be subjected to marine-derived MPs is through surface water, water column, and sediment (coastal or benthic). These primary pollutants originate from direct emissions of SMPs, and physical, chemical, and biological breakdown of PMPs are associated with human and aquatic organism hazard potential. The exposure assessment typically includes MPs fate in the aquatic environment and human exposure through various pathways (gastrointestinal tract ingestion, respiratory inhalation, and dermal infiltration: shown in Fig. 5). Important factors influencing the probability of human exposure to MPs are MP concentration (annual global waste generation), polymer density, and lifetime (degradability). In the hazard characterisation of MPs, the mean particle size of MPs and the potential hazard of polymers based on monomer toxicity are key factors to quantify MP toxicity to humans. MPs with different particle sizes have obvious differences in capability to accumulate on biofilms and may pose biological toxicity risk. Also, MPs may release or adsorb toxins based on their monomer composition (chemical toxicity) (Lithner et al., 2011). The combined information for risk characterisation will generate a complete risk assessment model to rank the hazard potential of MPs to human health from diverse polymers in the marine environment.

Methods commonly used in the RA of MPs include qualitative (broad categories: low, moderate, or high) and quantitative (numerical values: deterministic, probabilistic, or stochastic models) analyses (Piperagkas et al., 2019; Capillo et al., 2020). Also, semi-quantitative risk assessment has been used (Estahbanati and Fahrenfeld, 2016), in which the risk factor categories are first scored (0: good, 1: medium, 2: bad) and then calculated based on score arithmetic (using a probability-impact matrix). The final risk depends on the severity of the hazard, the dose, and the population vulnerability (human and animal) (FAO and WHO, 2021).

Extensive development of existing RA techniques and new detection and quantification techniques to address practical issues is necessary to ensure plastics' safe use and disposal. The methods and tools used in the RA of MPs include hazard identification models used to study the potential risks of MPs to the environment or humans. The development of a RA system makes it possible to elucidate the risks of MPs to humans and the environment directly from existing data in the literature without the need to repeat the original experiment. Table 7 lists some typical cases of MP RA studies conducted in recent years. Most MPs risk assessment models do not focus on specific polymer types; instead, MPs in different surroundings (marine organisms, sediments, water, and air) are evaluated. Common methodologies are usually based on published governmental regulations (REACH and EUSES), hazard ranking models based on the research of Lithner et al. (2011), ecological risk index method, pollution load index and the worst-case scenario based on data from previous studies (Liu et al., 2019; Lusher et al., 2013; Pan et al., 2021; Peng et al., 2018; Xu et al., 2018). Also, the main exposure routes investigated are direct ingestion, inhalation, and indirect ingestion through the food chain, while the key impact factors include MP concentration, size, shape, and polymer type. In addition, researchers have calculated different dose-response indicators for hazard characterisation according to their methodologies. Nevertheless, current studies need further improvement due to insufficient data on MPs (concentration, size, polymer type, and spatial-temporal distribution) and polymers (environmental lifetime, toxicity).

7. Discussion and conclusions

There is considerable work needed to develop the area of risk assessment of MPs in marine ecosystems. First, a clear definition of MP type and size range is required. Further standardisation and internationalization of MP sampling, processing, identification, and quantification standard operating procedures (SOPs) are required. Effective methods to reduce or eliminate MPs from marine ecosystems are also required, including developing MP emission prevention strategies and accelerating sustainable use and disposal of plastics. Fourth, reliable methods for assessing MP pollution levels are required, and new technologies to prevent the discharge of plastics into the aquatic environment include the development of eco-friendly polymers (starch-based plastic or polylactic acid) and 'green' additive chemicals (Karamanlioglu et al., 2017).

When developing SOPs for MPs in marine ecosystems, the regulation regarding the selection of sampling locations and duplicated sampling with uniform units is required. Also, a model for the content and distribution of MPs in marine ecosystems needs to be built to monitor hotspots and timelines comprehensively, estimate emerging anthropogenic pollutant loads (Ivleva et al., 2017), and establish the life cycle of MPs (Wagner and Lambert, 2018).

MP detection methods typically include combinations of spectroscopic analysis (RS and FTIR) and chromatography (TED-GC/MS). Both techniques identify polymer types and additives. RS and FTIR coupled to optical microscope are commonly used to provide information on particle size and number, while TED-GC/MS is used to derive the total mass fraction of polymer involved.

