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Review article

Microplastics in water systems: A review of their impacts on the environment and their potential hazards

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ABSTRACT

Microplastics, the microscopic plastics, are fragments of any type of plastic that are being produced today as plastic waste originating from anthropogenic activities. Such microplastics are discharged into the environment, and they enter back into the human body through different means. The microplastics spread in the environment due to environmental factors and the inherent properties of microplastics, such as density, hydrophobicity, and recalcitrance, and then eventually enter the water environment. In this study, to better understand the behavior of microplastics in the water environment, an extensive literature review was conducted on the occurrence of microplastics in aquatic environments categorized by seawater, wastewater, and freshwater. We summarized the abundance and distribution of microplastics in the water environment and studied the environmental factors affecting them in detail. In addition, focusing on the sampling and pretreatment processes that can limit the analysis results of microplastics, we discussed in depth the sampling methods, density separation, and organic matter digestion methods for each water environment. Finally, the potential hazards posed by the behavior of aging microplastics, such as adsorption of pollutants or ingestion by aquatic organisms, due to exposure to the environment were also investigated.

1. Introduction

The Iron Age is the last epoch of the three-age division (including the Bronze Age and the Stone Age) of the prehistoric period in human history to be named after the material that was mainly produced and used as a tool during that period [1]. Since then, eras have not been named after the materials used, but if we have to name the present era, it would not be an exaggeration to term it the Plastic Age. Plastics have proliferated worldwide since they were first discovered in the early 20th century, surpassed iron production after the 1990s, and are currently the most used material by humans [2]. After being commercially developed in the 1930s and 1940s, plastics have become increasingly dominant in the consumer marketplace due to their convenience and various advantages [3]; however, as the amount of plastic used increased, the problem of environmental pollution caused by plastic waste also emerged [4,5].

Prior to 2000, the focus was mainly on the biodegradability of synthetic polymer compounds such as plastic bags and disposable diapers. At the time, plastics were primarily concerned with waste emission and disposal, as well as the fact that plastics take decades or even hundreds of years to decompose [6]. In 2004, it was published in Science that studies have reported increasing microscopic plastics in the ocean [7]. Subsequently, the interest in the consequence of plastic accumulation in nature was heightened; not only in the marine environment but also in various environmental media such as air, soil, and freshwater.

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Microplastics, the microscopic plastics, are fragments of any type of plastic [8] that are less than 5 mm in length, according to the U. S. National Oceanic and Atmospheric Administration (NOAA) [9,10] and the European Chemicals Agency [11]. The United Nations Environment Programme (UNEP) defines microplastics as any solid plastic particle of 5 mm or less which are insoluble in water [12]. The International Organization for Standardization (ISO) ISO/TC 61 (Plastic)/SC 14 (Environmental Aspect) defines microplastics as any solid plastic particle insoluble in water with dimension between 1 μm and 1000 μm in the ISO/TR 21960:2020 standard terms and definitions [13]. There are two major sources for the occurrence of these microplastics; first, microplastics that are generated directly, and second, those generated from a secondary source, in that, microplastics generated when large plastic debris are broken down by weathering owing to physical and chemical effects in the natural environment. The production of primary microplastics such as microbeads has been prohibited in recognition of the seriousness of its impact on environmental pollution [14]. However, secondary microplastics have the potential to continue to arise from plastics that have already been discarded and exist in the natural environment [15]. Studies on microplastics have been mainly conducted in the marine environment, but now, it is necessary to investigate microplastics in various aspects that can exist in all environmental media, such as surface freshwater, groundwater, air, soil, and sediment. Moreover, although the harmful impacts of the microparticles on human health have not been established, it is evident that microplastic particles with a size close to nanometers are highly likely to be toxic, therefore warranting studies on the size of these microplastics [16,17].

In this review article, the current status and toxicity of microplastics in water systems have been intensively reviewed, focusing on their impacts on the entire process from exposure to the environment to analysis and comparing the literature on marine microplastics and microplastics in other water sources, and identifying the potential for direct human inhalation and exposure.

2. Methodology of the literature review

The literature search was performed in the following databases: Web of Science (https://www.webofscience.com/, Clarivate), Google Scholar (https://scholar.google.com/, Google), ScienceDirect (http://www.sciencedirect.com, Elsevier), and Scopus (https://www.scopus.com/, Elsevier). The search was performed using the filter function “Highly Cited in Field” and “Hot Papers in Field” provided by Web of Science. The publication year range of the research papers was set mainly to be within the last 10 years. The keywords used were microplastics, seawater, wastewater, and freshwater. This review article cites 184 peer-reviewed publications or reports.

3. Microplastics in water environments

Among the water environments, most studies have been conducted on the abundance and characteristics of microplastics in seawater. Only recently, studies on microplastics in various water environments, including freshwater, wastewater, and groundwater, are being conducted. In this study, although the number of papers on microplastics in seawater and freshwater was high in Asia, especially China, interest in microplastics was high in all continents based on the number of countries (Fig. 1(a)–(b)). On the other

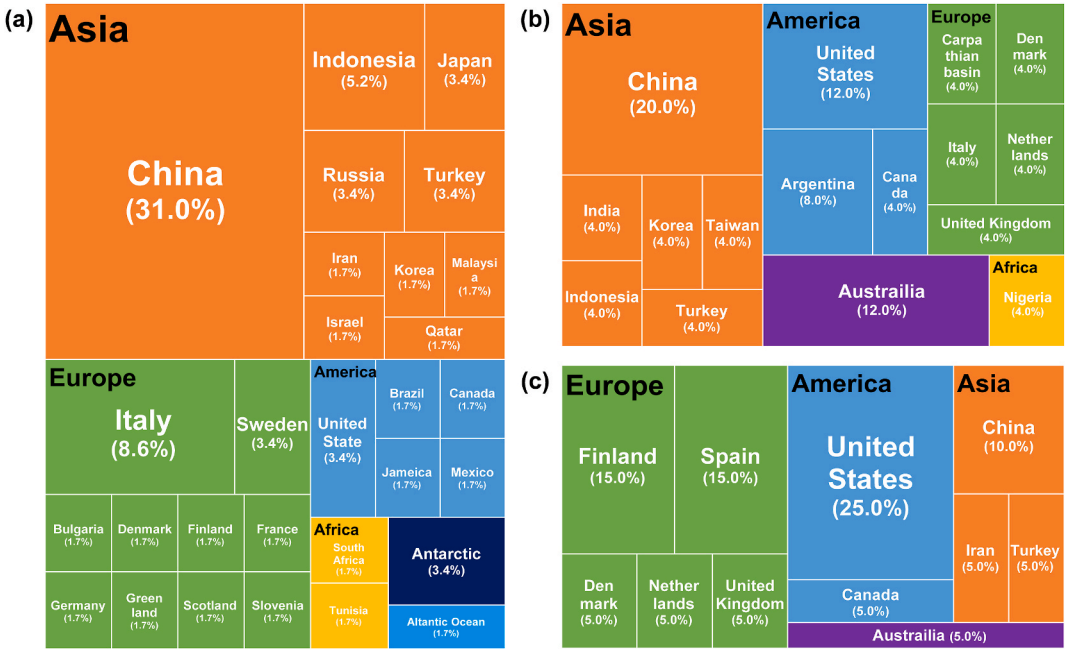

Fig. 1. Distribution of countries where the studies were conducted in (a) seawater, (b) freshwater, and (c) wastewater.

hand, the proportion of publications on microplastics in wastewater was relatively high in Europe (Fig. 1(c)). Since Europe has a relatively high population density and many countries share limited water resources, it is presumed that research on microplastics in wastewater has been actively conducted. Depending on the inherent properties of microplastics (density, hydrophobicity, recalcitrance) and environmental factors, microplastics can easily move between environmental media, and high-density microplastics can settle and accumulate at the bottom of the water environment. Due to this phenomenon, it is difficult to investigate the concentration and characteristics of microplastics in the overall water environments. Moreover, direct comparison between studies is difficult because the sampling and pretreatment methods are not similar. In this review, microplastics in the water environment were investigated and discussed based on the characteristics of the microplastics, differences in sampling method, and environmental influences.

3.1. Microplastics in seawater

3.1.1. Microplastics in seawater

Microplastics in seawater were among the first to be identified during the literature search and they yielded the most search results. The occurrence of microplastics in the ocean, which is representative regionally, is as follows: The Atlantic [18], Midwest Pacific [19], Mediterranean; Italy [20], Tunisia [21], Baltic Sea [22,23], Arabian Sea; Gulf of Oman [24], Indian ocean; Indonesia Bali [25], South China Sea [26,27], Korea Coasts [28], Arctic Ocean; Nordic Sea [29], Antarctic Ocean [30–32] (Fig. 2, Table 1).

In general, the concentration of microplastics is higher nearshore or in an estuary adjacent to land than in the open sea [33,34], and the concentration of microplastics is higher in the seas with geographical characteristics, such as in semi-enclosed bays, than in the open seashore [24,35,36]. The average plastic particle concentration in the same sampling area of the Ligurian Seas and Tyrrhenian Seas in Italy increased nine-fold in 2019 compared to 2018 [37]. Generally, the microplastic concentration tends to increase gradually over time.

According to the studies conducted in Goiana Estuary, Brazil, the concentration of microplastics was half that of fish larvae and about the same as the density of fish eggs [38]. Microplastics were found in oysters, mussels, and Manila clams living on the Korean coasts. In the oyster and mussel samples, the average microplastic concentration was 0.33 ± 0.23 n/g and 1.21 ± 0.68 n/individual, respectively. The annual intake of microplastics from bivalves was calculated as 587 n/person·year [28]. Some predictions have revealed that the weight of microplastics in the ocean can exceed the total weight of fish in the ocean without appropriate plastic waste management [39].

Notably, this severe plastic pollution problem is not limited to inhabited areas where plastic waste is generated; the Antarctic Ocean, considered to be relatively free of microplastics, contained plastic in the freshwater and Rose seawaters of Antarctica [30,31]. However, some research stations in Antarctica do not have wastewater treatment facilities, and improper waste management practices are presumed to have contributed to the generation of microplastics. Nevertheless, this can also be explained owing to a potential mechanism whereby plastic waste dumped in other areas spread in the region via wind or water current [32].

3.1.2. Results of the microplastic analysis and their influencing factors

3.1.2.1. Regional abundance of microplastics influenced by anthropogenic and environmental factors. Microplastics in seawater have various shapes and compositions depending on regional characteristics, types of industries, and the surrounding environment. Line-type composed of polyethylene (PE), which is used for nets and ropes, was detected in seawater where fishing is active [34,40],

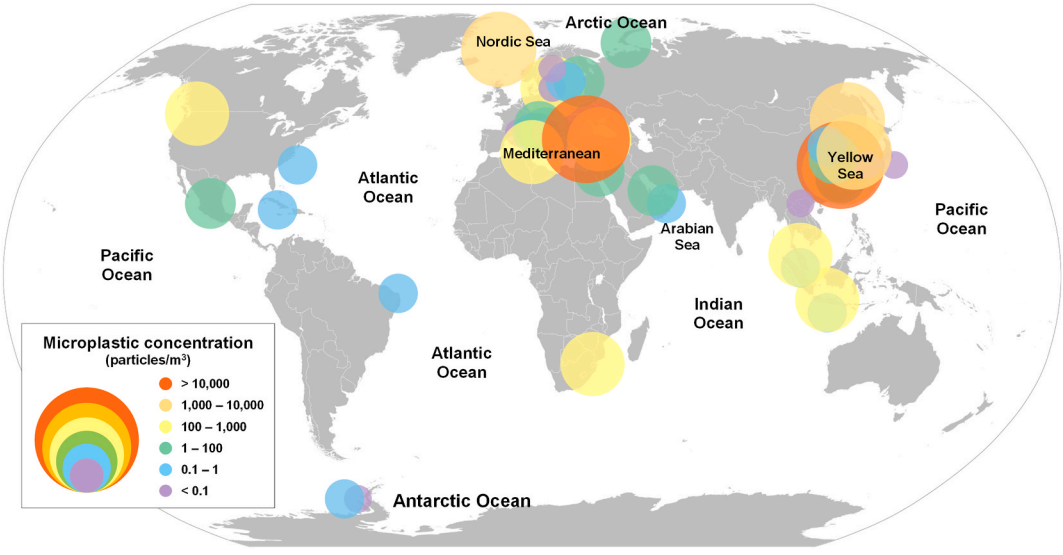

Fig. 2. Microplastics in the world oceans based on studies.

Table 1
Results of microplastic analysis in seawater.

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
Antarctic (Byers Peninsula)	two nylon drifting nets (333 μm and 100 μm)	33% H_2O_2, hypersaline solution (NaCl)	μ-FT-IR (micro-Fourier transform infrared spectroscopy)	fibers: 400–3546 μm (average 1118 μm), films: 10–1026 μm (average 199 μm)	0.47–1.43 items/1000 m^3 (0.95 items/1000 m^3)	[31]
Antarctica (Ross Sea)	pump system	–	FT-IR	detected particles: >60 μm filtration: >1 μm	0.0032–1.18 particles/m^3 (0.17 ± 0.34 particle m^3)	[30]
Atlantic Ocean	a flow pump system and a towed planktonic manta net	–	FT-IR	Net: >5 mm (30%), Pump: <2.5 mm	~60 items/m^3 (pump) ~1829 items/m^3 (net)	[18]
Brazil (Goiana Estuary)	a conical plankton net (300 μm)	–	stereomicroscope	300 μm–5 mm (2.23 ± 1.65 mm)	26.04 items/100 m^3	[38]
Bulgaria (Black Sea coast)	manta net (net opening 0.84 × 0.15 m; mesh size 0.3 mm)	3% H_2O_2	stereomicroscope	2.5–5 cm; 5–10 cm; 10–20 cm; 20–30 cm; 30–50 cm; >50 cm	4.62 × 10^4 items/km^2	[153]
Canada (Baynes Sound, Vancouver Island)	Jar (1 L glass jars) and Bucket (12 L stainless bucket) samples	potassium hydroxide (KOH) solution	FT-IR	–	0.69 MP/L (1 L samples) and 0.12 MP/L (10 L samples)	[132]
China (Yangtze Estuary and the coastal water)	surface water (Yangtze Estuary): 12 V DC Teflon pump, coastal waters (the East China Sea): neuston net with a 30 × 40 cm^2 opening and 333 μm mesh	30% H_2O_2, zinc chloride ($ZnCl_2$) solution	dissecting microscope	>0.5–1 mm, >1–2.5 mm, >2.5–5 mm and >5 mm	4137.3 ± 2461.5 n/m^3 (estuarine) and 0.167 ± 0.138 n/m^3 (sea samples)	[181]
China (Bohai Sea)	330 μm trawling net	aqueous 0.05 M Fe (II) solution ($FeSO_4{\cdot}7H_2O$) and 30% H_2O_2 solution	attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FT-IR)	floating large plastics (>2.5 mm) 7%, medium-sized plastics (0.5–2.5 cm) 38%, and microplastics (0.3–5 mm) 55%	0.33 ± 0.34 particles/m^3	[40]
China (Deep Bay, Tolo Harbor, Tsing Yi, Victoria Harbor, Hong Kong Island)	towing a plankton net (153 μm)	NaCl solution, 30% hydrogen peroxide (H_2O_2)	ATR-FT-IR	0.03–4.96 mm (water samples) and 0.01–4.7 mm (sediment samples)	51–27,909 particles/100 m^3 (local coastal waters) and 49–279 particles/kilogram (sediments)	[60]
China (the North Yellow Sea)	Niskin hydrophore with a 30 μm steel sieve	30% H_2O_2 in the presence of a Fe(II) catalyst, NaCl solution (1.2 g/cm^3), sodium iodide (NaI) solution (1.6 g/cm^3) * surface seawater: only NaCl	μ-FT-IR	<0.5 mm (35.7–83.5%) surface waters <0.5 mm (60.0–96.6%) sediment	545 ± 282 items/m^3	[64]
China (Changjiang Estuary)	pump with a stainless-steel sieve (mesh size: 70 μm)	30% H_2O_2 solution	μ-FT-IR	<5.0 mm (90%) 0.07–1.0 mm (68.4%), 1.00–5.0 mm (26.2%)	23.1 ± 18.2 n/100 L	[74]
China (Longjiao Bay)	pump with Nylon filter membranes (mesh size: 325 μm, 100 μm, and 4 μm) in a clean stainless-steel barrel	30% H_2O_2 solution, NaCl solution (1.2 g/cm^3)	μ-FT-IR	0.30 mm–5.00 mm (92.03%)	250–5150 particles/m^3 (mean 1594 particles/m^3)	[45]
China (Xiangshan bay)	plankton net tows (330 μm mesh and a diameter of 20 cm)	$ZnCl_2$ (ρ = 1.75 g/mL)	ATR-FT-IR	0.25–2 mm (average 1.54 ± 1.53 mm)	4.6 ± 0.5–20.1 ± 0.2 items/m^3 (8.9 ± 4.7 items/m^3)	[44]
China (the Nanxun Reef)	48 μm steel sieve	30% H_2O_2, KOH (10%, V/V), $ZnCl_2$	μ-Raman spectroscopy			[26]

