

Microplastics in aquatic environments: A comprehensive review of toxicity, removal, and remediation strategies



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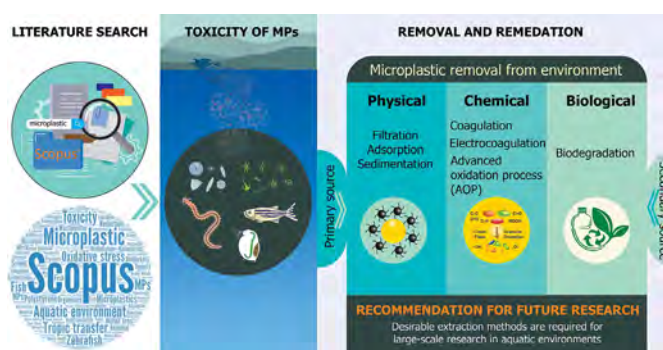
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HIGHLIGHTS

- Toxicity on fish, microalgae, zooplankton and polychaete, and remediation of MPs in aquatic environments were reviewed
- MPs combined with other contaminants could induce higher toxicity
- Biological and engineering processes for MPs removal were discussed
- Compared with engineering approaches, the biological process takes enough time to degrade MPs
- The density separation method as an engineering treatment exhibited excellent performance in MP removal

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of microplastics (MPs) in aquatic environments has been a global concern because they are toxic and persistent and may serve as a vector for many legacies and emerging pollutants. MPs are discharged to aquatic environments from different sources, especially from wastewater plants (WWPs), causing severe impacts on aquatic organisms. This study mainly aims to review the Toxicity of MPs along with plastic additives in aquatic organisms at various trophic compartments and available remediation methods/strategies for MPs in aquatic environments. Occurrences of

Abbreviations: APP, Antifouling paint particle; B[a]P, Benzo[a]pyrene; HDPE, High-density polyethylene; DEHP, Di(2-ethylhexyl)phthalate; DBP, Dibutyl phthalate; DDT, Dichlorodiphenyltrichloroethane; DAF, Dissolved air flotation; DM, Dynamic membrane; FTIR, Fourier transform infrared spectroscopy; LCC, Leaf-branch compost cutinase; LDPE, Low-density polyethylene; M-CNTs, Magnetic carbon nanotubes; MHET, mono(2-hydroxyethyl) terephthalic acid; MPs, Microplastics; MPSS, Munich Plastic Sediment Separator; MSF, Magnetic seeded filtration; NPs, Nanoplastics; NTU, Nephelometric Turbidity Unit; OFE, Oil film extraction; POPs, Persistent organic pollutants; PA, Polyamide; PAHs, Polycyclic aromatic hydrocarbons; PBDs, Polybrominated diphenyls; PDBEs, Polybrominated diphenyl ethers; PFAS, Polyfluoroalkyl substances; PAHs, Polycyclic aromatic hydrocarbons; PCBs, Polychlorinated biphenyls; PET, Polyethylene terephthalate; PLA, Polylactic acid; PP, Polypropylene; PPU, Polyurethane; PS, Polystyrene sulfonate; PUR, Polyurethane; PVC, Polyvinyl chloride; ROS, Reactive oxygen species; ST, Supplementary Table; SF, Supplementary Figure; TPTCl, Triphenyltin chloride; WWTPs, Waste water treatment plants.

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Plastic additives
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Wastewater treatment
Remediation

oxidative stress, neurotoxicity, and alterations in enzyme activity, growth, and feeding performance were identical in fish due to MPs toxicity. On the other hand, growth inhibition and ROS formation were observed in most of the microalgae species. In zooplankton, potential impacts were acceleration of premature molting, growth retardation, mortality increase, feeding behaviour, lipid accumulation, and decreased reproduction activity. MPs together with additive contaminants could also exert some toxicological impacts on polychaete, including neurotoxicity, destabilization of the cytoskeleton, reduced feeding rate, growth, survivability and burrowing ability, weight loss, and high rate of mRNA transcription. Among different chemical and biological treatments for MPs, high removal rates have been reported for coagulation and filtration (>86.5 %), electrocoagulation (>90 %), advanced oxidation process (AOPs) (30 % to 95 %), primary sedimentation/Grit chamber (16.5 % to 58.84 %), adsorption removal technique (>95 %), magnetic filtration (78 % to 93 %), oil film extraction (>95 %), and density separation (95 % to 100 %). However, desirable extraction methods are required for large-scale research in MPs removal from aquatic environments.

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1. Introduction

Plastics are ubiquitously used in daily life as they are light weighted, durable, and low price (Meng et al., 2020). As a result, global plastic production has increased notably since 1950 and has already exceeded 2791 million tons (Cai et al., 2022). If the scenario continues, the volume of plastic debris will be 12 billion metric tons by 2050 (Sobhani et al., 2020). Microplastics (MPs) are generally defined as those with sizes of 1 μ m to

5 mm, with any shape being either solid state or polymeric matrix (insoluble in water) (Frias and Nash, 2019). On the other hand, plastics with 1–1000 nm in size are regarded as nanoplastics, which can show colloidal characteristics (Gigault et al., 2018) and are considered more toxic to aquatic organisms (Gigault et al., 2021). In addition to their wide distribution in the ocean, MPs can gradually accumulate in seafood (Piyawardhana et al., 2022) and terrestrial and inland water systems (Chae and An, 2018). Since many plastics are difficult to degrade in the natural environment, MPs

recovery and remediation strategies have attracted increasing global attention over the past years (Eskandarloo et al., 2017).

MPs can be categorized based on their sources (Sharma et al., 2021). Primary MPs are manufactured in micro-scale ranges and are widely used in granulates, pellet manufacturing, cosmetics, and other personal care products, such as toothpaste and scrub (Wu et al., 2019). Secondary MPs result from their degradation in terrestrial and aquatic environments, such as discharged plastics, fishing nets, urban wastes, and synthetic fibers released from textiles and wastewater treatment plants (WWTPs) effluents (Anderson et al., 2016). The WWTPs contribute significantly to MPs abundance in the environment (Pivokonsky et al., 2018). Researchers have observed that about 35 % of MPs found in the ocean are microfibers derived from washing and laundry (Ziajahromi et al., 2016). In addition, wastewater commonly contains 70 % textile fibers from wastewater treatment plants (WWTPs) (Cristaldi et al., 2020). In addition to fibers, WWTPs use other types (films, fragments, pellets, and granules) of MPs based on their shape. Among them, fibers and fragments are common shapes, and fiber particles are difficult to remove due to their considerable length and width (Sol et al., 2020). The MPs are then transported from their sources, like WWTPs, to bio-solids at a significant amount in an aquatic environment (Wei et al., 2019; Wang et al., 2019a; Chen et al., 2022).

Also, MPs are able to act as a carrier for different contaminants like polybrominated diphenyl ethers (PDBEs) (Tanaka et al., 2015), heavy metals (Tang et al., 2020), polychlorinated biphenyls (PCBs) (Velzeboer et al., 2014), polycyclic aromatic hydrocarbons (PAHs) (Lo et al., 2019), dichlorodiphenyltrichloroethane (DDT) (Wang et al., 2018a), poly-fluoroalkyl substances (PFAS) (Scott et al., 2021) and pharmaceuticals (Ateia et al., 2020). MPs carried with these additional contaminants cause synergistic toxic effects and pose a significant threat to the organism and human health (Barboza et al., 2018a). Recent toxicity studies reported that MPs <150 µm in size can pass the cell membrane, translocate into organs, and cause serious health problems such as inflammatory impacts, oxidative stress, defects in offspring, and metabolic disorders in animals (Rahman et al., 2021). Other studies on human cell lines demonstrated that MPs exposure results in inflammatory reactions, limits cell growth, and changes in cell morphology and gut microbiota (Kutralam-Muniasamy et al., 2022). Additional studies reported that MPs could be transported throughout the body by blood circulation (Leslie et al., 2022). Moreover, MPs <2.5 µm generally remain in the lungs, which can pass through the respiratory wall and cause different lung diseases after exposure (Prata, 2018). MPs were also observed in various human biological samples such as saliva, liver, colectomy specimens, sputum, breastmilk, and feces (Kutralam-Muniasamy et al., 2022). In marine and freshwater organisms, MPs ingestion can damage the filtering mechanism of zooplankton, mussels, and other organisms, and generate inflammation, neurotoxicity, and genotoxic effects (Prokić et al., 2019).

To degrade or remove MPs from the environment, several physical, chemical, and biological treatment technologies have recently been adopted (Ahmed et al., 2021). Some physical treatments, such as coagulation and filtration, dynamic membrane filtration, agglomeration, electrocoagulation, advanced oxidation process (AOPs), primary sedimentation/grit chamber, adsorption removal technique, magnetic filtration oil film extraction, froth flotation, and density separation methods are widely used in WWTPs due to their efficacy, low cost, and low energy requirement. In addition, biological removal technology for MPs degradation has been focused on recently (Kushwaha et al., 2021). Researchers observed that some organisms, such as bacteria, fungi, and algae, can degrade MPs (Yuan et al., 2020). MPs biodegradation ability and associated degradation effects (e.g., degradation rate, weight loss) are significantly dependent on microbial characteristics and environmental factors (e.g., pH, temperature, and strain stability) (Yuan et al., 2020).

It is clear that MPs have been a frontier research area of interest for researchers. According to the Scopus database (<https://www.scopus.com>) by 17th August 2022, over 11,000 publications were found related to the search term *microplastics** between 2010 and 2022. Strikingly, the total

number of papers related to *microplastics** published in 2020 ($n = 2099$), 2021 ($n = 3011$), and 2022 ($n = 3364$) are over five times greater than the total number of articles published between 2010 and 2019 ($n = 1629$, Fig. 1). So far, there have been some reviews on the Toxicity of MPs in aquatic environments (e.g., Ateia et al., 2022; López et al., 2022; Luo et al., 2022b). In addition, many reviews are on the remediation and reduction of MPs from aquatic environments (Lu et al., 2023; Gong et al., 2023). For instance, Padervand et al. (2020), Katare et al. (2021), Zhang et al. (2021a), and Bhatt et al. (2021) conducted studies regarding the toxicity and remediation techniques of MPs. The interaction of different contaminants, different types of MPs toxicity, and some remediation methods were described by Ahmed et al. (2021). Mao et al. (2022) reviewed MPs toxicity in aquatic organisms with various nutrition levels. Although studies on MPs Toxicity in aquatic environments and MPs remediation are increasing, comprehensive reviews on MPs' Toxicity in aquatic environments and associated removal and remediation strategies are rare (Zhang et al., 2021b; Sussarellu et al., 2016). To fill this gap, this study comprehensively compiled and presented up-to-date information from recent studies regarding MP toxicity in aquatic environments along with notable remediation and reduction strategies for MPs removal from the environment. Therefore specific objectives are:

- To present a short bibliometric analysis of research papers related to MPs toxicity in aquatic environments and reductions/remediation of MPs from the environment. This bibliometric analysis will improve our understanding of the general structures in MP research, research trend, hot topics, and research gaps related to MP research.
- To determine the Toxicity of MPs in aquatic organisms among fish, phytoplankton, zooplankton, and polychaetes across different trophic levels. Particular focus was given to reviewing up-to-date research on the Toxicity of additives (both organic and inorganic) and the effects of MPs on aquatic organisms.
- To compile available MPs removal strategies, including both biological and engineered/chemical methods, including eleven treatments, i.e., dynamic membrane, filtration, coagulation, agglomeration, electrocoagulation, advanced oxidation process, primary sedimentation/Grit chamber, adsorption removal technique, magnetic filtration, oil film extraction, froth flotation, and density separation.
- To reveal research gaps and future research directions for MPs.

2. Literature search methods and a short bibliometric analysis

For this review, literature data were extracted using a broad combination of keywords (e.g., “microplastics* + toxicity”; “microplastics* + freshwater + toxicity”; “microplastics* + removal”; “microplastics* + reduction”; “microplastics* + remediation”) in several databases, including Google Scholar, Scopus, Pubmed and Web of Science. A total of 500 papers, mainly from the last five years, were evaluated, of which 340 papers are presented in this review.

A short bibliometric analysis was performed to evaluate the general structure of MPs research, especially related to their Toxicity in aquatic environments and their removal/remediation strategies (Zhang et al., 2020b). Specifically, the bibliometric analysis included: i) analysis of a network of collaboration established by co-authorship, ii) analysis of the co-authorship map of countries, and iii) keyword evolution analysis. Bibliometric analysis was performed using VOSviewer (version 1.6.13; van Eck and Waltman, 2010). Database of Scopus was accessed on 17th August 2022; search terms included title, abstract, and keywords (1980–2022). The following two different search strings were used for bibliometric review and keyword evolution analyses:

2.1.1. Search string for literature related to aquatic toxicity of MPs

For this purpose, search strings, such as “(microplastics* AND toxicity*) AND (aquatic* OR marine* OR freshwater*)” were used, resulting in 1984 documents.

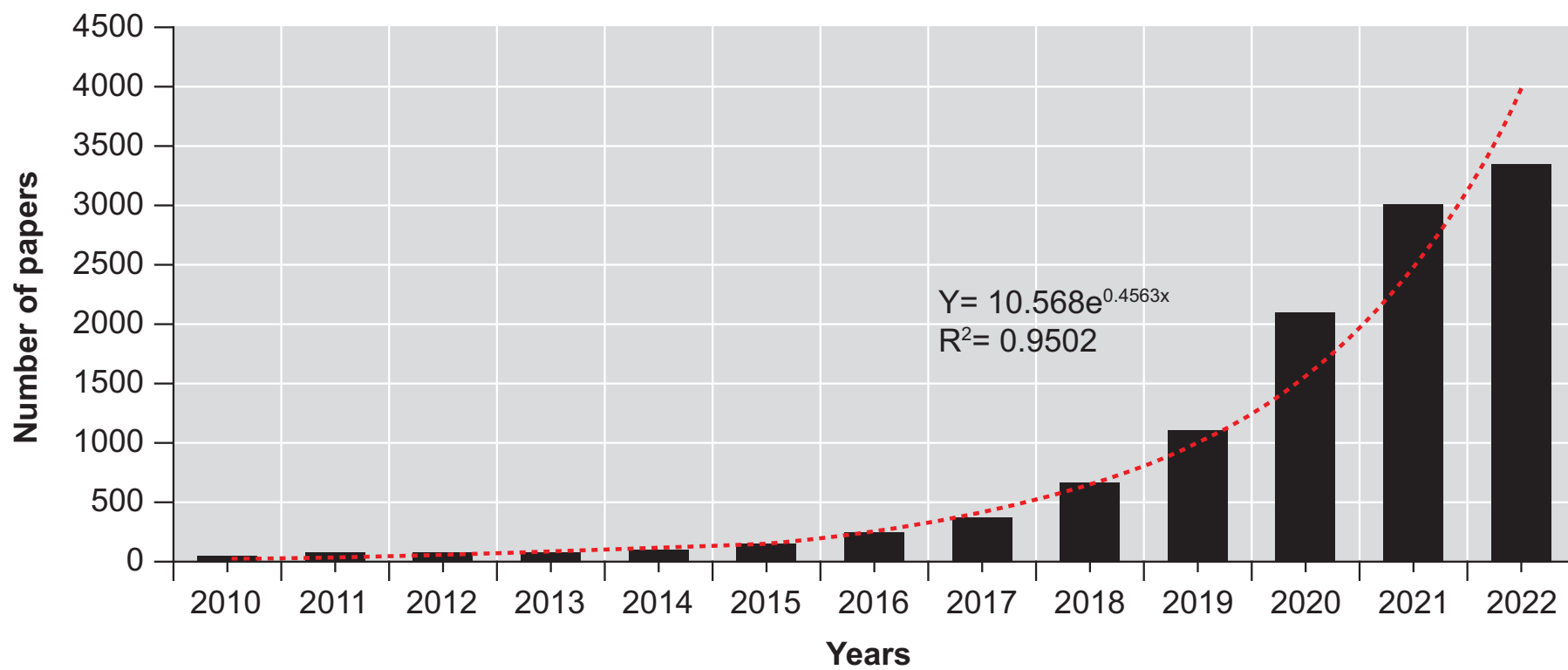


Fig. 1. The number of published papers per year (2010–2022) related to the MPs' research worldwide (data source: Scopus).

2.1.2. Literature related to MPs remediation and reduction strategies

In this case, we employed search strings like “microplastics* AND (removal* or remediation* or reduction*).” These search terms resulted in 3444 documents. Our search terms related to the reduction and remediation of MPs from the environment revealed 3444 documents.

2.2. Analysis of the contribution of authors

Over 4700 authors contributed to the field of the toxicity of microplastics in aquatic ecosystems, with 70 authors published at least 10 documents (SF 1). Results indicated that Wang J (53 documents), Li Y (authored 43 documents), and Zhang W (26 documents) were the most prolific authors (SF 1a). In the field of microplastic remediation in the

environment, 3264 authors were identified, with 34 authors published at least 10 documents. Authors such as Zhang Y (40 documents), Li Y (30 documents), Wang H (25 documents), Wang Y (22 documents), and Li J (15 documents) were found to be the most prolific authors in the field (SF 1b).

