

Review

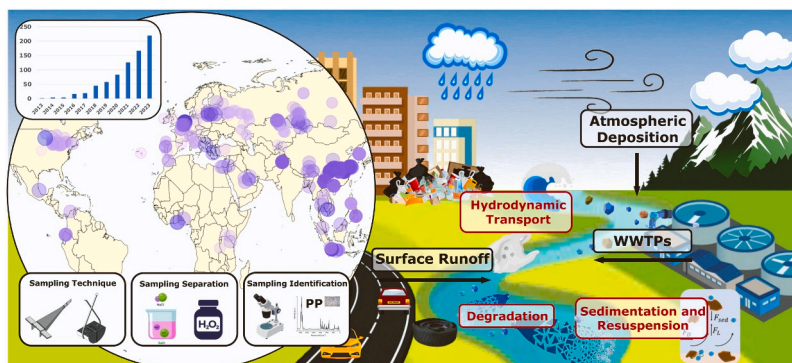
Microplastics monitoring in freshwater systems: A review of global efforts, knowledge gaps, and research priorities

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HIGHLIGHTS

- Microplastics (MPs) are widely distributed in global freshwater systems.
- Higher MP abundances are found in areas with intense human activities, such as United States, Europe, and China.
- The selection of the sampling methods and the size range will significantly affect the reported MP abundance.
- There is a pressing need for a standard analysis protocol for MP pollution in freshwater.
- Future studies should employ artificial intelligence for fast, accurate, and large-scale characterization of MPs.

GRAPHICAL ABSTRACT



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ABSTRACT

The escalating production of synthetic plastics and inadequate waste management have led to pervasive microplastic (MP) contamination in aquatic ecosystems. MPs, typically defined as particles smaller than 5 mm, have become an emerging pollutant in freshwater environments. While significant concern about MPs has risen since 2014, research has predominantly concentrated on marine settings, there is an urgent need for a more in-depth critical review to systematically summarize the current global efforts, knowledge gaps, and research priorities for MP monitoring in freshwater systems. This review evaluates the current understanding of MP monitoring in freshwater environments by examining the distribution, characteristics, and sources of MPs, alongside the progression of analytical methods with quantitative evidence. Our findings suggest that MPs are widely distributed in global freshwater systems, with higher abundances found in areas with intense human economic activities, such as the United States, Europe, and China. MP abundance distributions vary across different water bodies (e.g., rivers, lakes, estuaries, and wetlands), with sampling methods and size range selections significantly influencing reported MP abundances. Despite great global efforts, there is still a lack of harmonized analyzing framework and understanding of MP pollution in specific regions and facilities. Future research should prioritize the development of standardized analysis protocols and open-source MP datasets to facilitate data comparison. Additionally, exploring the potential of state-of-the-art artificial intelligence for rapid,

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0304-3894/© 2024 Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies.

accurate, and large-scale modeling and characterization of MPs is crucial to inform effective strategies for managing MP pollution in freshwater ecosystems.

1. Introduction

Plastics are the most common product in modern society. Since their initial commercial development in the 1950s, plastics production has increased dramatically with over 390.7 million tons produced globally in 2022[1]. Due to their widespread production and usage, plastic wastes inevitably enter the aquatic environment through various pathways and it was reported that around 9–14 million tonnes of plastic waste would enter aquatic ecosystems every year[2]. Among them, those very small plastics with a diameter of less than 5 mm in length are referred to as ‘microplastics’ (MPs) and have become a major concern in aquatic environments[3,4]. Many studies have reported the ubiquitous presence of MPs in aquatic ecosystems and their potential risks to the environment and human health through the release of harmful additives contained within MPs or bioaccumulation via the food chain[5,6]. Furthermore, the physical and chemical properties of MPs have been found to facilitate contaminant sorption to their surfaces which also makes MPs a vector of contaminants to organisms following ingestion [7].

Based on these facts, research on MPs as an emerging environmental contaminant is expanding rapidly due to increasing global concern. Previously, the investigation of MP pollution has mainly focused on the marine systems[8–11] and has been studied extensively since approximately 2005[12]. However, understanding of the occurrence and impacts of MP pollution in freshwater systems remains relatively underdeveloped and it was reported that only about 3.7 % of MP-related studies are associated with freshwaters[13,14]. Much of the existing studies only consider freshwater systems as sources and transport pathways of MPs to the marine systems[15] while ignoring their close relationships to humans and ecosystems (e.g., such as the ecological function as crucial drinking water sources). Recent findings suggest that streams and rivers have the potential to serve as MP sinks [16] and the prevalence and impact of MP pollution in freshwater systems might be as significant, if not greater, than in marine systems[17–20], which deserves more public attention.

Based on the above consensus, the studies of MPs in the freshwater system have grown dramatically since 2014 (Fig. 2(a)). Recent reviews of MP pollution in freshwater systems have focused on methodology for analyzing and detecting MPs[21,22], the occurrence and impact analysis of MPs[23–26], toxicity assessment[15,27,28], or specific regions [29–36]. Through their findings, the global presence of MPs in freshwater systems has been verified, demonstrated by their appearances from tropical rivers to remote mountain lakes with highly varying abundance in surface water and sediments[23]. The ecological impacts and chemical hazards of MPs in freshwater systems are discussed[15] and the current techniques used for MP sampling and detection are listed and compared[21,22]. However, most of these reviews only simply listed the information from the cited studies without carefully integrating and summarizing the results and failed to provide the potential explanations behind the data and additional conclusions. There is an urgent need to provide a more in-depth critical review with quantitative evidence of the latest global efforts on understanding MP pollution in freshwater systems, systematically summarize the standard workflow of analyzing MP pollution, explore the cause and sources of MP pollution, and discuss the needed actions and future opportunities for MP pollution in freshwater systems.

Therefore, the aim of this review is to systematically assess the current situation of global MP monitoring in freshwater environments, with a specific focus on surface water and partial studies of sediments. Reviewed publications were retrieved from Google Scholar for the last decade (from 2013). Preliminary search keywords included

“freshwater”, “lake”, “lakes”, “river”, “rivers”, “reservoir”, “reservoirs”, “wetland”, “wetlands”, “estuary”, “estuaries” * AND “microplastic” which generates a list of 755 publications (Fig. 2(a)). After the initial search, a screen was conducted by all the co-authors to filter publications beyond the scope of this review (e.g., studies focused on nano-plastics, biota, impact assessment, groundwater, drinking water treatment plants, commercial ponds) with a shorter list of 649 publications. Quality control was then conducted to remove publications without strict peer review (e.g., all preprint publications and part of the conference and journal papers), producing a shortlist of 557 studies. Among them, 328 of these studies were then selected based on their direct relevance to the scope of this review, focusing on MP monitoring in freshwater systems. Three key aspects are identified and summarized in this paper which are: methods used in MP analysis in freshwater systems, occurrence and characteristics of MP pollution, and factors affecting the spatial-temporal distribution of MPs. Finally, we reviewed the best practices and standard workflow for analyzing MP pollution and discussed the current knowledge gaps and future potential in this area.

2. Methods used in microplastic sampling, separation, and detection in freshwater systems

2.1. Sampling methods

The sampling methods for collecting MPs in freshwater are quite similar to those that are used in marine environments, which can be mainly summarized in two types, volume-reduced sampling and bulk sampling[37] (Table 1). In the volume-reduced sampling, the volume of the sample is reduced during the sampling period. While bulk sampling refers to sampling where the whole volume of the sample is taken without reducing it during the sampling process.



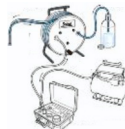

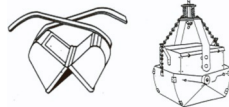

For water compartments, neuston plankton net and manta trawl are two commonly adopted volume-reduced sampling approaches to collect surface-water MPs in open water areas [38–52]. These trawls are designed with an opening to funnel the water into the net, a long net with a fine mesh opening to filter the water, and a cod end to retain the collected materials. During sampling, the trawl is usually attached to a boat through a rope system, submerged, and towed at a low speed over a predetermined distance or time to collect surface water samples from 0–50 cm (mainly based on the heights/diameter of the trawl and the submerged level)[44,46,53,54]. A mesh size in the range of 60 to 500 μm (300–335 μm is the most commonly used)[41,53,55] is typically used to concentrate surface water samples due to the lower size boundary of MPs of 333 μm suggested by the National Oceanic and Atmospheric Administration of USA[4]. The volume of water through the trawl (usually $>100\text{ m}^3$) is either measured using a flow meter[44, 56] or is calculated by multiplying the towing distance by the width and height of the trawl[54]. For bulk sampling, the most frequently used methods are grab sampling and pump sampling, which raises increasing attention in recent sampling campaigns after 2018 (53.5 %). Grab sampling method usually uses a stainless-steel bucket or glass bottle with fixed sizes to collect surface water samples from 0–50 cm[57–62]. Typical sample volume (single sample) is usually in the range of 1 to 30 L[63–67]. After initial collection, the samples were sieved using designed mesh sizes to concentrate the samples and keep those potential MP particles. Pump sampling refers to pumping water from 0–100 cm depth manually or using a motor through an inline sieve or filter [68–73]. Compared to grab sampling, pump sampling allows larger volumes of water samples to be collected (usually in the range of 20 to 50 L for a single sample, but can be up to 2000 L) and a deeper sampling depth which may be more suitable in areas with low MPs density. For

both of these two bulk sampling methods, taking multiple replicates (usually three or more) is highly suggested[57,74–80] considering the limited sampling area covered and the high variability of MP spatial distribution. Upon collection of the samples from the environment, the collected particle samples are usually preserved with 5 % methyl aldehyde[54,75,78] and stored at 4 °C before analysis[54,76] or fixed in 2.5–5 % formalin[76,81] or submerged in ~ 40 % ethanol (EtOH)[82].

For sediment compartments, bulk sampling is so far the dominant method and is selected based on the sampling location. For collecting samples in littoral zones and on beaches, manual grab methods utilize tools such as stainless-steel hand scoops/spades/trowels/shovels are quite common[83–88]. Sampling was usually conducted using a sampling quadrat (10 × 10 to 50 × 50 cm) and sediment samples were collected from the top 2–10 cm [40,83,85,89–91], which are consistent with those protocols used for marine environments[92,93]. However, the sample weight (0.025–10 kg)[94–96] or volume (1–12 L)[97–102] largely varies between studies, potentially affecting cross-comparison and representativeness. Once collected, the sample is usually sieved in the field using a stainless steel 5 mm mesh size to remove large debris [103]. While for deep sediment sampling of the beds for the water body, the sampling campaigns usually require a vessel and the use of

specialized equipment that is lowered to the bottom to collect the samples (e.g., grab sampler or corer)[104–107]. Among them, Ponar, Ekman, and Van Veen grab samplers (which can also be used for surface sediment sampling but are less commonly employed for this purpose) are frequently deployed to collect benthic sediment from 0–10 cm[76, 108–114]. The sample volume is mainly based on the designed size of the grab sampler which typically ranges from 3–8 L[113,115]. Besides grab sampler, core sampling is another crucial method for investigating the vertical distribution and historical accumulation of MPs in deep sediments[115–120]. This technique involves extracting cylindrical sections of sediment, typically using gravity corers or piston corers[118, 121], which preserve the stratigraphic integrity of the sample, allowing researchers to analyze MP concentrations at different depths (from 5–10 cm to up to over 2 m)[116,120,121] and potentially date the sediment layers to understand the temporal trends of MP pollution[120, 121]. The core samples were then sliced into 1–5 cm thick layers for further analysis [118,120,121]. After that, the samples are transferred using non-plastic containers (such as glass trays or steel-less buckets) or covered with aluminum foil[110,122–124]. While, in the case of plastic container utilization, blank control should be included to prevent potential contamination of the samples[125]. Need to mention, an

Table 1
Sampling methods of MPs in water and sediment of freshwater systems.