(*continued on next page*)

Table 1 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
		solution, Hydrochloric acid (HCl) solution	(micro-Raman spectroscopy)	<0.5 mm, 0.5–1 mm, 1–2 mm, 2–3 mm, 3–5 mm	1250–3200 items/m^3 (average of 1773 items/m^3)	
China (the Maowei Sea)	stainless-steel sampler	10% KOH solution	μ-FT-IR	<0.25 mm, 0.25–0.5 mm, 0.5–1 mm, 1–5 mm	1.2–10.1 particles/L (average 4.5 ± 0.1 particles/L)	[36]
China (Sanggou bay)	bucket with a 50 μm mesh-filter	Seawater: 1 M NaOH solution, Sediment: 30% H_2O_2, NaCl solution (1.2 g/cm^3), NaI (1.8 g/cm^3), Oyster: 30% H_2O_2, 65% HNO_3 (1:3, v/v), NaCl solution	μ-FT-IR	0.1–0.5 mm (36.7%), 0.05–0.1 mm (27.3%)	63.6 ± 37.4 items/L 89.5 ± 20.6 items/L (After the typhoons)	[79]
China (Jiaozhou bay)	a clean stainless-steel hydrophore with a 20 μm mesh-sized sieve	1.5 g/mL $ZnCl_2$ solution (sediment)	ATR-μ-FT-IR	3–4 mm particle size (35.71%), 0.5–0.99 mm (28.57%), and 1–1.99 mm (25.01%)	1–6 items/50 L (equivalent to 20 items/m^3–120 items/m^3)	[48]
China (seven small-scale estuaries, Shanghi)	stainless-steel apparatus	30% H_2O_2, NaCl	μ-FT-IR	<2 mm (99.5%) (0.02 mm–2.535 mm)	13.53 ± 4.6–44.93 ± 9.41 particles/L (27.84 ± 11.81 particles/L)	[49]
China (Bohai Sea)	manta net (mesh of 0.33 mm)	0.05 M Fe (II) solution and 30% H_2O_2	μ-FT-IR	<5 mm (69.49%)	0.49 ± 0.18 particles/m^3 and 217,639 ± 83,843 particles/km^2 (Four season)	[34]
China (Nansha Islands)	neuston trawl (333 μm mesh size)	30% H_2O_2, NaCl solution (1.20 g/cm^3)	μ-FT-IR	1–2 mm size (30.4%) 209 μm–4917 μm (2234.26 μm)	0.0556 ± 0.0355 n/m^3	[27]
China (mid-west Pacific Ocean)	manta trawl (333 μm mesh)	30% H_2O_2, $FeSO_4$, NaCl	μ-Raman	0.3–0.5 mm (18.5%), 0.5–1 mm (28.5%), 1–2.5 mm (35.1%), and 2.5–5 mm (17.9%)	6028–95,335 pieces/km^2 (34,039 ± 25,101 pieces/km^2)	[19]
China (the South Yellow Sea)	5 mm stainless-steel mesh and a 50 μm plankton net	H_2O_2 (30%), $ZnCl_2$ (1.6 g/mL) solution	FT-IR	50–500 μm (>75%) 1–2 mm (9.6%), 2–5 mm (4%) in January	6.5 ± 2.1 items/L (January),4.9 ± 2.1 items/L (April), 4.5 ± 1.8 items/L (August)	[80]
China (Ma'an Archipelago)	30 L stainless-steel bucket with stainless-steel sieves (5 mm, 0.3 mm, and 50 μm meshes)	0.05 M ferrous sulfate solution and 30% H_2O_2, NaCl	μ-FT-IR	500–1000 μm (48.9 ± 21.9%), 200–500 μm (21.5 ± 18.5%)	0.2 ± 0.1–0.6 ± 0.2 items/L	[33]
China (Qingdao)	20 L stainless-steel bucket with 20 μm mesh-sized sieve	KOH, NaCl, $ZnCl_2$ solution (density 1.50 g/cm^3) (sediment)	ATR-μ-FT-IR	100–330 μm (23.5%), 330–500 μm (13.9%), 500–1000 μm (31.1%), 1000–2000 μm (20.7%)	93.33–991.67 items/m^3 (446.81 ± 75.04 items/m^3)	[43]
Denmark (The South Funen Archipelago, Baltic Sea)	manta trawls, bulk sampling (Integrated Water Sampler; IWS)	HCl (37%), H_2O_2 (50%), Nile Red solution (1 mg/ml)	self-constructed photo box (Pentax K-30, Omnilux UV)	size-fractioned by sieving (mesh sizes 1.0 mm, 0.63 mm, 0.3 mm)	Trawl samples: 0.07 particles/m^3 (±0.02), 12,897 particles/km^2 (±3922) IWS samples: 1.03 particles/L (±0.80)	[133]
Finland (Baltic Sea)	100 μm plankton net (WP2), and a 30 L water sampler (Jussi; large Limnos-type water sampler)	H_2O_2 (30%), 1 ml chitinase enzyme and 30 ml pH 5 acetate buffer (sodium acetate and glacial acetic acid)	FT-IR	20 μm–724 μm (WP2)/8386 μm (Jussi)	Plankton net samples: 0–1.6 MP/m^3 and 0–766 ng/m^3 Water sampler (Jussi): 0.02–1.7 MP/L and 0–775 ng/L	[22]
France (Bay of Marseille)	manta net (mesh size 150 μm),	–	GC/MS		0–0.308 items/m^3 (0.051 items/m^3)	[170]

(*continued on next page*)

Table 1 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
	stainless-steel collector			150–500 μm, 500–1000 μm and >1000 μm		
Germany (the urban Kiel Fjord, southwest Baltic Sea)	260-cm long net (300 μm mesh size)	–	FT-IR	0.3–18.2 mm long (median size = 1.3 ± IQR 1.4 mm)	0.0–1.8 particles/m^3 (0.04 ± 0.06 particles/m^3)	[73]
Greenland (Nordic Sea)	pump and plankton net with a 5 mm stainless-steel sieve	30% H_2O_2, $ZnCl_2$ solution (1.6 g/cm^3)	FT-IR, SEM-EDS	2–5 mm, 1–2 mm, 0.5–1 mm and 0.1–0.5 mm	East Greenland Current: 1.19 ± 0.28 items/L Greenland Sea Gyre: 2.43 ± 0.84 items/L	[29]
Indonesia (Benoa Bay, Bali)	mini manta trawl-net (mesh size 300 μm)	H_2O_2 (30%)	μ-FT-IR	500–1000 μm (37.9%), >1000 μm (35.7%), 300–500 μm (22.1%), and <300 μm (4.3%)	wet season: 0.61 particles/m^3 dry season: 0.62 particles/m^3	[25]
Indonesia (the northern coastal waters of Surabaya)	sterile HDPE bottle with a 3-inch diameter stainless-steel filter (mesh sizes: 5 mm and 200 μm)	H_2O_2 (30%)	ATR-FT-IR	<300 μm (0.122%), 300–500 μm (45.478%), 500–1000 μm (48.539%), and >1000 μm (5.861%)	0.38–0.61 N/L (0.49 N/L)	[41]
Indonesia (Small Islands of Bintan)	Neuston net	1% of H_2O_2, 3 M of $ZnCl_2$	ATR-FT-IR	500 μm–1000 μm (19–35%), 300 μm–500 μm (15–36%), 100 μm–300 μm (15–40%)	0.46 ± 0.25 pieces/m^3	[135]
Iran (Chabahar Bay, Gulf of Oman)	neuston net (333 μm mesh size)	0.05 M Fe (II) solution with 30% H_2O_2, NaCl	ATR-FT-IR	100–500 μm, 500–1000 μm, 1000–3000 μm	0.07 ± 0.03–1.14 ± 0.27 particle/m^3 (0.49 ± 0.43 particle/m^3)	[24]
Israel (Mediterranean coast)	The manta net (333 μm mesh size)	4% formalin	stereomicroscope	<0.3 mm, 0.3–5 mm and 5 mm–2.5 cm	7.68 ± 2.38 particles/m^3 (1,518,340 particles/km^2)	[130]
Italia (major Italian river mouths as sources)	a manta trawl and a plankton net (WP2) (333 μm mesh net)	–	μ-FT-IR	–	0.641–0.119 items/m^3 (0.297 ± 0.044 items/m^3)	[50]
Italia (Ligurian and Tyrrhenian Seas)	manta trawl (330 μm mesh size)	–	ATR-FT-IR	<1 mm (1.7%), 1–2.5 mm (21.6%), 2.5–5 mm (41.6%), >5 mm (35%)	1009–122,817 particles/km^2 (28,376 ± 28,917 particles/km^2)	[57]
Italy (Ligurian Seas and Tyrrhenian Seas)	manta trawl (330 μm mesh)	–	ATR-FT-IR	<5 mm (88.7%); 2.5–5 mm (29.5%), 1–2.5 mm (55%), <1 mm (4.2%)	1286–3,814,018 particles/km^2 (255,865 ± 841,221 particles/km^2)	[37]
Italy (Tuscany coast)	manta trawl (330 μm mesh size), WP2 standard ring net (200 μm mesh size)	–	FT-IR	<0.5 (minor portion), 0.5–1.0, 1–2.5 (most abundant), 2.5–5.0 mm	41.1 g/km^2 and 69,161.3 items/km^2 (surface water) 0.26 items/m^3 (water column)	[20]
Italy, Slovenia, Croatia, Montenegro and Greece (five gulfs of the Adriatic Sea)	manta-nets (net opening of 330 μm)	H_2O_2 digestion	ATR-FT-IR	330 μm–1 mm (SMP, 34%), 1 mm–5 mm (LMP, 64%), >5 mm (Meso-plastics 2%)	315,009 ± 568,578 items/km^2 (217 ± 575 g/km^2)	[56]
Jameica (Kingston Harbour)	335 μ mesh manta trawl	0.05 M Fe(II) solution and 30% H_2O_2, NaCl solution (~5 M)	FT-IR	0.335–1 mm (24%), 1–2.5 mm (47%), 2.5–5 mm (22%), >5 mm (7%)	0.76 particles/m^3 (359,593.41 particles/km^2)	[59]
Japan (Hiroshima Bay)	a neuston net (mesh size of 350 μm)	sodium solution (1.7 specific gravity)	FT-IR, FE-SEM, X-ray CT	0.3–5 mm	0.004–0.06 pieces/m^2 (surface water)	[42]

(*continued on next page*)

Table 1 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
					552–9128 pieces/m^2 (bottom sediments)	
Japan (Island of Okinawa)	manta trawl (300 μm net)	–	μ-Raman	1.4 μm–47.8 μm (2.53 ± 0.85 μm (PS)–28.4 ± 9.4 μm (PVC))	–	[76]
Korea (Korean coasts)	stainless-steel beaker, portable hand nets (20 μm mesh size)	H_2O_2 (35%) and iron (Fe(II)) solution, lithium meta-tungstate solution (LMT; 1.6 g/cm^3)	ATR-μ-FT-IR	46–13,298 μm <300 μm (83%)	520 ± 130–2200 ± 760 n/m^3 (1400 ± 560 n/m^3)	[28]
Malaysia (Offshore and Estuary located in the Terengganu)	mobile water pump	4% formaldehyde	μ-FT-IR, SEM, microscope	fragments: 68.0–144.0 μm, fibers: 400.1–500 μm	421.8 ± 110 particles/m^3 (estuary), and 211.2 ± 104 particles/m^3 (offshore)	[161]
Mexico (four tropical bays)	neuston nets (250 μm mesh size)	–	Raman	<2 mm (>50%)	0.01–1.05 particles/m^2 (0.45 particles/m^2)	[53]
Qatar (northeastern section of Qatar's EEZ)	plankton tow-net (mesh size: 120 μm)	1 M NaOH	ATR-FT-IR	Granular: 125 μm–1.82 mm, fibrous: 150 μm–15.98 mm	0–3 particles/m^3 (~ 0.71 particle/m^3)	[142]
Russia (southeastern part of the Baltic Sea)	PLastic Explorer (PLEX) instrument (pumping system-bulk water sampling)	the Fenton's reagent, HCl solution	μ-Raman	>174 μm	32.2 ± 50.4 pcs/m^3	[23]
Russia (Barents, Kara and White Seas)	Manta trawl (330 μm mesh size)	–	FT-IR	–	28–963 thousand items/km^2	[47]
Scotland (Scottish Marine Regions, Offshore Marine Regions)	335 μm neuston net	–	μ-FT-IR	–	0–91,128 microplastics/km^2 (4565 microplastics/km^2)	[58]
Slovenia (Slovenian part of the Northern Adriatic)	Neuston (epineuston) net (mesh size of 300 μm)	–	near-infrared spectrometer (NIR)	2,69 ± 0,04 mm (area: 3,09 ± 0,08 mm^2)	472 × 10^3 ± 201 × 10^3 particles/km^2 (6,29 ± 2,68 particles/m^3)	[78]
South Africa (Port of Durban)	Plankton Pump with stacked sieves (5 mm, 500 μm, 300 μm, and 200 μm)	Nile red, 30% KOH: sodium hypochlorite (NaClO) solution	ATR-FT-IR, particle size analysis (PSA), and CTD multi-channel logger	200–300 μm, 300–500 μm, 500 μm–5 mm	highest: 145 particles/m^3	[46]
Sweden (Baltic Sea)	the manta trawl (standard mesh size of 335 μm and 80 μm net)	H_2O_2 (33%)	FT-IR	–	near central Stockholm: 4.2 × 10^5 plastics/km^2 offshore areas: 4.7 × 10^4 plastics/km^2	[68]
Sweden (Skagerrak/Kattegat, Baltic Sea and Gulf of Bothnia)	a manta trawl (mesh size of 333 μm) and an in-situ filtering pump (stainless-steel) with a filter stack	–	Near-infrared hyperspectral imaging	trawl samples: >0.3 mm (88%) pump samples: >0.3 mm (91%)	manta trawl samples: 0.04 particles/m^3 pump samples: 0.1 particles/m^3	[131]
Tunisia (Bizerte lagoon, Southern Mediterranean Sea)	water pump with a cylindrical stainless-steel filter (mesh size of 300 μm)	10% KOH solution, NaI (1.65 g/cm^3)	ATR-FT-IR	–	66.7–1766.7 items/m^3 (453.0 ± 335.2 items/m^3)	[21]
Turkey (southeastern coast of the Black Sea)	WP2 net with 200 μm mesh	4% borax-buffered formaldehyde	stereomicroscope	0.2–5 mm (92%)	1.2 × 10^3 particles/m^3 in November of 2014 0.6 × 10^3 particles/m^3 in February of 2015	[55]
Turkey (Küçükçekmece Lagoon)	12 V DC Teflon pump with stainless-steel sieve (mesh size of 50 μm)	4% formalin solution, H_2O_2 (30% v/v)	FT-IR, Raman, SEM-EDS, Py-GC-MS	–	33 particles/L (lagoons)	[35]

(*continued on next page*)

Table 1 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
United States (Chesapeake Bay)	The manta trawl (330 μm mesh net)	wet peroxide oxidation (WPO) method with an Fe (II) catalyst, NaCl	FT-IR	0.10–10 mm (average: 4 mm)	32,000 particles/km^2 (0.160 particles/m^3)	[51]
United States (Lower Hudson River Estuar)	333 μm Neuston net with a 1 L plastic bottle attachment	Fe (II) WPO process, ~5 M NaCl	stereomicroscope	–	2016: 243,772 particles/km^2, 2017: 143,204 particles/km^2, 2018: 830,762 particles/km^2, 2019: 244,142 particles/km^2	[52]

and paint chips from the surface of a ship and fall off into the water in a port with many ships and is subsequently found as a fragment/flake type microplastic [38]. Moreover, forms of polystyrene (PS) were found in marine farms that required a large number of buoys [41,42]. In coastal areas adjacent to densely populated cities, microplastics were mainly in the form of fibers under the influence of sewage effluent, and rayon was found the most on beaches near tourist destinations [36,43].