2.3. Analysis of the authorship map by countries

In the present study, a network analysis of the research on collaboration between different countries was carried out using complete counting methods. The minimum number of documents per country was set to 10. For this analysis, a link between two countries indicates a co-authorship network between authors of the institute in two countries (Fig. 2a and b). Further, a greater number of countries are involved in collaborative

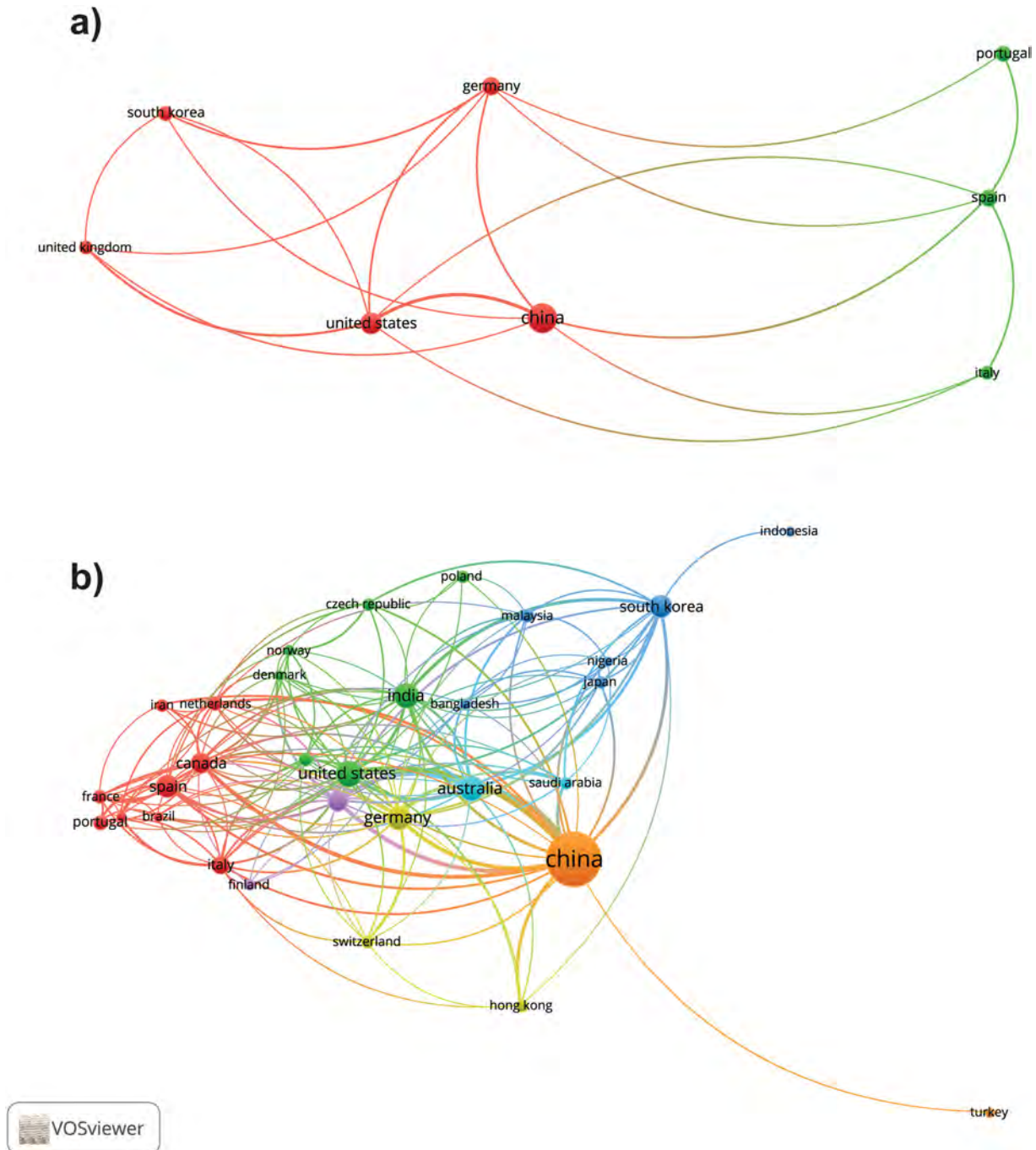


Fig. 2. VOSviewer co-authorship map of countries/regions involved in a) research related to the toxicity of the MPs in aquatic environments and b) research related to the remediation/reduction of the MPs in aquatic environments.

publications related to MPs remediation research (Fig. 2b) than in MPs toxicity in aquatic organisms (Fig. 2a). Our analysis (Fig. 2) shows that China, the United States, Germany, Spain, and Italy are among the top countries involved with many collaborative publications in MP research.

2.4. Keyword evolution analysis

Keyword evolution analysis was carried out to understand the research's major domain and elucidate recent research trends. Based on this analysis, future research directions can be recommended (Billah et al., 2022b; Zhang et al., 2020a). This analysis also provides views of essential knowledge elements, trends, research gaps, and frontlines in the subject areas (Zhang et al., 2020a). The appearance of a new keyword shows that our knowledge reaches a new domain. From these papers, keyword evolution analysis confirmed that 18 keywords were cited at least 15 times (Fig. 3). Most widely used keywords, such as combined toxicity, bioaccumulation, heavy metals, biofilm, and zebrafish, were considered research frontlines. Therefore, much research is needed on these issues. Based on the results in Fig. 3, three major research domains were confirmed:

- Acute and chronic toxicity of MPs: as indicated with the keywords *toxicity*, *microalgae*, *daphnia magna*, and *zebrafish*
- Bioaccumulation and biomarker studies: ecotoxicity and bioaccumulation studies of MPs as indicated with the keywords such as *bioaccumulation*, *zebrafish*, *biomarkers*, and *oxidative stress*

- Combined toxicity studies: as indicated with the keywords such as *heavy metals* (examples of the combined Toxicity of MPs with heavy metals are presented in Table 1)

According to our analysis, *zebrafish* and *Daphnia magna* are the most cited organisms related to MPs Toxicity (Fig. 3). These results can be explained by the fact that zebrafish (*Danio rerio*) has compelling advantages, including easy handling and keeping protocols in the laboratory, short life cycle, and high fecundity (100–200 eggs), making them successful model organisms in the ecotoxicology (Lu et al., 2016). The striking advantage of zebrafish in assessing MPs toxicity includes using embryos and larvae that can rely on yolk sacs for seven days. Thus, it is cost-effective to study the response of early developmental stage vertebrates to MPs toxicity using this species. Further, lower trophic chain organisms, such as copepod *Daphnia magna*, have long been considered prevalent model organisms in the field of ecotoxicology (Jonczyk and Gilron, 2005). As depicted in Fig. 3, the frequent citations for this copepod species in the scope of aquatic Toxicity of MPs are not unexpected (Table 1; Jemec et al., 2016).

Moreover, once in the aquatic environment, MPs can interact with organisms (e.g., fish), which leads to Toxicity to organisms, including oxidative damage (Table 1). As shown in Fig. 3, oxidative stress has received considerable attention in evaluating the Toxicity of MPs (Hoyo-Alvarez et al., 2022).

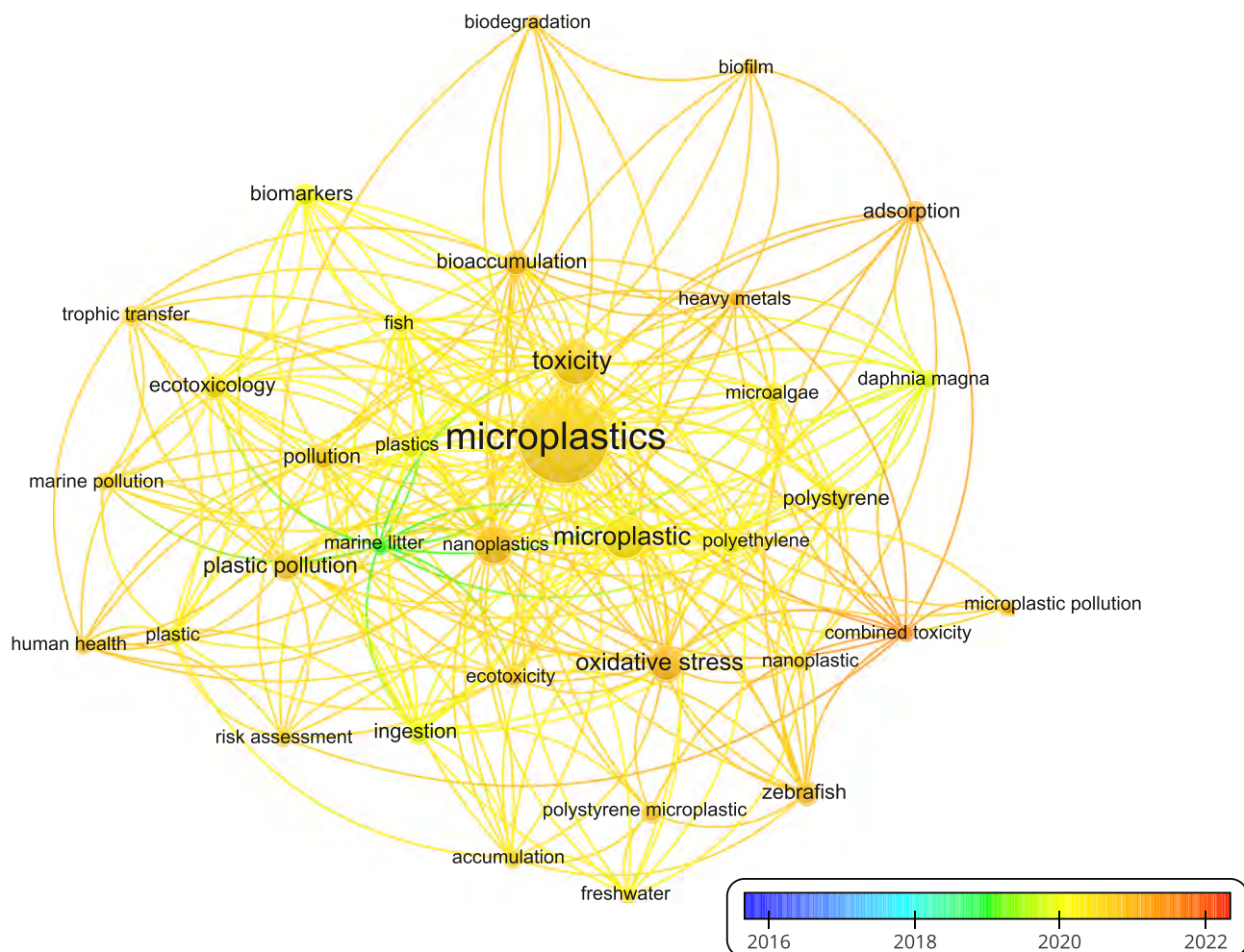


Fig. 3. VOSviewer overlay map regarding co-occurrences of author keywords analysis and research trend of toxic effects of MPs in various aquatic organisms. The circle size denotes the keyword frequency, and the colour reflects the average publication according to the year consideration.

Table 1

Experiments of the effects of MPs along with other chemical additives in different fish species.

Fish species	MPs			Contaminants	Concentration	Exposure route (via)	Exposure time	Toxicological/biological effects in fish	References
	Composition	Size (µm)	Concentration						
Green chromide (<i>Etroplus suratensis</i>)	PVC	80.72	N/A			Dietary	10 days	Antioxidant enzymes such as superoxide dismutase and catalase were impacted by MP exposure; red and white blood cells were decreased; some changes were observed in growth, acetylcholine esterase function, behaviour, and hematological biomarkers.	Vijayaraghavan et al. (2022)
Gilt-head bream (<i>Sparus aurata</i>)	LDPE	200–500	10 % in feed			Dietary	120 days	Although MP retention was absent in the 1st month, fish showed high feeding time.	Alomar et al. (2021)
Red tilapia (<i>Oreochromis niloticus</i>)	PS	0.3,5 and 70–90	100 µg/l			Dietary	14 days	Some vital occurrences like oxidative stress, ingestion, neurotoxicity, and metabolic changes were observed; 5 µm PS particles inhibit the acetylcholinesterase function.	Ding et al. (2020)
Ambon damselfish (<i>Pomacentrus amboinensis</i>)	PS	200–300	1.04 g/ml			Dietary	4 days	MP exposure changed behavioral activity and had a significant impact on survivability.	McCormick et al. (2020)
Glassfish (<i>Ambassis dussumieri</i>)	PE, PVC and PS	250–1000	0.01 g per tank			Dietary	95 days	MP exposure reduced growth performance and increased juvenile mortality;	Naidoo and Glassom, 2019
Zebrafish (<i>Danio rerio</i>)	PS and PP	4–100	10 µg/l			Water	21 days	Mast cells at the digestive epithelium, vacuolization, and cilia defects were increased; the volume of mucus in the intestine was decreased; Proteobacteria and Aeromonas were raised in the gut.	Qiao et al. (2019)
Silver barb (<i>Barbodes gonionotus</i>)	PVC	0.1–1000	0.2–1 mg/l			Water	96 h	Average thickness and proximal digestive tract increased at 73.4 % and 29.1 %, respectively. Trypsin and chymotrypsin functions were considerably increased.	Romano et al. (2018)
Goldfish (<i>Carassius carassius</i>)	PS, PA, and EVA	<5000	15–76 items/fish			Dietary	6 weeks	A significant amount of MPs were observed in the digestive tract; Fish weight was reduced by 17.5 to 21.5 %.	Jabeen et al. (2018)
African catfish (<i>Clarias gariepinus</i>)	PE	<60	50–500 µg/l			Water	96 h	High-density lipoprotein of blood was decreased considerably; the ratio of plasma albumin and globulin was increased	Karami et al. (2016)
Zebrafish (<i>Danio rerio</i>)	PS	0.07–20	20–2000 µg/l			Water	3 weeks	The abundance of MPs occurred in fish grills, liver, and gut; Infiltration, necrosis, and lipid droplets were found in fish hepatocytes; the Function of catalase and superoxide dismutase was increased.	Lu et al. (2016)

(continued on next page)

Table 1 (continued)

Fish species	MPs			Contaminants	Concentration	Exposure route (via)	Exposure time	Toxicological/biological effects in fish	References
	Composition	Size (µm)	Concentration						
European sea bass (<i>Dicentrarchus labrax</i>)	PVC	<300	0.1 % plastics			Dietary	90 days	Widening of lamina propria, shortening and swelling of villi, goblet increase, and enterocyte vacuolation occurred in 67 % of fish; Extreme inflammatory alterations, circulatory changes, hyperplasia, and morphological changes were observed at 50 %.	Peda et al. (2016)
Jacopever (<i>Sebastes schlegelii</i>)	PS	15	10 ⁶ items/l			Water	21 days	The abundance of MPs occurred in fish gills, liver, and digestive tract; Feeding time was increased; Movement rate and swimming speed decreased notably;	Yin et al. (2018)
Gilthead seabream (<i>Sparus Aurata L.</i>)	PVC	40–150	100–150 mg PVC items/kg feed			Dietary	30 days	Fish serum was increased in aspartate aminotransferase, creatine kinase, albumin, and globulin by 153.2 %, 368.5 %, 31.1 %, and 69.0 %, respectively; Fish cell respiration burst activity and phagocytic capability were notably increased.	Espinosa et al. (2017)
Common goby (<i>Pomatoschistus microps</i>)	PE	450–500	100 items/l			Water	96 h	Fish were substantially less effective and efficient predators.	de Sá et al. (2015)
Combined effect Blackspot seabream (<i>Pagellus bogaraveo</i>)	Proprietary polymer	1–5		Copper	10–810 µg/l	Water	9 days	MPs and copper can have an adverse effect on the fitness of fish larvae by increasing mortality and inducing oxidative stress, lipid peroxidation, and neurotoxicity.	Santos et al. (2022)
Rainbow trout (<i>Oncorhynchus mykiss</i>)	PS	21.89–466	30 or 300 µg/l	Chlorpyrifos	2 or 6 µg/l	Water	96 h	High levels of PS-MP and Chlorpyrifos caused significant histopathological abnormalities in the fish gut, including extensive necrosis, inflammatory cell infiltration, and villi tip loss; PS-MP enhances the toxicity in the presence of Chlorpyrifos in the gill tissue.	Karbalaei et al. (2021)
Red tilapia (<i>Oreochromis niloticus</i>)	PS	5	10 µg/l	sulfamethoxazole (SMX) and β-blocker propranolol (PRP)	50 µg/l	Water	14 days	Fish suffered from severe stress after exposure to PS-MPs and the selected drugs; Co-exposure to MPs and SMX induced the reduction of cytochrome P450 enzymes activity.	Huang et al. (2021a, 2021b)
Marine medaka (<i>Oryzias latipes</i>)	PS	10	0,2,20,200 µg/l	Phenanthrene	50 µg/l	Water	28 days	Decreased hatchability, delayed hatching, and reduced growth occurred due to high levels of MPs (20 and 200 µg/l) ingestion, whereas Phenanthrene caused malformations and inhibited proper hatching in larvae.	Li et al. (2020a)

Table 1 (continued)

Fish species	MPs			Contaminants	Concentration	Exposure route (via)	Exposure time	Toxicological/biological effects in fish	References
	Composition	Size (µm)	Concentration						
Red tilapia (<i>Oreochromis niloticus</i>)	PS	0.1	1–100 µg/l	Roxithromycin	50 µg/l/l	Water	14 days	Bioaccumulation of roxithromycin in fish was considerably accelerated by the presence of MPs; MPs reduced fish roxithromycin neurotoxicity; Fish liver cytochrome P450 enzyme activities changed after being exposed to both MPs and roxithromycin at the same time.	Zhang et al. (2019)
European sea bass (<i>Dicentrarchus labrax</i>)	Fluorescent red microspheres	1–5	0.26–0.69 mg/l	Mercury	0.01–0.016 mg/l	Water	96 h	After being exposed to MPs and mercury combinations, fish's swimming speed and resistance time decreased by a maximum of 80–87 % and 52–64 %, respectively, compared to the controls; After exposure, fish began to show erratic and lethargic swimming.	Barboza et al. (2018b)
Discus fish (<i>Symphysodon aequifasciatus</i>)	PS	32–40	50–500	Cadmium	50 µg/l	Water	30 days	MPs decreased bioaccumulation of cadmium and glutathione; After being exposed to MPs and cadmium simultaneously, fish showed enhanced activity of catalase, lysozyme, acid phosphatase, and alkaline phosphatase; Fish had a higher protein carboxyl content.	Wen et al. (2018)
Zebrafish (<i>Danio rerio</i>)	PVC	200–250	400 mg/l	Phenanthrene & 17 a-ethinylestradiol	Phenanthrene: 0.1–0.5 mg/l; 17 α-ethinylestradiol: 0.001–1 µg/l	Water	96 h	MPs reduced phenanthrene & 17 a-ethinylestradiol by a maximum of 33 % and 48 %, respectively.	Sleight et al. (2017)
Common carp (<i>Cyprinus carpio</i>)	PE		1–2 mg/l	Paraquat	0.2–0.4 mg/l	Water	24 h	After co-exposure to MPs and paraquat, glucose and creatinine levels in fish increased by 31.3–50 % and 3–3.5 folds, respectively; Following co-exposure, fish contained less total protein, globulins, cholesterol, and triglycerides by 24–36 %, 44–84 %, and 32.6–50.9 %, respectively. Activities of aspartate aminotransferase, alkaline phosphatase, and creatine phosphokinase were significantly increased.	Nematdoost Haghi and Banaee (2017)
Common goby (<i>Pomatoschistus microps</i>)	PE plastic microspheres	1–5	0.184 mg/l	Cefalexin	1.3–10 mg/l	Water	96 h	Predatory performance was reduced after exposure to MPs and cefalexin; MPs increased cefalexin toxicity, and	Fonte et al. (2016)

(continued on next page)

Table 1 (continued)

Fish species	MPs			Contaminants	Concentration	Exposure route (via)	Exposure time	Toxicological/biological effects in fish	References
	Composition	Size (μm)	Concentration						
Common goby (<i>Pamatoschistus microps</i>)	PE	1–5	12 mg/l and 0.012 mg/l.	Chromium (VI)	5.6–28.4 mg/l	Water	96 h	the combination of these substances decreased juvenile fitness. Simultaneous exposure reduced the predatory performance and inhibited acetylcholinesterase activity.	Luís et al. (2015)

PS: Polystyrene; PVC-Polyvinyl chloride; PP-Polypropylene; PE-Polyethylene, LDPE: Low-density polyethylene.