Compartment	Type	Technique	Sampling volume	Lower detection boundary	Sampling depth	Potential Issues
Surface water	Volume-reduced sampling	Manta trawl and neuston plankton net  [126,127]	> 100 m ³	60 to 500 μm	0 –50 cm	Underestimation of MPs in smaller size; Limited by the sampling environment; Disturbance of volume measuring by turbulence generated by the movement of the ship.
	Bulk sampling	Grab sampling 	1 to 30 L	0.45 to 100 μm	0 –50 cm	High variability due to relevant small sample size; Lack of representativeness.
		Pump sampling  [128]	20 to 2000 L	20 to 100 μm	0 –1 m	Potential contamination by the apparatus; Limited application scenarios; Lack of representativeness.
Sediment	Bulk sampling	Manual grab samplers (for surface sediment in the bank/shore of the water body) 	0.5 to 5 kg	0.45 to 500 μm	2 –5 cm	High variability due to sampling area and depth selection; High subjectiveness for the selection of sampling area.
		Grab samplers (for benthic sediment in the beds)  [129]	1 to 4 kg or 0.5 to 1 L	0.45 to 500 μm	0 –10 cm	High variability due to fluctuation of sampling depth; May cause disturbance in the sampling area.
		Corer (for vertical sampling of benthic sediment)  [130]	/	0.45 to 500 μm	From 5 –10 cm up to 2 m	Limited application scenarios.

accurate estimation of MP concentration in sediment samples requires a clear definition of sampling depth and number of replicates to guarantee the representativeness of the results[104].

2.2. Sample separation

Upon collection of the samples from the environment, the MPs contained within that sample need to be separated from all other organic and inorganic materials to ensure that the MPs can be quantified or positively identified (Fig. 1)[131]. These processes are similar for the surface water and sediment samples, while the later ones require more effort due to the much greater solid contents retained by the initial sieving. Therefore, the dry sediment samples usually require an additional step to be stirred with certain dispersant solutions (e.g., potassium metaphosphate solution or sodium hexametaphosphate solution) to disaggregate sediment before the sample separation[59,132].

For the initial separation after volume reduction, density separation is the most often used approach (~65 % of studies implementing this process), which involves the mixing of the sample with salt-saturated solutions with known density (Fig. 1(a))[133]. The saturated sodium chloride (NaCl) solution with a density of 1.2 kg/L is the most commonly (~68 % of the cited studies with density separation) used to achieve this separating process due to its low cost and no toxicity to humans[61,78,134–140]. While, the relatively low density of saturated NaCl solution could cause the loss of heavier polymers (e.g., polyethylene terephthalate (PET) and polyvinylchloride (PVC)) and occasionally result in low recovery rates (<90 %) and larger error bars[141]. As a result, other saturated salt solutions such as sodium iodide (NaI, 1.6 kg/L)[141–144], zinc chloride (ZnCl_2 , 1.6–1.8 kg/L)[58,72,74,81,98,102,112,145–148], calcium chloride (CaCl_2 , 1.4 kg/L)[149], potassium formate (HCO_2K)[150,151], and sodium tungstate dihydrate ($\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$, 41 % w/v; 1.4–1.6 kg/L)[152–154] are emerging as new trends due to their good recovery rates during lab experiments (99 %) and tight error bars. After mixing, the suspension is stirred for a specified time before being left to settle. This step allows low-density particles, including MPs, to rise to the upper layer, while high-density materials descend to the bottom layer. Through this process, interfering inorganic materials (such as inorganic clay) could be largely removed and MPs could be recovered from the supernatant. Besides traditional density separation using salt-saturated solutions, elutriation and oil-based separation are some other efficient alternatives to separate MPs in freshwater systems. Elutriation uses a stream of fluid flowing in the opposite direction to the centrifugal force to separate MPs from the settling organic matter and sediment based on their size, shape, and density (Fig. 1(b)) [155–157]. These methods are suitable for sediment

samples and could also be applied as an effective way to reduce the volume of brine solution required for density separation[158]. However, this method is not suitable for samples with high concentrations of organic matter and sometimes still needs to be applied with salt solutions[159]. Meanwhile, elutriation needs precise control of flow velocity which was investigated through numerical modeling based on hydrodynamic equations which can be technically challenging and restricts its applications[160]. Oil-based separation technique exploits the oleophilic property of MPs to separate them from their surrounding environmental matrix to the upper oil layer and segregate the non-MP suspended solids in a separation funnel (Fig. 1(c))[82]. However, oil-based separation has not been widely applied so far due to its unstable efficiency and accuracy [161] and the reported recovery rates could be less than 75 % [162]. Especially, the lipophilic properties of MPs may be altered due to the adherence of other contaminants or organic matters which may lead to the loss of MPs in the oil-water interface. Meanwhile, the residual oil might further interfere with the following identification of MPs by Fourier transform infrared (FT-IR) spectroscopy and thus need additional cleaning steps[163]. Overall, the approach still needs further development before large-scale applications.

After that, the samples collected need to experience a digestion process to remove organic material such as algae from the samples. This process can be divided into two main categories of chemical degradation and enzymatic degradation[23]. For chemical degradation, the hydrogen peroxide (H_2O_2 , 30 %) solution (usually in the presence of an iron(II) catalyst, i.e., Fenton's reagent) is the most popular oxidizing agent (~82 % of the cited studies which involves the digestion process) considering its efficiency in digesting organic matter (24 h) and negligible damage to the MPs[39,41,46,53,57,73,75,118,136,164–166]. Acid (e.g., HCl and H_2SO_4) and Alkali (e.g., KOH) digestions are also used in many studies, while the high risk of destroying or discoloring MPs raises concerns for these techniques[40,42,72,80,97,155,164,167–169]. There are also studies trying to use sodium hypochlorite (NaOCl, also written as NaClO) along with the above chemicals for the further digestion process of organic-rich freshwater samples [69,148,167,170]. In addition to chemical degradation, enzymatic degradation is gaining attention as an emerging environment-friendly approach method that is less likely to induce damage to MPs[171–173]. During the degradation process, MP samples are incubated with a blend of industrial enzymes such as protease, cellulase, and chitinase to remove different types of organic matter[174,175]. However, the application of enzymatic degradation is still limited on small scales due to its high price, relatively long time cost, and the fluctuation of enzyme efficiency due to different organic compositions in the samples[176]. Sometimes,

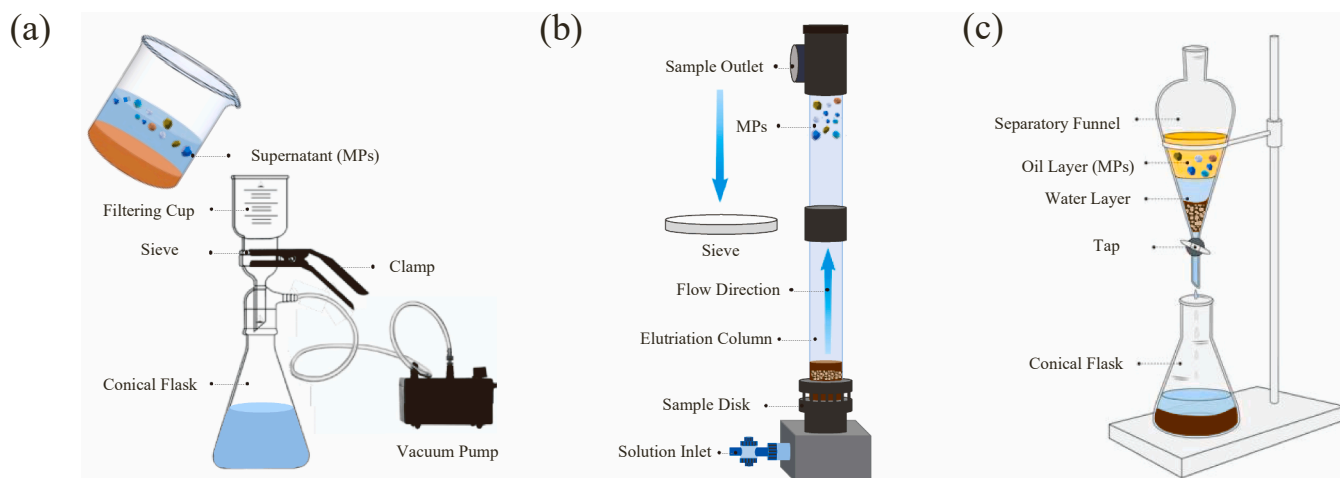


Fig. 1. Typical sample separation techniques for MPs in freshwater systems. (a) Density separation using salt-saturated solutions. (b) Elutriation separation techniques. (c) Oil-based separation.

enzymatic degradation still needs a following treatment with H_2O_2 to remove undigested debris[104].

2.3. Sample identification and quantification

After the MP-containing sample is treated, identification and quantification techniques are applied to determine the abundance and composition of the MP in the sample (Table 2).

The microscopic technique is the regular process used for the visual inspection of MPs for the monitoring studies, which simply means visually selecting and classifying the suspected MPs through a microscope (from normal optical and ultraviolet microscopes to more advanced electron microscopes such as scanning electron microscope (SEM)) based on physical characteristics[123,164]. The distinct advantage of microscopic inspection is that it can determine the shape, color, and size of the MPs at the same time. However, this method is criticized as highly subjective as different observers may record different proportions of MPs (like 67.3–81.3 % reported in the study of Lavers et al.)[177], leading to either overestimation (e.g., misclassification of

organic matters and dusts)[175] or underestimation (e.g. missing transparent particles) of certain types and colors of MPs. And an error rate of up to 70 % is reported when compared with baseline spectroscopy in relevant studies[178]. To improve the results provided by visual inspection, staining methods were applied to provide researchers with a simple and time-effective tool to facilitate the identification and quantification process. Currently, Nile Red is the most effective dye material based on its low cost, lower detection limit, high recovery rates (96.6 %), and less interaction with biogenic material[40,179–181]. After staining, the Nile-Red-stained MPs are visible under a fluorescent microscope, which can easily be counted[170]. Currently, the main drawback of Nile Red is that it can still stain some natural organic matter [23]. Hence, pre-purification is required to guarantee the accuracy of counting.