Microplastics from mariculture were detected in the marine industry. According to a study conducted in Xiangshan bay, China, the composition of microplastics derived from mariculture were PE, PP, PS, and rubber [44]. Polyethylene terephthalate (PET) and PE were found predominantly in mariculture from structures including waterproof membranes or shed films [45]. The proportion of microplastics derived from mariculture in seawater was 36.8–55.7%. Microplastics in seawater are influenced by the surrounding environment and have a complex causal relationship [44]. In the Port of Durban, South Africa, sewage overflow, rainwater drainage, port operational activity, and rivers all contributed to microplastic pollution [46].

In addition to the marine industry, plastic waste generated from the land flows into seawater via rivers, contributing to seawater microplastic pollution. Studies suggesting that the Severnaya Dvina River may play an essential role in transporting microplastics into the White Sea support the notion that rivers may be channels for microplastics [47]. Polymer-type microplastics were consistent in the bay than in the estuary [48], and the fiber-type was dominant in the vicinity of many nearby wastewater treatment plants (WWTPs) [49].

Plastic waste correlates with population density [50]. Microplastic concentrations are high where large rivers or tributaries near major cities meet the Chesapeake Bay, United States. Among cities, regions with high population density and large impermeable surfaces contributed enormously to microplastic pollution [51]. High concentrations of microplastics were found in offshore samples associated with sewage overflow sites [52], and sewage outlets were identified as the significant cause of microplastic dispersal into the water environment [50,53].

It was estimated that 80–90% of marine plastic pollution originates from terrestrial sources [54], and the need for plastic waste treatment strategies in urban areas has been emphasized [51]. A high concentration of microplastics was found around Suwung landfill, Indonesia, and the landfill was suspected to be the primary source of microplastic pollution [25]. Floating waste in seawater is associated with terrestrial pollution and river inflow [20]. Therefore, plastic waste from terrestrial sources cannot be ignored.

As microplastics accumulate in relatively closed areas [35], geological characteristics should be considered as they affect the concentration of microplastics in seawater. The Black Sea is known as one of the most polluted semi-enclosed seas globally [55], and the semi-enclosed bay of Maowei Sea, China, shows high concentrations of microplastic pollution due to poor circulation of ocean currents [24,36]. The Adriatic waters, Italy, are also heavily influenced by WWTPs as they are surrounded by land [56]. In contrast, the Kara Sea remains relatively clean as it is separated from the Atlantic influent [47]. In addition, the distance between the land and the sea can be a factor that affects the microplastic properties. However, microplastics detected in the mid-western Pacific Ocean away from the land were contributed by fishery activities but not from terrestrial sources [19].

3.1.2.2. Shapes of microplastics found in seawater. Microplastics found in seawater exist in various forms; from the initial form of the plastic during the manufacturing process to the decomposed form after being discarded and exposed to the environment. Fragments are one of the common forms of plastic waste found after the plastic has been broken down by ultraviolet light and mechanical forces from the wind and waves [57]. Approximately 70% of microplastics in the Scottish marine regions between 2014 and 2020 were fragmented plastics [58]. Fragments were the most abundant form of microplastics found on the Tuscany coast, Italy [20], Benoa Bay, Indonesia [25], Kingston Harbor, Jamaica [59]; in the Lower Hudson River Estuary, USA, 70% of the microplastics found from 2016 to 2019 were fragmented [52].

Pellet-type microplastics were found predominantly in the sea around Hong Kong Island. It is suggested that they may originate from terrestrial sources, including industrial activities, stormwater/wastewater discharges, illegal disposal, and accidental runoff [60]. Specific shapes, such as pellets and lines, are likely to exist under the direct influence of different industries. For example, small pieces of line or net exist through wear and tear on line-type equipment used in fishing [61]. Additionally, pellets are a form of precursor used to make plastic-based products; the presence of these substances in the natural environment may result from poor material handling in the plastics industry [62].

Foam- or bead-type plastic products are used for specific industries or purposes. Foamed polystyrene (FPS, expanded polystyrene)

are used to float mariculture facilities above surface water [42], and microbeads are used in the production of exfoliating scrubs [63]. In the surface waters of the Nanxum Reef in the South China Sea, blue microbeads were detected as the main type of microplastics [26], and pink beads made of polypropylene (PP) were found in Scotland's seas [58].

Microplastics in the film form are caused by the fragmentation of plastic bags but may have originated from vinyl used in agriculture [64]. Microplastics in the form of fibers may have originated in the fishery sector from the use of fishing nets and ropes [65]. Fibrous microplastics derived from laundry washing are propagated through freshwater and municipal sewage discharge [23,66].

By analyzing the composition of microplastics found in the central Mexican Pacific according to their shape, it was confirmed that the fragments were composed of PP or PE; the fibers were composed of PE or polyester (PES), and the films were made of PE [53]. However, not all compositions are identical depending on the shape of the microplastics. Both the shape and composition may vary depending on the manufacturing process and end-product of the plastic.

3.1.2.3. Microplastic compositions and properties. Microplastics found in seawater are composed of PE and PP [20,27,34,40,44,60]. Both types of plastics account for 60% of global plastic production. They are low-density plastics with a density of less than 1 g/cm^3; thus, they float on the sea surface [67,68]. PE and PP were predominantly detected in the surface waters of the Bohai Sea, China [40]. The PE found was in the form of faded green or blue lines. PP, a component of food packaging, has brittle properties. Both PE and PP can be attributed to the wear and tear of fishing gear, which are generated due to high-intensity fishing activities [26,27]. Moreover, PE and PP are used in various fields on land. PE is used in agriculture, food packaging, and the manufacture of plastic bottles and bags. PP is used in plastic containers, food packaging, carpets, and pipes. Both components, which have a lower density than water, originate from land and can be transported to the ocean through ocean currents [64]. PE has a relatively high impact strength compared to PP but its disadvantages include lower working temperature and tensile strength [69] than those of PP. As PP has low resistance to UV and oxidation, it ages faster in marine environments and breaks down more easily into smaller particles [70,71], producing many fragments over two orders of magnitude than does PE [72].

FPS was the most abundant, after PE and PP in the Hiroshima Bay, Japan. FPS is mainly used as a floating material to maintain oyster farming facilities at sea level. The results of this study are considered to be influenced by the local industry [42]. In South Korea, the marine environment has been polluted by buoys made of such porous PE [28]. On the northern coast of Surabaya, Indonesia, PS, commonly found in Styrofoam products, was the most dominant type of polymer (58.44%) [41]. Styrofoam made of FPS is mainly used in thermal insulated and food packaging buffers foam boxes, as well as in aquaculture [34]. PS with a porous structure is sensitive to frictional force [44]. It is easily broken into small pieces but is difficult to decompose and biodegrade naturally. Moreover, the small pieces are difficult to recover once dispersed in the water environment [34]. When exposed to the environment, the FPS ages and the internal pores of the FPS collapse, resulting in a different specific gravity and can sink to the sea bottom sediment. An overwhelming amount of FPS was found on the beaches and bottom sediments of the Hiroshima Bay, Japan. The shape of the FPS found on the beach was thick and round, while it exhibited a thin and complex shape in the sediments. The average size of FPS particles in bottom sediments was smaller than that of the beach FPS particles, but with a higher specific surface area [42].

PET, polyvinyl chloride (PVC), and cellophane microplastics were not found or found in small amounts in the surface water due to their relatively high densities but their impacts must be considered in the marine environment. PET has a negative buoyancy of 1.29–1.40 g/cm^3 and sinks to the seafloor immediately after reaching the sea [73]. PVC is a microplastic polymer most often found in the estuary of Changjiang, China [74]. Cellophane is an organic cellulose-based polymer with a high density (1.50–1.52 g/cm^3) and is mainly used for food packaging. When discharged into the environment, it is prone to sinking due to its high density and is found in sediments and fish samples [33]. It also accounts for 21% of microplastics found in sediments in the Port of Durban, South Africa [46].

Additionally, microplastics of different compositions have been found: polyamide (PA), known as Nylon, used in clothing, packaging, and fishing materials [75], and some rare polymers and waxes used in food and dentistry [56]. Although constituting only 3% of the microplastics found, rubber fragments were found in the Baltic Sea and were postulated to be caused by car tire abrasion [73]. In the marine water of Qatar, copolymers and alkyd resins were also found, which were deemed to have arisen from ship hulls and ballast water tanks.

3.1.2.4. Environmental factors affecting microplastic variability. The abundance of microplastics was influenced by anthropogenic factors, as well as environmental factors such as the surrounding environment, currents, and weather conditions [26,64]. Intensive economic activity deteriorates water quality, and it was hypothesized that the concentration of microplastics is affected by economic activity based on the correlation between low water quality and high concentrations of microplastics [49]. Some particles may have migrated from a distant location, but the correlation between microplastics and population density indicates that the plastic particles originated from terrestrial sources. In particular, the abundance of small microplastics was highest in regions related to more intensive anthropogenic activities [76]. Moreover, the highest concentrations of microplastics in the sea surface waters near Scotland were found around the most urbanized and industrialized areas. However, even in regions with a low population density, high concentrations of microplastics were detected at peripheral points in industrialized areas [58]. In general, the level of pollution decreased as the distance from the estuary to the open sea increased; the concentration of microplastics decreased as the distance from Yangtze Estuary to the East China Sea increased [33]. In contrast, on the Tuscany coast of Italy, the amount of floating microplastics increased with distance from the coast [20].

A study on the seasonal distribution characteristics of microplastics on six beaches along Qingdao, China, revealed that the geographic variation of microplastics was related to coastal currents [43]. The amount of microplastics on the six beaches increased from east to west, which coincided with the direction of the ocean current. Ocean currents and winds are significant factors governing

the dispersion and distribution of microplastics in the surface microplastic pollution in the four tropical bays of the central Pacific coast in Mexico [77]. Based on the findings of the study on sea surface microplastics in the Slovenian part of the Northern Adriatic, the factors affecting the concentration of microplastics in the ocean were strong wind, wind mixing, and sea surface currents [78]. Ocean currents transport microplastics [19,43], reducing the concentration of microplastics [33] and concentrating or depositing microplastics at the center of the ocean circulation [29]. In addition, high-density microplastics have been found in surface waters that have possibly been influenced by factors that affect turbulence and vertical mixings, such as ship movement, tides, wind, and storm events [19].

Weather changes, such as rainfall, snowfall, snowmelt, and typhoons, among other environmental factors, also affect the abundance of microplastics [34,38,73,79]. In the Marmara Sea, Turkey, the highest microplastics concentration was detected in autumn and the lowest in summer [35]. In contrast, in the southwest Baltic Sea, high concentrations of microplastics were found after rainfall and snowmelt. Although the mechanism by which microplastics enter the marine environment due to snowfall is unclear, rainwater drains are an important source of microplastics entering the marine environment [73]. In the Southern Yellow Sea of China, the abundance of microplastics was highest in January, the winter season, with the fiber shape being prominent. In August, the rainy season, the ratio of PP, PA, and PET polymer increased resulting in the most remarkable change in microplastic properties [80]. Although rainfall causes a change in the composition of microplastics, typhoons that have the potential to cause violent agitation and release microplastics from sediments, cause more significant fluctuations than rainfall. In the Sanggou Bay, China, before typhoons, microplastics in the form of fibers were dominant in seawater, sediments, and oysters. After the passage of a typhoon, the average microplastic concentration increased by ~ 40%, and the concentration of the fragment-type increased significantly. In addition, the ratio of PP, PS, and PET polymer also increased. In particular, after the typhoon, the microplastics in oysters more than doubled from 19 to 71 to 43–164 per individual [79]. Meteorological conditions must be considered in the study of marine microplastics; bad weather can cause changes in the composition of microplastics and can transfer microplastics from land to sea. However, instead of only analyzing the effects of the seasons on microplastics, the potential complex effects that the weather can have on microplastic abundances, such as anticyclonic gyres, isolated vortices, winds, water currents, and terrestrial pollution dispersion sources, should be considered [20].

Microplastic abundance on beaches is associated with, but is not limited to, ocean currents, wind, and weather events [60]. Due to the inherent properties of microplastics, such as hydrophobicity and specific gravity, they are not evenly distributed in the water column [74]. Owing to these properties, the seawater salinity gradient affects the abundance of microplastics. There were more microplastics in the seawater than in surface waters in the Baltic Sea owing to the differences in the density, salinity, and seawater stratification, which affects the sinking rate of microplastics [22]. In the Black Sea, it was suggested that the sinking rate of microplastics may vary due to the difference in density between brackish water, surface water, and seawater. Furthermore, a permanent halocline within a depth of 100 m was identified that significantly limits the exchange between the surface and deep water [55]. Seawater is classified by density due to its salinity gradient; therefore, based on the salinity gradient, microplastics continue to flow into some sections while keeping it relatively clean in other sections [47]. In the water column of the Baltic Sea, Russia, it was demonstrated that the vertical thermohaline water structure contributed to the role of microplastic sink buffer. Fibrous microplastics appeared predominantly in the subsurface layer, especially at 6.9 m, and decreased as the depth increased. This pattern is similar to the thermohaline structure, further reinforcing the notion that temperature and salinity gradient influence the distribution of microplastics. The thermohaline circulation in the world ocean, such as polar water transport, surface currents, deep-sea backflow, upwelling, downwelling, and other processes, is due to the temperature and salinity stratification of the world ocean, which are natural forces that circulate the vertical world oceans [23].

In addition to physicochemical factors, biological factors influence microplastic abundance and distribution. Among the low-density microplastic particles (which are less dense than the ocean density), the fibrous form remains afloat on the surface for 6–8 months, and the spherical particle floats for 10–15 years and then sinks as it loses buoyancy through biological contamination [53]. Microplastics do not sink, even with the density of pure microplastics, because of the formation of biofilms on the surface of microplastics that could influence their sinking rate [22]. However, owing to various biological actions, microplastics in seawater eventually sink into the deep sea over time, increasing the concentration of microplastics in sediments. Abundant microplastics were found in regions where there are residual currents and sediments. Moreover, the number of microplastics in seawater showed a positive correlation with the number of microplastics in sediments [48].

Microplastics entering the water system from the atmosphere cannot be ignored. Seawater samples from the Port of Durban, South Africa, are affected by atmospheric deposition. In particular, it is more concentrated in the vicinity of intensive anthropogenic activity. Potential atmospheric microplastics are also observed outside of the Port [46]. A study conducted in 2014 showed that global warming could affect the abundance of microplastics by releasing microplastics bound to ice in the Arctic Ocean [29]. In addition to these factors, it is difficult to predict the transport, distribution, and abundance of marine microplastics because several factors such as ocean hydrology, weather, physicochemical properties of microplastics, and biological processes influence the behavior of ocean microplastics [80]. The Pearson correlation analysis conducted in the Bizerte lagoon and the Sbibiouthern Mediterranean Sea, Tunisia, confirmed that environmental factors such as salinity, wind speed, water depth, pH, and temperature have different effects on each type of microplastics. Microplastic distribution depends on various environmental factors [21]. As the sources of microplastic pollution are diverse, it is difficult to attribute them to a specific source or cause [51], thus, more data are needed and more factors need to be considered to clarify the relationship between the abundance of microplastics and the factors influencing them [52].

3.2. Microplastics in wastewater

3.2.1. Microplastic abundance and its removal rate in wastewater treatment

Microplastics are discharged into the aquatic environment from the plastic industry or domestic wastewater, such as the use of personal care plastic-containing products and the washing of synthetic clothing [81]. According to the analysis of microplastics in the influent and effluent of seven WWTPs in Xiamen, a typical coastal city located in southeast China, 79.3–97.8% of the microplastics were removed by the WWTP. Based on the daily effluent and removal rate of microplastics, the non-negligible amount of microplastics discharged was $\sim 6.5 \times 10^8$ MPs from the seven WWTPs of Xiamen city each day [82]. However, it was considered a significant source of microplastics [83]. Among the WWTPs, the removal rate of microplastics in the membrane bioreactor (MBR) was ~ 60% better than that in the conventional activated sludge (CAS) process, but the removal rate of microplastics smaller than 0.25 mm was low [84]. Secondary Anaerobic-Anoxic-Oxic (A2O) treatments exhibited high microplastic removal rate (>90%) [85]. However, most microplastics were removed during primary treatments including solids skimming and sludge settling processes. Notably, the importance of effluent filters was also evaluated as low [86]. As the microplastic removal rate is affected by its size, shape, and type in wastewater treatment [82], the removal rate of microplastics may vary among studies. However, most microplastics tend to be removed through the WWTP. In contrast, 98% of microplastics were removed from the WWTPs in Denmark, and 3 t of microplastics were discharged per year. However, compared to the total annual microplastic emissions of 600–3100 t, microplastic emissions from the WWTPs were non-significant [87], suggesting that there are other sources of microplastics pollution (e.g., marine industrial waste, land-based plastic litter from rainfall, worn tire tread from the atmosphere) that can enter the aquatic environment. If an average of 5–13 million tonnes of plastic enters the ocean every year, it is estimated that by 2050, the weight of plastics discharged will exceed the weight of all fish in the ocean [39]. Therefore, even if a large amount of microplastics is being removed through wastewater treatment, remaining low concentrations of microplastics cannot be ignored.