Our search terms related to the reduction and remediation of MPs from the environment revealed 3444 documents. Analysis of the author keywords analysis from these papers (at least 15 times cited) revealed 20 keywords related to the remediation of MPs from aquatic environments (Fig. 4); the most cited five keywords included *microplastics* ($n = 448$), *microplastic* ($n = 98$), *removal* ($n = 47$), *nanoplastics* ($n = 53$), and *wastewater* ($n = 53$). Importantly, keywords, such as *adsorption*, *removal efficiency*, *coagulation*, and *nanoplastics*, seemed to be the research frontiers in MPs (Fig. 4). The keyword analysis reflected two main research themes:

- i) Keywords such as *microplastics*, *polystyrene*, *nanoplastics*, and *plastic pollution* indicated the research papers from the characterization, abundance, and diversity of MPs in environmental matrices

- ii) The remaining keywords (e.g. *coagulation*, *adsorption*, *removal efficiency*) were related to the removal or reduction strategies. For example, coagulation is considered an appreciable strategy for MP reductions in water plants, thus becoming a hot topic in recent studies (e.g. Patil et al., 2022)

It is further inferred from our analysis that *adsorption*, *coagulation*, and *WWTPs* are among the recently cited keywords indicated by the light-yellow colour in Fig. 4, implying the marked increasing recent interest in the removal of MPs with the adsorption and coagulation techniques from WWTPs. Thus, more research is needed to investigate the efficiency of these techniques in removing MPs from WWTPs.

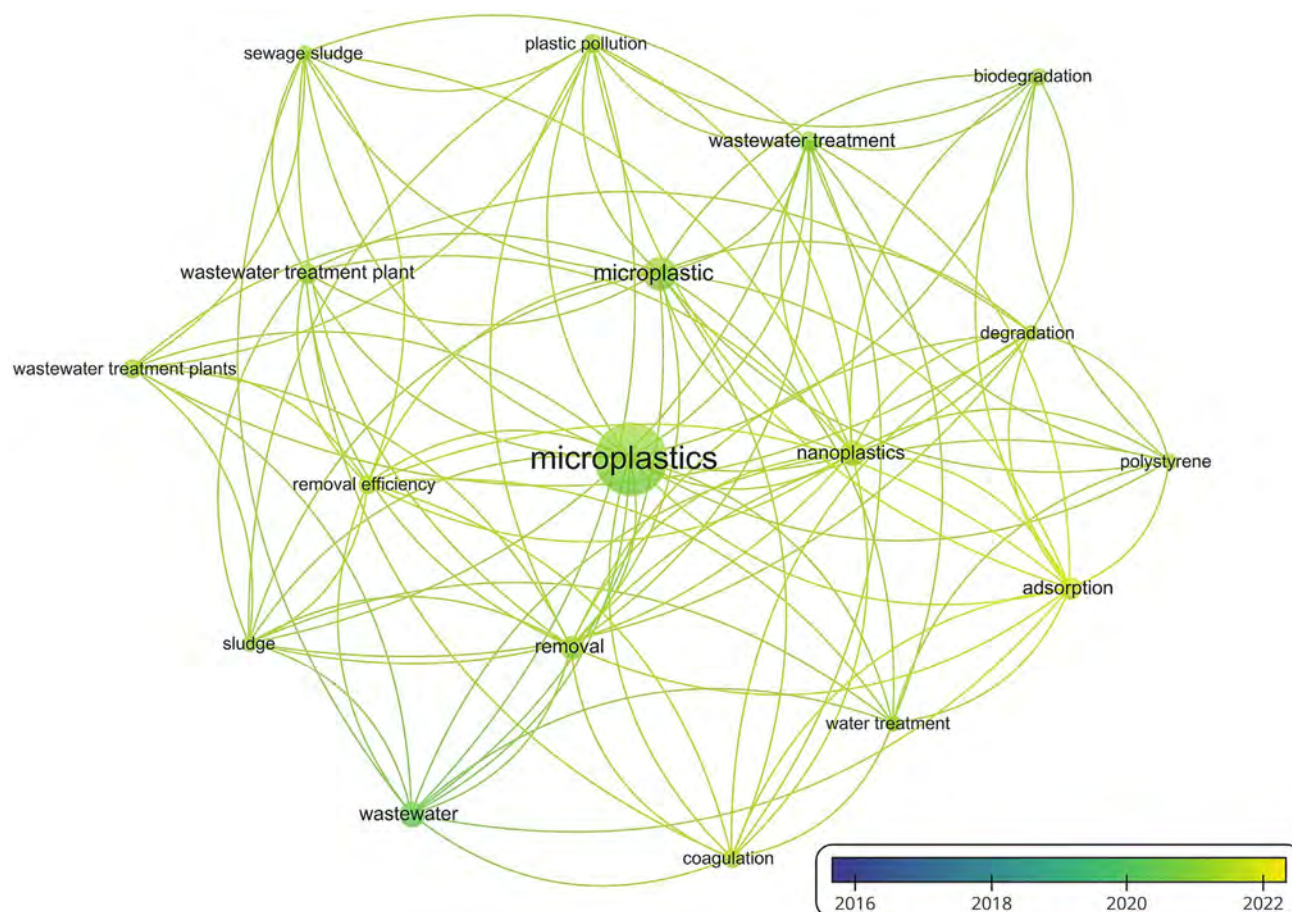


Fig. 4. VOSviewer overlay map regarding co-occurrences of author keywords analysis and research trend of remediation technique of MPs in the aquatic environment. The circle size denotes the keyword frequency, and the colour reflects the average publication according to the year consideration.

3. Microplastic toxicity

3.1. Toxicity induced by the chemical composition of MP

The possible Toxicity of MPs emerges from unreacted oligomers, monomers, and other chemical additive substances used in plastic (Thompson et al., 2004). As MP concentration residues reach a certain threshold, monomers and oligomers can migrate from food packaging materials, and humans take in MPs through various ways, including air, skin, and ingestion of food (Piringer and Baner, 2008). Residual polystyrene in food items can cause tremendous health problems where living tissues can absorb epoxy resins comprised of bisphenol A, interfering with cell division function. Chemical additives have been extensively used in polymer formation to upgrade product performance. Some functional additives like heat stabilizers, antioxidants, plasticizers, and colorants fillers are notable among them. Such additives can cause synergistic Toxicity along with the MP polymers (Luo et al., 2022a). For instance, researchers reported that a significant range of phthalates from baby bottles was 50–150 µg/kg of food materials after contacting for 120 min at 70 °C (Simoneau et al., 2012). In addition, the release level of bisphenol A from different food packaging plastic materials was investigated in the range of 100–800 ng/l (Padervand et al., 2020). Under specific conditions, the concentration was growing to µg/l scale in some phthalates (Fasano et al., 2012). These additives are to be quickly and easily released, and most of them do not have a chemical bonding ability with bulk plastic structures. Such released additives cause more risk to human health rather than raw MPs (Rist et al., 2018). However, the leaching of chemical substances significantly depends on the surrounding media of MPs accumulation and the exposure routes. Hahladakis et al. (2018) reviewed the migration, release, fate, and potential environmental toxic impacts of additive compounds on health. They concluded that potential Toxicity could arise from the additives due to inappropriate and uncontrolled discharge and recycling processes. In addition, some chronic health problems could occur by releasing volatile substances like benzene, ethylbenzene, toluene, methylene chloride, and styrene from plastic debris (Andrady, 2017).

3.2. Toxicity induced by MPs' physical properties

The physical properties of MPs include colour, shape, and size, and every property can exhibit different adverse effects (Ma et al., 2020). For example, specific surface areas, a colloidal property, have a significant contribution to the extent of MPs toxicity. High-specific surface areas help to absorb hydrophobic pollutants and other metal contents in aquatic environments and subsequently transport this to different habitats (Setälä et al., 2014). Based on biomarker response, juvenile African catfish revealed considerable tissue changes in the liver and brain for the effect of phenanthrene-loaded low-density PE glycol MPs (Karami et al., 2016). The shape and texture of MPs also influence the Toxicity and absorption capacity. Au et al. (2015) reported that polypropylene components cause more toxic effects in freshwater amphipod, *Hyalomma azteca*, than spherical polyethylene. Considering the size, MPs smaller than 40 µm caused the negative impacts of PLA and PUR, but smaller PVC particles (mainly 20 µm) had no effect on *daphnia magna* (Zimmermann et al., 2020). However, according to Sjollem et al. (2016), polyvinyl chloride (PVC) had detrimental impacts on growth, chlorophyll content, and photosynthesis at sizes of 1 µm but not for 1 mm. Similarly, nano-sized PS of 0.05 µm in *D. magna* caused a more significant decrease in cell density than 0.5 and 6 µm (Zhang et al., 2017). Fiber MPs that are present in the gut for a long time alter the food processing capacity, significantly impacting sub-lethal endpoints (Au et al., 2015; Padervand et al., 2020).

3.3. Toxicity induced by MPs and chemical additives

The Toxicity of additives, along with MPs, has been vividly investigated. Nobre et al. (2015) used assays of elutriate and pellet-water interface to simulate the leaching of chemical additives into the water column and

interstitial water and investigated the effects of raw MPs on the development of *Lytechinus Variegatus* embryos. They also reported that raw MPs showed more Toxicity, accelerating anomalous embryonic development in 58.1 % and 66.5 %, considering the former and latter investigation methods, respectively. Moreover, MPs act as a vector for other coexisting pollutants causing detrimental health problems (Zhao et al., 2020). For instance, MPs associated with organic compounds such as phenanthrene, 4,4'-DDT, and PBDEs can magnify the bioavailability in the food chain and eventually enter the human body (Wardrop et al., 2016). It was reported that the adsorption of PAHs on MP particles was essentially in the same order of magnitude as that for metals, and the adsorption capacity for heavy metals was marginally higher (Gao et al., 2019). The high level of heavy metals in the MPs' surface indicates that MPs can act as an alternative source of heavy metals in aquatic environments. The most dangerous additive types are phthalates, lead compounds used as heat stabilizers, and brominated flame retardants (Sendra et al., 2021). Some brominated flame retardants, including PBDEs, have structural similarities to polychlorinated biphenyls (PCBs), well-known environmental pollutants that accumulate in aquatic animals' adipose tissues (Darnierud, 2003). They can have neurotoxic effects and change the proper function of thyroid hormone (D'silva et al., 2004). Phthalates are chemicals with estrogenic properties that can impact in the reproductive system and endocrine function (Mariana et al., 2016). Potential toxic effect in fin whales has already been observed due to the high-level of phthalates ingestion (Guzzetti et al., 2018). Moreover, in *Balaenoptera physalus*, it has been demonstrated that exposure to high concentrations of phthalates may change antioxidant defense and other systems that prevent cell damage caused by oxidative stress and potential abnormalities of endocrine function (Fossi et al., 2016). Studies have revealed that phthalate plasticizers, particularly those with smaller molecular weights, are acutely and chronically hazardous to a wide range of aquatic microorganisms, invertebrates, algae, and fish (Warren et al., 2003; Roy, 2021). Also, pharmaceuticals may become more bioavailable after being consumed by MPs, which can accelerate their bioaccumulation and biomagnification via the food web and regulate their harmful effects (Santos et al., 2021). However, there was limited consensus regarding the impact of MPs on exposure to the coexisting contaminants (Wang et al., 2021). To better understand this, more studies are necessary, especially for the functional role of joint Toxicity of coexisting pollutants. The following sections present the toxic effects of some common chemical additives:

3.3.1. Effects of MPs and pharmaceutical additives

Pharmaceutical compounds generally exhibit pH-dependent sorption on various MPs (Wang et al., 2022). According to previous studies, pharmaceutical compounds generate cationic species at low pH that increase the electrostatic attraction to MPs surface (Atugoda et al., 2021). On the other hand, neutral as well as zwitterionic pharmaceutical compounds interact with non-polar MP surfaces through van der Waals and hydrophobic forces that accelerate the sorption during high pH (Atugoda et al., 2021). After forming anionic species, the electrostatic repulsive interaction with MPs surface can be enhanced, and the sorption rate can be reduced (Atugoda et al., 2021). By dissolving various salts, such as NaCl, MgCl₂, CaCl₂, and Na₂SO₄, to stimulate the ionic strength of natural seawater, the effect of ionic strength on contaminant sorption dynamics has been studied (Liu et al., 2019d). For toxicological concern, the impact of pharmaceuticals and MPs on microalgae *Tetraselmis chuii* were closely investigated by Prata et al. (2018). They observed that pharmaceuticals combined with MPs exerted more Toxicity in *T. chuii* compared with pharmaceuticals alone. The possible reason is that the interaction between microalgae and MPs facilitated the uptake of other contaminants by the cell wall (Prata et al., 2018). They also reported that the EC₅₀ values for the various combination were 125 g/ml (MP-Procaimide) and 11 g/ml (MP-Doxycycline). In addition, a minimum inhibitory concentration of 41.5 g/ml without pharmaceuticals was observed. These findings demonstrate how different mixtures can have various harmful

behaviors. Similarly, [Qu et al. \(2020\)](#) conducted a combined study using methamphetamine and PS and observed that the endpoint factor growth's EC₅₀ was 0.32 µg/ml. They also reported that the adverse effect of methamphetamine in *Chlorella pyrenoidosa* increased due to the presence of MPs. The result also showed that the oxidative damage shifted from 19.9 to 36.8 nmol mg/protein, apoptosis induced almost 2.17 folds, and EC₅₀ shifted from 0.77 to 0.32 µg/ml ([Table 2](#)). Another combined impact of MPs and chlorpyrifos (CPF) on the algae, *Isochrysis galbana*, was investigated by [Garrido et al. \(2019\)](#). They demonstrated that the IC₅₀ value was 2.57 µg/ml, and the EC₅₀ value for single MP exposure was more significant compared to single and combination scenarios. Meanwhile, higher risk at low concentration was observed

with a lower EC₅₀ value during exposure to combined MPs. Therefore, clarifying the impact of MPs and organic matter in algae is crucial.

Considering all the issues, more attention is urgent regarding the mixed effect of pharmaceuticals, as such compounds are ubiquitous and exhibit Toxicity at a low concentration. For example, some drug groups, such as doxycycline and anesthetic procainamide are pervasively used in animals and humans that are widely found in aquatic ecosystems ([Parsai et al., 2022](#)). Such drugs induce more Toxicity to microalgae being at a low concentration ([Prata et al., 2018](#)). As these types of compounds' Toxicity greatly depend on the type and size of MPs, more studies are required in this direction ([Sheng et al., 2021](#)). Moreover, such studies need specific reference points to estimate ecological risk correctly.

Table 2

Toxic effects of single MPs, combined impact of MPs, and different contaminants on various aquatic microalgae. EC₅₀ denotes 50 % effective level; IR₅₀ denotes 50 % growth retardation, Cd: Cadmium; Cu: Copper; CPF: Chlorpyrifos.