To further determine the chemical composition of the detected MPs, FT-IR (52 % have been applied in the cited studies, one study can apply more than one technique) and Raman spectroscopy (21 % of the cited studies) are prevalent spectroscopic methods for identifying MPs[91, 150,173,182–188]. These methods utilize radiation at specific

Table 2
Summary of commonly used identification and quantification techniques for MPs.

Type	Technique	Details	Potential Issues
Visual Method	Microscope	Visually selecting and classifying the suspected MPs through a microscope based on physical characteristics.	<ul style="list-style-type: none"> • Time consuming; • Highly subjective and may result in either overestimation or underestimation of the abundance for certain types of MPs; • Extremely inaccurate in determining the polymer types.
	Staining method (with microscope) SEM/EDS	Use of specific stains and dyes to enhance contrast of MPs with other particles under a microscope. Utilize a focused electron beam to scan the sample surface, generating high-resolution images of topography and morphology, while simultaneously producing characteristic X-rays that are analyzed to determine the elemental composition of the sample.	<ul style="list-style-type: none"> • Potential confounding effect of staining other organic material in the MP samples. • Struggling with detecting light elements (H, He, Li, Be), which are important components of most polymers; • The technique provides bulk elemental composition, which may not distinguish between similar polymer types; • Contamination or surface coatings can interfere with accurate analysis.
Spectroscopic Method	FT-IR spectroscopy	Infrared radiation (IR) is passed through the MP sample. Specific frequencies of IR radiation are absorbed and transmitted, causing peaks in an IR spectrum to identify and quantify the molecular composition and structure of a sample.	<ul style="list-style-type: none"> • High fixed costs for the instruments; • Time-consuming for whole particle identification; • Limited resolution, samples below 20 μm may not yield enough interpretable spectra; • Certain requirements for the thickness, regularity, and transparency of the samples under different operating modes.
	Raman spectroscopy	Irradiated laser light interacts with the molecules and atoms of the MP sample. The molecules will reflect back-scattered light in a different wavelength and generate a molecular fingerprint spectrum to characterize the chemical components of the sample.	<ul style="list-style-type: none"> • High fixed costs for the instruments; • Time-consuming for whole particle identification; • Fluorescence interference with biological residual and other chemicals leads to difficulties in the identification of polymer types; • Limited spectra database.
Chromato-graphic Method	Pyrolysis GC/MS	MP sample is heated to decomposition to produce smaller molecules that are separated by gas chromatography and detected using mass spectrometry.	<ul style="list-style-type: none"> • Destruction of the samples; • Limited identification of polymer types; • Provides no information regarding number, size, or shape; • Less applicable for mixtures with high impurities; • Requirement of well-trained and experienced operators.
	TED-GC/MS	Employ thermal desorption to volatilize compounds from a sample, which are then separated by gas chromatography and identified using mass spectrometry.	<ul style="list-style-type: none"> • Destruction of the samples; • Provides no information regarding number, size, or shape; • Time-consuming and high cost • Requirement of well-trained and experienced operators.
	Liquid chromatography	MP sample is dissolved in a selected solvent and size exclusion chromatography for polymer characterization.	<ul style="list-style-type: none"> • Destruction of the samples; • Provides no information regarding number, size, or shape; • Limited resolution and high fixed costs for the instruments; • Sample preparation requires careful handling, which can be time-consuming and may introduce additional errors.

wavelengths to excite samples, producing vibrations unique to the structure of the material. The produced characteristic spectra were compared with the known plastic polymers in the spectral library to allow the identification of the material[189]. The FT-IR and Raman spectroscopy are accurate, non-invasive, and complementary techniques. Currently, various FT-IR techniques have been employed for MP characterization. For large MPs with sizes $> 100 \mu\text{m}$, attenuated total reflection FT-IR (ATR-FTIR) is effective, offering enhanced identification capabilities for MPs that are irregular, thick, or opaque[42,57,72,74,75,168,190]. For smaller MPs (down to $20 \mu\text{m}$), the micro-FT-IR spectroscopy facilitates simultaneous visualization, mapping, and collection of spectra[74,168,175,191], but has certain requirements for the thickness, regularity, and transparency of the samples[72,192,193]. On the other hand, Raman spectroscopy, with its finer laser beam, can detect MPs as small as $1 \mu\text{m}$ [102], surpassing the capabilities of FT-IR spectroscopy which is limited to MPs larger than $20 \mu\text{m}$ [194]. Also, the non-contact analysis of Raman spectroscopy allows MPs to remain intact for subsequent examinations. However, Raman spectroscopy is extremely time-consuming and needs to take 24 h for Raman mapping [195]. Also, as Raman spectroscopy is based on the methodology that the fluorescent samples are excited by the laser, the contaminants by biological residual and others (such as additive and pigment chemicals) would interfere with the spectra, leading to the difficulty in the identification of polymer types[149,196]. For both techniques, given the large number of particles counted and time considerations, usually only a representative subset of samples (by color/category, in the range of 10 % to 100 % with a total number of around 100–200 particles) are chemically identified across all samples[42,46,55,79,149,165,167,168,197]. Particles achieving a synthetic polymer hit quality index (HQI) of > 60 –80 % were considered reliable and assigned to the tested samples [42,44,70,71,74,82,167].

Besides these spectroscopic methods, Scanning Electron Microscopy coupled with Energy Dispersive X-ray Spectroscopy (SEM/EDS) is another powerful analytical technique widely used in MP identification [41,46,78,198–200]. SEM employs a focused beam of high-energy electrons to scan the surface of a sample, producing high-resolution images of its topography and morphology. When combined with EDS, the electron beam interacts with the sample and generates characteristic X-rays specific to the elements present [41,46,199,201]. Although these techniques could determine the elemental composition, which is particularly valuable for distinguishing between plastic particles and other materials, they only provide qualitative analysis (not confirmatory tests) to identify particles that are similar to plastic (e.g., distinguish between organic and inorganic materials) [21]. Therefore, this method could not provide detailed polymer information which strongly restricts its application (~ 4 % of the cited studies).

In addition to spectroscopic methods, chromatographic methods such as pyrolysis-gas chromatography-mass spectrometry (Py-GC/MS), thermal extraction desorption-gas chromatography-mass spectrometry (TED-GC/MS), and liquid chromatography offer valuable mass-based insights into MP composition [53,111,183,202–204]. Py-GC/MS employs controlled thermal degradation to pyrolyze the polymer under an inert atmosphere by breaking its chemical bonds. This process breaks down the macromolecules into lower molecular weight molecules, which can be separated chromatographically by GC and detected by their mass spectrum through MS[104,205,206]. TED-GC/MS, a less destructive alternative to Py-GC/MS, uses lower temperatures to volatilize compounds from the sample[207–210]. By analyzing their thermal degradation products and comparing their mass spectra with those of spectral libraries or databases or by comparing the pyrograms obtained with the reference pyrograms generated by known virgin polymers, these methods could provide detailed mass-based information on polymer composition and additives, allowing for quantitative analysis of different polymer types[211]. In addition, results from these methods usually with high accuracy and greater sensitivity, since it is less affected by impurities that may be in the matrix (such as organic matter)

compared to spectroscopic techniques[24]. Liquid chromatography, on the other hand, involves dissolving MPs in a selected solvent and size exclusion chromatography for polymer characterization[179]. While this method typically requires substantial sample volumes, it offers a mass-based chemical characterization of bulk samples, improving representativeness. However, it should be noted that these chromatographic methods are destructive and need to be applied with caution, which may be the primary reason limiting their current applications (~ 10 % of the cited studies) [212,213].

Need to mention, considering the complex nature of environmental MPs (such as the different sizes, shapes, polymer types, and interfering materials) and the different features for various identification techniques. No single method can achieve comprehensive analyses of MPs and it is necessary to use multiple complementary analytical tools for accurate identification and characterization. In real-world practice, potential MPs are typically visually identified and categorized using stereomicroscopes for morphology features. Staining techniques like Nile Red may be employed to enhance the visibility of potential MPs. Following this preliminary sorting, chemical identification techniques are applied to confirm the polymer composition. Non-destructive spectroscopic methods, primarily FT-IR or Raman spectroscopy, are commonly used techniques to provide polymer-specific spectral fingerprints for chemical identification. For bulk samples or when investigating additives, destructive techniques such as Py-GC/MS or TED-GC/MS may be employed to provide additional mass-based information. This combination of visual and chemical analytical approaches allows the overcoming of limitations for individual methods and enables cross-validation of the results, ensuring a more comprehensive and reliable identification of MPs in environmental samples.

3. Microplastic occurrence in freshwater

3.1. Distribution and abundance of microplastic in freshwater

In the past years, an increasing number of studies have been identifying MPs in varied freshwater systems across the world in 72 countries, with most of the efforts in North America, Europe, and East Asia (Fig. 2 and Fig. 3). While after 2020, more efforts have been put into those underrepresented areas, such as Balkans (Albania[214], Greece [215], Romania[151], Montenegro[216], Turkey[120]), Central Asia (Kazakhstan[217], Uzbekistan[218]), South Asia (Nepal[219], Bangladesh[220], Pakistan[136,221]), West Asia (Saudi Arabia[222,223], Iran[224], Jordan[225], Iraq[226]), Africa (South Africa[80], Botswana[227,228], Ghana[229], Kenya[229], Nigeria[230], Egypt [231], Tanzania[154], Morocco[232], Tunisia[233], Namibia[234]), Oceania (New Zealand[97,235], Fiji[236]) and Central and South America (Mexico[237], Brazil[238], Ecuador[239], Colombia[240], Chile[241], Guatemala[242]). Based on the reviewed papers, MPs have been found in the freshwater systems on all continents, even in the stream of Antarctica[243], while the abundance of MPs is highly varying between different regions.