3.2.2. Results of microplastics analysis and factors influencing microplastics

Based on the microplastic analysis of wastewater, fiber-type microplastics were found the most, followed by the fragments and film types [81,88]. Fiber-type microplastics are defined as having a length between 3 nm and 15 mm and a length-to-diameter ratio greater than three. According to the Annex XV restriction report for intentionally added microplastics by the European Chemicals Agency [89], the Granule type was also among the most found microplastics and is thought to be due to industrial wastewater [82]. However, in most studies, the fiber-type was enumerated the most, and 90% of microplastics found in wastewater effluent consist of microfibers [84,90]. While most of the fibers found in WWTPs are anthropogenic and derived from textiles, some of the fibers found in wastewater may not be plastic, such as cellulose-based, and are derived from other anthropogenic sources [91]. The size of the fiber-type microplastics is smaller than the fragment-type; therefore, they occur in high abundance in the sludge [85]. Most low-density plastics produce fiber-type microplastics or films, and high-density plastics produce microplastics as fragments and flakes. There is a possibility that high-density plastics may not have been sufficiently considered in the analysis of microplastics, which use low-density separation solution such as NaCl [85,92]. PET and PE are most often found in the composition of microplastics [81,84], and PP or PVC tends to vary depending on the sampling location and surrounding environment [88,93]. Although the color may vary depending on the dye used in the plastic, the color of the microplastics found at a WWTPs located in Cartagena, Spain, was beige (36.9%), followed by white (23.6%), black (7.8%), blue (7.0%), and green (3.9%) [94]. Indigo, indigo carmine, or cellulose has been identified as a constituent of most blue fibers. One of the reasons why these fibers are found in wastewater is the washing of blue denim jeans [90]. Fibers generated from laundry have an average length of 360–660 μm and an average diameter of 12–16 μm. They can pass through the WWTPs and pose a threat to marine organisms owing to their elongated structural characteristics [95]. According to the study on microplastic occurrence in wastewater, as fiber-type microplastics are predominant compared to other types, they can pose more severe problems; therefore, the occurrence and fate of microplastics that can flow into water bodies should be intensively studied [84,96].

Microplastic analysis is influenced by the inherent properties of microplastics and environmental factors. The density of microplastics affects their buoyancy and vertical distribution. Compared to the density of water, the low density of certain microplastics can be correlated to that of suspended solids (SS), which float on the water surface, while high-density microplastics tend to settle in deep water and appear in benthic organisms [82,96]. The concentration of microplastics is also affected by environmental factors. According to a study conducted in Toronto, Canada, stormwater, which accounts for 22% of tire wear and asphalt sealant particles, flows into the water bodies and increases the concentration of microplastics originating from cities [90]. In contrast, during periods of low rainfall, microplastics in wastewater have similar characteristics to microplastics found in domestic sewage [97]. The efficiency of microplastic removal in WWTPs is not affected by seasonal variation [94]. Furthermore, the microplastic concentration and fiber-type are influenced by the population density [92,98]. The microplastic concentrations in the form of fragments, foams, pellets/beads, and film were strongly correlated with urban-related watershed characteristics and were higher during rainfall conditions than during periods of low rainfall [99]. These results indicate that fiber is mainly generated through domestic wastewater and is affected by the population density. Moreover, other types of plastics, including fragments, foams, pellets/beads, and film, are affected by urban-related watershed factors. However, it is considered that this may also depend on the surrounding environment.

3.2.3. Migration of microplastics from water bodies to the soil

The sewage sludge from WWTPs is often used for agriculture, and microplastic concentrated in the sludge accumulates in the soil. According to a study on microplastics in sewage sludge from the WWTPs in China, $1.60–56.4 \times 10^3$ of particles per kg were concentrated in sewage sludge. Therefore, it is estimated that 1.56×10^{14} particles are accumulated in the soil when the sludge is used

in agricultural fields [92]. As high-density microplastics are only partially discharged and continue to accumulate, on average, 12% of microplastics in wastewater and 88% of microplastics in sludge are present in the WWTPs [88]. Soils treated with sewage sludge have, on average, 256% higher microplastic particles than non-treated soils [93]. Thus, agricultural sludge is also a significant source of microplastics [85]. However, owing to the absence of natural blank samples in the analysis of soil samples, there is no set validation method [93]. Of note, in addition to the microplastics present in sewage or sludge, the concentration of microplastics can be overestimated due to the leaching of microplastics from procedural contamination such as sampling and pretreatment processes.

Reusing sludge from the WWTPs can cause a link between environmental media (water bodies) and the soil. Wastewater is rich in nutrients and provides an optimal environment for microorganisms to reproduce. Furthermore, the microplastics discharged can create habitats for other bacteria with natural substrates and transport bacteria downstream, creating colonization opportunities for pathogenic bacteria [100,101]. Moreover, there is a higher concentration of microplastics downstream. In addition to measuring the concentration of microplastics in sewage, a survey of sediments, sludges, and the shorelines where microplastics finally reach should be conducted. Ultimately, the fate of microplastics should be investigated, and strong institutional support is needed to minimize the discharge of microplastics into water bodies.

3.3. Microplastics in freshwater

While extensive research has been conducted on seawater microplastics, there is a lack of information on freshwater microplastics [102]. Nevertheless, research on the abundance and behavior of microplastics in freshwater is gaining momentum. The importance of freshwater cannot be underestimated, given that it is a source of water that is supplied to households through water treatment.

3.3.1. Abundance of microplastics in freshwater

3.3.1.1. Current status of microplastics in freshwater affected by the land-based environment. Inland freshwater bodies, in and around areas where plastics are produced, are directly affected by the surrounding environment, reflecting the microplastics present in the freshwater bodies. Studies have also been conducted on the abundance of microplastics in sediments by place or season to understand the impact of the surrounding environment on freshwater (Table 2). Microplastics (13.3 items/L), more than twice the amount in the reference area, were detected in the largest textile industrial area in Asia, located in China. Owing to the production and trade activities of the nearby textile industry, high levels of microplastic contamination have been detected in the local freshwater and sediments [103]. In the surface waters of Nigeria's Ox-Bow Lake, 73.1% of the microplastics found were beaded, and it was suggested that the large number of beads was probably caused due to the cultural heritage involving production of beads and use of beads as fishing gear [104]. The Taihu Lake in the Taihu Basin had a relatively high concentration of microplastics (3.4–25.8 items/L); this can be attributed to the industry and agriculture providing 14% of China's gross domestic product. The dominant particle was cellophane, which is classified as a typical semi-synthetic material [105]. Stormwater treatment wetlands at Gold Coast in Australia are artificial wetlands to treat runoff and stormwater while collecting pollutants along the way. Microplastics were detected in both the water and sediments of stormwater treatment wetlands; synthetic rubber particles potentially from automobile tires were found in the sediments [106]. According to the findings of the analysis of water and sediment samples in the ponds of the European Carpathian basin (region of Hungary), the concentration of microplastics was always lower at the pond outlet than at the inlet. It can be hypothesized that the pond acts as a storage for microplastics [107]. Land-based sources are essential contributors to environmental microplastics and stormwater retention ponds also play a role in transporting microplastics from land to the aquatic environment [108].

3.3.1.2. Potential sources of microplastics based on their shape. Land-based microplastics found within nature can be inferred from potential sources through their shape [109]. The pellet form is a precursor material commonly used to make plastic-based products, usually in the plastics industry [62]. Microbeads may have originated from facial cleansers, while line-shaped microplastics may have come from fishing lines, clothing, or other fabrics. Flakes are a common form of broken plastic with various inferred sources. The flake-shaped microplastics found in the Ofanto river in Italy were black PE of irregular shape and were characterized by brittle and weathered edges. This may be related to the agricultural activities around the Ofanto river [109]. Fiber morphologies were abundantly found in all environments, while other morphologies, such as fragments, films, and foams, were found in regions with high anthropogenic influence [110]. The microplastics found in the water and soil of the Netravathi River in India were predominantly in the form of fibers, but the fragment-type was found in the sediments. Material packaging and fabric fibers are the main sources of microplastics owing to poor solid waste management and outdated wastewater treatment facilities [111].

3.3.1.3. Hydrological influence on the abundance and distribution of microplastics. With freshwater bodies of different sizes and with different topography in different regions, the hydrological process is one of the critical factors that influence the abundance and distribution of microplastics. Microplastics tend to increase downstream in the Suzhou and Huangpu rivers, China, and microplastic pollution is high in urban centers and estuaries. In contrast, the amount of microplastics in the form of fibers tends to decrease from small urban water bodies to the sea. The concentrations of microplastics may have decreased owing to a dilution effect as they moved into the ocean. Small urban waters are more likely to be affected by adjacent pollutants, whereas transport of microplastics in large rivers is more likely to occur through hydrological processes [112]. The finding that the abundance of microplastics is higher downstream than upstream is supported by a study conducted on the Netravathi River in India, which shows the impact of urban population growth and anthropogenic activities on the abundance of microplastics in the river [111]. Similarly, in the Nakdong River

Table 2
Results from the analysis of microplastics in freshwater.

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Composition	Concentration	
Argentina (Río de la Plata estuary)	bucket, plankton net (36 μm)	Fe (II) 0.05 M and 30% H_2O_2, NaCl (5 M)	Stereomicroscope	–	139 MP m^{-3}	[124]
Argentina (Patagonia)	net of 38 μm mesh size	30% H_2O_2	Stereomicroscope, μ-Raman	Indigo Blue (44.1%), PET (38.3%), PU (11.8%), PS (2.9%) and PP (2.9%)	0.9 ± 0.6 MP m^{-3} (0.3–1.9 MP m^{-3})	[121]
Australia (Cooks river)	14 L metal bucket through 37 μm plankton net	NaCl solution (1.2 kg L^{-1}), 30% KOH: NaClO, Nile red	microscope	–	400–17,383 particles/m^3	[126]
Australia (Goulburn River)	Grab sampling (5 L food-grade blue PP jars)	sodium hydroxide (NaOH)	Stereomicroscope, ATR-μFTIR	PES (30.6%), PA (12.9%), rayon (8.1%)	0.40 ± 0.27 items/L	[182]
Australia (Gold Coast)	sampling device consists of four removable stainless-steel mesh screens (25, 100, 190, and 500 μm)	30% H_2O_2, NaI with a density of 1.59 g/mL	dissecting microscope, ATR-μFTIR	polystyrene-co-ethylacrylate (black fragments) followed by PP, nylon, and PES	- inlet: 0.9 ± 0.3 microplastic particles/L - outlet: 4.0 ± 2.4 microplastic particles/L	[106]
Canada (Lake Ontario)	stainless-steel bucket with PP rope	saturated calcium chloride solution ($CaCl_2$, ρ = 1.4 g L^{-1})	stereomicroscope, μ-Raman	Cellulose, Unknown, Anthropogenic (Dye-Based, unknown), PET, PE, PVC, and PVC additive	13.3 ± 15.5 particles L^{-1} in wastewater effluent, 0.9 ± 1.3 particles L^{-1} in agricultural runoff, 15.4 ± 7.9 particles L^{-1} in stormwater runoff, 0.8 ± 0.7 particles L^{-1} in the lake	[90]
China (Taihu Lake)	steel sampler	H_2O_2 (30%, v/v)	Stereomicroscope, μ-FT-IR microscopy, SEM/EDS	cellophane	0.01×10^6–6.8×10^6 items/km^2 in plankton net samples, 3.4–25.8 items/L in surface water	[105]
China (Suzhou River and Huangpu River)	metal pail (nylon filters of 20 μm pore size), air lift pump	KOH solution (10% w/v)	Stereomicroscope, ATR-μFTIR	fibrous PES (27.7%), rayon (14.4%), PP (8.7%)	0.08 items/L–7.4 items/L	[112]
China (Tibetan Plateau)	2 L stainless-steel bottle	0.05 M Fe (II) solution and 30% H_2O_2	stereomicroscope, Raman	PP (32.69%), PE (29.81%), PS (13.46%) and PET (9.62%)	277.33 ± 95.36 number/m^3 (66.67 number/m^3–733.33 number/m^3)	[123]
China (China Textile City stream)	metal pail	H_2O_2(30%, v/v), NaCl	stereomicroscope, μ-FT-IR	PES (65.7%), rayon (6.7%), PP (6.5%), non-plastic (10.5%)	6.8 items/L in surface water (2.1–71.0 items/L)	[103]
China (Yellow river)	stainless-steel bucket	NaCl solution (1.3 g/mL), H_2O_2 (30%)	optical electron microscope, ATR-FT-IR	PE, PP, PS	wet season: 497 (380–582) items/L dry season: 930 (623–1392) items/L	[183]
Denmark (Ponds)	filtering device (10 μm stainless-steel mesh)	30% H_2O_2 with catalyst, followed by an enzyme treatment (Cellubrix, Viscozyme, Alcalase), NaOH, $ZnCl_2$ solution (1.7 g/cm^3)	μFT-IR microscope	number concentrations PP (71.5%), PE (9.1%), PVC (7.4%), PES (5.5%) and PS (2.2%) mass concentrations PP (48.9%),PVC (39.1%),PES (5.0%), PE (3.9%), and PS (2.2%)	490–22,894 item m^{-3}/85–1143 μg m^{-3} (1409 item m^{-3}/231 μg m-^{3})	[108]
Europe (Carpathian basin)	Developed mobile sampling system. (Pump, PVC hose, filter (2 mm, 300 μm, 100 μm), flowmeter)	30% H_2O_2	ATR-FT-IR	PP, PE	13.79 ± 9.26 particles/m^3 (3.52–32.05 particles//m^3)	[107]

(*continued on next page*)

Table 2 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Composition	Concentration	
India (Netravathi River)	stainless-steel bucket of 10-liter volume	H_2O_2; (30%) in the presence of ferrous solution (Fe (II); 0.05 M), $ZnCl_2$	Stereozoom Microscope ATR-FTIR	PE (58.33%) and PET (28.57%), PVC	288 pieces/m^3 (56–2328 pieces/m^3)	[111]
Indonesia (Ciwalengke River)	grab sampling method	–	light binocular microscope Raman	PES and nylon fiber	5.85 ± 3.28 particles per liter	[114]
Italy (Ofanto river)	plankton nets (333 μm)	30% H_2O_2 in the presence of an iron (II) catalyst, NaCl solution (1.2 g/cm^3)	digital microscope, Py-GC/MS	PE (76%), PS (12%), PP (10%), PVC (0.7%) and TDI-PUR (0.35%)	0.9 ± 0.4 p/m^3–13 ± 5 p/m^3	[109]
Korea (Nakdong river)	stainless-steel beaker, submersible pump	35% H_2O_2 and Fe (II) solution, lithium meta-tungstate solution (1.6 g/cm^3)	ATR-FT-IR	PP (41.8%), PES (23.1%)	293 ± 83 (upstream, February 2017)–4760 ± 5242 (downstream, August 2017) particles/m^3 in water	[113]
Netherlands (Rhine and Meuse Rivers)	glass jars	–	FT-IR	–	100 L^{-1} (48–187 L^{-1})	[115]
Nigeria (Ox- Bow Lake)	clean Teflon pump (stainless-steel mesh)	H_2O_2 (30%, v/v)	micro-FT-IR	Dry season (water, sediment): PET (72.63%) and Plasticised PVC (10.9%) Raining season: Plasticised PVC (81.5%) and low-density polyethylene (LDPE; 4.2%)	Dry season: 1004–8329 items·m^{-3} Raining season: 201–8369 items·m^{-3}	[104]
Taiwan (Fengshan River)	hemp sling with a stainless-steel bucket hanging (50 μm, 297 μm, and 5000 μm)	$ZnCl_2$ solution (density: 1.8 g/cm^3)	ATR-FT-IR, gas chromatograph/mass spectrometer	PE, PET, PA, and PES	Water: 334–1058 items/m^3	[125]
Turkey (Urban Crater Lake)	glass bottles (0.045 mm)	–	μ-Raman, SEM	PE, PP	–	[128]
United Kingdom (Trent catchment)	the metal bucket on a telescopic pole	30% H_2O_2	dissection microscope, ATR-FT-IR	PE (12), PP (3), PS (2), PVA (2), UA (1)	178 plastic particles	[122]
United States (Gallatin River watershed)	grab sample (average of 1.3 L)	–	stereomicroscope, μFT-IR	Semi-synthetic cellulose (30%), PES (20%), PET (13%), Cotton (5%), PP (4%), PA (4%), Urethane (4%), etc.	1.2 microplastics L^{-1} (0–67.5 microplastics L^{-1})	[129]
United States (Hillsborough River)	Neuston net (500 μm)	–	μ-Raman	PE, (51%), PP (24.7%) and PS (5.9%), other polymers (10.5%) including PET (1%) and false positives (7%)	Surface samples: mean concentration 2.36 #/m^3 and 2.36 mg/m^3 water column: mean 0.94 #/m^3 and 0.34 mg/m^3 Bottom sample:July (2.71 #/m^3), August (2.08 #/m^3); July (3.42 mg/m^3), August (2.08 #/m^3 and 14.06 mg/m^3)	[120]
United States (Lake Mead)	microplastics net (100 μm)	30% H_2O_2 and an iron catalase, lithium meta-tungstate (1.6 g/mL)	stereoscope	–	0.44–9.7 particles/cubic meter	[110]

in Korea, the concentration of microplastics tends to increase as it goes downstream, with increase in the proportion of fibers [113].