Microalgae species	Polymer type	Assessment parameter	Respective value	Endpoints	References
<i>Chlorella vulgaris</i>	Virgin PVC	IR	IR _{maximum} = 30.03 % (150 µm, 10 mg/l)	■ Growth	Wang et al. (2021)
	Aged PVC		IR _{maximum} = 16.72 % (150 µm, 10 mg/l)		
<i>Chlorella vulgaris</i>	Virgin PS	IR	IR _{maximum} = 36.84 % (250 µm, 10 mg/l)	■ Growth	Wang et al. (2021)
	Aged PS		IR _{maximum} = 29.10 % (250 µm, 10 mg/l)		
<i>Chlorella pyrenoidosa</i>	PS-COOH (1 µm)	EC ₅₀	EC ₅₀ > 50 µg/ml	■ ROS formation ■ Growth retardation	Zhang et al. (2020a)
<i>Chlorella pyrenoidosa</i>	PS (Fe)-COOH (1 µm)	EC ₅₀	EC ₅₀ > 50 µg/ml	■ ROS formation ■ Growth retardation	Zhang et al. (2020a)
<i>Chlorella pyrenoidosa</i>	PS-NH ₂ (1 µm)	EC ₅₀	EC ₅₀ > 50 µg/ml	■ ROS formation ■ Growth retardation	Zhang et al. (2020a)
<i>Chlamydomonas reinhardtii</i>	Virgin PVC	EC ₅₀	EC ₅₀ = 104.93 µg/ml	■ Growth	Wang et al. (2020a)
	UV-age PVC		EC ₅₀ = 63.66 µg/ml		
<i>Raphidocelis subcapitata</i>	PVC	EC ₅₀	1.62 %	■ Growth	Capolupo et al. (2020)
<i>Skeletonema costatum</i>	PVC	EC ₅₀	34.6	■ Growth	Capolupo et al. (2020)
<i>Raphidocelis subcapitata</i>	PS	EC ₅₀	23.5 %	■ Growth	Capolupo et al. (2020)
<i>Skeletonema costatum</i>	PS	EC ₅₀	>100 %	■ Growth	Capolupo et al. (2020)
<i>Raphidocelis subcapitata</i>	PP	EC ₅₀	64 %	■ Growth	Capolupo et al. (2020)
<i>Skeletonema costatum</i>	PP	EC ₅₀	18.1 %	■ Growth	Capolupo et al. (2020)
<i>Skeletonema costatum</i>	PE (74 µm)	IR	IR = 25.3 % (100 µm)	■ Growth	Zhu et al. (2019a, 2019b)
<i>Skeletonema costatum</i>	PVC (74 µm)	IR	IR = 29.3 % (100 µm)	■ Growth retardation	Zhu et al. (2019a, 2019b)
<i>Skeletonema costatum</i>	PVC800 (1 µm)	IR	IR = 36.8 % (100 µm)	■ retardation	Zhu et al. (2019a, 2019b)
<i>Skeletonema costatum</i>	PS	IR	IR = 24.7 % (100 µm)	■ Growth	Zhu et al. (2019a, 2019b)
<i>Phaeodactylum tricornutum</i>	PE (1–100 µm)	–	No effect	■ Growth	Gambardella et al. (2019)
<i>Isochrysis galbana</i>	PE (2–6 µm)	–	No effect	■ Growth	Gambardella et al. (2019)
<i>Scenedesmus obliquus</i>	PS	IR ₅₀	IR ₅₀ = 61 µm/ml (0.1 µm) IR ₅₀ = 7.5 µm/ml (0.5 µm) IR ₅₀ = 33 µm/ml (1 µm) IR ₅₀ = 22 µm/ml (2 µm)	■ Photosynthesis ■ Growth ■ ROS formation	Liu et al. (2019b)
<i>Scenedesmus obliquus</i>	PS-NH ₂	IR ₅₀	IR ₅₀ = 24 µm/ml (0.1 µm)	■ Photosynthesis ■ Growth ■ ROS formation	Liu et al. (2019b)
<i>Chlorella vulgaris</i>	Virgin PVC	IR	IR _{3 days} = 28.25 % (10 µg/ml)	■ Enzymatic functions	Fu et al. (2019)
	Aged PVC		IR _{4 days} = 35.26 % (10 µg/ml)	■ Growth	
	Aged PVC-copper (Cu)		IR = 16.05 %		
<i>Chlorella pyrenoidosa</i>	PS	IR	IR _{maximum} = 38.5 % (0.1 µm)	■ Growth	Mao et al. (2018)
<i>Skeletonema costatum</i>	PVC	IR	IR = 39.7 % (50 µg/ml)	■ Growth	Zhang et al. (2017)
<i>Dunaliella tertiolecta</i>	PS	IR	IR _{57%} = 250 µg/ml (0.05 µm) IR _{13%} = 250 µg/ml (0.5 µm)	-Photosynthesis	Sjollem et al. (2016)
				■ -Growth	
Combined effect of MPs and contaminants to microalgae					
<i>Chlorella vulgaris</i>	Aged PVC (150 µm, 10 mg/l)	IR	IR _{PVC+Cd} = 32.51 %	■ Growth	Wang et al. (2021)
	Aged PS (250 µm, 10 mg/l)		IR _{PVC+Cu} = 18.89 %		
	Cu (2 mg/l)		IR _{PS+Cd} = 27.55 %		
	Cd (2 mg/l)		IR _{Cd} = 85.14 % IR _{Cu} = 19.50 %		
<i>Chlorella pyrenoidosa</i>	PS-Methamphetamine (0.7 µm)	EC ₅₀	EC ₅₀ = 0.32 µg/ml	■ Growth	Qu et al. (2020)
<i>Isochrysis galbana</i>	PE-CPF (pesticide)	IR ₅₀	IR ₅₀ = 2.57 µg/ml	■ Growth	Garrido et al. (2019)
<i>Tetraselmis cguui</i>	MP	–	Reduced Chl-a at 46 % (0.9 µg/ml) No considerable impacts up to 41.5 µg/ml	■ Chlorophyll ■ Growth	Prata et al. (2018)
	MP-Doxycycline	EC ₅₀	EC _{50 Chl-a} = 7 µg/ml EC _{50 Growth} = 11 µg/ml		
	MP-Procainamide	EC ₅₀	EC _{50 Chl-a} = 31 µg/ml EC _{50 Growth} = 125 µg/ml		

3.3.2. Impact of MPs and heavy metals

Researchers showed the affinity of MPs towards heavy metals, which are persistent pollutants in the environment (Khalid et al., 2021). The MP polymeric matrix may contain heavy metals as functional additives, recyclates, or reaction byproducts (Turner et al., 2020). The quantity of heavy metals that each type of polymer adsorbs from the environment and the amount they naturally contain varies (Li et al., 2020b). The weathering/aging improves the MPs' adsorption capacity towards heavy metals since smaller MPs particles provide more surface area to volume ratio (Wagner et al., 2014). Additionally, the charged or neutral portion of MPs surfaces can directly adsorb cations and complex metals (Ashton et al., 2010). Metals such as Ag, Cd, Co, Ni, Pb, and Zn adsorption increase along with the increasing pH of river water (Turner and Holmes, 2015). According to Acosta-Coley et al. (2019), the concentration of heavy metals in MP particles is 10 to 100 times higher than that of the bulk materials in the environment. A significant percentage of metals in water and sediment can be bioavailable (generally called bioavailable fractions of metals) and thus bioaccumulate in organisms (Billah et al., 2019). Therefore, the additive effects of MPs with metals are potentially interesting to ecotoxicologists. Fu et al. (2019) demonstrated that copper with PVC-MPs (0.5 mg/l and 10 mg/l, respectively) inhibited the growth rate of *Chlorella vulgaris* more than single PVC-MPs. However, metals showed a decreased toxic effect on *C. vulgaris* when metals were combined with aged PS and PVC polymers (Wang et al., 2021). Such aged MPs exerted a high adsorption ability for metals, reducing the organism's bioavailability of coexisting heavy metals (Wang et al., 2021).

3.3.3. Effect of MPs and polyaromatic hydrocarbons (PAHs)

Organic contaminants such as PAHs are also widespread, carcinogenic, and mutagenic and can be transferred to the food chain (Kim et al., 2013). PAHs are sometimes regarded as polynuclear aromatic hydrocarbons containing multiple aromatic rings along with the basic form of two rings (naphthalene) and three rings (phenanthrene/anthracene) (Verla et al., 2019). US-EPA (US environmental protection agency) categorized 16 priority PAHs (also called legacy PAHs depending on their carcinogenic and mutagenic potentials), for which Toxicity and environmental abundance have generally been found (Hämer et al., 2014; Karami et al., 2016). In terms of pollution sources in the aquatic environment, PAHs can be petrogenic (e.g., oil spills, leakage from ships/boats) or pyrogenic (e.g., combustion-related sources) (Billah et al., 2022a).

Due to the large surface area and improved portioning between PAHs and MPs, adsorption at the nano/microscale would be faster than that of the same polymer in the bulk form (Zhu et al., 2019a). When PAHs interact with MPs in water, they combine to generate PAHs-enriched MPs, altering the fate, distribution, and level of Toxicity of MPs for aquatic species (Koelmans et al., 2016). The adsorption rate of PAH can be influenced by several factors, such as a high surface area to volume ratio, hydrophobicity, crystallinity, and physical (such as particle size) and chemical (such as functional groups) features of MPs (Herbort and Schuhen, 2017). Several investigations have confirmed the additive toxicological effect of MPs with PAHs (e.g., Browne et al., 2013; Paul-Pont et al., 2016). Paul-Pont et al. (2016) examined that fluoranthene-loaded MP could be accumulated to a high extent rather than pure fluoranthene in the species of *mytilus* spp. (marine mussel). The study also confirmed that a considerable level of antioxidant markers and histopathological abnormalities were present for the primary issue. Moreover, the interaction between the cell wall substances (e.g., p-glycoprotein) of the species involved in contaminant excretion and MP surfaces was clearly described (Paul-Pont et al., 2016).

Along with benzo[a]pyrene (B[a]P), PVC exhibited more Toxicity after co-exposure. Using studying with sand lugworm (*Acarochloris marina*), Browne et al. (2013) observed that 5% of previously preserved PVC with phenanthrene, triclosan, nonylphenol, and PBDE-47 can be transferred to the gut, and the accumulation rate in the tissue of these contaminants was higher than 5 % of PVC microsphere alone. Compared to the same chemicals in dissolved form in the surrounding water, higher

concentrations of organic pollutants on ingested MP may make them less bioavailable to aquatic organisms (Sørensen et al., 2020).

4. Toxicity of MPs to aquatic organisms

Different sizes of MPs can enter aquatic organisms through filter feeding and ingestion (Liu et al., 2020). The most common species, such as fish, phytoplankton (microalgae), zooplankton, and polychaetes that are prone to MPs toxicity, are summarized below.

4.1. MPs toxicity to fish

The effects of MPs on various fish species are presented in Table 1. Lu et al. (2016) investigated the impact of polystyrene MPs exposure on zebrafish, corroborating MP toxicity's size dependency. They observed that small-sized fragments with a large surface area could also absorb additional chemicals that can change the physical property of the plastic, causing Toxicity (Andrady, 2017). They also reported that, after seven days of exposure of the organism to 5 µm MP, MP substances accumulated in the liver, gut, and gill, whereas 20 µm MPs remained in the gill and gut. The potential toxic effects included liver inflammation and fat buildup, as well as oxidative stress and adverse changes to the metabolism profile of the fish liver.

In most cases, MPs reduce growth performance and increase fish mortality (Naidoo and Glassom, 2019). Ingested MPs accumulate in fish's intestinal tract, creating a blockage in the digestive tract and reducing the feeding rate due to sanitation (Wright et al., 2013). The blocked digestive track may lead to nutritional imbalance and growth inhibition in fish (Jabeen et al., 2018). For instance, Yin et al. (2018) reported that *Seabastes schlegelii* lost its growth rate, weight, and gross energy at 65.9 %, 65.4 %, and 9.5 %, respectively, after exposure to MPs at 10⁶ items/l. Moreover, the intake of MPs could accelerate inflammatory responses in fish (Lu et al., 2016) and alter the metabolic function (Lu et al., 2016), and inhibit the proper immune function (Greven et al., 2016). Fine-shaped MP could translocate to different vital organs like gills and the liver and destroy the organs after continuous deposit and transportation of MPs (Yin et al., 2018). The toxic effects of MPs are notable when the MPs and other associated chemicals enter the biological matrix of fish, presented in Table 1 (Cole et al., 2011). It was observed that the desorption rate of POPs from MPs in gut conditions was shown to be 30 times higher in laboratory simulations compared to that in seawater alone (Bakir et al., 2014).

4.1.1. Toxicity of plastic-additives additives to fish

Leaching of plastic additives can expose aquatic organisms to dangerous chemicals that can lead to endocrine disruption (Chen et al., 2019). Rochman et al. (2013) investigated that the bioaccumulation rate of some additives, including PAHs, polychlorinated biphenyls (PCBs), and polybrominated diphenyls (PBDs) increased at 2.4, 1.2, and 1.8 folds higher, respectively, due to the presence of MPs in Japanese medaka (*Oryzias latipes*). This accumulation rate of POPs could destroy liver function, involving glycogen decrease, fatty vacuolation, and single-cell necrosis (Rochman et al., 2013). In another investigation, Barboza et al. (2018b) observed that Juvenile European seabass (*Dicentrarchus labrax*) exposed to a mixture of MPs and Hg had Hg concentrations in their gills and livers that were up to 2.0 and 1.6 times greater than those of fish exposed to the same quantities of Hg alone. Batel et al. (2016) created an artificial aquatic food chain for brine shrimp (*Artemia* sp.) nauplii and zebrafish (*Denio rerio*) and reported that MPs might be used as a media of transferring benzo(a)pyrene from nauplii to zebrafish. Thus, fish species are prone to additive chemicals through trophic transfer (Batel et al., 2016).

Recently, some studies suggested that MPs are a suitable carrier of the bacterial pathogen (*Aeromonas salmonicida*) for fish (Viršek et al., 2017). However, more research is required to determine whether fish populations could become infected by the virus when they ingest MPs. Again, most studies concerned the Toxicity of microbeads, while fiber and films were

dominated types of MPs in the environment. Less consideration was given to the effect of morphological characters of MPs in fish bodies.

4.2. MPs toxicity to phytoplankton (microalgae)

Phytoplankton and microalgae are vital elements of aquatic ecosystems as they are the primary producers (Nolte et al., 2017). As primary consumers depend on microalgae, aquatic organisms can ingest MPs along with microalgae. Microalgae can also interact with MPs in aquatic environments depending on their size, species, and MP types, level of MPs, and surface characters (Parsai et al., 2022). The toxicity evaluation in microalgae is complex; however, the section will discuss the intrinsic interactions between MP polymers and microalgae.

The Toxicity of MPs in different algae species was presented in Table 2. Regarding photosynthesis, the adverse impact of MPs has been investigated in the marine algae *Skeletonema costatum* by Zhang et al. (2017). They reported that the highest inhibition of growth ratio occurred up to 39.7 % after 96 h exposure to 1 μm MPs. In addition, more adverse effect was observed when chlorophyll content and photosynthetic capability decreased at 20 % and 32 %, respectively, due to a high concentration of MPs (50 mg/l). Morphological observations using scanning electron microscopy (SEM) suggested that the most likely cause of Toxicity was the adsorption and aggregation of MPs on the outer surface areas of microalgae.

Mao et al. (2018), Prata (2018), and Wang et al. (2020a) accorded that the significant effect of MPs on any microalgae species was growth inhibition (Table 2). Ultimately, this effect also induced other impacts, such as photosynthesis depression and ROS formation, causing oxidative stress effects. In general, the size of algal cells was <20 μm , which is smaller than most of the single MPs. However, MPs could directly damage the algae cell and result in a cytotoxic effect on the algae (Liu et al., 2019c). Studies reported that MPs could damage microalgae by reducing their availability of light in the water column, which could impair their photosynthesis capability (Wang et al., 2020a; Liu et al., 2019c). After absorbing small-sized MPs (<1 μm) on algal surfaces, MPs might decrease cell membrane fluidity (Yi et al., 2019). The low fluidity of cell membranes impaired the material transport between cells and the surrounding environment and increased the adverse effect on cell structure (e.g., vacuolation, plasmolysis) (Mao et al., 2018). Moreover, ROS production due to MPs absorption paved the way for hetero-aggregation (microalgae-MPs) through the excretion of extracellular polymeric substance (EPS). Hence, many scholars supported that excreted EPS was responsible for reducing photosynthesis rate and cytotoxicity (Nolte et al., 2017). However, the detailed interaction pathways and mechanism of MPs with microalgae should be thoroughly investigated to improve understanding of MPs Toxicity to microalgae.

4.2.1. Toxicity of other compounds with MPs to microalgae

Pharmaceuticals and MPs are common contaminants in aquatic environments. Aquatic organisms, especially microalgae, are likely to be impacted by the co-existing pollutants (Fonte et al., 2016). The biological Toxicity from exposure to the mixture of pharmaceuticals and MPs might be significantly different due to the binding characteristics of pharmaceuticals with MPs and toxicological interactions (Fonte et al., 2016). Marine microalga *T. chuii* was exposed to pharmaceuticals (procainamide and doxycycline) and MPs, showing significantly higher Toxicity, including growth retardation and photosynthesis disruption for pharmaceuticals and MPs mixture than pharmaceuticals alone (Prata et al., 2018).

However, organic pollutants such as triclosan (TCS) induce less Toxicity with PVC than PS and PE due to the high adsorption rate of TCS on PVC (Zhu et al., 2019b). In addition, Yi et al. (2019) observed that the triphenyltin chloride (TPTCl) toxic levels were reduced by 15–19 % in the presence of 0.55 μm PS. They also reported that despite the decreased bioavailability of TPTCl, PS increased the uptake of TPTCl, causing TPTCl toxicity in green microalgae, *Chlorella pyrenoidosa*, which was more susceptible to TPTCl Toxicity due to cell destruction and higher TPTCl absorption.

Humic acid (HA) or humic substance is one of the major components of natural organic matter (NOM) in aquatic environments (Xu and Guo,

2017). HA considerably reduces small-size MPs' Toxicity by creating a corona on their surfaces, decreasing microalgae's affinity (Liu et al., 2019c). Similar mitigative effects of NOM on the Toxicity of nanoparticles and metals (Hg) were also observed in embryonic zebrafish (Kteeba et al., 2017; Li et al., 2019). There is some evidence regarding the combined Toxicity of one type of MPs with other ionic species/HA, but there is no information considering the combined Toxicity of different-sized MPs to microalgae (Parsai et al., 2022).

Examples of combined impacts of MPs with different contaminants are presented in Table 2. Wang et al. (2021) reported that aged PS and PVC in *Chlorella vulgaris* exhibited more substantial growth retardation than virgin polymers. Capolupo et al. (2020) evaluated the chemical composition and the effect of leachates containing different plastic materials like PP, PVC, PS, PET, and car tire rubber (CRT) on the growth of freshwater microalgae, *Raphidocelis subcapitata*, and marine microalgae *Skeletonema costatum*. They demonstrated that CRT exhibited more toxic effects than other compounds in freshwater ($\text{EC}_{50} = 0.5 \%$) and marine species ($\text{EC}_{50} = 19 \%$).

4.3. MPs toxicity to zooplankton

Zooplankton is a potential element in the food chain. They typically consume MPs that could cause a trophic transfer of MPs via the food chain resulting in MP bioaccumulation negatively impacting higher trophic organisms. (He et al., 2022; SF 2). There are many studies regarding MPs ingestion by zooplankton, and most of the MPs were fibers, pellets, and fragments forms (do Sul and Costa, 2014). It has been observed that some viruses, bacteria, and other microorganisms could create a biofilm on MPs' surfaces, leading to MPs' entry into organisms and, thus, increasing Toxicity (He et al., 2022). Table 3 summarizes recent studies regarding the Toxicity of MPs in zooplankton under laboratory conditions.

Previous studies demonstrated that the uptake of MPs accelerated the intestinal tract damage of zooplankton, feeding abnormalities, hatching problems, adverse impact on gene expression, suppression of reproduction efficiency, and mortality (Tang et al., 2019). The thioredoxin (TRx) system plays a crucial role in cellular antioxidant defense by removing ROS (He et al., 2022). In *daphnia sp.*, PS may pose a toxic effect on gene expression. Using quantitative real-time PCR, the expression of permease, arginine kinase (AK), and thioredoxin reductase (TRxR) in *daphnia sp.* was investigated after exposure to four different concentrations (0 (control), 2, 4, and 8 mg/l) of 1.25 μm PS microbeads for ten days. Their results showed that permease, *daphnia* TRxR, mRNA, and AK expressions were reduced significantly under 8 mg/l PS. Moreover, the molecular effect of MPs was detected when MPs regulated extracellular material transport, oxidative defense, and energy production (Tang et al., 2019). Rehse et al. (2016) reported that MPs could induce feeding abnormality in zooplankton when they are exposed to 1 μm pristine MPs. In another study, ephyrae jellyfish health was impacted by short-term MPs exposure, affecting their behaviour and survivability (Costa et al., 2020).