In the surface water, the MP abundance could vary from 0.003 particles/ m^3 (Northern Dvina River, Russia, in the size range of 0.5–5 mm, the observations in Antarctica are excluded considering their uniqueness)[244] to 3622,000 particles/ m^3 (Kinnickinnick River, U.S.A., in the size range of 0.01–5 mm) [65]. When considering the same sampling methods, for both surface water and sediment samples, the highest MP abundance was found in the area with the highest human economic activities, such as the Laurentian Great Lakes of North America, lakes and rivers in many European countries, and China (details can be found in the Supporting information, SI). Take grab sampling as an example, the abundance of MPs in surface waters of freshwater environments worldwide varied greatly from almost none to hundreds of thousands per cubic meter[245]. High MP abundance was found in residential areas and industrial areas with intense human activities. For example, high MP abundance is observed in the Amsterdam

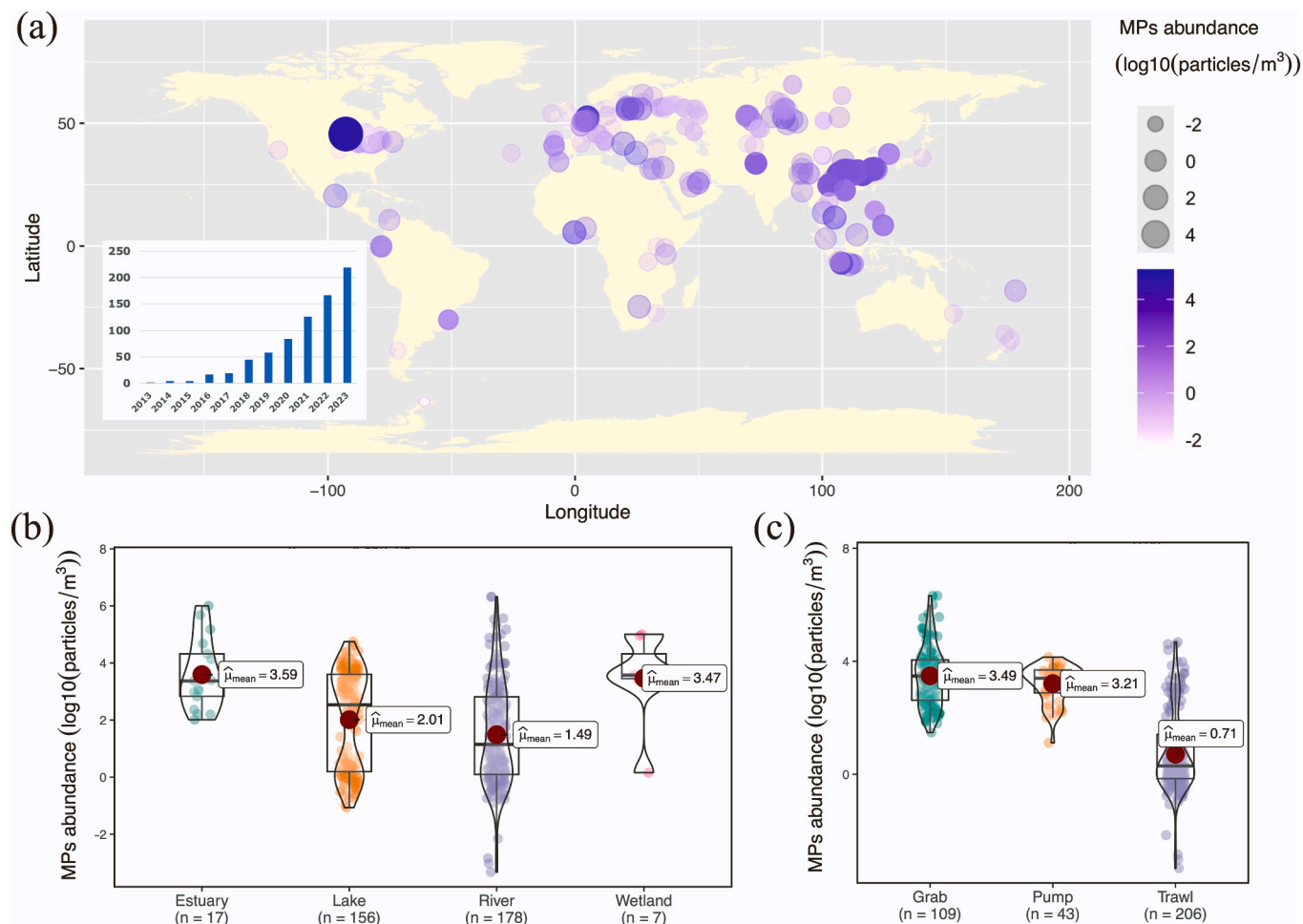


Fig. 2. MPs occurrences and characteristics in the surface water of global freshwater systems from the selected studies. (a) MPs abundance and distribution in global freshwater systems and the recent study trends. Only the studies with detailed latitude and longitude have been included in this figure. (b) MPs abundance in different water types. (c) MPs abundance from different sampling methods.

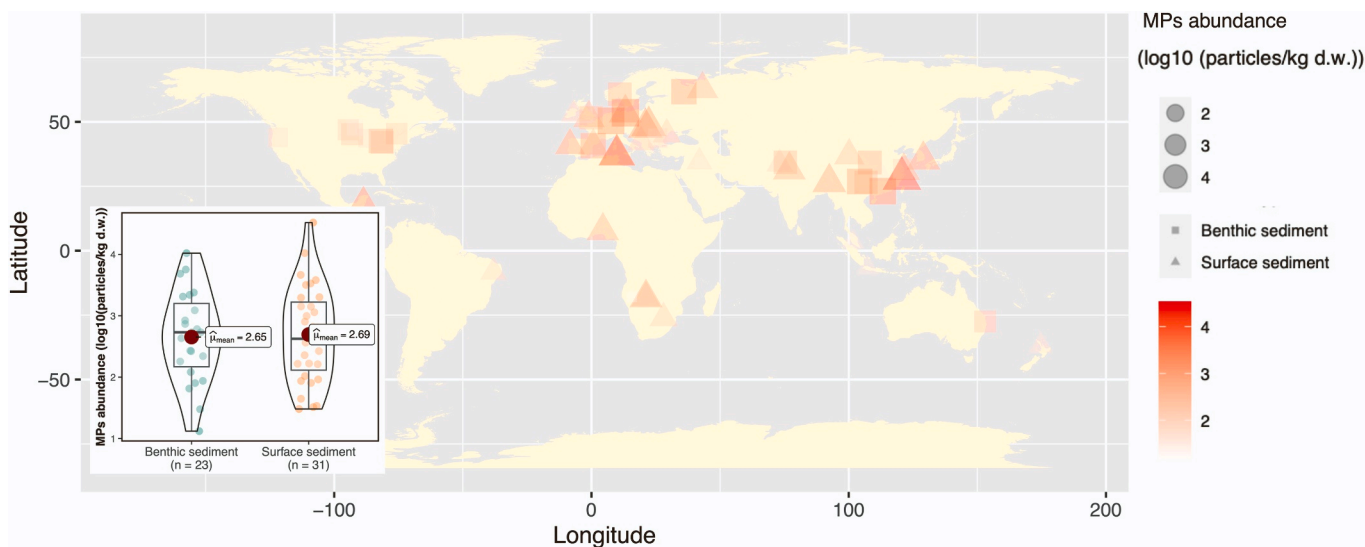


Fig. 3. MPs occurrences and characteristics in the sediments of global freshwater systems from the selected studies.

canals, Netherlands (48,000 to 187,000 particles/m³, in the size range of 0.01–5 mm) [145], Saigon River, Ho Chi Minh City, Vietnam (172,000 to 519,000 particles/m³, in the size range of 0.05–5 mm) [58], and Yellow

River estuary, China, where the abundance could reach 623,000–1392,000 particles/m³ (in the size range of 0.05–5 mm) [57] where larger residential areas, industrial and commercial centers exist. Opposite of

these, areas with less economic activity may have a relevant low MP pollution level, such as Lake Victoria in Uganda (0.73 particles/m³, in the size range of 0.3–5 mm)[42].

Regarding different water types (Fig. 2(b)), the estuaries exhibit notably higher MP abundance. This can be attributed to their unique position as convergence zones where rivers meet the sea. These dynamic environments experience frequent hydraulic exchanges, which inhibit MP settlement and promote resuspension, resulting in a higher abundance of MPs in the surface water[246]. As one of the most common water body types, the river has the lowest average abundance varying in a wider range. These results may be due to the constant movement of water (i.e., less residence time) to transport MPs downstream, potentially reducing local concentrations. This result may indicate that the rivers are more likely major transport pathways of MPs rather than sinks. Compared with rivers, lakes have a slightly higher average abundance. The potential explanation behind these results is that, as semi-closed systems with varying and weaker hydrodynamic conditions, MPs would stay longer in lakes and accumulate more readily[150,247]. Surprisingly, the wetlands, that usually away from intense human activities, have the second-highest average abundance. There are two potential reasons for this result. First, wetlands could act as natural filters, where complex vegetation structures, shallow water depth, and slow water movement may impede MP transportation and facilitate the long-term accumulation of MPs[197,248,249]. Second, based on the complex sampling environment, grab sampling is usually the only accessible method to collect the environmental samples in wetlands (85.7 % of these studies (6 of 7)) which results in higher recovery and counting rates (the reason is discussed below).

Also, there is a huge difference among different sampling methods (Fig. 2(c)). Trawl sampling is so far the dominant sampling method used in freshwater systems. The reported abundances using trawling sampling lie in a wide range which demonstrates the heterogeneity among different sampling areas. However, as discussed earlier, due to the relatively large mesh size (e.g., 300–335 µm is the most commonly used), trawl sampling will inevitably neglect small particles (<300 µm) which results in a lower MP abundance and the potential underestimation of the MP pollution levels. The highest MP abundance is reported when applying the grab sampling method. There are several potential explanations for this phenomenon. First, the samples for grab sampling are typically filtered through a membrane or glass filter with small pore sizes (typically 0.45 µm). Therefore, this method is capable of capturing small particles. Second, the grab sampling is suitable for locations without open water which may facilitate MP accumulation. Third, the selection bias from the researcher is also a potential reason. The researchers may have prior knowledge about the sampling areas and would typically expect a certain particle distribution (e.g., dominant of small MPs) and select the method accordingly. Besides these two methods, pump sampling seems to be a compromise option that could guarantee enough sampling size while also capturing the MPs in the smaller size range, leading to moderate MP abundance. While the adoption of pump sampling is currently limited with the least number of applications in the reviewed studies.

In addition to surface water, sediment is another important “sink” of MPs in freshwater environments[99] (Fig. 3). Throughout the world, the abundance of MPs in freshwater sediments ranged from dozens to tens of thousands per kilogram dry weight (d.w.)[87,100,132,250–254]. For example, the MP abundance in the sediments of the Lagoon-Channel of Bizerte in North Tunisia reached 3000–18,000 particles/kg d.w. (in the size range of 0.3 to 5 mm)[233]. While, the average abundance values per unit of dry weight in China could range from 34 (in the size range of 0.3 to 5 mm) to 32,947 particles/kg d.w. (in the size range of 0.02 to 5 mm)[31,255,256]. The distribution of MPs on sediments is uneven, largely influenced by their properties, human and environmental factors, such as winds, currents, and distance to the wastewater treatment plants (WWTPs) and industrial areas. Also, the results will be largely dependent on the sampling area (e.g., high tide line, intertidal areas,

transects) and sampling depth since some areas may contain higher concentrations of MPs[257–259]. For instance, MP content in different depth sediments shows significant differences, with a descending trend from the surface 0–5 cm layer to the 15–20 cm layer (54.7 % of that in 0–5 cm layer), and ultimately disappears when the depth exceeds 40 cm [258]. Similar results are also found in other studies[118,259]. Besides, the collection of sediments on the tide line, the high accumulation area for MPs, may result in overestimation[260]. In general, there is no significant difference between the MP abundance in the surface sediment and the benthic sediment globally (Fig. 3). However, when evaluated in the same or close sampling spots, MP concentrations found in the benthic sediments are typically higher than those observed in sandy beaches[99,113], which supports the hypothesis that benthic sediments might be an important accumulation pool for MP pollutants. Although there is more consistency in the selection of sampling tools for sediments compared to that for surface water, the abundance of sediment is still incomparable among different studies due to the different selection of units and the size range[59,261].