3.3.2. Factors affecting the distribution of microplastics in freshwater

3.3.2.1. Inherent properties of microplastics. Microplastics present in freshwater vary in abundance and distribution due to their inherent properties along with environmental factors. Analysis results of actual environmental samples from stormwater ponds in Denmark confirmed that small particles are more abundant than large particles due to the properties of the microplastics. In addition, according to the Stroke Law, the terminal velocity of a particle is proportional to the square of the particle's diameter; therefore, large particles tend to either float on the water surface or sink to the bottom of the pond [108]. Water samples from the Ciwalengke River in Indonesia contained many small microplastic particles, while large particles were found in sediments [114]. The density of the polymer, as well as the terminal velocity due to the microplastic particle size, can influence the sedimentation of the plastic particles. Floating particulate matter along the Rhine and Meuse Rivers in the Netherlands showed an abundance of microplastics; moreover, large amounts of microplastics were found in estuary sediments [115].

It is natural for high-density microplastics to settle into sediments over time, but low-density microplastics also often settle down. This is owing to the interaction of microplastics with aggregates, biofouling, and fecal matter that can improve sedimentation by reducing the buoyancy of microplastics and increasing particle density [116–119]. Downstream of the Nakdong River in Korea, 2827 times more microplastics were found in the sediment than in the surface layer [113]. Particles with large particle sizes and rough surfaces easily form biofilms and are more affected by degradation [120]. Due to the inherent properties of microplastics and interactions with surrounding substances, microplastics are not uniformly distributed in the water system. This is supported by the varying distribution of microplastics in water and sediment samples obtained using plankton nets from the Taihu Lake, China [105].

3.3.2.2. Influence of the surrounding environment on the distribution of microplastics. The distribution of microplastics is susceptible to environmental influences, primarily anthropogenic and hydrodynamic factors [111]. In nine lakes across Patagonia in Argentina, microplastics identified were mainly from urban settlements, textiles, and fisheries [121]. Microplastics found in Lake Ontario, Canada, also correlated with proximity to urban areas, indicating the vital contribution of the urban areas in producing microplastics. Dye-based anthropogenic substances and various chemical compositions such as cellulose, PE, PET, and PVC originate from various sources, including the plastics industry, dumping, roads, and effluents. The chemical composition that can specifically confirm the impact of urban centers is cellulose acetate (CA), which is mainly contained in the disposed commercial cigarette butts [90]. In support of this, the spatial distribution of microplastics detected in the Suzhou and Huangpu Rivers in China showed a tendency to increase in cities and estuary waterways [112]. The concentration of microplastics in Lake Mead in the U.S. was high at locations with direct and artificial use and input [110]. It is widely known that sewage reflects all urban characteristics and directly affects freshwater. However, WWTP effluent from three rivers within the Trent catchment in the UK did not significantly increase the concentration of microplastics, confirming that there may be differences in the sewage composition [122]. In general, commercial and industrial areas emit more microplastics than residential and highway areas [108]. Notably, PE was found on the Tibetan Plateau in China, which was shown to have originated from sheets used for mulching to cover the surface of cultivated soils during crop cultivation [123]. As the composition of microplastics can vary depending on the land use around freshwater, it has been suggested that the surrounding land use should be considered when analyzing microplastic loads [108]. An innovative factor that reflects the intensity of anthropogenic activity is the night light index, which can consider tourists and residents [123].

In the Río de la Plata estuary in Argentina, the distribution of microplastics differed significantly by sampling location. High concentrations of microplastics were found in the most urbanized areas with high sewage discharge and turbidity. These results support that microplastics are affected by anthropogenic activity and also have a significant correlation with habitat quality. However, no correlation was found with the measured environmental factors [temperature, pH, turbidity, conductivity, salinity, dissolved oxygen (DO), total dissolved solids (TDS), or wind intensity] [124]. In the Fengshan River in Taiwan, microplastics in the water and sediment strongly correlated with the water quality factors such as river pollution index, chemical oxygen demand (COD), SS, flow velocity, and polymer types [125]. As there are few studies on the correlation between microplastics and water quality parameters, additional research is needed.

3.3.2.3. Weather changes and microplastic migration routes. Weather is one of the factors that can significantly influence the distribution of microplastics, along with the intrinsic properties of the microplastics and the influence of the surrounding environment. In the Crooks river, Australia, the abundance of microplastics increased by 40 times after a storm. The concentration of the microplastics peaked for 2 days after heavy rain and then decreased rapidly over the next 5 days [126]. Heavy rain introduces pollutants, including land-based microplastics, into the river, and the high flow rate caused by the blown water resuspends and transports microplastics deposited on the river bed [127]. In this process, the concentration of microplastics increases rapidly. Rainfall also affects the composition of microplastics. A study conducted in three rivers within the Trent region in the UK found that 22% of particles detected in stormwater consisted of tire and road wear particles [90]. Rainfall is a strong vector for transporting microplastics distributed in the land, water, and environmental media. A study conducted in the Nakdong River, Korea, found that 70–80% of the annual microplastic load is moved during the rainy season and 92% of microplastics are moved through the water column [113].

As stormwater runoff can transport microplastics from land to sea via freshwater, the need for it to be included in the microplastic pathway has been emphasized [108]. Agricultural irrigation in the Tibetan Plateau, China, has become the main route for microplastic transfer through runoffs from rivers to soil [123]. The spatial distribution of microplastics in water and sediments is due to the flow

dynamics and tidal exchanges. Moreover, the microplastic density and anthropogenic discharges account for the distribution of microplastics [125]. In addition, microplastics are transported over long distances by the wind and rain. At the Urban Crater Lake, at an elevation of 2380 m above sea level in Turkey, microplastics that were broken down into small pieces were detected in five locations far from arable land, with less dense microplastics transported into the sea via streams and rivers. It was shown that the wind could move the microplastics to other areas [128]. The possibility of atmospheric deposition was also confirmed by discovering tire particles in a pond near the highway [108]. As aquatic organisms can be exposed to these distantly migrated microplastic particles, the importance of stormwater management strategies to minimize microplastic inflow into the aquatic ecosystem was emphasized [126].

A study measuring flux fluctuations with fluid flow conditions in the Hillsborough River in Tampa, USA, found that under calm flow conditions, advection fluxes were 10 times greater than that of lateral and vertical fluxes; under turbulent conditions, the lateral and vertical plastic fluxes increased 3–10 fold [120]. When turbulence and mixing occur owing to typhoons or heavy rain, the concentration of microplastics can change by the mixing of microplastics in the water column and sediment, regardless of the density of the microplastics. Larger and irregularly shaped plastic particles were more affected by turbulence. In general, vertical flux can increase the concentration of microplastics in places where microplastic pollution has aggravated, but findings from a different study showed a different perspective that microplastics can be diluted depending on the size of the aquatic environment [129]. The concentration of microplastics in the Netravathi River, India, showed a strong correlation with the water level. This indicates a relationship between the concentration of microplastics and the amount of water, suggesting that microplastics originating from the land migrate to the freshwater environment [111].

Under the influence of the weather and the surrounding environment, land-derived microplastics flow into freshwater and consequently affect aquatic life. The abundance of microplastics in fish inhabiting the Taiwan Fengshan River was correlated with SS, pH, and conductivity, and these water quality parameters affected the bioavailability of microplastics [125]. In addition, it has been suggested that the accumulation of polycyclic aromatic hydrocarbons (PAHs) in fish may be due to contaminated microplastics, highlighting the dangers of microplastics in freshwater.

4. Methods for microplastic extraction and identification

4.1. Sampling and pretreatment methods for microplastics analysis

4.1.1. Seawater sampling and pretreatment methods

4.1.1.1. Trawl-net and bulk/pump sampling methods. Seawater sampling has been performed mainly by the trawl-net sampling method using the manta net and WP2 plankton net, while the bulk/pump sampling method was used in some studies. Plastic particles present in seawater are not equally distributed in the water column, unlike other suspended particles [74]. Due to the heterogeneous properties of microplastics, large patches were found on the Mediterranean coast of Israel; one of the patches contained a large number of plastic particles (324 particles/m^3) [130]. The trawl-net sampling method is more suitable for analyzing the heterogeneous distribution of microplastic pollution because it collects samples from a large area of the surface water and has the advantage of being a representative sample [131]. However, it is important to note that plastic particles with positive buoyancy are mainly collected while high-density plastic particles are omitted, and plastics (especially fiber types) smaller than the mesh size of the sampling device pass through; therefore, the amount of microplastics may be underestimated [18,37,58,132,133]. In addition, a reduction in microplastic particles was observed with increasing wind speed [131], and some microplastics were underestimated due to vertical redistribution of mixed-layer particles by wind or wave action [134]. In the net-based sampling method, the sample volume is dependent on calculations; therefore, if the manta trawl-net is partially submerged due to the surrounding environment, such as the waves and wind, the actual sampling volume is likely to be smaller than the calculated volume [131,133]. The risk of contamination may be greater in trawl-net sampling than in bulk sampling due to exposure to the atmosphere during the rinsing procedure and transferring the net to the container [131]. In order to solve the shortcomings of the manta trawl method with one mesh size, a "Simultaneous grading of microplastics size sampler" with a filter device connected to the rear end of the net was developed, and sampling was performed [135]. However, the limitations of the trawl-net sampling method could not be overcome.

The bulk sampling method is more practical than the trawl-net sampling method when collecting point source samples [46]. Furthermore, it is possible to collect samples from various depths [18,131]. In addition, microplastics can be selectively extracted by controlling the mesh size of the filtration device, and microplastic particles can be classified by size by configuring the filter at multiple stages. However, the concentration of microplastics in a small volume of the bulk sample is not representative of the actual concentration of microplastics. The minimum sample volume for analysis should be determined based on the local water quality characteristics, such as organic or microplastic contamination. In particular, when low microplastic concentrations are expected, it is recommended to set the minimum sample volume so that the results are not biased due to insufficient sample volume [133].

According to a study that performed both trawl-net sampling and bulk sampling in the field, more microplastics were found in bulk sampling [136,137]. In another study conducted in the sea around Sweden, 10 out of 11 sampling points had higher microplastic concentrations in the pump sample than in the trawl sample. The difference in the average concentration of microplastics between the trawl sample and the pump sample was the highest at the sampling point in Kattegat, where the difference was 700 times [131]. It is suggested that small microplastic particles were not considered because many studies have used neutron meshes with mesh sizes larger than 333 μm to avoid the risk of mesh clogging [55].

In general, plastic particles are gradually fragmented in the marine environment due to mechanical actions such as photooxidation

and biodegradation [138], and the amount of fragmented microplastics tends to increase geometrically as the size decreases [27,131,139]. When the same seawater sample near Vancouver, Canada, was continuously filtered, the microplastic concentration after using an 8 μm mesh size for filter filtration was 8.5 times higher than that when a 65 μm mesh size was used for filter filtration [132]. Therefore, the mesh size difference between the manta net mesh size and the bulk sampling mesh size is a decisive point in comparing the number of microplastics. To circumvent this issue, the need to integrate a small mesh size into the microplastic sampling method was established [131], and the use a filter smaller than 10 μm was suggested to prevent loss of microplastics [132].

However, not only the mesh size but other factors also affect the measured microplastic concentration. Therefore, even when using a sampling method with the same mesh size, the abundance of microplastics can differ by several orders of magnitude [131]. For instance, other influencing factors include the traveling speed for sampling, wave height, wind speed, inherent properties of the microplastics, salinity, geographic characteristics, and environmental influences [22,26,68,74,78]. In order to investigate the correlation between microplastics, fish larvae and eggs, and environmental variables, the microplastic concentration was evaluated by separating the river and sea into three regions (upper, middle, and lower) [38]. In addition, to evaluate the characteristics of microplastics at different depths of seawater, a sampling device (PLastic Explorer) was developed to collect seawater samples at different depths [23].

Another essential factor that adversely affects microplastic concentration is airborne contamination introduced during sampling. In particular, microplastics in the form of fibers (<50 μm) were not considered in the estimation of the concentration of microplastics because of the risk of airborne contamination during field sampling [73]. During the pretreatment process for microplastic analysis in the laboratory, it was difficult to prevent contamination of the sample from the air; therefore, the fiber-type was excluded from the analysis [42]. In the blank sample, it was confirmed that most microplastics were induced by airborne contamination, and the possibility of reducing the inflow of microplastics was achieved by controlling the causes of microplastic contamination in the field environment [23].

Many factors can influence the microplastic analysis during the sampling process. The sampling method should be established, including the optimization of the composition and specification of the sampling device and the control of the sampling environment. In addition to the methodological approach, ocean circulation, weather conditions, and various sources of waste from anthropogenic activities can affect the homogeneity of microplastic analysis [20]. Furthermore, environmental factors must be considered.

4.1.1.2. Pretreatment method for microplastic analysis: digestion and density separation. The pretreatment method for analyzing microplastics in seawater can be divided into two significant steps: the digestion (organic oxidation) step that removes organic matter attached to plastics to improve the precision of the analysis; the density separation step for extracting microplastics from removed organic matter or contaminants. Samples from a sea area with little organic matter, such as Antarctica, are analyzed directly by a non-invasive method without additional treatment [30]. However, as seawater generally contains marine organisms and organic substances, a pretreatment method is required to increase the precision of the analysis.

Hydrogen peroxide (H_2O_2) is generally used for the digestion of seawater samples because it is effective in oxidizing organic matter, does not affect plastic polymers in the environment, and shows a high recovery rate (70–95%) of microplastics after pretreatment [140]. After hydrogen peroxide treatment, the microplastic samples contain high amounts of carbon, oxygen, and chlorine, which are identified as non-biological organics [141]. However, when hydrogen peroxide is not sufficient to oxidize organic matter, Fe(II) solution ($FeSO_4{\cdot}7H_2O$) is added (Table 1).

Various chemicals such as Nitric acid (HNO_3), Potassium hydroxide (KOH), Perchloric Acid ($HClO_4$), Formic acid (CH_2O_2), Sodium chlorate ($NaClO_3$), and Proteinase K are used to digest microplastics ingested by fish and invertebrates [140]. KOH is widely used to analyze microplastics in marine organisms owing to its powerful basicity and strong decomposition efficiency for tissues of marine organisms [26,36]. However, because KOH decomposes polycarbonate (PC) and PET among plastic polymers, it is necessary to optimize its concentration or use a different digestion solution suitable for the oxidation of organic matter. In seawater rich in organic matter, robust oxidation solutions or bleaching reagents are used, making the original color of microplastics fade or reducing their durability [140]. It is suggested to select a digestion solution according to the characteristics of the type of microplastics to be analyzed. For plankton-rich Qatar seawater, a microplastic extraction technology was developed and optimized using Sodium hydroxide (NaOH) [142].