Furthermore, nylon MPs can lead to premature molting in young copepods despite MPs' bioavailability and Toxicity greatly depending on the MP's size and chemical structure (Cole et al., 2019). Cole et al. (2013) investigated the impact of MPs on plankton uptake using a bio-imaging procedure and reported that MPs decreased the copepod's food intake. For instance, the copepod *Calanus helgolandicus* consumed fewer algae after exposure to MPs that had the same size and shape as the algae. Such impediments to copepod feeding might exhibit a prolonged decrease in the amount of carbon biomass, impacting an adverse impact on the food web (Cole et al., 2015). In another study, Jeyavani et al. (2022) reported that increased mortality and disruption of antioxidant biomarkers occurred due to swimming behaviour change after exposure *Artemia salina* to PP for 14 days.

Spawning and progeny of zooplankton are also impacted by MPs, resulting in increased mortality. For example, Parthenogenetic larvae are prone to health danger after ingesting 10 μm PS microspheres (Wang et al., 2019b). Several studies reported the impacts of PS MPs on various intertidal barnacle life stages within and between generations, showing no effect

Table 3

Toxicological/biological effects of different types of MPs on zooplankton.

Zooplankton species	MPs type	MPs sizes (µm)	Concentration	Exposure route (via)	Exposure time	Biological/toxicological effects	References
<i>Artemia salina</i>	PP	11.86–44.62	1, 25, 50, 75, and 100 µg/mL	Water	2, 7, and 14 days	Alteration in homeostasis; Increased mortality; Change in swimming behaviour and disrupts antioxidant biomarkers	Jeyavani et al. (2022)
<i>Acartia tonsa</i> , <i>Temora longicornis</i>	Tire wire plastics (TWP)	8–20	10–10,000 TWP/l	Dietary	19 days	No particular toxicity in the species; Impact on feeding behaviour and pellet production; Influence on food web transfer efficiency.	Koski et al. (2021)
<i>Aurelia</i> sp.	PE	1–4	0.01–10 mg/l	Dietary	24 and 48 h	Impact on health in short-term MPs exposure; Change in survivability and behaviour; Impact in immobility and pulse frequency.	Costa et al. (2020)
<i>Amphibalanus amphitrite</i>	PS	1.7, 6.8 and 10.4	1, 10, 100, and 1000 items/l	Dietary	77 days	Inhibit growth and increase mortality at 1000 items/l MPs.	Yu and Chan (2020)
<i>Brachionus plicatilis</i> , <i>Acanthocyclops robustus</i>	PE	≤ 1000	140–180 items/m ³	Water	–	Potential risk to zooplankton community.	Alfonso et al. (2020)
<i>Daphnia magna</i>	PS	1.25	8 mg/l	Water	10 days	Uncontrolled permease, AK, mRNA and daphnia-TRxR.	Tang et al. (2019)
<i>Calanus helgolandicus</i>	Nylon	1 µm–5 mm	100 items/ml	Water	24 h	The feeding rate was 6 % decreased; Change in the fecal pellet sinking rate.	Coppock et al. (2019)
<i>Calanus finmarchicus</i>	Nylon	10–30	100 items/ml	Dietary	24 h	Accelerate premature molting; Feeding rate reduced at 40 %; Accumulation of lipid.	Cole et al. (2019)
<i>Euphausia superba</i>	PE	27–32	400 ng/l	Dietary	25 days	Acute exposure of MPs showed mortality; Change in feeding behaviour.	Dawson et al. (2018)
<i>Ceriodaphnia dubia</i>	PE	1–4	0.125–4 mg/l	Dietary	48 h, 8 days	Dimorphism of carapace and antenna observed by scanning electron microscope.	Ziajahromi et al. (2017a, 2017b)
<i>Parvocalanus crassirostris</i>	PET DEHP	5–10	0.11–1 ng/l (DEHP) 10,000–80,000 items/ml (PET)	Dietary	24 days (PET), 48 h (DEHP)	Egg production was reduced; DEHP induced reproduction problems that could be inherited by generations.	Heindler et al. (2017)
<i>Daphnia magna</i>	PET (fiber)	62–1400 (length) 31–528 (width)	12.5–100 mg/l	Dietary	48 h	Mortality increased when daphnia was not pre-fed with algae; No particular toxicity after pre-feeding algae.	Jemec et al. (2016)
<i>Daphnia magna</i>	PE	1, 100	400 mg/l	Dietary	96 h	Induced immobilization according to dose and time	Rehse et al. (2016)
<i>Calanus helgolandicus</i>	PS	20	75 items/ml	Dietary	7 days	Decreased reproductive activity; Reduced energy; Reduction in egg carbon biomass.	Cole et al. (2015)
<i>Centropages typicus</i> , decapod larvae, chaetognaths	PS	0.4–30.6	635 items/ml	Dietary	24 h	Reduced algae feeding; Copepods consumed fecal pellets laden with MPs; MPs adhered to exposed zooplankton's exterior carapace and appendages.	Cole et al. (2013)

within a generation but effects in between generations (He et al., 2022). For instance, after exposure to PS of parent larvae, considerable mortality was observed in offspring (Yu and Chan, 2020). Heindler et al. (2017) reported a reproduction problem that could be inherited from the Calanoid copepod offspring after exposure to di(2-ethylhexyl) phthalate. An experimental study supported that fibers impose more adverse reactions in daphnia than PE beds and malformations of the carapace and antenna (Ziajahromi et al., 2017a). On the contrary, there was no particular toxicity of *Acartia tonsa*, *Temora longicornis* after exposure to tire wire plastics (Koski et al., 2021). However, the impact on feeding behaviour, pellet production, and foodweb transfer efficiency might be significant under extreme doses and exposure periods (Koski et al., 2021).

Jemec et al. (2016) first investigated the MP textile fiber effect on the feeding behaviour of freshwater daphnia and reported that mortality increased after exposure to fibers for 48 h without pre-feeding on algae. In crustaceans, a delayed lethal impact was observed within 24 h. Some studies revealed that crustaceans were prone to ingest different MPs (Jemec et al., 2016). In line with this, other studies reported that daphnia generally

could ingest 2, 20, and 50 µm PE that can accumulate in their gut, generating immobilization and impeding lipid metabolism. However, all tested MPs did not extensively harm zooplankton. For example, planktonic sea urchins directly ingest PE MPs along with a hydrophobic organic compound (4-n-nonylphenol), and there was no induced toxic record (Beiras et al., 2019). This result is a hypothetical challenge to MPs serving as organic pollutant vectors in marine food webs. Nevertheless, further investigations are required to determine how MPs are metabolized in zooplankton, the presence or absence of residual MPs in excreta, the chronic impact of MPs on reproduction, and to elucidate the combined effect of MPs with other contaminants, what types of MPs are safe for zooplankton and other creatures.

4.4. MPs toxicity to polychaete

Polychaetes are abundant in aquatic ecosystems and are one of the key organisms in the coastal and estuarine food chain (Rodrigues et al., 2011). Also, polychaetes are the potential food source for many aquatic

communities (e.g., fish, birds, crustaceans) and act as a substantial vector of many contaminants through bioaccumulation (Pires et al., 2022). These organisms are exposed to pollutants directly in benthic sediment and pore water (Banta and Andersen, 2003). Therefore, they are considered a good model organism for contamination evaluation, being a sentinel species (Dean, 2008). So far, several studies have focused on the toxic effects of MPs and other chemical additives on polychaetes (e.g., Green et al., 2016; Soares et al., 2021). Former studies reported that polychaete is responsive in MPs and other associated chemical additives even at a low concentration level which can be observed in the endpoints (Pires et al., 2022). Most

studies used mainly PS, PP, PVC, and MPs mixtures in different polychaete species, and oxidative stress generation and energy reservation were the common endpoints.

The effects of PVC exposure to different polychaete species are presented in Table 4. Green et al. (2016) reported that the survivability and biomass of *Acaryochloris marina* were not significantly harmed after 31 days of exposure in the concentrations (0.02, 0.2, and 2 % of wet sediment weight). However, metabolic rates increased due to the presence of MPs and higher O₂ consumption at 2 % of PVC, which was justified as one of the stress responses. Wright et al. (2013) investigated with same

Table 4

Effects of MPs along with other additives in polychaetes exposed in laboratory conditions.

Polychaete species	Contaminants	Sizes	Concentration	Matrix	Exposure time	Biological/toxicological effects/endpoints	References
<i>Hediste diversicolor</i>	LDPE	63–75 µm and 300–355 µm	Not measured	Sediment	28 days	Occurred neurotoxicity causing 60 % lower AChE activity; Reduction SOD, CAT, and GST enzyme activity.	Urban-Malinga et al. (2022)
<i>Hediste diversicolor</i>	PA, PE, HDPE, PP, and polyethylene vinyl acetate (PEVA)	<100 µm	100 mg/kg	Sediment	7 and 14 days	Increase energy metabolism, amino acids, and osmolytes metabolism with induction of autophagy process; Destabilization of the cytoskeleton that caused tubulin loss in body tissue	Missawi et al. (2022)
<i>Hediste diversicolor</i>	APP (historic and modern)	0.06–1 mm	0–18.8 g/l	Sediment	5 and 18 days	Modern APPs decreased feeding rate, weight, and burrowing ability; High level of metallothionein-like protein for historic APPs	Muller-Karanassos et al. (2021)
<i>Hediste diversicolor</i>	PA, PE, HDPE, LDPE, PP and PEVA	1–5 mm and 300 µm – 1 mm	0, 10, 100 mg/kg	Sediment	1,3,7 and 14 days	Reduced survivability and growth.	Missawi et al. (2021)
<i>Hediste diversicolor</i>	PS nano particles	100 nm	0.005–50 mg/ml	Water	28 days	Inhibit cholinesterase activity; Increase burrowing time; Induce oxidative damage by increased protein oxidation	Silva et al. (2020b)
<i>Hediste diversicolor</i>	PP and PE	0.4–400 µm	10 and 100 µm/l in water; 10 and 50 mg/kg in sediment	Water and sediment	98 h	Decrease coelomocytes viability; Increased immunotoxicity	Revel et al. (2020)
<i>Hediste diversicolor</i>	PS nanoparticles	100 nm	0.005–50 mg/ml	Water	28 days	Reduced regenerative capacity	Silva et al. (2020a)
<i>Hediste diversicolor</i>	APP	500 µm and >2 mm	0.2 g/l	Water	–	Increase Cu accumulation; Inhibit the organisms to control Cu accumulation in body tissue	Muller-Karanassos et al. (2019)
<i>Perinereis aibuhitensis</i>	PS	8–12 µm and 32–38 µm	100 beads/ml and 1000 beads/ml	Dietary	28 days	Smaller particles were more detrimental to health than larger particles;	Leung and Chan (2018)
<i>Arenicola marina</i>	PS, PA	500 µm and 1000 µm	20 g (PS) and 10 g (PA) in 10 L bucket	Sediment	106–240 days	Occurred sediment bioturbation	Gebhardt and Forster (2018)
<i>Hediste diversicolor</i>	PVC, PVC + B[a]P	250 µm	PVC: 200 and 2000 items/kg, PVC + B[a]P: 1 mg/l	Sediment	10 and 28 days	Induced stress syndrome; Increased bioaccumulation of contaminants	Gomiero et al. (2018)
<i>Galeolaria caespitosa</i>	Dibutyl phthalate	–	0.02, 0.2, 2 and 20 mg/l	Water	15 min–2.5 h	Disrupted embryonic development; Inhibited sperm SOD; Increased lipid aldehyde accumulation; Damage sperm centrioles	Lu et al. (2017)
<i>Marenzelleria spp</i>	PS (fluorescent), Dibutyl phthalate	10 µm	PS: 5, 50, and 250 beads/ml Dibutyl phthalate: 0.02–20 mg/l	Water	24 h	Impact on feeding	Setälä et al. (2016)
<i>Laeonereis acuta</i>	BDE-47	–	0.17–410 ng/g dw	Sediment	14 days	Induce bioaccumulation; Impact on oxidative stress response	Díaz-Jaramillo et al. (2016)
<i>Arenicola marina</i>	HDPE	2.5–316 µm	0.02–2 %	Sediment	31 days	PVC, HDPE, and PLA decreased biomass in the sediment; A high concentration of PVC (2 %) changed metabolism activity; Impacted burrowing activity	Green et al. (2016)
<i>Arenicola marina</i>	PVC PLA PS	8,7–478 µm 1.4–107 µm 10 µm and 30 µm 90 µm	0.02–2 % 0.02–2 % 50 items/g 10 items/g	Sediment	14 days	Impact on energy metabolism; No adverse effect at high concentration	Van Cauwenberghe et al. (2015)
<i>Perinereis aibuhitensis</i>	Bisphenol A (BPA)	–	10, 50, and 100 µg/l	Water	4,7, 14 days	Induced the expression level Pa Gα mRNA; High-level BPA increased the high rate of mRNA transcription.	Zhao et al. (2014)

species and demonstrated that worms exhibited considerably lower eating activity after chronic exposure (4 weeks), whereas those acutely exposed for 48 h displayed extended gut residence time at 5 % of unplasticized polyvinyl chloride (UPVC). The gross energy reserve in 1 % and 5 % UPVC exposed worms were substantially decreased along with a maximal 50 % reduction to 5 % UPVC. Gomiero et al. (2018) conducted a study with *Hediste diversicolor*. Their experiments included spiked sediment with pristine PVC at 200 and 2000 items/kg for 10 and 28 days. The exposure time and MP concentrations were actively linked with the immune response induction of the susceptible species, and the mitochondrial activity decreased. However, DNA damage, lysosomal membrane integrity, and oxyradical generation were not impacted. Meanwhile, *A. marina* showed behavioral impairment and lower egestion rate at acute exposure (0 and 5 % dw), and chronic exposure at the same concentration level reduced feeding activity (Wright et al., 2013). The result was in line with Browne et al. (2013) and Green et al. (2016), where immune system response and metabolism rates of *A. marina* and *H. diversicolor* were considerably impaired after chronic exposure to PVC (0–5 % and 200–2000 items/kg) (Gomiero et al., 2018). The impact of MPs on polychaete can be different depending on the plastic composition and specific polymer (Missawi et al., 2021). The effects of PS exposure to various polychaete species are represented in Table 4. Silva et al. (2020b) studied oxidative stress, its impact on metabolism, neurotoxicity, and borrowing capability of *H. diversicolor* using various concentrations of PS nanoplastics (NPs). They claimed that enzyme superoxide dismutase (SOD) function increased at higher exposure, whereas the role of glutathione S-transferases (GSTs) and catalase (CAT) were inhibited, which might cause excess protein carbonylation. Moreover, the increased metabolic activity was closely related to validating the antioxidant defense system for PS, although the borrowing capacity was decreased. In addition, after exposure ten times lower than the previous PS NPs concentration, the regeneration capacity of *H. diversicolor* was reduced. However, it was explained that there were no behavioral changes at a higher concentration as PS NPs tended to aggregate alongside time and concentration, producing larger-sized plastics that might be less bioavailable (Silva et al., 2020b).

Size-related studies were conducted by Leung and Chan (2018) to determine the effect of PS on the regeneration rate of *Perinereis albuhiensis* for four weeks of exposure time. Using MP particle size at 100 µm and a concentration of 1000 items/ml, the study reported that PS MPs reduced the posterior segment regeneration rate, creating more mortality. Moreover, feeding smaller particles to the species revealed that smaller particles were more detrimental than larger ones. In another study, a positive relationship was identified between the concentration of MPs in the sediment and weight loss of *A. marina* when it decreased the feeding response at the highest dose (7.4 % dw) (Besseling et al., 2013). The study also reported that the highest dose (7.4 %) reduced the bioaccumulation of 19 polychlorinated biphenyls (PCBs), while a small PS dose (0.074 %) increased PCBs bioaccumulation in the species. Even at an exceptionally high concentration (110 MP/g of sediment) with three different sizes of PS (10, 30, and 90 µm) revealed no significant toxicity to the organism in short-term exposures (14 days) (Van Cauwenberghe et al., 2015).

Setälä et al. (2016) designed an experiment for *Marenzelleria* sp. to investigate the ingestion of 10 µm PS at 5, 50, and 250 beads/ml. The authors reported that the worm's consumption rate increased along with the concentration. Particles with smaller sizes (100 nm and 8–12 µm in diameter) showed relatively higher effects on the organisms regarding behaviour, oxidative status, regeneration ability, and defense efficiency (Silva et al., 2020a). On the contrary, *Arenicola marina* (lug-worm species) showed a negative feeding response (7.4 %) and weight loss for large-sized MPs (400–1300 µm) with high concentrations, which ultimately affected health, growth, and survivability (Besseling et al., 2013). Green et al. (2016) determined the effect of HDPE and polyvinylchloride in *A. marina*. After 31 days of exposure, no impact on the organisms was observed, especially in behaviour, survivability, and biomass. However, organisms were to be affected by the high concentration of HDPE in terms of respiratory rate.

4.4.1. Toxicity of MP additives to polychaete

Over the past few years, several studies have been conducted regarding the impact of plastic additives associated with MPs in aquatic systems, including polychaetes (Pires et al., 2022). Examples regarding the Toxicity of plastic additives in polychaetes are given in Table 4. In these investigations, growth, behaviour, and survivability were featured as endpoints (Shin et al., 2014; Browne et al., 2013).

An experiment was employed by Muller-karanassos et al. (2021) to investigate acute and chronic Toxicity (5 days and 18 days, respectively) of three identical APPs on *H. diversicolor* at a normal concentration (0–18.8 g/l). They showed that the influence of contemporary APPs was greater than that of historic and silicone APPs, leading to a reduction in body weight, feeding response, and burrowing activity. The possible reason is that modern APPs had high Cu concentrations (approximately 2.6 times higher than historical ones) that posed more threats at normal concentrations. Moreover, further analysis supported that there was a relationship between APP ingestion and Cu accumulation in body tissue, while silicone APPs were the least harmful (Muller-karanassos et al., 2021).

Using contaminated sediment from a field site or spiked sediment under laboratory-controlled conditions with deca-BDE, penta-BDE, and PCB 209, the polybrominated flame retardants' bioaccumulation and Toxicity of the flame retardants diphenyl ethers (PBDEs), including the specific congener PCB 209, were examined in *Nereis virens* (Klosterhaus et al., 2011). The 28-day sediment exposure (ranging from 0 to 3000 ng/g dw depending on the congener) was intended to measure bioaccumulation, survival, energy stores (lipids), and biomass.