Need to mention, that almost all the studies reported the MP abundance using number-based units (i.e., number of particles) rather than weight-based units (~4 %)[67,183,204,256,262,263] primarily due to the special characteristics of MPs, that is MPs with smaller sizes may be more abundant and with higher environmental impacts but contribute negligibly to the total mass due to their tiny size.

3.2. Characterization of microplastic in freshwater

Although MPs have generally been defined as plastic debris between 1 and 5000 µm[22], there is still some deviation from the typical size classification of MPs, especially at the lower end of that scale[264,265]. The smallest size of MPs reported is mainly affected by the methodology applied to collect, extract, and detect MPs from environmental samples. In practice, the minimum size of MPs reported in water samples varied from 5 µm to 500 µm depending on i) the size of the mesh/filter used to sample water or perform filtration and ii) the resolution capability of the instrument used to identify the MP polymer type. Only 30.8 % of reviewed studies reported the smallest sizes of MPs detected or targeted, which leaves the cross-comparison between studies extremely challenging. In addition, it is also difficult to compare the concentrations and characteristics of MPs within different size classes across studies because there is no consistent size binning. There are 34 different size classes reported among the reviewed studies (SI), such as a typical five size categories of < 1 mm, 1–2 mm, 2–3 mm, 3–4 mm and 4–5 mm[76] and a more coarse size category of 0.02–0.25 mm, 0.25–1 mm, and 1–5 mm [78]. This discrepancy has made the comparison of MP concentrations by size very challenging.

Although there is no standard size classification binning for MP reporting, a number of studies have reported a trend of increasing concentration by decreasing the size of MPs in both water and sediment samples[97,142,266–269]. For example, Hu et al.[266] reported that MPs smaller than 500 µm were the most abundant in both water and sediment samples from the Yangtze River Delta, China, and that the abundance of MPs decreased with increasing MP size. The same trend has also been reported in the study of Yuan et al.[270] and others[199, 223,271]. In general (Fig. 4(a)), among the reviewed studies for surface water, MPs smaller than 1 mm are dominant (62 %). This emphasizes the importance of detecting smaller-size MPs to avoid underestimation of MP concentrations in freshwater environments because of the potential bioavailability to a wider range of organisms[272].

For the polymer types (Fig. 4(b)), Polyethylene (PE) (31 %) and polypropylene (PP) (27 %) were the dominant types of MPs reported in water samples across studies reviewed in this study. Further, polystyrene (PS), polyethylene terephthalate (PET), polyamide (PA), and polyvinyl chloride (PVC) accounted for 9 %, 8 %, 6 %, and 2 %, respectively. The abundance of polymer types found would reflect the global plastic demand as PE and PP are highly produced polymers

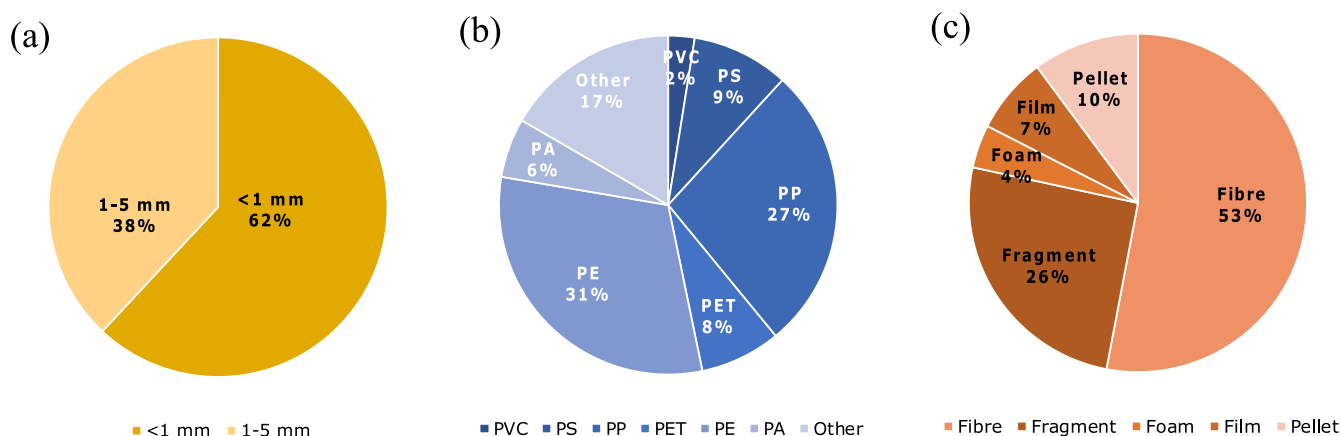


Fig. 4. Composition of MPs in size (a), polymer type (b), and shape (c). The detailed data can be found in the SI.

around the world and are widely used in packaging, personal care products, and containers[57,124,150,270]. In addition, PET and PA are commonly used plastic polymers in synthetic fabric which may be derived from WWTPs discharge[273].

Based on the morphological characteristics, MPs can generally be divided into fiber, fragment, pellet/granule, film, and foam based on the forms and definitions suggested by GESAMP[274]. Shapes can, to a large extent, indicate the parent materials of the MPs. Textiles are considered a major contributor of MP fibers, and washing is an important pathway that releases them into the environment[275]. Film mainly originates from plastic bags and packing materials, and foam and irregularly shaped fragments can originate from the breakdown of plastic containers or other plastic products[54]. Granules can be virgin pellets spilled during transportation and processing [276] or microbeads used as sandblasting media and abrasives in consumer products[199,261]. Based on the aforementioned studies, fibers, and fragments account for the overwhelming majority[277,278]. As shown in Fig. 4(c), fibers account for 53 %, probably because of a large amount of laundry wastewater discharge[251,279–282], and it is a concern because it is not removed by the current wastewater treatment process[283]. Fragments account for 26 %, and this can be because of the impact of runoff on the crushing

of large pieces of plastic[284]. In addition, pellets, films, and foams have also been found in freshwater in proportions < 10 %, of the total pollutants.

4. Factors affecting the spatial-temporal distribution of microplastics

4.1. Sources and pathways of microplastics to freshwater

MP pollution was strongly affected by a variety of factors, such as those related to human activities[43], geographic characteristics[247], and seasonal variation[57,98]. Understanding the sources and sinks of MPs can help to design targeted mitigation strategies (Fig. 5).

Considerable evidence has demonstrated that the effluents from WWTPs are a major pathway of MP pollution in freshwater systems, especially in developed urban areas [59,76,172,238,267,283,285–288]. Despite the fact that advanced WWTPs (like tertiary treatments) are capable of removing up to 98 % of MPs over 10 μm [147,203,287,289, 290], there are still millions to trillions pieces of MPs being discharged into the receiving water per year via WWTPs considering their giant volume[111,145,286,287,291,292–297]. Studies have reported that the

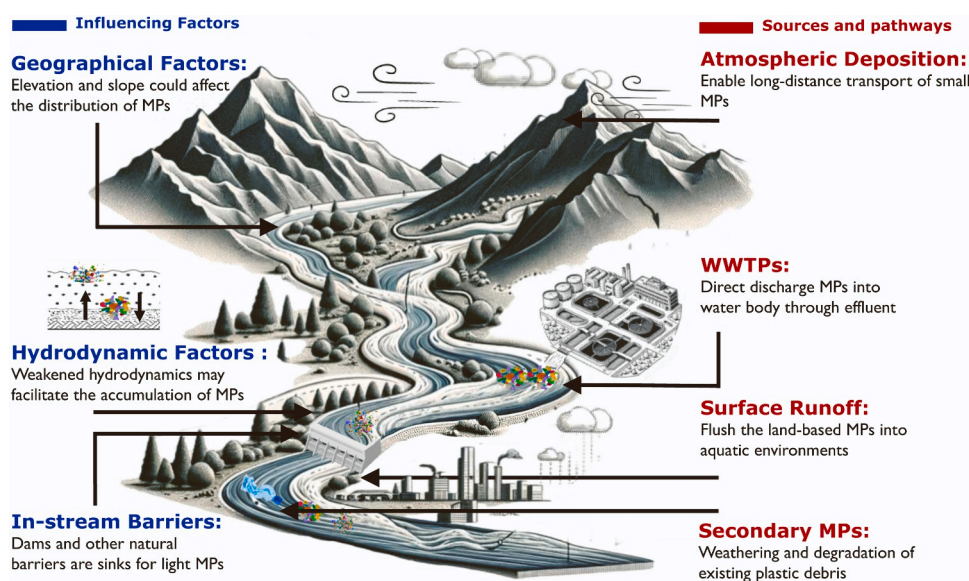


Fig. 5. Sources, pathways, and fate of MPs in freshwater systems. The direct pathways include effluents of the WWTPs, surface runoff, and atmospheric deposition. After entering the water bodies, the existing large plastic debris could degrade to secondary MPs through the weathering and degradation process. After that, these MPs would be transported along the freshwater systems which are affected by geographical and hydrodynamic factors.

MP abundance in the effluents could reach a very high level (e.g., 51,000–81,000 particles/m³ reported by Leslie et al. [145] in the size range of 0.01–5 mm) and the MP abundances were highest in sediment samples closest to the discharge site of WWTP effluents (e.g., 4400 ± 620 n/kg d.w. reported from Viitala et al. [153] in the size range of 0.063–5 mm) compared to other sites. Besides, there are also studies found that the MP abundances at sites downstream of WWTPs could be nearly ten times higher than that in upstream [298], which indicates that WWTPs (especially those areas without advanced treatment techniques where the MP abundance could be 4000 times higher than that in upstream) [239] serve as an important pathway of MPs to freshwater environments [299]. However, the level of pollution they contributed may still vary based on their scale, location, and type of influents [300–302]. Besides, the composition of the MPs in the effluents can also be a clue to determine the original sources of the MPs. For example, MPs in the effluent of a sewage WWTP were very similar to those in toothpaste formulations regarding color, shape, and size, indicating that MPs in personal care products may be potential sources of MPs in freshwater environments [290].

Besides point sources such as WWTPs, MPs can also enter the freshwater system through non-point sources through surface runoff and atmospheric deposition [303]. For surface runoff, one typical pathway is through precipitation which may flush the MPs or larger debris from terrestrial environments (e.g., tire and road wear particles, leachate from waste landfill, and agricultural plastic mulching) into aquatic environments [149,304–312] or cause the combined sewer overflows [313]. High abundances of MPs in surface waters (could be up to 2 magnitudes higher than the normal abundance) have been observed after rain events [55,149,291,304,308,314–317] and these abundances (e.g., 1100 to 24,600 particles/m³ reported by Werbowski et al. in size range of 0.125–5 mm and 15,400 particles/m³ reported by Grbić et al. in size range of 0.025–5 mm) could be even higher than those in nearby WWTPs effluent [149,318]. Besides, large amounts (with a maximum abundance of 660 particles/kg) of large-size (1–4 mm) MPs in sediments downstream of storm drainage outlets have also been discovered [15,100]. Based on the composition of the MPs, tire and road wear particles [318], industrial overflows [313,315], leachate from waste landfills [311], and agricultural runoff [53,149] are all identified as direct sources of MPs in surface runoff. All this evidence suggests urban surface runoff is a major source of MPs, especially for areas away from WWTPs.