Low-density microplastics are composed of polymers such as PE (density: 0.91–0.96 g/cm^3) and PP (density: 0.85–0.94 g/cm^3), with a lower density than of seawater (density: 1.02 g/cm^3); thus, they float on the seawater surface [64,143]. NaCl solution (density: 1.2 g/cm^3) was used for density separation of low-density microplastics from contaminants in seawater samples. However, high-density microplastics such as PVC (density: 1.41 g/cm^3) and PET (density: 1.29–1.40 g/cm^3) also can float to the surface due to vertical redistribution caused by changes in the weather, vertical structure of tidal currents, and salinity gradient [74,79,80]. Using a NaCl solution for density separation may cause high-density microplastics such as PVC and PET present on the seawater surface to be omitted from the analysis [60]. PE and PP make up more than 60% of the plastics produced; hence, it is not surprising that they are detected more than other plastics [67]. However, when extracting microplastics using zinc chloride ($ZnCl_2$) solution with a density of 1.5 g/mL, PET, which is denser than seawater, accounted for 56.25% of the microplastics [48]. In order to prevent the underestimation of high-density microplastics, it is necessary to either select a density separation solution that can separate various types of microplastics at once or perform multiple density separation processes that can separate high-density microplastics individually. Microplastics can be underestimated not only because of density separation methods but also because of unestablished pretreatment methods, including digestion methods [55]. Therefore, a standardized pretreatment method is urgently needed.

4.1.2. Analysis of microplastics in wastewater: wastewater sampling and pretreatment methods

Wastewater samples are collected using an electric pump constituting a filtering device made of metal such as stainless-steel or aluminum [81,84,144], and a glass bottle for storage for the analysis of microplastics in wastewater [87,94,115]. However, the size of the filter varies across studies, such as 11, 37, 125, 333, 500 μm, and 1 mm. If a sampling tool with a relatively large pore size such as a manta net of 333 μm is used, microplastics with smaller sizes may be excluded from the analysis. Due to the abundance of organic matter in wastewater, the oxidation process is essential during the pretreatment step, and density separation is carried out to separate microplastics from a complex matrix composed of organic matter. Hydrogen peroxide (H_2O_2) or Potassium hydroxide (KOH) is mainly used for the oxidation process to remove organic matter. However, for effective oxidation, either iron (II) sulfate is added for the Fenton reaction or a cellulase enzyme is used in a biological procedure. The oxidation process with hydrogen peroxide is effective, but if the pretreatment time is prolonged, microorganisms may grow, and it may be unsuitable for filtration [145]. Density separation is performed using reagents with a density greater than 1 g/mL, such as sodium chloride (NaCl; density: 1.2 g/mL), calcium chloride ($CaCl_2$; density: 1.3 g/mL), and zinc chloride (density: 1.6 g/mL) [146]. High-density microplastics (MPs) such as PVC (density: 1.14–1.56 g/mL) or PET (density: 1.32–1.41 g/mL) cannot be separated using a relatively low-density solution such as NaCl (1.2 g/mL); therefore, high-density MPs are excluded from the analysis [92]. To verify the quality assurance of the microplastic extraction process, sampling and pretreatment were performed using samples without microplastics [88]. However, it may not be possible to consider environmental factors that cannot be controlled, such as plastic-containing facilities installed in WWTPs [87,147]. As the wastewater sampling and pretreatment methods for microplastic analysis are not the same for each study, it is difficult to compare data between studies; therefore, there is a need to establish a standard sampling and pretreatment method (Table 3).

4.1.3. Freshwater sampling and pretreatment methods

4.1.3.1. Sampling method for the freshwater environment. The plankton net, commonly used to extract microplastics in seawater, is challenging to use in freshwater owing to the different conditions prevailing in that environment. The plankton net cannot be used unless the water body is large or deep enough to accommodate manta trolls. In addition, facilities such as boats and bridges are required. Moreover, the measured filtration amount may not be accurate even when measured with a flow meter [107]. In three rivers within the Trent catchment in the UK, the grab sampling method was used instead of manta and Neuston nets because sampling was limited to small freshwater bodies [122]. A sampling method suitable for freshwater sampling environment should be selected.

The grab sampling method is commonly proposed because it can collect small microplastics, unlike the general net sampling method, which, however, can represent a variety of polymer types and capture microplastics at great depths [129]. The mesh size is one of the crucial factors in selecting the sampling method. On the Ofanto river in Italy, when plankton nets with a mesh size of 333 μm were used for sampling, small particles passed through the net, resulting in an underestimation of the microplastic concentration [109]. Based on a study on the difference in microplastic concentration according to the mesh size used, up to 100,000 times more microplastic particles were detected when an 80 μm mesh size was used compared to that when a 450 μm mesh was used [99]. As the mesh size may impact the concentration of microplastics in the field, care must be taken in selecting the appropriate mesh size.

4.1.3.2. Digestion: limitations of wet peroxide oxidation in the analysis of microplastics. Digestion using acidic and basic solutions has been used to remove organic matter in samples, but because they dissolve polymers and are not effective in removing some organic substances, an alternative method using oxidation treatment has been proposed [148,149]. Wet peroxide oxidation (WPO; Fenton's reaction) digestion does not significantly affect the polymers and is effective in removing organic matter. However, owing to the high temperature (60–100 °C) generated during the oxidation process, some polymers, such as PA and PE, melt or their weight decreases [110,149,150]. Owing to the oxidation reaction of WPO as well as the environmental weathering conditions, the microplastics are decolorized, leading to the findings of transparent particles in the microplastic analysis [151]. When choosing an oxidizing agent for removing organic matter, its effect on the temperature rise should be considered so that the temperature can be maintained below 60 °C, and further research should be conducted to limit the discoloration of microplastics.

4.1.3.3. Factors for selection of density separation solution. Although denser solutions are better at recovering microplastics from samples, several factors, such as toxicity and cost, must be considered. Zinc chloride has a density of 1.7 g/cm^3 but has the disadvantages of being corrosive and toxic. Sodium tungstate dihydrate ($Na_2WO_4{\cdot}2H_2O$) and sodium polytungstate ($3Na_2WO_4{\cdot}9WO_3{\cdot}2H_2O$) contain barite and have a high density of 1.4–1.6 g/cm^3, but they are costly. As NaCl has a density of 1.0–1.2 g/cm^3, it is challenging to separate high-density microplastics, but it is low-priced and exhibits high solubility and low toxicity. For this reason, in many studies, NaCl was selected as a density separation solution. For effective separation of microplastics, an appropriate density separation protocol should be developed.

4.2. Microplastic identification

Several analytical instruments have been introduced to identify microplastics in water systems (Fig. 3). In Fig. 3, the upper limit of the detection range was set to 100 μm, a size that most microplastic analysis instruments can identify except for the microscope. Although SEM can analyze the surface of microplastics at the nanoscale, the detection range was not expressed because it cannot analyze the types of microplastics. At the beginning of the research, quantitative evaluation of microplastics was performed with the naked eye or using a microscope. The frequency of using infrared-based analysis instruments has increased for the qualitative

Table 3
Results of microplastic analysis in wastewater.

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
Australia (Sydney)	four removable stainless-steel mesh screens	30% H_2O_2 solution, NaI solution	FT-IR spectroscopy	60–125, 125–250, 250–500, and >500 μm	2.2 microplastics/L in the primary effluent, 0.28 microplastics/L, after tertiary treatment 0.21 microplastics/L after RO	[81]
Canada (Toronto)	automated ISCO 6712 samplers, grab sample	$CaCl_2$ solution	μ-Raman	>125 μm	0.8 particles L^{-1} in lake samples, 15.4 particles L^{-1} in stormwater, 13.3 particles L^{-1} in wastewater, 0.9 particles L^{-1} in agricultural runoff	[90]
China (11 provinces)	–	NaCl, H_2O_2 solution (30%)	Scanning electron microscopy (SEM), μ-FT-IR	>37 μm	22.7 ± 12.1 × 10^3 particles kg^{-1} dry sludge	[92]
China (Xiamen)	multi-use pump	30% H_2O_2 and 0.05 M $FeSO_4$ solution, saturated sodium chloride solution	μ-Raman	43–5000 μm	Influent: 1.57–13.69 items/L, Effluent: 0.20–1.73 items/L	[82]
Denmark	glass bottles	Sodium dodecyl sulfate (SDS) as a surfactant, cellulase enzyme, H_2O_2 catalyzed by iron (II) sulfate	Focal Plane Array (FPA)-based FT-IR imaging	500 μm–1 mm, 1–2 mm, and >2 mm	Influent: 7216 particles/L, Effluent: 54 particles/L	[87]
Finland	filtering device with an electric pump	–	imaging FT-IR spectroscopy	>300 μm, 100–300 μm, and 20–100 μm	MBR: 6.9 to 0.005 MP L^{-1} (99.9%), Rapid sand filter: 0.7 to 0.02 MP L^{-1} (97%), dissolved air flotation: 2.0 to 0.1 MP L^{-1} (95%), disc filter: 0.5–2.0 to 0.03–0.3 MP L^{-1} (40–98.5%)	[184]
Finland (Mikkeli)	10-L stainless-steel bucket attached to a metal wire	0.05 M Fe(II) solution, 30% H_2O_2, and cellulase enzymes	FT-IR and Raman	<6 mm, >0.25 mm (250 μm)	MBR permeate: 0.4 MP/L, final effluent of the CAS process: 1.0 MP/L	[84]
Finland (Viikinmäki, Helsinki)	designated filter with an electric pump	–	Stereomicroscope, imaging FT-IR	>300 μm, 100–300 μm, and 20–100 μm	effluent: 2.0 × 10^8 to 7.9 × 10^8 particles per day	[147]
Iran (Bandar Abbas)	water pump and filtered using 333 μm plankton net	KOH, $ZnCl_2$ solution	stereomicroscope, fluorescence microscopy, a field emission scanning electron microscope (FE-SEM),FT-IR	0.003–0.05 mm, 0.05–1.0 mm, 1.0–1.5 mm in wastewater effluent 0.003–1.0 mm, 1.0–2.5 mm, 2.5–5.0 mm in sludge	wastewater: 70.66 (±14.12, SD) MP.35 L^{-1}, sludge: 6070 (±807.25) MPs.kg^{-1}	[88]
Netherlands (Rhine and Meuse Rivers)	glass jars	NaCl, NaOH, H_2O_2	FT-IR analysis	>300 μm (i.e. 300–5000 μm) or <300 μm (i.e. between ca. 10 and 300 μm)	Raw sewage influents: 68–910 L^{-1}, effluents: 51–81 L^{-1}, sludge: 510–760 kg^{-1} wet weight (ww)	[115]
Spain (Valencia)	soil auger	distilled water, NaI mixture.	μ-FT-IR analysis	11 μm–5 mm (<5 mm, >11 μm)	Sewage sludge: 18,000 ± 15,940 light density MPs kg^{-1} and 32,070 ± 19,080 heavy density MPs kg^{-1}	[93]
Spain (Madrid)	Two-liter Pyrex glass bottles	H_2O_2 (33% w/v), NaCl, 1.2 kg L^{-1}	stereomicroscope, FT-IR	25–104 μm, 104–375 μm, and >375 μm, <5 mm	effluent: 12.8 ± 6.3 particles/L, WWTP mixed sludge: 183 ± 84 particles/g, heat-dried sludge bore: 165 ± 37 particles/g	[85]
Spain (Cartagena)	glass bottles with metallic lid	120 g L^{-1} NaCl (2.05 M)	Stereomicroscope, FT-IR analysis.	<200 μm, 200–400 μm, 400–600 μm, 600–800 μm, 800–1000 μm,	3.20 (±0.67) MP L^{-1}, 2.59 (±0.85) MP L^{-1}, 2.13 (±0.38) MP L^{-1}, and 0.31	[94]

(*continued on next page*)

Table 3 (*continued*)

Country (site)	Sampling	Pretreatment	Analysis	Results		Reference
				Size	Concentration	
				1–2 mm, 2–3 mm, 3–4 mm, and 4–5 mm	(±0.06) MP L^{-1}, for GGR, PCL, BRT, and EFF, respectively.	
Turkey (Mersin Bay)	standard zooplankton sampling net (mesh diameter 26 μm)	H_2O_2 (35%)	digital optical microscope, Spectra-Tech IR-Plan microscope coupled to an FT-IR	>26 μm, <5 mm	Influent: 2.8 particles/L at Karaduvar, 3.1 particles/L at Tarsus and 1.5 particles/L at Silifke Effluent: 1.6, 0.7, and 0.6 particles/L in Karaduvar, Tarsus, and Silifke, respectively.	[83]
United Kingdom (River Clyde)	Steel buckets (10 L) attached to steel wire	–	FT-IR	>65 μm	Influent: 15.70 (±5.23) MP·L^{-1}, Effluent: 0.25 (±0.04) MP·L^{-1}	[144]
United States (Chicago)	two neuston nets of 333-μm mesh	0.05 M Fe(II) and 30% H_2O_2, NaCl	SEM	>333 μm	1.94 (±0.81) m^{-3} upstream, 17.93 (±11.05) m^{-3} downstream. 730,341 (±279,341) km^{-2} upstream and 6,698,264 (±3,929,093) km^{-2} downstream	[100]
United States (nine rivers in Illinois)	neuston nets of 333-μm mesh	0.05 mol/L Fe(II) and 30% H_2O_2, NaCl	py-GC-MS	>333 μm	upstream of WWTP effluent sites: 2.355 (±0.375) no./m^3, downstream: 5.733 (±0.850) no./m^3	[101]
United States	extraction pump	30% H_2O_2 in the presence of an iron (II) catalyst	dissection microscope	125–355 μm, >355 μm	Effluent: 0.05 ± 0.024 microparticles/L effluent	[98]
United States (29 Great Lakes tributaries)	neuston net (mesh size: 333 μm)	30% H_2O_2 in the presence of an iron (II) catalyst	dissection μ-FT-IR and Raman	0.355–0.999 mm, 1.00–4.749 mm, and ≥ 4.75 mm	maximum concentration: 32 particles/m^3, median: 1.9 particles/m^3	[99]
United States (Southern California)	125 μm filtering assembly	8.25% sodium hypochlorite (NaClO)	FT-IR spectroscopy	45–180 μm, 180–400 μm, and >400 μm 20–45, 45–180 μm, 180–400 μm, and >400 μm (WRP 1)	Centrate thickening system influent: 51/100 mL, Final effluent: 373/4.23 × 10^5 L	[86]

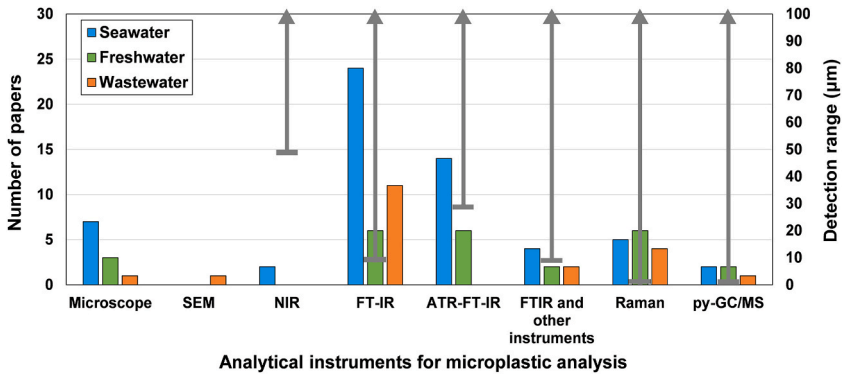

Fig. 3. The number of studies in which microplastic analysis instruments were used and the detection range of microplastics by analytical instruments.

evaluation of various sizes and types of microplastics. In addition, the Attenuated Total Reflectance mode has been applied, and additional analysis such as scanning electron microscopy (SEM)-energy dispersive spectroscopy (EDS) or X-ray CT has been performed. Recently, a Raman spectrometer has been used, with a lower detection limit for microplastics than that of infrared-based analysis methods. However, it is still challenging to identify microplastics at the nanoscale level due to the limitations of spectroscopic analysis, so pyrolysis–gas chromatography-mass spectrometry (Py–GC-MS) is proposed.

4.2.1. Identification of microplastics in seawater

4.2.1.1. Plastic debris (floating marine litter) using the visual transects method. Floating marine debris larger than 2.5 cm is quantified using the visual transects method [152]. Two observers are stationed on each side of the ship, investigating the shape and size of marine debris, and sorting them into bottles, fragments, packaging, and bags [153]. The identified microplastics are defined as potential microplastic particles due to the uncertainty of whether particles identified with the naked eye are plastic or not, and the degree of non- or semi-synthetic anthropogenic origin is high [132].