The impact of a specific PBDE congener on *Capitella* sp. I larval settling (24-h test with spiked sediment at 0.5 and 3 ppb BDE-47) was investigated under normoxia and hypoxia conditions (Shin et al., 2014). Moreover, the effects of bioaccumulation and oxidative stress were analyzed in *Laonereis acuta* adults subjected to polluted sediment (0.17–410 ng/g dw) for 14 days using the same congener BDE-47 (Díaz-Jaramillo et al., 2016). Perron et al. (2012) conducted a study regarding triclosan (TCS) bioaccumulation in *N. virens* under spiked sediment for 28 days. They revealed that after 14 days of depuration, the TCS concentration declined to 2.5 g/g lipid with a little increase to 4.5 g/g lipid after 28 days. The biota sediment accumulation factor (BSAF) contained 0.23 kg organic carbon/kg lipid that was reduced by 50 % after the depuration period.

Eom et al. (2019) adopted an emerging biocide (Sea-Nine™ 211) in *P. albuhiensis* to evaluate the biological response after the alteration of intracellular malondialdehyde contents (MDA), glutathione (GSH) and enzyme function of superoxide dismutase (SOD), glutathione S-transferase (GST), glutathione reductase (GR), AChE and glutathione peroxidase (GPx) at 0.1, 1 and 10 µg/l for 14 days. They found that the MDA substance considerably increased according to time and dose, indicating oxidative damage caused by lipid peroxidation. Moreover, after being exposed to 10 g/l Sea-Nine, there was a high level of intracellular GSH and antioxidant defense-related enzyme (SOD, GR, CAT, GST, and GPx) activity. However, AChE activity was reduced after exposure to a maximum concentration of 10 µg/l. The study concluded that Sea-Nine was hazardous to marine polychaetes at a sublethal concentration as it might cause lipid peroxidation, cholinergic system control, and oxidative stress.

The molecular-level effect of bisphenol A (BPA) waterborne exposure in *P. albuhiensis* was evaluated at 10, 50, and 100 µg/l over 4, 7, and 14 days, respectively (Zhao et al., 2014). They showed that BPA enhanced Pa Gα (G protein alpha response to various stimuli involving medications, hormones, and environmental variables), and it was positively correlated with exposure dose and duration. Moreover, the BPA endocrine-disrupting effects in the organisms might be related to changes in the G protein pathway.

Phthalates are one of the crucial functional groups of plastic additives. Previous studies focused on dibutyl phthalate (DBP), which was experimented with different embryonic stages of *Galeolaria caespitose*. After 30-min DBP exposure in water (from 0.02 to 20 mg/l) exhibited sperm dysfunction as well as embryogenesis abnormality (Lu et al., 2017). Additionally, DBP inhibited SOD activity, and oxidative stress

was found primarily in DBP-treated cells. However, it was a challenge to identify the actual toxicity of MPs with other emerging contaminants and pharmaceuticals and how polychaetes ingest them. Recently, some studies (e.g., Knutsen et al., 2020; Pequeno et al., 2021) examined the bioaccumulation of MPs in different polychaetes, demonstrating the little biological effect. To confirm these seemingly contradictory results is a potential research gap.

5. MPs-remediation strategies for aquatic environments

The potential sources, locations, and routes of MPs in terrestrial, atmospheric, and other aquatic branches such as estuaries, rivers, oceans, soils, and atmosphere were summarized in SF 3. It is clear that urban actions and other anthropogenic activities, such as WWTPs' activities, contributed significantly to the attribution of MPs in aquatic environments (Lv et al., 2019). Regardless of dry or wet deposition, the suspended MPs in the atmosphere can transport to other land, rivers, and oceans (Leslie et al., 2017). Some strategies are discussed below to control and mitigate MP pollution in aquatic environments.

5.1. MPs source control

Control of MPs' sources is the vital preliminary approach, including production, implementation, and discharge. As more plastic production and usage will raise plastic abundance in the environment, every step must be focused on reducing plastic pollution. In addition, enterprises and governments must work together to tackle the issue of MPs' contamination by laws and regulations that are either mandatory or supportive.

5.2. Production step

The growing recognition of microbeads and single-use plastic bags as sources of marine plastic pollution has led to expanding limited policies (Xanthos and Walker, 2017). Restricted manufacturing of MPs will reduce the release of billions of MPs into the aquatic medium (Rochman et al., 2014). Some polymers, such as PP, PET, PE, and PS, considerably tolerate light, chemicals, temperature, and hydrolysis due to strong C—C or C-heteroatom backbones (Zhang et al., 2021b). Moreover, stability plays a crucial role in reducing MPs degradation in aquatic systems (Bansal et al., 2021). The high volume of biodegradable materials consists of recycling (Sander, 2019). Hence, it is advisable to reverse the trend towards employing biodegradable polymers as environmentally friendly (Kubowicz and Booth, 2017). For instance, Bagheri et al. (2017) studied the degradation of non-degradable PET and bioplastic and observed that only poly (lactic-co-glycolic acid) was degraded in 207 days. In this regard, bio-resource could be a potential alternative to biodegradable plastic production (Shruti and Kutralam-Muniasamy, 2019). However, “Biodegradable” labels on plastic products can mislead consumers, which might encourage them to use more plastics. Even biodegradable plastics can be broken down eventually in an aquatic system that may be unable to achieve sustainable economic goals (Kubowicz and Booth, 2017).

5.3. Implementation stage

Taking consideration to the preventive measure of controlling MPs through the application of laws and regulations is increasing. For instance, Lv et al. (2019) proposed that banning plastic bags and microbeads, isolating microfibers from laundry and styrofoam products, and eliminating MPs from WWTPs are fruitful approaches for MPs' source control. Actually, there are several legislations around the world to limit the use of plastic products. However, enforcing legislative frameworks regarding MPs management remains in its infancy (Xu et al., 2021). When there is no proper legislation by local governments, some constructive frameworks can be forced to decrease MPs' release. To tackle MPs' pollution, the US government has banned MPs from regular cosmetic items since 2017 (Picó and

Barceló, 2019). Other countries, such as Canada, Australia, and European Union (EU), have also contributed to action plans regarding this issue. Considering the source of MPs, strict efforts have been established against the sale and consumption of single-use plastics as well as plastic bags and straws (EU, 2015). Evidence showed that MPs used in cosmetics are potential sources of MPs in the environment. Recently, a comprehensive strategy has been established throughout the EU as a significant part of the circular economy. The main focus is to reduce plastic consumption and production and to make plastics recyclable by 2030 (EU, 2015). Such schemes are to be publicly implemented to minimize the significant quantity of MPs in the open environment.

5.4. Discharge step

Due to the small particle size and ultralow concentration, MPs are extremely difficult to remove once they are discharged into aquatic systems (Sun et al., 2019). In this regard, WWTPs are one of the multiple sources of releasing MPs in the environment (Schöpel and Stamminger, 2019).

MPs may not be successfully removed by WWTPs facilities, allowing MPs to be released to aquatic environments (Estahbanati and Fahrenfeld, 2016). There are three MP treatment steps (preliminary, primary, secondary, and tertiary). In an MPs treatment plant, the concentration of MPs in the influent was 1–10,044 items/l, whereas the effluent concentration was 0–447 items/l (Sun et al., 2019). Incomplete removal of MPs during treatment processes (70–100 %) could exhibit significant plastic pollution in aquatic environments (Estahbanati and Fahrenfeld, 2016; Zhang et al., 2021b). It was estimated that MPs could be removed in preliminary and primary steps at 35–59 % and 50–98 %, respectively, by trapping MPs in solid flocs, skimming light floating MPs, and settling heavy MPs (Zhang et al., 2021b). Moreover, the primary treatment could eliminate large MPs (1–5 mm) by 38 %, while small MPs (0.1–0.5 mm) increased by 37 % (Dris et al., 2015). At the secondary treatment stage, biological treatments and clarification were included, and the MPs could either be trapped by organisms' ingestion or accumulation with sludge flocs and extracellular polymers (Murphy et al., 2016). Along with this, several investigations have been done regarding remediation strategies that can be employed for MPs' removal from the aquatic environment.

5.5. MPs removal and remediation techniques

5.5.1. Biological removal processes

The biodegradation method is environmentally friendly and cost-effective compared with other abiotic treatments, has low energy input, and has carbon footprint optimization (Hu et al., 2021a). Some microorganisms like fungi and bacteria can degrade MPs primarily as these organisms use MP particles as nitrogen sources, habitat, and proliferate (Montazer et al., 2020). Such functions provide an excellent era for MPs degradation facilities and purification.

Certain bacteria, such as *Rhodococcus* sp 36, *Kocuria palustris* M 16, and *Bacillus* strains, are capable of degrading MPs, although the degradation rate was low, resulting in 1–10 % weight loss between 30 days to 1 year (Roager and Sonnenschein, 2019). For example, *Ideonella sakaiensis* 201-F6 can degrade most PET particles within six weeks at 30 °C (Yoshida et al., 2016), and PET hydrolases enzyme is the primary agent to degrade PET polymers (Bååth et al., 2021). The structural analysis of the enzyme revealed that these enzymes represent α/β folds and disulfide bonds, providing thermal balance during PET degradation (Fecker et al., 2018). In the degradation procedure, agar plates are used for isolating *I. sakaiensis* as a single colony from the concentrated microorganisms. For this, the Yeast extract-sodium carbonate-vitamin (YSV) medium is used to dilute the microbial consortium, and the cultured microbes serially are monitored through PCR. The degradation of PET film can be measured by weight loss, CO₂ production from PET catabolism, and surface morphological changes in films which can be detected by scanning electron microscopy (SEM) (Yoshida et al., 2021). PET at first is degraded as bis(2-hydroxyethyl) terephthalate (BHET), mono (2-hydroxyethyl) terephthalate (MHET), and

TPA (Palm et al., 2019). The procedure is facilitated for cell growth by PETase secreting and integrating enzymatically released substances (terephthalic acid (TPS) and mono(2-hydroxyethyl) terephthalic acid (MHET) (Knott et al., 2020). Moreover, the PETase enzyme has exceptional hydrolytic activity and prefers PET as a substrate (Chen et al., 2018). According to Tournier et al. (2020), the performance of PET hydrolase and leaf-branch compost cutinase (LCC) led PET to degrade to monomers by almost 90 % within 10 h (Fig. 5a, b). The degraded polymers were further used to form PET for recycling. High degrade capability involves low PET crystallinity and high chain mobility (Yang et al., 2016). Another study reported that microbial consortiums (single bacterium) could degrade plastic due to bioaugmentation and acclimatization (Roager and Sonnenschein, 2019). Some genera, like *Thermobifida* and *Thermomonospora* could degrade PET (Ru et al., 2020). However, the degradation of non-hydrolyzable polymers such as PP and PE that are widely available in the environment remains a significant challenge as these particles are resistant to biodegrading (Krueger et al., 2015).

Fungi can degrade petroleum-based plastics by secreting enzymes, as they use MPs as energy and carbon sources (Fig. 6). There is evidence that some plastics like PP, PVC, PA, LDPE, HDPE, polystyrene sulfonate, and polyester polyurethane (PPU) are degradable by fungi. The combination of the intracellular process for fungal adaptation, detoxification, and extracellular enzymatic procedures are credentials for the degradation capability of fungi. The ultimate release of hydrolase enzyme can degrade MPs (Sánchez, 2020). In addition, fungi exhibited relatively better performance in PE degradation than bacteria (Muhonja et al., 2018). For instance, PE can be effectively degraded by marine fungus *Zalerion maritimum* (Paço et al., 2017). A typical reason is that fungi have taken advantage of adaptability as they are found in terrestrial and aquatic environments (Raghukumar, 2017). In one experiment, *Z. maritimum* was grown in a growth medium with 20 g/l of glucose, 20 g/l of malt extract, and 1 g/l of peptone as well as 35 g/l of sea salts (Sigma-Aldrich, 2016). After that, stirring was continued for 5 days before MPs removal assays. For culture, a batch reactor with 0.130 g of MPs and 50 ml of 10 folds diluted minimum growth medium was inoculated along with 0.5 g of

filtered *Z. maritimum*. The batch reactors were incubated at 120 rpm for 28 days. The fungus biomass and MPs were separated from the medium through filtration with glass fiber filters. The biomass was closely inspected to determine the extraction of attached MPs. Fourier transform infrared-attenuated total reflectance spectroscopy (FTIR-ATR), NMR, and optical, and electron microscopes were used for further analysis (Paço et al., 2017).

Fungi facilitate plastic degradation as hydrophobic substrates can synthesize hydrophobin to attach hyphae to them. Some additional strategies like decreasing biocides, adding pro-oxidant specimens, and an antioxidant stabilizer in plastic production have been suggested for effective biodegradation by fungi (Sánchez, 2020). Notably, studies regarding functional enzymes for plastic degradation are very limited. Therefore, more studies are needed considering these aspects. Although enzyme biocatalysis successfully depolymerized only PET with hydrolyzable linkages, such as amides and esters, PA and PU have not yet been successfully depolymerized (Zhu et al., 2022; Wei et al., 2020). Moreover, due to the lack of functional groups and the high molecular weight of the polyolefin chain, studies on biodegradation are also limited. Hence, abiotic treatment is essential before polyolefin (Cowan et al., 2022).

It was reported that some algae could interact with MPs through adsorptive actions and physical entanglement (Peller et al., 2021). Sundbæk et al. (2018) conducted a study on the adherence behaviour of fluorescent MPs on microalgae (*Fucus vesiculosus*) surface using ~20 µm PS MPs. Very narrow microchannels were present in the plant cells of the sorbent to prevent PS MPs translocation into the tissue. The study reported that near the cut surface area of the microalgae showed the highest sorption rate (~94.5) of MPs. It was regarded that alginate substances from the cell wall with gelatinous features played a crucial role in the cut areas. The effectiveness of the surface charge of MPs, the surface features of microalgae, and the sorption of tiny MPs by microalgae were highlighted in many previous studies. For instance, Nolte et al. (2017) investigated the adsorption of 20–500 nm PS on single-cell green algae (*Pseudokirchneriella subcapitata*). At first, all particles (apart from the non-functionalized PS particles) were characterized by hydrodynamic size and electrokinetic potential in deionized water

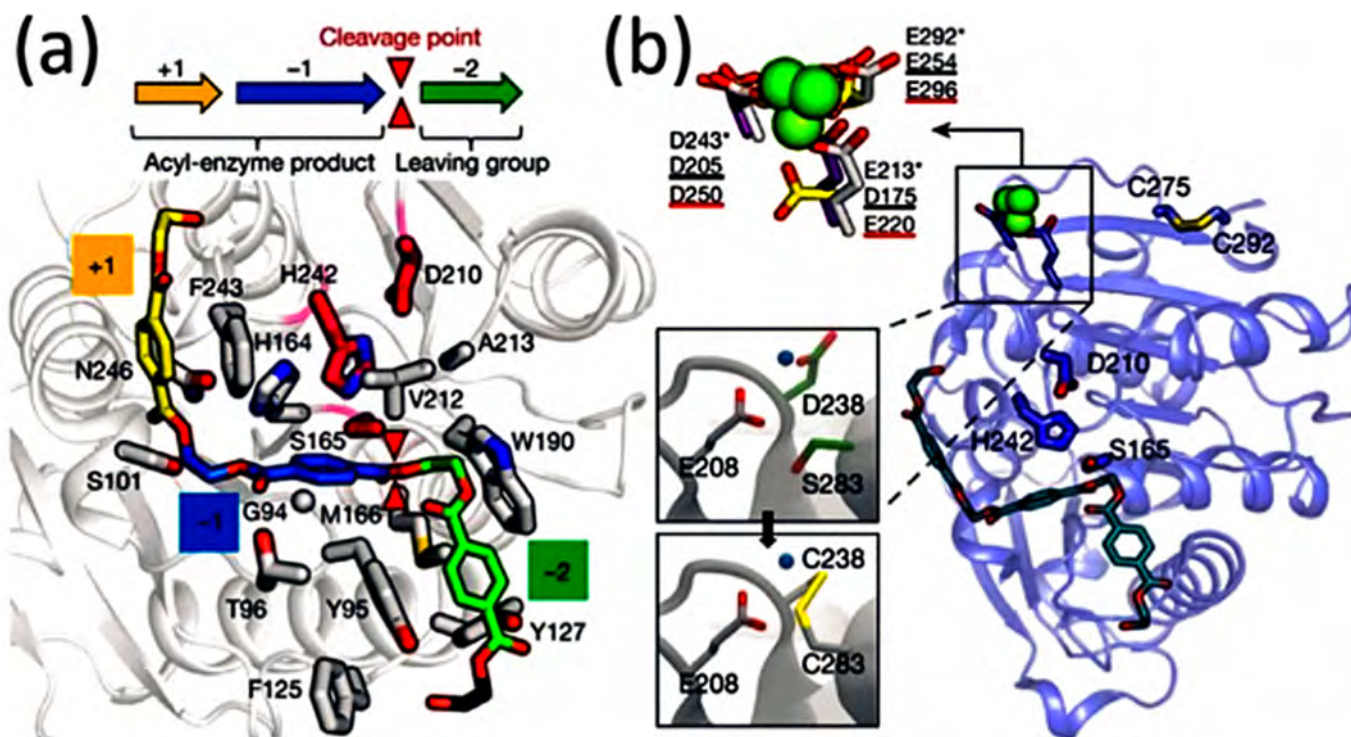


Fig. 5. a) Structural model of 2-HE(MHET)3 docked in wild-type LCC (grey ribbon), which enhanced LCC's specialized PET-depolymerisation activity b) LCC thermostability is increased by using a disulphide bridge. From Tournier et al., 2020 and Hu et al., 2021a.