Another important source of MPs in freshwater was atmospheric transport and deposition [45,319–321]. Wind can blow away light MPs from the ground and deposit them in areas far from their original sites (could also be facilitated through precipitation) [322]. A study based on the monitoring of the wet and dry deposition in a remote Pyrenean mountain location found that MPs can be transported through the atmospheric deposition to reach and affect remote, sparsely inhabited areas [323]. This conclusion is further verified by the fact that large amounts of secondary MPs were found along the shores of sparsely populated mountain lakes, where there was scarce primary MP pollution [89,321,324]. Another study also monitored the MP abundance in the rainfall (700 to 6000 particles/m³ reported by Zhang et al. in the size range of 0.1–5 mm) [325], which could account for 24.0 % to 77.4 % of the total surface runoff to the receiving water.

Besides these direct sources and pathways, weathering and degradation of existing plastic waste (mainly from mismanaged municipal solid waste like open dumping and inadequate landfilling) [119,326] in water bodies also provide great contributions [12,168,327]. Plastics in freshwater systems undergo physical (e.g., mechanical degradation from the action of waves or sand friction) [328] and environmental degradation (such as oxidative weathering from exposure to UV-B or biodegradation such as by the action of hydrocarbon-degrading microorganisms) [328,329]. In general, the overall degradation patterns of MPs in freshwater were found to be similar to those in the marine environment: cracks, pits, and adherent particles [330,331], despite milder physical forces than in marine environments [11]. The degree of weathering to

the surface of MPs can be used to track the history of the particles which indicates most of the MPs in the natural waterbodies are secondary MPs. However, the rate of fragmentation and degradation of plastics is still unknown in freshwater environments [24].

Unlike many other pollutants, MPs can continuously break down into smaller particles through degradation which significantly increases the abundance and the difficulty of removal once they are released to the aquatic environment [332–334]. Therefore, due to this special characteristic, it is extremely valuable to understand the potential sources of MPs and implement effective mitigation strategies at its source. Among them, the primary mitigation strategy focuses on effective plastic waste management [335–338], such as the restriction of open dumping, promotion of effective plastic recycling, and conduct of strict leachate management. Besides, interception strategies such as implementing advanced wastewater treatment technologies (e.g., tertiary treatments, membrane bioreactors) is a key step to capture a large proportion of MPs before they enter freshwater systems [339–341]. Additionally, the implementation of green infrastructure in urban areas, such as bio-retention systems and constructed wetlands, can help trap MPs in stormwater runoff before they reach water bodies [342–344]. Recently, research efforts have been shifted to degrading environmental MPs into nontoxic intermediates or highly valuable products through techniques such as advanced oxidation processes and biodegradation [345–349]. Although satisfying degradation performance has been achieved under lab conditions, there is still a long way before large-scale application in aquatic environments [350]. Consequently, source control will remain a cornerstone of MP management strategies for the foreseeable future.

4.2. Transport of microplastics in freshwater

Understanding the transport and potential pathways of MPs in freshwater systems is crucial for shaping effective management strategies. Many studies have demonstrated the critical role of rivers for MP transportation and the annual load/flux could vary from hundreds of billion to trillion particles per year [281,314,351] or dozens to thousands of tons per year [44,71,352–355]. For instance, the annual load of MPs carried by the Nakdong River, South Korea was 53.3–118 tons by weight, with 81 % transported during the wet season [352]. While, the annual MP load from the Yangtze River, China to the East China Sea could reach 7020 tons [353]. There is also another study trying to estimate the MP loads from all European rivers and the results indicated that in total of 14,400 tons of MPs were exported from point sources to the North Sea, Baltic Sea, Black Sea, Mediterranean Sea, and the European river basins draining into the Atlantic Ocean in 2000 [356]. All these results highlight the substantial contribution of river systems to marine plastic pollution. Lakes and reservoirs present a different dynamic, often acting as sinks for MPs. A modeling study conducted for Switzerland found that 33 % of all MPs are retained in lakes, of which 99 % are retained in the 15 biggest lakes in Switzerland [357]. However, the number of studies on the lake is still highly limited and requires extra effort in the future. In general, these loads and fluxes are typically estimated through process modeling that combines field survey data with source information. MP fluxes may be inaccurately estimated if the input data are not sufficiently comprehensive, which hinders cross-comparisons between studies and regions [356].

While it is widely acknowledged that freshwater systems serve as critical pathways for MPs, their transport is governed by a complex interplay of factors. Among these, geographical characteristics play a significant role in determining the distribution and fate of MPs in aquatic environments [167]. Various geographical characteristics (e.g., elevation, slope, bend, surface water areas) could influence the transport of MP, which further affects its abundance distribution. Correlation and regression analysis have been conducted to demonstrate that the MPs seems to accumulate in downstream areas where higher abundances and lower elevations are observed when there are no other direct emission sources upstream [79,80,154,167,358]. Similarly, bivariate regressions

also find there is a positive relationship between the slope of the watershed with its MP abundance[149]. In addition, the narrowing of the water surface may increase the concentration of buoyant MPs, since there is less surface area for these particles to be distributed across[68, 73]. Regarding sediment, studies have shown that MPs tend to be deposited in the sediment along the bends of rivers[257]. However, only very few studies have directly addressed these links and additional efforts are urgently needed for research in this area.

Besides the geographical factors, hydrodynamic factors also play a vital role in MP transport[53,359]. There is evidence that longer water residence time, lower flow rates, and weakened hydrodynamics may facilitate the accumulation and deposition of MPs due to the inhabitation of the turbulence and resuspension of settled particles to the surface of the water column and the facilitation of the settlement to the sediment through the biofouling, adsorption of natural substances and gravity [68,186,256,270,360–365]. For instance, MP abundances are typically lower in the center of river channels and higher along river banks[366, 367] and higher MP abundances may be present in sediment than in surface water during the dry season[352,368,369]. Meanwhile, volume changes (usually due to seasonality such as floods in wet seasons) could also affect the vertical transport of MPs [360,370,371]. Such intense hydrological activities could cause MPs to be less apt to settle or to remain trapped in surface sediment, and even resuspend from benthic sediments in the water column due to sediment mobilization[117,369, 372,373], especially for small-sized MPs (50–500 μm)[374].

Besides these, in-stream barriers, such as dams or aquatic vegetation, have been found to be sinks for light plastic particles, which would also drive the spatial variability of MPs along rivers [361,375–379]. As a result of water impoundment, floating materials on the water surface cannot pass the dam. Previous studies have reported the accumulation effects of dams on MPs by comparing the concentrations in upstream rivers and within reservoirs [54,380–383], indicating that accounting for dams may be important when modeling global riverine MP transport.

5. Limitations in current studies

5.1. Lack of harmonized and standardized analyzing framework

The first issue that needs to be addressed in order to investigate the general impact of MPs in the freshwater system is the harmonization of MP analysis protocols. Currently, no unified methods exist for sampling, pre-treatment, and detection of MPs. Therefore, researchers are applying different analysis protocols in different studies which hinders data comparability.

For example, based on the reviewed publications, current studies have used trawl sampling (51 %, more than one technique can be used in a

study), grab sampling (37 %), and pump sampling (11 %) to collect the MPs in the surface water. Trawl sampling offers advantages in covering large sampling areas and reducing sample volume, but it will inevitably neglect small particles, especially fibers[57,70,71]. In contrast, grab and pump sampling could mitigate MP loss during sampling and are more suitable for diverse geographical and hydrological conditions, such as wetlands and low-flow rivers. However, these techniques are constrained by practical limitations on sample volume. Therefore, due to the nature of various sampling methods and the selection of different mesh sizes (Fig. 6(a)), the reported MP abundance is provided in different units (such as particles/ km^2 for trawling methods and particles/L for bulk sampling methods) or with huge differences in the magnitude of the reported results (Figs. 6(b), up to 3 to 4 magnitudes) in same studies, and even for the same sampling sites[59,69,75,77,170, 196,384]. Such inconsistencies have also been witnessed in the sediments where the units can vary from particles/kg d.w., particles/L, to particles/ m^2 [63,83,89,110,123,150]. In the meantime, few studies have conducted direct comparisons of different analysis techniques in the same environmental matrices which leads to a lack of understanding of the selection criteria for these techniques[385,386]. In addition, there is currently no standardized reporting format for MP studies which may cause the missing of certain important information in the reported results. As mentioned earlier, about 70 % of the studies didn't report the size range of the MPs or in very rough and inconsistent size categories (such as <1 mm, 1–2 mm, and 2–5 mm). Even for the same samples, the MP abundance could have a difference with up to two magnitudes when reporting in different size ranges[228]. Such inconsistency makes the cross-comparison of different studies extremely difficult which hinders the understanding of the status of MP pollution in ecosystems and the development of appropriate mitigation strategies.

5.2. Lack of monitoring in specific regions and facilities

Although there is an increasing number of studies focusing on MP pollution in freshwater systems, data on MP occurrences in freshwater systems is still fragmented and there is a severe spatial imbalance in relevant studies across the globe with a significant underrepresentation of results from Africa, South America, and North Asia. The lack of monitoring data and the geographical disparity create blind spots in the understanding of global MP contamination, potentially overlooking regional nuances in MP sources, distribution patterns, and environmental impacts (Fig. 7). Additionally, most studies lack long-term, replicate sampling, and continued measurement which impedes the validation of regional MP pollution levels and hinders the tracking of potential pollution sources and influencing factors[23]. Besides, there is an acute absence of regular monitoring data from important industrial

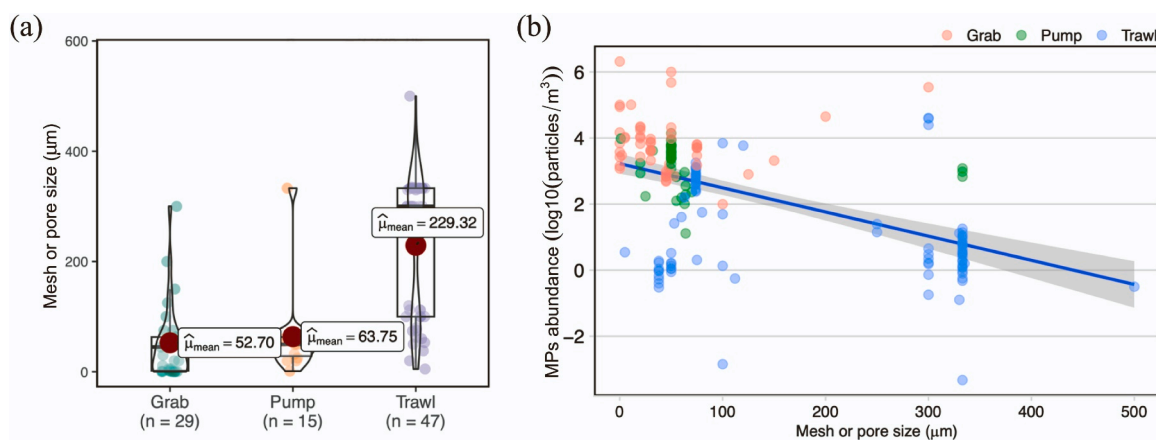


Fig. 6. Relationship among the mesh size, sampling methods, and the detected MPs abundance. (a) Mesh or pore size applied for different sampling methods. (b) Scatter plot for the applied mesh or pore size with the reported MPs abundance.

and infrastructural facilities (e.g., industrial companies and WWTPs) that are likely major contributors to MP pollution. The lack of suitable methods for cost-effective real-time monitoring and regular monitoring data results in no clear picture of the magnitude of MP pollution and hampers the ability to develop targeted strategies for MP management.