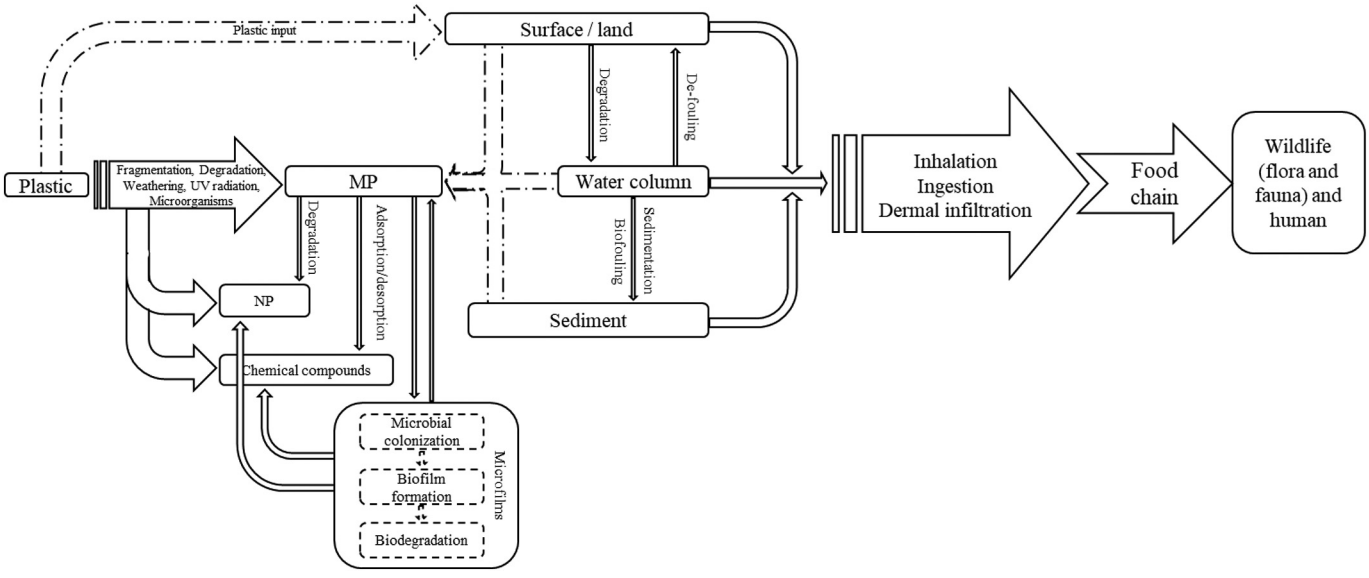


Fig. 5. Potential routes of exposure, uptake, distribution, and degradation of MPs after intentional or unintentional release into the aquatic environment.

Table 7
Risk assessment studies on marine MPs with details of polymers assessed, source, pathways, exposure, dose-response characteristics, and sensitivity of key parameters of the model.