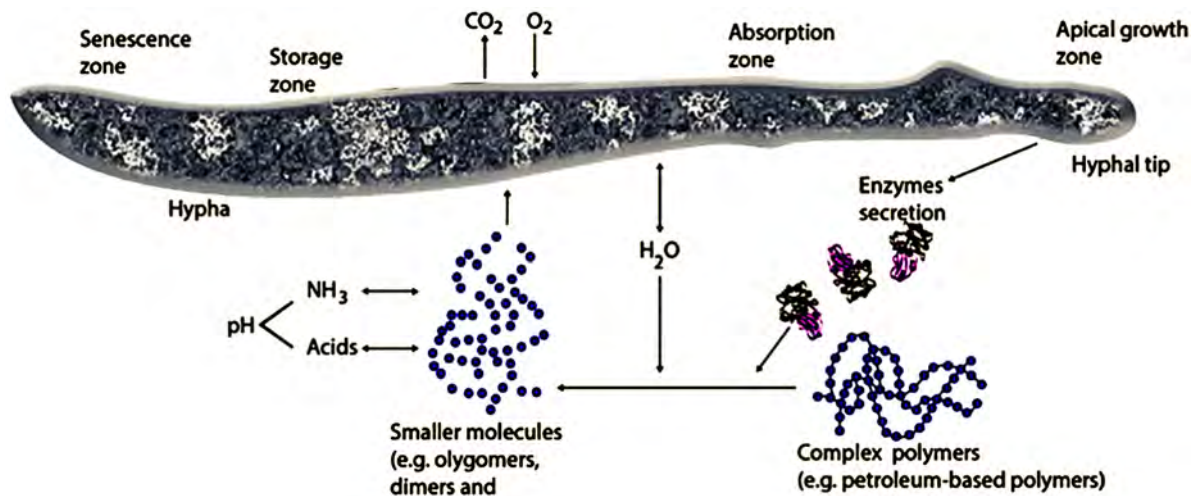


Fig. 6. Polymer degradation diagram through the fungal cell (hypha). From Sánchez, 2020 and Hu et al., 2021a.

with a stable pH and suitable medium using a Zetasizer. Measurements of dynamic light scattering (DLS) and electrokinetic were conducted with 10 mg/l of de-ionized water and freshwater algal medium. Meanwhile, the algal test preparation was performed following ISO guidelines (ISO, 2012), and all experiments were carried out in freshwater. Prior to the adsorption and growth inhibition test, the test media was mixed with premeasured particle stock suspension, and then the alteration of mean hydrodynamic diameter was recorded at 5 s intervals for 15 min at 20 °C. They reported that positively charged PS was more effectively adsorbed on algae surfaces than those with negative charges. The study concluded that positively charged MPs exhibited a higher tendency to be sorbed due to algae's anionic polysaccharide chemical structure. In addition, some strategies such as, counting strategies might be chosen based on the types of MPs and exposure level (SF 4). Counting MPs on

the entire surface of microalgae might be more acceptable for assessing more environmentally realistic levels.

5.5.2. Engineering/chemical processes

5.5.2.1. Dynamic membrane (DM). At present days, DM treatment has become more prevalent in remediation from sludge (Liu et al., 2016), oily water (Yang et al., 2011), industrial wastewater (Zhang et al., 2010), surface water (Yi et al., 2012) and municipal wastewater (Ma et al., 2013a, 2013b). This method can efficiently remove MPs from synthetic wastewater (Lei et al., 2020). Li et al. (2018) set up a basic dynamic filtration system with a storage (feed) tank, a filter set-up, and an effluent collection tank (Fig. 7). Diatomite (AR, Tianjin BASF) and tap water were used to prepare the synthetic wastewater used as the feed water. An electric agitator was

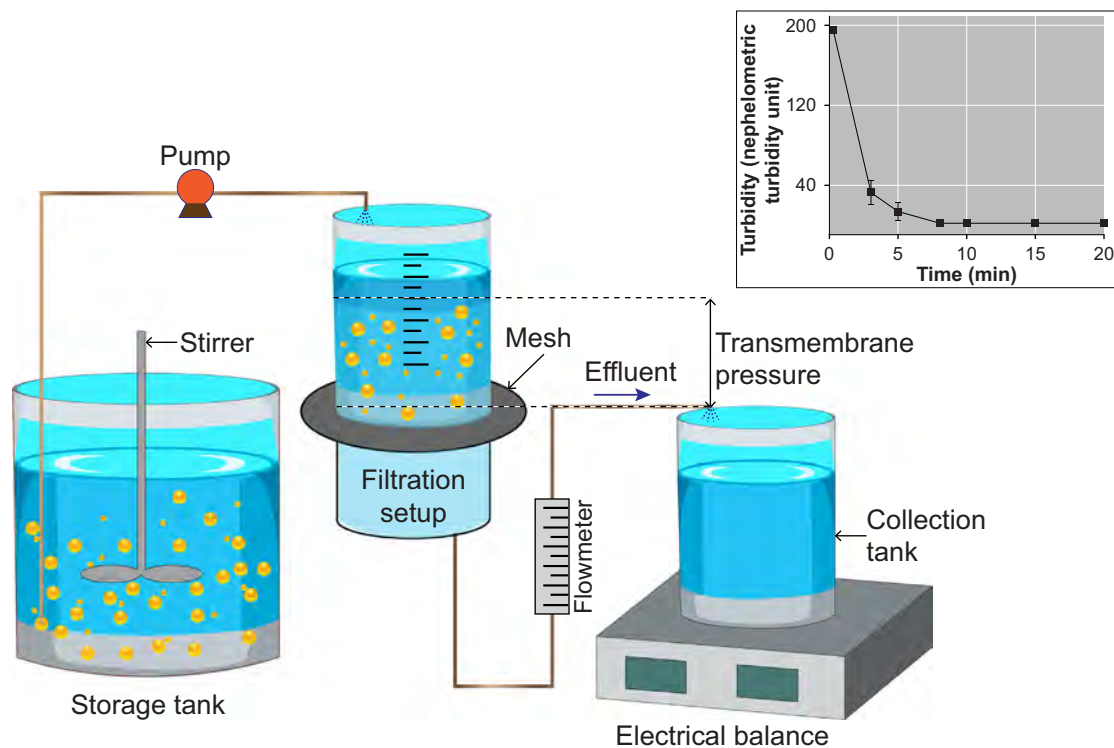


Fig. 7. A schematic diagram for a membrane bioreactor setup shows a graph of the turbidity's decline over time as MPs are eliminated. TMP means transmembrane pressure, whereas NTU is a nephelometric turbidity unit. From Li et al., 2018.

used to continuously mix the prepared and stored synthetic wastewater. The transmembrane pressure (TMP) was measured across the membrane while the synthetic wastewater was continuously injected into the filtration system. An electronic balance was used to weigh the gathered, filtered effluent in a collection tank and confirm the permeate flux. The experimental data were used to analyze the DM's performance and repeated three times for each experimental trial to ensure the data validity.

Ersahin et al. (2017) carried out a study regarding MPs removal through a submerged and external anaerobic dynamic membrane bioreactor (AnDMBR) at a laboratory scale. In this method, peristaltic pumps were utilized to collect permeate from the membrane modules and feed substrate into the anaerobic bioreactors. TMP was measured using a pressure sensor, and to regulate the thickness of the DM layer and TMP on the surface of the woven cloth, biogas sparging and backwashing were utilized. The impact of MPs level and influent flux on the removal efficiency of DM were notable, produced on a diatomite platform oriented with a supporting mesh of 90 μm while synthetic wastewater filtration process. They reported that decreasing turbidity from 195 NTU in the influent and <1 NTU in the effluent exhibited excellent filtration results. In addition, higher MP concentration and influent fluxes stimulated the formation of DM (Horton and Dixon, 2018).

In another study, Ward (2015) evaluated the MPs removal efficiency with a noble design of polymer coating as an elongated mesh screen. He reported that the platform was easily constructed from readily available components and had the benefit of having exceptional durability. Other benefits of tools involved the minimal requirement of mechanical equipment and the absence of electric power.

Recent studies showed membrane bioreactors have a higher capacity for removing micro-sized plastics than basic DM (Lares et al., 2018). Knoblock et al. (1994) coupled the biological process with porous membranes to study the purifying potential of a linked system. The wide range of successful removal of MPs from industrial wastewater using membrane bioreactors demonstrates the aptitude of this technique for treating high-strength pollutants like polymeric debris (Gurung et al., 2016). Inspired by this, the removal of several types of MPs from WWTPs was studied by Talvitie et al. (2017) using cutting-edge final-stage treatments, such as membrane bioreactors, quicksand filters, disk filters, and dissolved air floating. They reported that the removal efficiency rate of the membrane bioreactor was as high as 99.9 %, with MP levels decreasing from 6.9 to 0.005 items/items/l. They also demonstrated that any sized MPs, even the smallest size (20–100 μm), could be removed using fast sand filtration, dissolved air floating, and membrane bioreactors. Notably, the removal

effectiveness was not dependent on MP shape; in particular, the removal of textile fibers, which predominated in both influents and effluents during the treatment process, was successful. Moreover, the FTIR analysis of the samples revealed that the membrane bioreactor significantly reduced the amounts of polymer in the final effluent, highlighting the sorption setup's effectiveness in trapping MPs with different chemical structures (SF 5). However, MPs' influent flow, size, and concentration greatly influenced the removal efficacy of this method. In this regard, porous membranes in biological processes could increase the removal efficacy to 99.9 %. Nevertheless, further study is required to identify the proper efficiency rate under different concentrations and sizes of MPs.

5.5.2.2. Coagulation and filtration. The removal process of large-sized MPs through coagulation is made easily accessible. Through ligand exchange and complication, the methods enable coagulants to bind tiny particles (Ma et al., 2019a). Skaf et al. (2020) experimented with coagulation treatments adopting alum ($\sim 5\text{--}10\text{ mg/l Al}$) with initial turbidity of 16 NTU. They reported that the turbidity was reduced to <1 NTU, and sulfuric acid (H_2SO_4) associated with Al-containing species increased due to additional alum, which affected pH and zeta potential. It was suggested that sweep flocculation could remove microspheres with 20 mg/l surfactants that did not impact coagulation efficiency even at a lower alum dose. However, the efficacy would be impaired due to the high loadings of MPs and alum. Although surfactants effectively impacted PE microfibrils' stability, the fibers could be removed profoundly through coagulation.

MP coagulation by some coagulants like Fe-based coagulants, Al-based coagulants, and synthetic organic coagulants was vividly explored (Ma et al., 2019b; Zhang et al., 2020c). For instance, Ma et al. (2019a, 2019b) investigated PE removal from drinking water using coagulation coupled with ultrafiltration (Fig. 8). In this process, a flat-sheet polyvinylidene fluoride (PVDF) membrane was utilized, and all the membranes were submerged in deionized water for 1 day to eliminate all impurities. UF stirred cell (Millipore 8400, USA) was adopted during the tests, and pressure was controlled at 0.1 MPa with N_2 . $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ stock solutions were diluted with feed water (300 in the stirred UF cell with 0.1 M NaHCO_3) to produce the test solution. Rapid (300 rpm) and slow (100 rpm) mixing phases were allowed for floc growth. Samples were obtained with a hollow glass tube below the suspension surface to capture photos of PE particles and flocs during the coagulation test.

In this method, $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ exhibited better performance than $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ in PE removal. Because the greater zeta potential of Al-based flocs compared to Fe-based flocs made it easier to neutralize negatively

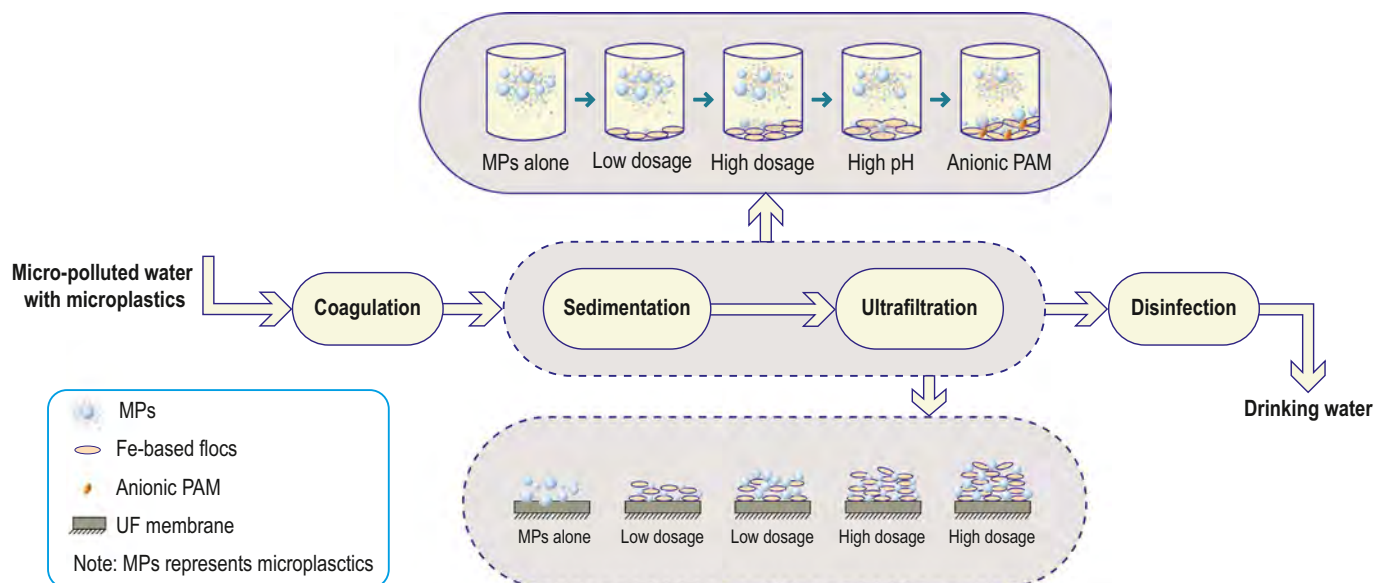


Fig. 8. A schematic diagram of MPs through coagulation associated with the ultrafiltration technique. From Ma et al., 2019a.

charged PE MPs and promoted subsequent sweep flocculation (Mahmud et al., 2022). For the smaller-sized PE MPs, the removal rate was higher. After employing a high dose of AlCl_3 (270–405 mg/l) for the <0.5 mm MPs, a relatively higher removal efficiency (~40 %) was measured. However, natural organic matter, ionic strength, and turbidity level had little impact on the removal efficiency. Studies also reported that pH 6 exhibited high removal efficiency while low removal efficiency was observed at 15 mM Al-based salt dose (below 40 %). Although there was little effect on ionic strength and turbidity level, additional anionic polyacrylamide generated positive Al and Fe-based flocs in the neutral condition, which was one of the key features, but the removal efficiency for all sized MPs through FeCl_3 was considerably low (<15 %). Considering this, researchers employed ultrafiltration and observed that MPs might be removed entirely under a minor membrane fouling. The primary drawback of the coagulation process is that more additional chemicals are needed, which is not environmentally friendly. For example, Al-salt residues in drinking water due to excessive use of Al-based coagulation had an adverse impact on living beings (Sharma et al., 2021).

On the other hand, achieving the high efficiency of traditional filtration of minute-size MPs from a large volume of water and other highly concentrated contaminants remained a challenge (Misra et al., 2020). Thus, ultrafiltration provides low energy consumption, high removal efficiency, and the use of compact plant size (Minténig et al., 2018). Enfrin et al. (2020) investigated MPs and nanoplastic-related fouling of a poly(sulfone) commercial ultrafiltration membrane and observed that >25 % of MPs were adsorbed on the membrane surface after 48 h. However, hydrophilic interactions might have reduced the adsorption rate along with the surface-repellent actions. Indeed, coagulation coupled with filtration is considered one of the primary techniques for WWTPs (Liu et al., 2019a).

5.5.2.3. Agglomeration. A macromolecular network can be generated by silanes through hydrolysis and condensation that can effectively remove MPs from aquatic environments. The fundamental diagram regarding the concept is presented in Fig. 9. A high pH value is preferable for faster hydrolysis and condensation rates. Herbolt et al. (2018a) experimented with subsequent MPs removal along with the formation of alkylsilanetriol

agglomeration. They found that n-alkyl-based chlorosilanes produced hydrochloric acid and silanols for the highly reactive feature while actively contacting with water. Their approach was based on the hypothesis that hydrophobic and Van der Waals interactions between precursors and MPs particles caused the induced production of an inclusion compound with particle growth. The coagulation and inclusion units in an aggregation method commenced the molecular self-organization of MPs. However, a macromolecular hybrid silica network was created through the condensation of silanol groups associated with siloxane bonds for low silanol stability and the catalytic effect of hydrochloric acid. They also found that medium-chain (C3–C5) trichlorosilane was more effective in trapping MPs than their short-chain (C1–C2) and long-chain (>C8) molecules.

The pH-induced sequence made the sol-gel technique applicable to industrial effluent treatment (Zhang et al., 2021a). MPs can be trapped with agglomeration extracts that are likely to be separated from the water and undergo a long-lasting thermal recovery (Zhang et al., 2021a). Herbolt et al. (2018b) experimented with synthesizing functionalized molecular precursors in an inert nitrogen environment to create bio-inspired alkoxy-silyl hybrids that provided linkage regents between the MPs. Hence, agglomeration of the MPs using the sol-gel involved 3-D agglomerates could be removed through filtration process. In addition, the author investigated different bioinspired molecules and concluded that trialkoxysilyl functionalized triazacyclohexane exhibited the best performance to eliminate MPs in the agglomeration process. Since the molecules were not ecotoxicologically relevant, Sturm et al. (2020) assessed the alkyltrichlorosilanes performance using several linear and alkyl-branched groups. However, due to inductive and steric effects, effective hydrolysis and condensation of branched alkyltrichlorosilanes were relatively time-consuming for MPs removal.