5.3. Lack of understanding of microplastics in finer size ranges and size classifications

Despite the acknowledgment that smaller MPs (<100 μm) pose significant environmental hazards[24], the majority of current studies have still focused on relevant larger particle sizes (>300 μm), primarily due to the limitations of the selected sampling (i.e., limited by the mesh or sieve size) and detection methods (i.e., limited by the resolution limit for the selected detection methods). This may lead to potential underestimation of the concentrations and risks for those small particles (Fig. 7). Nowadays, nanoplastics, in particular, present a novel challenge due to their size and potential for pervasive environmental penetration. The limitations of current analytical techniques in identifying and characterizing small MPs call for urgent development in more advanced analysis methods. By extending the detectable range of MP sizes, researchers can gain a more accurate understanding of their distribution and risks, which is essential for effective environmental risk assessment and the development of mitigation strategies.

5.4. Lack of studies to model the distribution and transport of microplastics

As research in the field of freshwater MPs is still in its infancy, much is still unknown regarding their spatiotemporal distribution and transport characteristics (Fig. 7). Most of the current studies are still simple monitoring studies that report the occurrence of MPs in a single or few water bodies and their corresponding characteristics. Only a small number of studies focus on modeling while limited to examining MP concentrations as a function of either spatial or temporal factors, with very few addressing both and across scales[387]. Some studies tried to use hydrodynamic modeling (e.g., Lagrangian transport model and others) to simulate the transport of MPs in different water bodies[356, 388–390]. However, these models are highly computation intensive and require massive input data, which involves the consideration of hydrodynamic processes (such as advection, dispersion, aggregation, sedimentation, and resuspension) and various emission sources. Thus, most of the current studies are limited in either spatial scales (usually a small section of a single water body) or can only consider a few types of point sources. There is a lack of modeling studies to systematically understand the distribution of MPs on a larger spatial scale and explore the interactions of multiple influencing factors with MP pollution to facilitate the identification of hotspots and guide policy design.

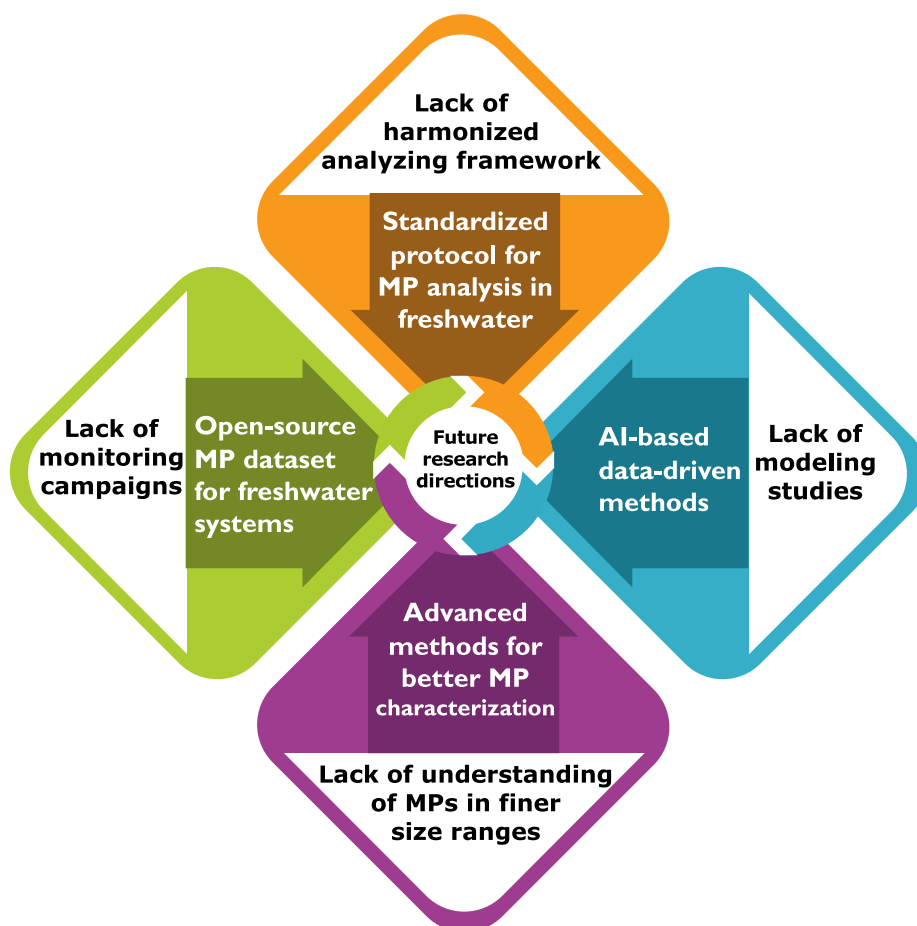


Fig. 7. Limitations in current studies and future research opportunities.

6. Future research directions

6.1. Standardized protocol for microplastic analysis in freshwater systems

To fill critical gaps in MP research in freshwater systems, there is a pressing need for a universally accepted standard analysis protocol to harmonize the selection of sampling, separation, and detection techniques. A concerted global effort could facilitate the standardization of future MP analysis practices and guarantee the quality and comparability of the provided results, which leads to a more complete and comprehensive understanding of MP pollution. To be part of this action, here, we summarized the minimum requirements for such protocols based on the current scientific findings.

For MP sampling in freshwater systems, considering the obvious flaws of trawl sampling as (1) its serious underestimation (a difference of more than three orders of magnitude) of small particles ($< 300\ \mu\text{m}$) and particles with specific shapes (usually fibers), [57,70,71] (2) infeasibility in specific geographical and hydrological conditions, and (3) difficulty to estimate the exact volume of water being filtered (as the net's immersion depth changes constantly with waves and boat movement), trawling methods, which are widely used in marine MP sampling, should no longer be considered as a preferred method in MP sampling in freshwater systems [23,196]. For two types of bulk sampling methods, call for a minimum volume to be set to improve the representativeness and comparability of studies is urgently needed. Based on the current comparison studies, an absolute minimum volume of 500 L per sample is highly suggested based on the typical MP abundance in water bodies [22]. Besides, considering the significant environmental hazards of MPs with smaller sizes, we recommend setting the mesh size for the sieves or the pore size for the filter papers as $50\ \mu\text{m}$ or lower to include more particles in those ranges [391]. For all three types of sediment sampling, it is necessary to report the sampling volume (e.g., wet weight, volume, or sampling area) and sampling depth to guarantee representativeness and cross-comparison. Samples are recommended transferred using non-plastic containers (such as glass trays or steel-less buckets), covered with aluminum foil to avoid additional efforts for blank control tests.

Regarding sample pre-treatment, during the initial density separation process, although saturated NaCl solution with a density of $1.2\ \text{kg/L}$ is previously recommended by both the MSFD technical subgroup (2013) [92] and NOAA [392], considering the low recovery of higher density polymers (i.e., resulting in an underestimation of the MP abundance), this solution is no longer suitable for efficient MP separation. The same reasoning also applies to CaCl_2 and sodium tungstate dihydrate ($1.4\ \text{kg/L}$, not high enough). Solutes such as ZnCl_2 , though reusable, are often expensive, hazardous, corrosive, and flotation is often performed using a centrifuge, where space is limited [158]. For the current situation, we propose to use higher density solutions such as NaI ($1.6\ \text{kg/L}$) due to its high density, safety, and the possibility of reuse, and possibly in combination with separation columns or the use of oil to improve recovery rates [141]. For the purification of organic matter, acid and alkali digestion should be avoided or at least used with caution since it may lead to the underestimation of MPs in environmental samples due to its destruction of certain particles.

For sample identification and quantification, based on the editorial from the journal of *Science of The Total Environment*, the use of only simple microscopic techniques or visual inspection for both sample identification and quantification is highly questionable and is no longer recommended in future scientific reports [393]. In fact, it is difficult to quantify and qualify MPs from complex environmental samples using a single analytical method. Thus, the use of complementary staining methods and the combination of multiple methods is preferred, which strongly depends on the sizes of MPs [23]. When the sizes of MPs fall in the range of $< 1\ \text{mm}$ and the minimal cut-off size is tens of microns, the combination of the microscopic analysis with spectroscopic analysis is highly recommended. Regarding ease of handling, analytical time, and number of polymers to be analyzed, $\mu\text{-ATR-FTIR}$ spectroscopy is

currently the method recommended for routine analyses of environmental samples. If the minimal cut-off size is in a range of a few microns, Raman spectroscopy is preferred as it is the only technique to obtain better spectra from particles $< 20\ \mu\text{m}$ in size. Thermal analysis (e.g., pyrolysis-GC-MS) and automated mapping spectroscopy (e.g., FPA-FTIR) may be suitable for laboratory experimental samples of known polymer types. Although it has some advantages over $\mu\text{-ATR-FTIR}$ and Raman spectroscopy, small MP particles may still be missed in complex environmental samples with various unknown types of weathered polymers. These methods are not recommended for routine monitoring studies at present but may be useful for screening analyses of bulk samples or further complementary analyses of MPs that have not been fully characterized by spectroscopy [179]. For the final reported results, we recommend using concentrations by volume (particles/ m^3) for surface water samples and concentrations by mass (particles/ kg d. w.) for sediment. Meanwhile, we suggest using detailed 7-size classes or finer ones (i.e., $20\text{--}50\ \mu\text{m}$, $50\text{--}100\ \mu\text{m}$, $100\text{--}200\ \mu\text{m}$, $200\text{--}500\ \mu\text{m}$, $500\ \mu\text{m}\text{--}1\ \text{mm}$, $1\text{--}2\ \text{mm}$, $2\text{--}5\ \text{mm}$) for easy and fair comparison among precedent and future studies.

Here, we summarized the best practices of the main procedures used in MP analysis so far based on the current studies (Fig. 8). We hope this result could accelerate the pace for the improvement of current protocol for MP analysis in freshwater systems, like the recently released one from UNEP [394] and those that have previously designed for the marine systems (MSFD [92], NOAA [392], and GESAMP [274]). Meanwhile, as new techniques are still emerging, this protocol could be further improved in the near future.