4.2.1.2. Marco/microplastic particle analysis. The shape and size of plastics at the macro (>100 μm)/micro (100 μm–100 nm) level, which are difficult to analyze with the naked eye, are analyzed using a stereomicroscope. The equipment used for in-depth analysis of the composition and characteristics of microplastics includes Micro-Fourier Transform Infrared Spectroscopy (μ-FT-IR), Attenuated Total Reflectance-Fourier Transform Infrared Spectroscopy (ATR-FT-IR), Near-infrared hyperspectral imaging (NIR), micro-Raman spectroscope (μ-Raman), and SEM-EDS (Table 1); those with the prefix "micro-" are equipped with a microscope to analyze microplastics, and the Attenuated Total Reflection (ATR) mode is chosen for surface analysis of microplastic particles [74].

To confirm the spectra match between the samples and data library, plastic products that are presumed to be the source of microplastics, such as food packaging (PE), bottles (PP), tableware (PS), and fishing tools (PA), are collected and analyzed [74]. In another study, plastic products used in mariculture, such as fishing nets, plastic foam, and plastic film, were collected to confirm the source of microplastics detected at the site [44].

4.2.1.3. Limitation of plastics analysis. In the analysis of microplastics using ATR-FT-IR, most fragments or line-shaped microplastics can be identified as plastics; however, only certain fibers or pellets can be identified [60]. Analysis using μ-FT-IR requires microplastic particles of at least 50 μm [33]. In quality assurance experiments, 100% recovery of particles and colored plastic fibers was observed, whereas the recovery of white and translucent plastic fibers was 60% [68]. The color of microplastics is mainly transparent because most disposable products are transparent, and the majority of colored particles tend to enter the surface water and lose their color [35]. To prevent the underestimation of transparent particles predominantly present in water environments owing to the limitations of spectroscopic analysis, Nile Red (9-diethylamino-5H-benzo [a]phenoxazine-5-one), which selectively dyes microplastics with high hydrophobicity, is used [154].

In addition to the physical properties of microplastics, such as shape, size, and color, the deformation of microplastics under the environment and manufacturing processes influence the spectrum results of the analysis. Most of the microplastics collected from the environment and analyzed were identified as plastics of a specific polymer-type, such as PE and PP, and some were determined to be of other polymer types or mixtures of polymers [68]. Natural weathering and chemical modifications caused by dyes, copolymers, plasticizers, and other plastic additives added during the manufacturing process of plastic can affect the results of FT-IR analysis [155]. The Raman analysis revealed additional Raman peaks according to size reduction, which could be related to structural deformation caused by natural weathering or embedding in the organic matter [76]. Cellulose, which is reported to make up 30% of fibers, has almost identical FT-IR spectra as rayon, a semi-synthetic polymer; thus, arbitrarily excluding these particles may lead to inaccurate results [29].

Thermal analysis, an alternative to spectroscopic analysis, is often used for sewage microplastic analysis because a large amount of

microplastics is required to meet the detection limit [154]. Spectroscopic analysis is mainly performed in seawater. However, in the case of spectroscopy, validation of microplastic identification is missing; relying solely on visual classification can lead to the overestimation of the microplastic concentration as the false-positive rate can be potentially high [153]. In addition, as the number of microplastics is affected by the subjective estimation of the observer, the method of counting non-uniform microplastics should be standardized and is considered an urgent priority in microplastic policy-making [49].

4.2.2. Analysis of microplastics in wastewater

Microplastic analysis method can be divided into spectrometry and thermal analysis. Microplastics of 1–5 mm can be distinguished with the naked eye, and those that are hundreds of μm in size can be analyzed using an optical microscope. FT-IR and Raman spectroscopy are mainly used to analyze microplastics that are at a scale of tens of micrometers. Thermal analysis, such as Py–GC-MS or thermal extraction desorption-gas chromatography-mass spectrometry (TED-GC-MS), is recommended for nano-scale particles [156]. Compared to spectrometry, which analyzes each particle, thermal analysis requires more microplastics to meet the detection limit, and the analysis cost is high. In addition, in complex environmental samples with various unknown plastic composites, small-sized microplastics may be missed [154]. Spectroscopy is time-consuming as it must be validated for every particle suspected of being a microplastic. Black pigmented fibers or clear fragments are often underestimated as they cannot be identified [115,147]. They can also be overestimated because non-microplastic particles are sometimes identified as microplastics, or microplastic particles break into smaller particles during identification [81,144]. In order to clearly distinguish microplastics from non-microplastic particles, microplastics were either dyed using the Rose-Bengal method [81], or the identification and digestion processes were improved, or a bleaching reagent was used to remove any organic substances attached to the microplastics [86].

4.2.3. Constraints of analytical interpretation of microplastics in freshwater

The size of the mesh used for sampling is progressively getting smaller, from the mesh used in the plankton net used to collect microplastics from a large area rich in organic matter to the mesh used in the filter used for grab sampling to analyze small microplastics. However, in the spectroscopic analysis that can confirm microplastic particles' shape, size, and composition, the false-positive rate increased as the particle size decreased [157]. When the size of the microplastic particles was larger than 100 μm, 83% of the particles were visually identified as plastic by spectroscopy, whereas the confirmation rate was only 63% when the size was less than 50 μm [110]. A fibrous black substance found in the Rhine and Meuse Rivers in the Netherlands was identified as a microplastic under the microscope but was not identified by FT-IR [115]. The typical size of microplastics for polymer identification by ATR-FT-IR is $>$ 500 μm [122]. Even the successful extraction of microplastics from freshwater environments may cause constraints in their interpretation owing to the limitations of the analytical equipment. Due to this, small-sized microplastics may be underestimated; thus, care must be taken in interpretation according to the size of microplastics.

5. Potential hazards of microplastics in the environment

5.1. Aging and fragmentation of microplastics due to environmental influences

In general, microplastics in the marine environment are gradually fragmented through physical abrasion, UV irradiation, and biodegradation, and the number of fragmented microplastics increases geometrically as the size decreases [64]. In the surface waters of the Nordic Seas, Greenland, microplastics detected in samples had a flaky and weathered appearance. The fragmented microplastics had irregular cracks, and the surface of the microplastics, in the form of a film, was cracked like onion skins [29]. After the passage of a typhoon, microplastics found in seawater samples showed more wear marks, folds, and breaks than those in non-stormy weather conditions [79]. The aging of microplastics was confirmed through these surface changes, and it was considered a result of the mechanical forces of the wind and waves and the weathering caused by UV rays [37]. Flow rate is also a factor influencing microplastic fragmentation. A strong hydrological process with high flow velocity in the flood season is considered to further contribute to the fragmentation of microplastics [49].

Microplastics are constantly being decomposed in the marine environment under physical, chemical, and biological influences. The rate of microplastic decomposition can vary depending on several environmental factors such as temperature, sunlight, depth of water, and the presence of bacteria [21]. For example, differences in UV irradiation intensity and physical abrasion by waves can lead to different weathering rates of plastic waste in the sea and on beaches. Microplastics are fragmented by waves, high UV intensity, and long stays on the beach. On the contrary, microplastic particles in seawater take longer to fragment because the UV intensity is relatively lower than on the beach [60].

Aging and fragmented microplastics become smaller in size. The toxicity of the microplastics is size-dependent, and small microplastics are more toxic to marine zooplankton than large-sized microplastics [64]. In microplastics found in the coastal surface water of the subtropical island of Okinawa, Japan, it was demonstrated that small microplastics, less than 20 μm in size, have the highest potential to harm marine ecosystems [76]. Particles smaller than 20 μm may pass through cell membranes [158], and micro/nanoparticles smaller than 1.5 μm in diameter can directly damage cells [16]. In addition, scanning electron microscopy/energy dispersive spectroscopy analysis showed that microplastics with many weathered and exfoliated areas adsorbed heavy metals [29]. Microplastic particles found on the beach have a higher degree of oxidation and it is more difficult to trace their sources owing to their worn and deformed morphology [60]. These results indicate that the potential risks of aged and fragmented microplastics to the marine ecosystem cannot be ignored.

5.2. Microplastics in the aquatic food chain

Microplastics generated from plastic waste are easily moved and diffused by tides and currents and are unevenly distributed in the water column due to their inherent properties, such as hydrophobicity and density [45,74]. Microplastics exist in various sizes in the water column, of which, those 0.335–1 mm in size are similar to the size of zooplankton (*Acartia tonsa, Penilia avirostris, Temora turbinata, Paracalanus* spp.), the primary food for fish [59]. Microplastics colored similar to food, such as white, brown, and yellow, are easily mistaken for food and ingested by fish [40]. As predators tend to capture microplastics that are colored similar to their prey [159], colored particles are relatively more ingested by aquatic organisms than transparent plastics [160].

The microplastics ingested by fish are mainly in the form of fibers and are primarily transparent or blue [26,36]. Blue particles may be more attractive than other colors to marine organisms, and transparent particles are more likely to be consumed by fish as they are mistaken for zooplankton and tiny jellyfish. Furthermore, zooplankton which is a food source for fish, ingest microplastics. Most of the microplastics detected in Malaysia were fibers, with an average intake of 0.104–1 particles per individual [161]. Microplastics in the form of fragments were mainly found in bivalves such as oysters and mussels, and their size was smaller than 300 μm. The size, shape, and polymer-type of microplastics were similar to those found in nearby seawater. The microplastic intake of oysters/mussels was evaluated considering the microplastic concentration in seawater, seawater filtration rate of oysters, and high tide time. In Korea, the estimated annual microplastic intake was 587 n/person [28]. Microplastic contamination was also confirmed in gastropods inhabiting tropical estuaries in Malaysia. The characteristics of the detected microplastics were predominantly in the form of fibers with black color, and the main polymers were polyethylene-propylene-diene (PE-PDM) and PES. PE-PDM was presumed to be derived from the fragmentation of automobile parts [162].

In the Nansha Islands in the South China Sea, the concentration of microplastics on surface waters was 1733 items/m^3, and 3.1 items of microplastics per individual were found in fish samples [26]. Rayon, PES, and PP microplastics were commonly found in fish from the Maowei Sea, a typical aquaculture bay in China. They were also found in oyster tissue [36]. White microplastics were mainly detected in the gastrointestinal tract and gills of fish, and the fibrous form was dominant; similar findings were reported in oysters, whereby the fiber form accounted for 69% of microplastics [36].

In addition to being mistaken for food and ingested by marine organisms, the bioavailability of microplastics also increases owing to environmental factors. The halocline effect may limit the transport of microplastics, increasing the bioavailability of microplastics during the vertical migration of zooplankton such as copepods [55]. Microplastics exposed to the marine environment, regardless of their density, become denser than seawater and sink due to biofilm formation caused by marine animal excrement and organic matter [33,76]. Benthic species might ingest microplastics deposited as biofilms, resulting in higher levels of microplastics in benthic species than in pelagic species [33,36]. The distribution of microplastics and bioavailability of marine organisms are affected by habitats; therefore, it is necessary to analyze not only seawater samples but also sediment samples for microplastics. Organisms at the bottom of the food chain that feed on small fragments are migrators of microplastics [38], and bioaccumulation of pollutants released into the ocean, including microplastics, has led to the human consumption of microplastics accumulated in marine organisms.

The presence of microplastics in aquatic systems and the ingestion of microplastics by aquatic life are common. The plastics in Australia's *Paratya australiensis* (shrimp) were more diverse than those in freshwater. Shrimp in this area did not consume other types of microplastics and only consumed the fiber form. Perhaps because of the shrimps' small size and eating behavior, they tended to consume relatively small microplastics [18]. The discovery that shrimps ingest certain forms and colors of microplastics has the potential to be a promising biomarker of microplastic contamination. A study on the Rhine and Meuse Rivers in the Netherlands found that mussels and other benthic organisms accumulate microplastics in their bodies [79]; Demersal fish from these rivers had higher microplastics than did benthopelagic fish, and long-fiber microplastics were ingested at the bottom of the river, whereas large fragments of microplastics were ingested on surface water [49]. Although aquatic organisms may be suitable biomarkers of microplastics through their ingestion tendency, such as shrimp and some Demersal fish, microplastic absorption in clams from the Taihu Lake in China is negatively correlated with microplastics in sediments; therefore, careful attention is required in the selection of indicators [132].

5.3. Inherent toxicity of microplastics and adsorbed harmful substances

In 2011, a study evaluated the environmental and health risks of plastic polymers based on their chemical composition. Fifty-five plastic polymers were ranked and evaluated on a monomer basis. Polymers ranked as the most dangerous are made from monomers classified as mutagenic or carcinogenic, and they belong to the polymer families of polyurethanes (PURs), polyacrylonitrile, PVC, epoxy, and styrenic copolymers [163]. In the plastic manufacturing processes, synthetic polymers are mixed with additives to improve plastic products, which can be toxic and have adverse effects through ingestion, inhalation, or contact [164].

Microplastic risk assessment was conducted in surface water in the Changjiang estuary, China, using the hazard scores of plastic polymers of a previous study and the percentage of microplastic polymer types collected at each sampling station [74]. The study area contained PVC with high hazard scores and was assessed as having a high risk. PVC leaked into the marine environment could have a fatal effect on marine organisms by the release of carcinogenic monomers and intrinsic plasticizers [165], and the surface of PVC can readily adsorb contaminants such as persistent organic pollutants (POPs), causing complex toxic effects [166].

In the marine environment, microplastics are prone to adsorb contaminants such as POPs and heavy metals [64]. A small amount of Zn attached to microplastics was detected in a sample from the Nordic Sea, Greenland [29]. Plastics can become a carrier of hazardous chemicals to fish, causing hepatic stress and altering endocrine function [167,168]. Microplastics combined with pollutants can significantly affect coral reefs [169].

High concentrations of sigma7PAE were detected in zooplankton present in the Mediterranean surface waters, France [170]. POPs such as polychlorinated biphenyls (PCBs), PAHs, dichlorodiphenyltrichloroethanes (DDTs), OPEs, and PAEs were found in the northern part of the Yellow Sea, China [171,172]. Plastics can increase the bioavailability of bisphenol A and cause neurotoxicity in fish [173]. Investigation of microplastic pollution in the nearby seas of Italy revealed dominant black microplastics, which adsorbed higher concentrations of PCBs and PAHs than that absorbed by other colors [50].

The toxicity of plastics depends not only on their composition or the hazardous chemicals attached to them but also on their shape. A study on the bioaccumulation and toxicity of microplastics ingested by fish has shown that the accumulation of microplastics in the form of fibers is more toxic than in the form of fragments or microbeads [174]. Aged plastics in the form of pellets absorb high levels of organic chemicals [175].

5.4. Suggestions to limit the potential hazards of microplastics

Freshwater closest to land is the first to be affected by land-based waste, while wastewater is the result of anthropogenic activity, and seawater is affected by the marine industry and is the final destination of inland waters. Therefore, any aquatic medium is significant. A large amount of plastic waste is generated daily, and the waste is discharged into water environments without adequate waste treatment processes. The characteristics of the discharged plastic waste depend on the anthropogenic activities of the surrounding area and environmental factors.

The discharged plastic debris exponentially increases as it decomposes into smaller particles due to mechanical effects such as UV rays and weathering. Degraded plastic particles are disseminated due to their inherent properties, such as hydrophobicity and density, and environmental factors, such as ocean currents, weather, and salinity gradients.

Microplastics of several hundred micrometers similar to the size of zooplankton are mistaken for food and ingested through the mouth of aquatic organisms. In the case of oysters and mussels, some microplastics ingested through pseudofeces are discharged, but the microplastics that enter the marine organisms are usually accumulated and adversely affect their nervous system or digestive system. Microplastics in the food chain can pose a threat to human health.

Plastics released into the environment are not in a manufactured form but are usually corroded and fragmented owing to environmental factors. Aged microplastics absorb high concentrations of organic chemicals. Therefore, toxicity evaluation should be performed using aged microplastics rather than manufactured plastics. Additional toxicity evaluation for harmful substances such as heavy metals and POPs, which are easily adsorbed by aged microplastics, is essential.

One of the most significant limitations of microplastic research is that there is a lack of studies on plastic waste in sediments. According to a UNEP report, the sum of plastic waste found on beaches and floating on water is ~ 30%, whereas most of the marine waste, 70%, settles in sediment [176]. Marine waste that sinks into the sediment is challenging to recover and poses a threat to marine ecosystems as it is potentially toxic. In particular, benthic species can readily ingest plastic waste found on the seabed. When a change in ocean current or a weather event occurs, there is vertical mixing of the seawater layer, and the plastic waste gets freed and moves around.