Country or region	Hazard identification: contaminants	Methodology	Source	Pathway	Exposure (MPs concentration)	Hazard characterisation: dose-response	Critical parameters	Reference
Worldwide (mainly European countries)	LDPE: polychlorinated biphenyls (PCB), polycyclic aromatic hydrocarbons (PAH), dichlorodiphenyltrichloroethane (DDT), bisphenol A (BPA), and polybrominated diphenyl ether (PBDE)	Worst case scenario based on data from previous studies.	Bivalves (seafood)	Oral intake	Annual 4% of MPs	Less than 0.1% of the total dietary intake (no notable effect).	Inaccuracy assumptions; particle size distribution.	Lusher et al., 2013
Asia, Europe, and North America	MP (ecotoxicological RA)	Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) guidance.	Pelagic and benthic freshwater and organisms.	Ecosystem	Data with different sizes, shapes, and materials particles.	Endpoints: survival, growth, reproduction, and significant metabolic changes: causing 50% growth inhibition and lethality.	Data homogeneity, lower cut-offs, data are missing on some of the most polluted rivers.	Adam et al., 2019
Worldwide	MPs	Simulate future exposure fate of MPs using deterministic experiments data by meta-analysis; species sensitivity distribution.	Surface water, seafloor, beach sediment, and marine species (bivalves).	Ingestion, inhalation, and food chain.	Data in the table from previous literature.	Risk characteristic ratios >1: ambient concentration exceeds the safe concentration, 5% hazard concentration for species (33.3 particles l^{-1}).	Sampling location and sample size; High spatial, temporal, and species population variability in different sea areas.	Everaert et al., 2018
Changjiang Estuary and the adjacent East China Sea	MPs	The hazard scores of plastic polymers proposed establishing an index of MP polymer types and the pollution load index (PLI).	Surface water	Hydrological dynamics and human activities.	Concentration, shape, size, colour, and composition types of MPs.	The ratios of PLI_{zone} and high-risk stations to determine the risk in the region: significant correlation between the polymeric risk index and PLI ($P = 0.02 < 0.05$).	Research methodology, objectives, thermodynamic properties of plastics, and geographic locations.	Xu et al., 2018
EU	MP	European Union System for the Evaluation of Substances (EUSES) model (estimation of the fate and distribution of MPs in the environment); REACH.	Water, sludge, sediment, and organisms (fish, filter-feeders, and direct soil ingestion) in marine and freshwater.	Hydrological dynamics, human activities, and food chain.	Using the obtained MP release data modelling predictions to indicate human exposure to MPs in a specific product or process.	Many detailed tables (in the reference) depend on experimental exposure conditions, choice of material and indicators.	Simplified assumptions, exposures, distribution ratio between water and sediment, distribution and decontamination capacity of sewage plants, and temporal and spatial variability.	Scudo et al., 2017
Dongshan Bay in southern China	MP (ecological RA)	Hazard-ranking model based on the United Nations' Globally Harmonized System of Classification and Labelling of Chemicals; PLI.	Surface water	Ingestion, inhalation, and food chain.	Range: 0.23 to 4.01 particles m^{-3}, mean: 1.66 ± 1.41 particles m^{-3}.	MPs-induced risk index (H estuary = 12.94), pollution load index ($PLI_{estuary}$ = 17.34), and potential ecological risk index from combined MPs polymers ($RI_{estuary}$ = 21.5); the overall risk of MPs pollution is Hazard Level II (minor risk).	MPs concentration and diversity of polymer composition and the resulting hazards caused by individual MPs polymer.	Pan et al., 2021
Shanghai, China	Suspended atmospheric microplastic (SAMP)	Ecological risk index (RI) method and hazard score of the plastic polymer.	Sediments, water column, and aerosol.	Ingestion, inhalation, and food chain.	Filtered air: 0 to 4.18 particles m^{-3}.	RI is low (0.36 to 4.03), and the distribution of risk indices is correlated with altitude and geography.	Toxicity factors for polymers lacking cellulose, rayon, and alkyd resins may underestimate RI.	Liu et al., 2019
Shanghai, China	MP	Ecological risk index (RI) method and hazard score of the plastic polymer.	Sediments from the river and tidal flat.	Ingestion, inhalation, and food chain.	The abundance of MPs in rivers near densely populated areas is one to two higher orders than in tidal flats in rural areas.	Depends on MPs origin, deposition, fate and distribution, migration, persistent organic pollutants, attached biofilms, and ecotoxicological effects.	Uniform sampling/quantification methods and clearly defined environmental relevant concentrations of MPs.	Peng et al., 2018
Manas River Basin, China	MP	Hazard score of the plastic polymer; PLI.	Inland freshwater (surface water).	Hydrological dynamics and human activities.	Average MPs abundance in April (17 ± 4 particles l^{-1}) is higher than in July (14 ± 2 particles l^{-1}).	Season affects the abundance of MPs, but not their shape, size, and type.	Compounds and concentration to affect the hazard index.	G. Wang et al., 2021; T. Wang et al. (2021)

Small plastic particles (<150 μm) can be absorbed by biota tissue, organs, and even cells, causing adverse effects on human health (Lusher et al., 2017). Microplastics may pose several toxicity concerns, including acute and chronic toxicity (cytotoxicity, immunotoxicity and reproductive toxicity), carcinogenicity, and developmental toxicity. Nanoplastics may pose chronic toxicity (cardiovascular toxicity, hepatotoxicity, and neurotoxicity), genotoxicity, and developmental toxicity. MPs toxicological properties and general quantitative and qualitative analysis methods used in MPs Risk Assessment (RA) are summarised. A robust dose-response model for MPs requires further investigation.

This study focused on state of the art regarding the definition and characterisation of MPs, levels found in marine and food ecosystems, detection methodologies, and risk assessment strategies, thus providing a reference point for future studies into their impact on human ecosystem health. Also, it is necessary to prioritise and direct future research into the most harmful MPs by carrying out appropriate risk assessment approaches.

CRediT authorship contribution statement

Zhihao Yuan: Conceptualization, Methodology, Formal analysis, Software, Data curation, Visualization, Investigation, Writing – original draft. **Rajat Nag:** Conceptualization, Visualization, Supervision, Writing – review & editing. **Enda Cummins:** Conceptualization, Resources, Supervision, Project administration, Writing – review & editing.

Declaration of competing interest

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

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