6.2. Advanced analysis methods for better microplastic characterization

Increasing demand for MP pollution monitoring at national and global levels requires the improvement of existing methods and the development of novel methodologies to reduce identification time and effort [179]. The technological frontier in MP research lies in the development of advanced pre-treatment and detection methods for better efficiency and accuracy. Promising research directions lie in almost every step of MP analysis which include, for example, the further discovery of novel enzymatic degradation methods with lower cost, higher recovery rates, and shorter processing time for sample pre-treatment and advanced spectroscopy or imaging techniques to enable fast, real-time detection of MPs with lower detection limit and reasonable cost. Besides, the fast development of artificial intelligence (AI) also enables the realization of fully- or semi-automated analysis of MPs that incorporate image analysis methods (e.g., computer vision and other deep learning methods) to obtain the physical (size, shape, and color) and chemical (polymer type) characteristics of the MPs [395–406].

6.3. Open-source microplastic dataset for freshwater systems

An open-source, standardized MP dataset would catalyze a paradigm shift in this research area, allowing for unprecedented levels of collaboration and data-driven decision-making. Currently, there are growing efforts to build global MP datasets for public decision-making and scientific discoveries. However, most of the current achievements are for the marine environment, such as the global MP database from the National Centers for Environmental Information (NCEI) [407] and the Global Microplastics Initiative from the Adventure Scientists [408]. Besides, considering the ununified methods for MP sampling and analysis, there are still certain concerns about the quality of the data collected from crowdsourcing. There is an urgent need to construct a comprehensive (i.e., including varied freshwater environments from remote lakes to urban waterways), accessible (i.e., open-source), transparent (i.e., the data source is trackable), and standard (i.e., data are calibrated and validated to guarantee comparability) database for MP pollution in freshwater systems. Such efforts would improve the future modeling of

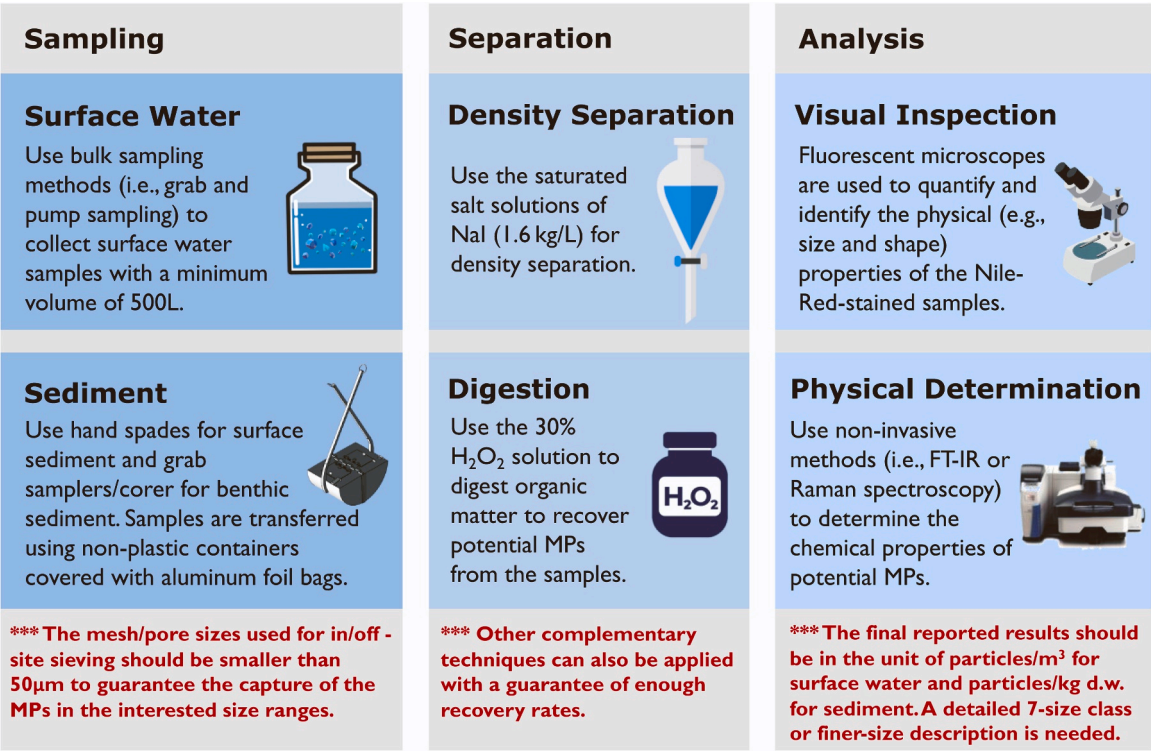


Fig. 8. Best practices of the main procedures used in MP analysis so far.

MP contamination, facilitate cross-study comparisons, and provide a robust evidence base to inform and influence global policy decisions.

6.4. AI-based data-driven methods to understand the global distribution and driving factors of microplastics

Developing quantitative methods that accurately represent the

global distribution and elucidate the driving factors behind MP pollution is critical. The current methods are either limited to a few influencing factors or in relatively small spatial scales. This is an urgent need for a more fancy and sophisticated modeling approach that can simultaneously consider multiple influencing factors (such as flow volume, human activities, and industrial output) and map the MP distribution with higher spatial and temporal resolutions. As mentioned earlier,

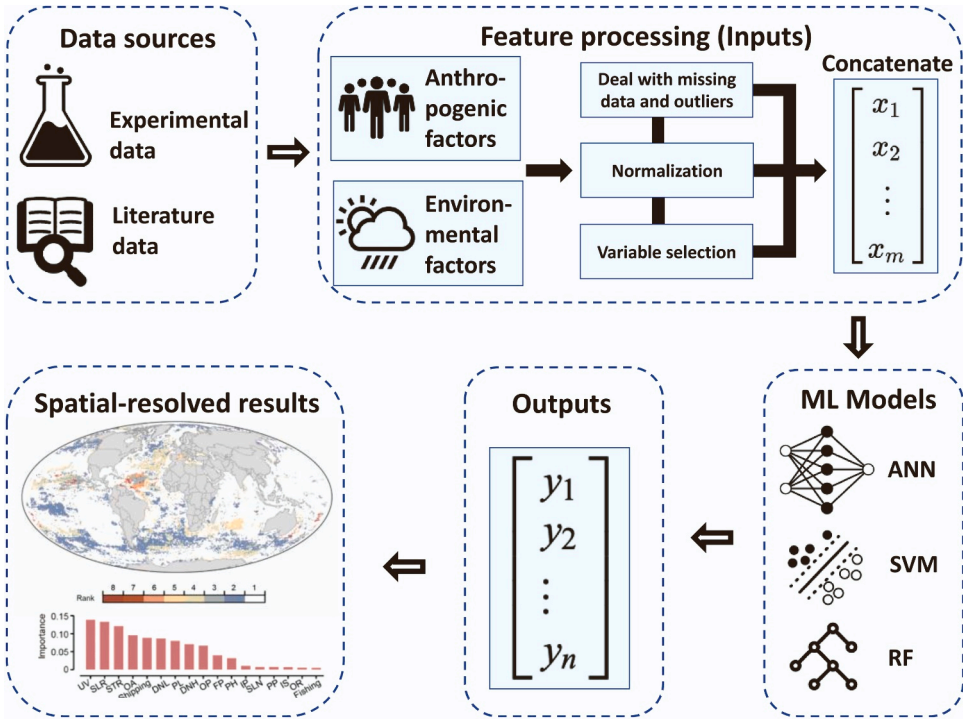


Fig. 9. Basic modeling framework for the AI-based data-driven prediction models for MP analysis.

although in its infancy, the fast development of AI provides new insights into this challenge [409,410]. Machine learning (ML) models have been adopted to achieve rapid and large-scale estimation of MP abundance in different environmental media [411–417], with only two of them focused on the freshwater environment [416–418]. In all of these studies, supervised learning models are trained based on the collected empirical data from the literature or from lab experiments to characterize the complex nonlinear relationship between the MP abundance with the selected predictors [411–415,417] (Fig. 9). The common predictors include both anthropogenic (e.g., population density) [412,414, 415,417] and environmental factors (e.g., meteorological information and physicochemical conditions of the sampling area) [411–415,417] which may directly or indirectly affect the MP distribution. The provided results demonstrate that ML models have great potential to provide reasonable baseline estimation for MP pollution, identify potential influencing factors, and provide predictions for future trends. These efforts could provide quantitative evidence and transformative insights to support sustainable MP management through the lens of a circular plastics economy and better facilitate the understanding of how manufacturing processes, waste management practices, and consumer behavior contribute to MP pollution in order to develop effective, systemic pollution prevention strategies.

7. Conclusions

MPs have become one of the emerging contaminants in the aquatic environment and have caused increasing public concern. However, there is still a lack of sufficient knowledge about MPs in freshwater systems. This review evaluates the current understanding of MP monitoring in freshwater environments by systematically summarizing the progression of analytical methods, and examining the distribution, characteristics, and sources of MPs, alongside the existing issues and future research directions. Currently, the presence of MPs in freshwater systems has been reported in every continent around the world with highly varying pollution levels. Higher MP abundance was found in the areas with intense human economic activities and there is great heterogeneity in the physical and chemical characteristics of the MPs among different regions. Different MP abundance distributions have been witnessed among different water bodies (e.g., rivers, lakes, estuaries, and wetlands), while sampling methods and size range selections could significantly influence the reported MP abundances. Based on the current findings, the major sources and direct pathways of MPs in the freshwater environments are the effluents of the WWTPs and surface runoff, while the secondary MPs generated from the existing large plastic debris also provide great contributions. After that, these MPs would be transported along the river systems into lakes or oceans. So far, MP pollution has no unified sampling, pre-treatment, and detection methods, which caused significant difficulty in comparing the pollution levels for different studies. Most studies lack long-term, replicated sampling, and continued measurement and there is a critical data gap in specific regions and facilities. There is an urgent need to construct a standardized protocol for MP analysis in freshwater systems and advanced analysis and modeling techniques are required to enable a better understanding of MP pollution and facilitate the development of targeted strategies for MP sustainable management.

Environmental implication

Microplastics (MPs), as an emerging environmental pollutant, have raised great global concerns due to their ubiquitous presence in aquatic ecosystems and their potential risks to the environment and human health. This paper provides an updated and more in-depth review to systematically summarize the current understanding of MP monitoring in freshwater environments by examining the distribution, characteristics, and sources of MPs, alongside the progression of analytical methods with quantitative evidence. We also discussed the current knowledge

gaps and research priorities for MP pollution in freshwater systems to facilitate a better understanding of MP pollution.

CRediT authorship contribution statement

Bu Zhao: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Ruth E. Richardson:** Writing – review & editing, Validation, Project administration, Methodology, Investigation, Formal analysis. **Fengqi You:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.135329.

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