As the water systems are the final destination for microplastics, it is necessary to regulate various industries and all types of plastic products manufactured and discharged on land to prevent the increase in plastic waste in the water systems. There is a need for efficient waste management and improvement of drainage systems on land. This cannot be solved by regulating one or two countries, but a global coalition and regulation will be an excellent start to solving the problem. In addition, microplastic removal technology should be developed for each water environment.

6. Discussion on microplastic research categorized by water environments

6.1. Comparison of microplastic quantification protocols in water environments

Based on the studies reviewed in this paper, the characteristics of each water environment are summarized and shown in Table 4.

In general, Trawl nets are designed to scrape the bottom in large areas and collect widely distributed fish species [177]. The sampling method is also used to collect irregularly and widely distributed microplastics. Trawl-net used in seawater has a relatively large mesh size (200–333 μm). Many studies conducted so far have mainly used the trawl-net method. Grab sampling or pump sampling was used for wastewater due to the relatively poor water quality and process structure. In freshwater, trawl-net and bulk/pump sampling methods were used respectively or simultaneously depending on the water system's size or the study's purpose. However, the mesh size of nets used to collect microplastics in water systems is much larger than that of microplastics, which can harm living organisms. Studies considering micro/nanoparticles in size range smaller than 20 μm, which are known to pass through cell membranes, and smaller than 1.5 μm, which can directly damage cells, must be conducted.

The pretreatment method for microplastic analysis is divided into two types. One is via digestion to remove organic matter, and the other is a density separation method to extract microplastics from contaminants. As the digestion step has already been studied in seawater, the same or improved method was applied in wastewater and freshwater. The amount of organic matter determines the strength of the reagent used for digestion. As a density separation solution, NaCl, which is easy and low in toxicity, is mainly used for the separation of low-density microplastics collected from surface water. When the ocean currents change or during extreme weather conditions, the seawater layers are vertically mixed, and the deposited high-density microplastics are resuspended and detected on the surface of the seawater. Hence, to not omit high-density microplastics, high-density separation solutions such as $ZnCl_2$, NaI, and LMT are beginning to be used. It is desirable to apply various pretreatment reagents to extract microplastics from materials that interfere

Table 4
Summary of reference-based microplastic quantification protocols in water environments.

		Seawater	Wastewater	Freshwater
Main sampling method		Trawl-net (Nylon, plankton, neuston, manta net)	Bulk/Pump (Stainless-steel bucket, automated samplers, electric pump)	Both trawl-net and bulk/pump (Plankton nets, metal bucket, steel sampler)
Pretreatment	**Digestion**	H_2O_2, Fenton, NaOH, KOH, HNO_3, HCl	H_2O_2, Fenton, NaClO, NaOH, KOH, cellulase enzymes	H_2O_2, Fenton, KOH: NaClO, NaOH, KOH enzyme treatment
	Density separation	NaCl, $ZnCl_2$, NaI, LMT	NaCl, ZnCl2, NaI, CaCl2,	NaCl, $ZnCl_2$, NaI, $CaCl_2$, LMT
Analysis		spectromicroscope, FT-IR, Raman	spectromicroscope, FT-IR, Raman, py-GC/MS	spectromicroscope, FT-IR, Raman, py-GC/MS
Influence factors		Marine industries and port (mariculture, fishing), geological characteristics (semi-enclosed seas)	Anthropogenic activities, population, domestic sewage, urban-related watershed attributes	Manufacturing industries, agriculture, highways and roads, hydrological process, WWTPs
Microplastics	**Shape**	Fragments, lines, fibers, granules	Fibers, fragment, films, beads	Fibers, fragments, films, beads, pellets
	Composition	PE, PP, PS	PES, PA, alkyds, cotton, Anthropogenic (Dye-Based), PP, PE, PET, PS	PES, nylon, Semi-synthetic cellulose PP, PE, PS, PET
Medium features		Where secondary microplastics can be mostly produced due to strong UV rays and waves.	The result of anthropogenic activities, where it is treated and discharged.	A water source necessary for human activities.
		Final destination where all water comes together.	Transfer between media using sewage sludge (from water to soil)	Places that are primarily affected by land-based waste.

with the analysis. However, depending on the characteristics of the pretreatment reagent, microplastics may be damaged or underestimated. Therefore, a pretreatment reagent that does not damage microplastics while removing substances that interfere with analysis should be selected, and an optimal protocol should be developed. In addition, the developed protocol should be applied to all microplastic detection studies aimed at the quantitative evaluation of microplastics.

The marine industries and geographic structure highly influence the characteristics and abundance of microplastics in seawater. The detected plastics are mainly PE, PP, and PS, the most used polymers in the marine industries and daily life. Fragments were the most common type of microplastics to be found because of the intense UV rays and waves affecting the microplastics, generating secondary microplastics. As the sea is the final destination for all water flow, microplastics in various shapes and compositions have been discovered.

Microplastics in wastewater directly reflect the anthropogenic activity. Correlations between microplastics in wastewater, urban-related watershed attributes, and population were shown. As the proportion of wastewater generated from washing is high, microplastics in the form of fibers and PES and PA polymers that make up clothes were detected in domestic wastewater. In addition, when sewage sludge is applied to agriculture, microplastics contained in the sludge are transferred from sewage to the soil, promoting the movement of microplastics between environmental media.

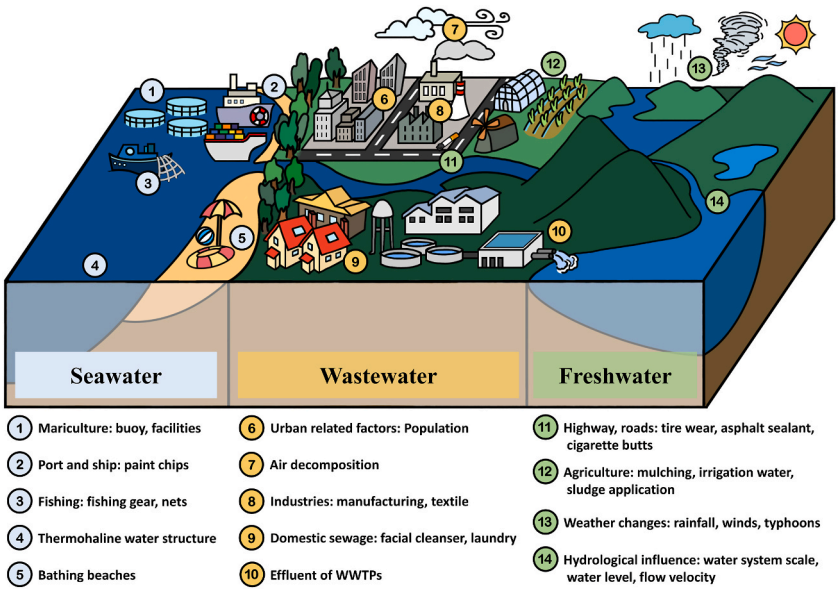

Fig. 4. Results of microplastic detection in water systems affected by the surrounding industries and environment.

Microplastics in freshwater have been affected by agriculture and manufacturing industries such as textiles, including worn tire material on highways and roads. Further, they were affected by hydrological processes and wastewater by anthropogenic activities, and were sensitive to the surrounding environment; a large amount of fiber-type PES and nylon was found in freshwater adjacent to cities. In addition, PE, PP, and PS, the commonly used polymers, were detected.

The results of microplastic detection in water systems affected by the surrounding industries and environment are summarized and shown in Fig. 4. Microplastics at the sampling point are mostly affected by adjacent anthropogenic activities. However, microplastics are detected even far from the sampling point in the circulating water environment. In order to understand the behavior of microplastics, it is also essential to consider the factors that can influence their properties.

6.2. *Factors affecting microplastic analysis*

Factors affecting the abundance and distribution of microplastics in water environments can be divided into two types. These are microplastics' inherent properties such as hydrophobicity, specific gravity, and size, and environmental factors such as biological interactions in the aquatic environment, meteorological phenomena, and industrial facilities near the water system (Fig. 5).

Plastic waste exposed to intense UV rays, waves, and wind in the environment goes through the aging process and decomposes into smaller sizes, spreading all over the earth. Microplastics down to the micro level can be transported through the atmosphere. Microplastics were also found in the Alps and Antarctica as the wind easily transports them [178]. Low-density microplastics can be deposited due to interactions with aggregates, biofilms, and excrement. Additionally, the deposited microplastics are resuspended under the influence of weather conditions and are redistributed in the aquatic environment. Occasionally, plastic waste enters the water system owing to the lack of proper waste management at industrial facilities near the sampling area.

Although the correlation between microplastics in the water environments and their sources has seemed to be revealed, there have been contradicting results owing to variable weather changes. Notably, the vertical distribution, influenced by the specific gravity and biological interaction of microplastics, and the horizontal distribution, caused by environmental factors, have increased microplastic variability through weather changes. Weather is a factor that must be considered in studying microplastics in water environments. It is difficult to measure the distribution of microplastics in a study area with one-time sampling. Long-term monitoring is required for accurate microplastic analysis.

Aside from these environmental variables, mesh size remains the biggest challenge because it differs across studies. In addition, there are limitations in the interpretation of the results because the sampling and pre-processing methods are different. As a top priority, sampling and pretreatment methods for microplastic analysis should be established. However, because the digestion reagent and density separation solution can affect specific polymers in different ways, it is recommended that a pretreatment protocol for each polymer analyzed and an optimal protocol be developed depending on the polymer.

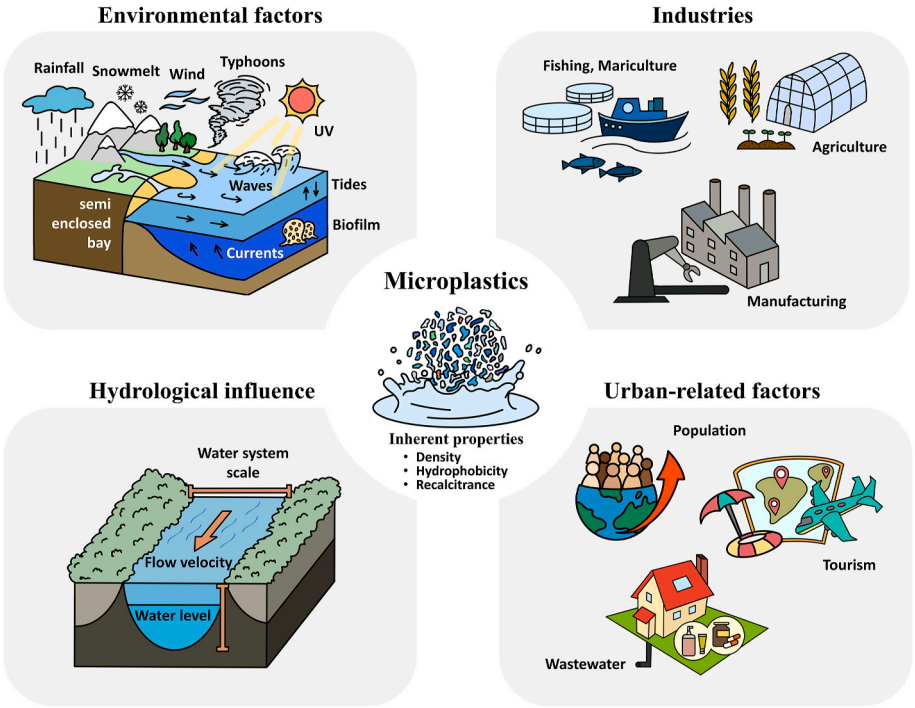

Fig. 5. Factors affecting microplastic analysis.

6.3. Threats of microplastics and the appropriate countermeasures

The uncontrollable impacts of microplastics on the environment is a major reason for the emerging importance of research in this field. Although plastics are constantly being manufactured and disposed, proper waste management is not being carried out. The plastic waste released into the environment is broken down into small pieces and passed through the food chain by small creatures at the bottom of the food chain, posing a potential threat to human health. Microplastics decomposed into micro-levels have been found to enter the human lungs through respiration as well as ingestion [179]; therefore, the threat of microplastics to humans cannot be underestimated.

The size of microplastics that can directly damage cells by passing through cell membranes is known to be less than 1.5 μm. Since plastic particles at a few micro to the nanoscale cannot be analyzed by spectroscopic analysis, the analytical method using pyrolysis must be applied. Another factor as crucial as the analysis method is controlling the pretreatment environment since plastic particles introduced due to airborne contamination must be excluded from the analysis results. As the plastic particles to be analyzed approach the nanoscale, the priority of research on pretreatment environmental control to prevent contaminants should increase. In addition, in-situ analysis is suggested to prevent any contamination that may occur during sample transfer.

Although many studies have been conducted on the toxicity of microplastics, the small size of microplastics is challenging to control, making it an obstacle to conducting toxicity assessments. In addition to the adverse effects caused by the inherent physical/chemical properties of microplastics, toxic substances present in the environment that easily attach to microplastics pose an even greater threat. Most studies conducted so far have used plastic materials to control the experimental factors. However, the chemical properties of aged microplastics may change due to various environmental factors [180]. Studies on plastics whose chemical properties have changed due to aging in the environment will need to be further investigated.

Plastic wastes that have already been disposed into the environment continue to decompose and threaten life on earth. To prevent further aggravation of the situation, comprehensive waste management must be devised to stop the reckless discharge of waste into the environment. For accurate studies on microplastics, sample collection and pretreatment methods should be first established; then, a nano-level microplastic identification method should be developed. Toxicity assessment of microplastics and attached contaminants and all environmental factors that can affect microplastics should be studied. Finally, it is necessary to investigate the behavior of microplastics, control the path they take to enter the human body, and minimize the concentration of microplastics that can enter the human body. If plastic waste cannot be controlled realistically, it is suggested that studies be conducted on the allowable amount of microplastics in the human body without impacting human health.

7. Conclusions

This review summarized the abundance and distribution of microplastics in the current water environments based on recently published and highly rated papers. Furthermore, the environmental factors affecting microplastics and the toxic effects of microplastics were intensively discussed. Although many studies have been conducted on microplastics in water systems, there are few studies on microplastics smaller than 1.5 μm in size. This is partly because of the limitations of the analysis and because the microplastics introduced during sampling and pre-processing cannot be sufficiently controlled. In addition, standard sample collection and pretreatment methods for microplastic analysis have not been established, making it difficult to compare study results. However, the results of previous studies have specified the environmental factors that can affect microplastics and helped suggest future research directions. It is necessary to derive systematic research results through standardization of microplastic analysis methods and develop micro-and nano-level plastic analysis technology. There is still little research on the toxicity of microplastics in the environment. Studies on microplastics whose properties have changed due to aging and contaminants attached to these microplastics should be conducted. As microplastics are easily changed by the influence of the surrounding environment, monitoring rather than one-time sampling is required, and attention should be paid to the advancement of plastic waste management methods and treatment methods.

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Abbreviations

A2O, anaerobic-anoxic-oxic; ATR-FTIR, attenuated total reflectance-Fourier transform infrared spectroscopy; CA, cellulose acetate; CAS, conventional activated sludge; COD, chemical oxygen demand; DDT, dichlorodiphenyltrichloroethane; DO, dissolved oxygen; ECHA, European Chemicals Agency; EDS, energy dispersive spectroscopy; FE-SEM, field emission scanning electron microscopy; FPS, foamed polystyrene; HDPE, high-density polyethylene; ISO, International Organization for Standardization; LDPE, low-density polyethylene; MBR, membrane bioreactor; NIR, near-infrared hyperspectral imaging; NOAA, National Oceanic and Atmospheric Administration; PA, polyamide; PAHs, polycyclic aromatic hydrocarbons; PC, polycarbonate; PCB, polychlorinated biphenyls; PE, polyethylene; PE-PDM, polyethylene-propylene-diene; PES, polyester; PET, polyethylene terephthalate; POP, persistent organic pollutant; PP, polypropylene; PS, polystyrene; PUR, polyurethane; PVC, polyvinyl chloride; Py-GC-MS, pyrolysis–gas chromatography-mass spectrometry; SEM, scanning electron microscopy; SS, suspended solids; TED-GC-MS, thermal extraction desorption-gas

chromatography-mass spectrometry; UNEP, United Nations Environment Program; WPO, wet peroxide oxidation; WWTP, wastewater treatment plant; μ-FTIR, micro-Fourier transform infrared spectroscopy; μ-Raman, micro-Raman spectroscopy.

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