5.5.2.4. Electrocoagulation. Electrocoagulation has become a desirable technique, particularly in wastewater treatments (Abbasi et al., 2022). It has been widely employed to treat wastewater that contains dairy waste (Akansha et al., 2020), phosphate (Tian et al., 2017), heavy metals (Pan et al., 2016; Kim et al., 2020), herbicides (Raschitor et al., 2020), PFAS (Ryan et al., 2021) and others. The technique incorporates the advantage of flotation, coagulation, and electrochemical approaches (Kim et al., 2020; Akansha et al., 2020). This method has three stages: electrolytic reaction occurring on the electrode's surface, coagulants' formation in the medium and the contaminant adsorption by the coagulants, and particle separation through the assistance of hydrogen bubbles in sedimentation (Akansha et al., 2020). Fe or Al electrodes are used as sacrificial anodes during treatments, and oxidation occurs in this metal anode when current is applied (Kim et al., 2020). Moreover, metal hydroxide coagulants start forming so that pollutants' surface charge becomes unstable and colloids disintegrate (Perren et al., 2018). A weak atomic force, Van der Waals, appears while coagulants and pollutants come closer. The coagulants then trap suspended particles, creating a sludge layer and oxidation of water to generate O_2 and H_2 gas. The primary reactions are presented in Eqs. (1)–(5):



Although this is an effective treatment, there is little focus on the electrocoagulation of MPs from wastewater treatment. Perren et al. (2018) experimented with removing MPs through electrocoagulation in a 1-l stirred-tank batch reactor for 60 min adopting wastewater containing PE microbeads with various concentrations. The electrodes were placed in

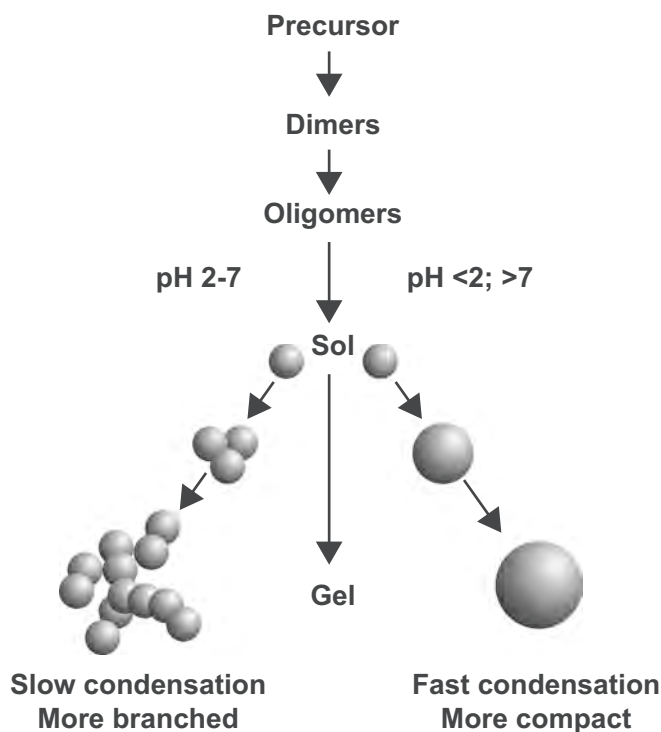


Fig. 9. Schematic diagram of the reaction steps of the sol-gel technique. From Bögershausen, 2004 and Dorcheh and Abbasi, 2008.

the parallel position of the reactor (Fig. 10). Each experiment involved running the reactor for 60 min before turning it off, allowing the contents to settle for 16 h. They observed an influential MPs removal occurred in the pH range of 3 to 10, and the maximum reduction was observed at pH 7.5. The impact of conductivity was also investigated by employing concentrated NaCl (208 g/l representing a conductivity of 7.44–13.75 mS/cm) in the sample. They suggested that the impact of Cl^- concentration on MPs removal was negligible, which was contradictory to the relay on dye-removal efficiency on Cl^- ions (Pirkarami and Olya, 2017). Although HOCl formation aided dye-degradation, it took more time (43 to 60 min) to degrade microbead particles significantly. The electrocoagulation cell's removal efficiency was unaffected by current density. However, operating the cell at least at a current density of 11 A/m² increased the cell's energy efficacy. The energy consumption, $E(t)$, could be calculated where the stable voltage was maintained using a direct current (DC) power supply. Moreover, the specific mass removal per unit energy, $X_s(t)$ (g/kJ) of the cell, could be calculated from $E(t)$ and absolute removal, $M_{\text{abs}}(t)$, using the following Eqs. (6)–(7):

$$E(t) = VIt \quad (6)$$

$$X_s(t) = M_{\text{abs}}(t)/E(t) \quad (7)$$

According to reactor working cost analysis, lower energy was correlated with higher water conductivity, and the process was simple to adopt as electrodes were set up to form coagulants electrically (Perren et al., 2018). Moreover, electrochemical procedures were economically affordable for tertiary treatment due to the less reliance on chemicals and microorganisms (Sharma et al., 2021). In addition, being versatile and energy efficient, this strategy had the lowest potential for secondary contamination. However, electrocoagulation has some drawbacks, including the requirement of sacrificial anode replacement and the electricity demand (Song et al., 2017). Since this method has significant scope, future research is essential for treating MPs from wastewater through electrocoagulation.

5.5.2.5. Advanced oxidation process (AOP). In the AOP technique, UV-oxidation and chlorination are the familiar approaches for MPs removal

from WWTPs (Liu et al., 2021a). In this method, chlorine is pervasively used as a disinfectant since MPs are not entirely resistant to chlorine (Kelkar et al., 2019). The adaptation of chlorine in AOP increases the MPs concentration due to MP cracking (Lv et al., 2019). The chlorination diagram is shown in Fig. 11a. Kelkar et al. (2019) conducted a study on MPs chlorination where extreme circumstances were used to observe the physical and chemical changes. They reported that plastic particles could go through the worst-case situations, which were neither practical nor indicative of the conditions in a designed water environment. Reagent-grade water and sodium hypochlorite solution (12.5 % Cl_2 by weight) was added to obtain the predetermined concentrations, and the solution pH was maintained between 6 and 7. Plastics from virgin and environmentally weathered were subjected for 30 min to a Cl_2 concentration of 5 mg/l at a concentration-time regime (CT value) of 150 mg min/L (WWTP conditions). They also reported that chlorination could break down the existing chemical bonds and generate new ones. Moreover, in their experiments, HDPE exhibited a unique chemical structure containing, C-C-C symmetrical chain, CH_2 bend, C-C-C asymmetrical chain, and CH_2 twist due to compression force (Kelkar et al., 2019; Eichhorn et al., 2001). In chlorination, the newly generated (Cl- CH_2 -C-H) might induce hydrophobicity and toxicity resulting in MPs' accumulating and adsorbing harmful contaminants (Wang et al., 2018a). Although chlorination could be employed in aromatic and aliphatic molecules, a considerable Raman shifting occurred in the aliphatic C—H compartment (from 2901 cm^{-1} to 2940 cm^{-1}). Such shifting indicated the compartment bond's compression force in the direction of increased energy (Eichhorn et al., 2001). In addition, chlorination also altered MPs' physical and chemical characteristics due to the powerful oxidizing feature. However, the alteration of the chemical bond of PP during chlorination treatment was rare. Hence, chlorination was ineffective for PP treatment even at a high dose and with a long exposure time (Kelkar et al., 2019). Along with this, the coexistence of additional contaminants, biofilms, and microbes might change the chemical structure of MPs for chlorine quenching and competitive reactions.

Another MPs treatment is UV-oxidation, which can alter MPs' chemical properties and topography. In an experiment by Cai et al. (2018), a UVA340 lamp was used to simulate natural sunlight to reproduce sound and natural sunlight. Initially, the samples were evenly distributed on glass dishes and

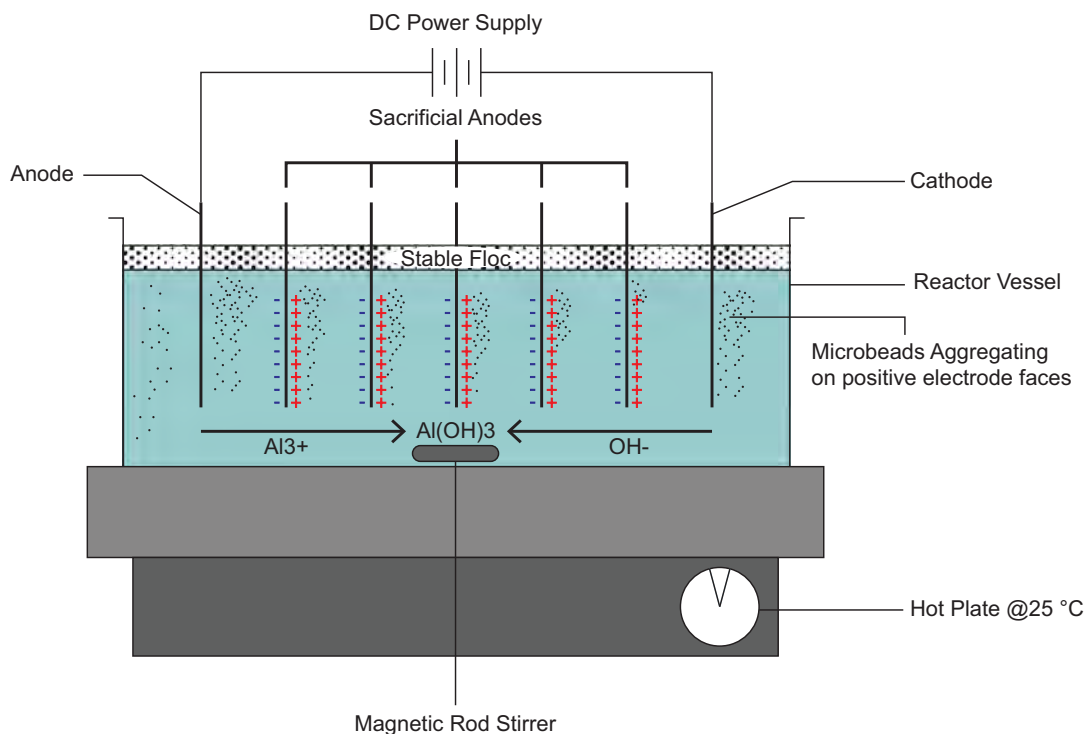


Fig. 10. Diagram of the bench-scale reactor system that was employed in electrocoagulation process. From Perren et al., 2018.

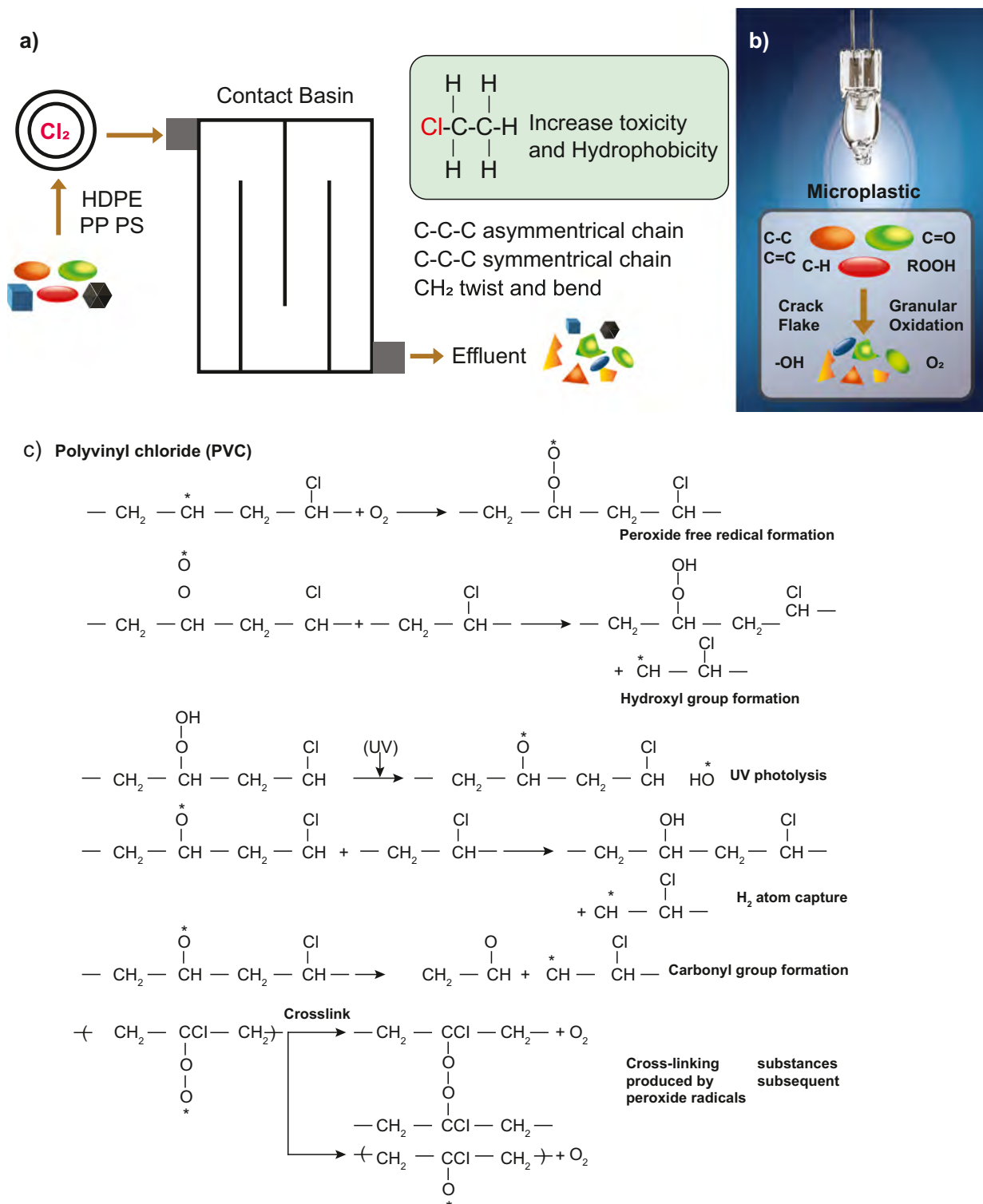


Fig. 11. a) Schematic diagram of chlorination technique. From Kelkar et al., 2019; b) Basic UV-oxidation steps for MPs removal. From Cai et al., 2018; c) UV-oxidation reaction on PVC. From Shi and Zhang, 2006.

immersed in the simulated seawater or ultrapure water solution. This aging experiment was carried out through continuous UV irradiation over three months (each month was considered a cycle). After employing UV-oxidation, the surface of virgin MPs became rough apart from compact texture and homogeneous nature. For the PE, PS, and PP, cracks/flakes, granular oxidation/flakes, and flakes were the frequent situations of degradation (Fig. 11b). They also observed that MPs with fissures and flaws were

comparatively simple to break, producing smaller particles at a nanoscale. Cracks were regarded as stress concentrators and fracture sites as they were an extension of fractures. Flake embrittlement developed on the brittle surface areas of MPs (Cooper, 2012). Under UV irradiation, cleavage of C—H bonds and C—C bonds generated peroxy free radicals (Fig. 11c; Cai et al., 2018). Moreover, in the MPs surface area, UV irradiation could initiate chromophore groups (carbonyl groups (C=O), untreated monomers

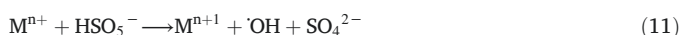
(C=C) and hydroperoxide groups (ROOH) and hydroxyl groups (OH) to produce oxygen-containing free radicals and started chain reactions (Cai et al., 2018; Wang et al., 2020b). These peroxy free radicals generated by UV irradiation would go through subsequent reactions to create crosslinking substances.

Meanwhile, the carbonyl group-containing molecular chain would be degraded to a relatively lower molecular mass (Cai et al., 2018). Yet, the toxicity and intermediates of UV oxidation were less understood. Therefore, an in-depth investigation is required to determine the effects of UV irradiation and environmental variations on MPs breakdown. It is also necessary to understand how salinity and dissolved organic matter influence MPs degradation in WWTPs and natural waters.

One of the most investigated AOPs for environmental remediation is UV/H₂O₂, which produces •OH through UV and H₂O₂ reactions like Eqs. (8) and (9) (Kim et al., 2022a). It was widely used for decomposing recalcitrant pollutants in various studies due to excess redox potentials of •OH of 1.89–2.72 V (Nguyen et al., 2022). Liu et al. (2021b) observed that physicochemical alterations occurred due to UV/H₂O₂. In addition, changes in the PE surface morphology, hydrophobicity, carbonyl index (CI), glass transition temperatures, crystallinity, embrittlement, and average molecular weight were also observed. As anticipated, the chemical oxidation caused by •OH increased surface roughness, crystallinity, and embrittlement. As the samples' surface colour changed, surface oxidation was also observed. Compared to pristine PE, CI rose about two times greater. However, due to chain scission processes and the development of various functional groups (such as polar carbonyl groups) on the surface of treated PE, the average molecular weight and hydrophobicity decreased.



In one stage, peroxymonosulfate (PMS, HSO₅[−]) was activated by heat and catalysis to produce radicals for the MPs treatment. This approach successfully could have the two primary radicals SO₄^{•−} and •OH, which were generated in the following reactions, Eqs. (10) and (11) (Stefan, 2017). The energy from the heat was used to make ions more reactive, and the energy also helped catalysts to activate PMS for distributing major radicals.



Kang et al. (2019) employed the hydrothermal catalytic PMS system to observe the degradation process of cosmic MPs. Through one-pot pyrolysis, they fabricated new nanocomposites by encasing manganese carbide nanoparticles (Mn@NCNTs) in N-doped carbon nanotubes. Under the hydrothermal condition, Mn@NCNTs activated peroxymonosulfate to degrade MPs. The synergistic impact of N-doping, Mn encapsulation, and sturdy helical structure significantly enhanced the catalytic activity of catalysts. Notably, the MPs concentration, catalyst dosage, peroxymonosulfate dosage, and hydrolysis temperature were crucial factors in reaching the removal efficiency at 50 wt% in the Mn@NCNTs/peroxymonosulfate system. For the decomposition of ultrahigh-molecular-weight polyethylene, Hu et al. (2021b) developed a hydrothermal coupled Fenton system where MPs pretreatment was employed to remove the hydroxyl and acid groups from ultrahigh-molecular-weight PE MPs. For this treatment, MPs were added in 2 M NaOH; after vacuum filtration, the obtained MPs were to be dry. For the Fenton oxidation process, the oxidation was performed in a Teflon autoclave, and certain types of MPs, such as HDPE, PVC, PP, LDPE etc., were dispersed in ultrapure water at 1 g/l through vigorous magnetic stirring (1500 rpm). To avoid sludge production and facilitate weight loss measurement along with MPs characterization, the solution was adjusted to acidic condition ($c(\text{H}^+) = 0.2 \text{ M}$). FeSO₄·7H₂O (4 mM) and H₂O₂ (200 mM) were added to the solution to accelerate the reaction and MPs were collected through vacuum filtration. The method achieved 95.9 %

weight loss in 16 h and 75.6 % mineralization efficiency in 12 h. The combination of hydrothermal hydrolysis, a proton-rich environment, and a lot of hydroxyl radical generation was causative factors for the high effectiveness.

Photocatalysis was suggested as a desirable alternative in terms of sustainable development (Ricardo et al., 2021). Photocatalysis is a light-mediated method where photoreactions occur with the assistance of catalysts. Jiao et al. (2020) developed a universal photoinduced C—C cleavage and coupling pathway to transform different waste plastics into high-energy-density C₂ fuels. The method was employed under simulated natural environments such as air, atmospheric pressure, room temperature, pure water, and simulated one-sun irradiation. In this method, plastic particles, such as PE, PP, and PVC, were selectively photoconverted into C₂ fuels without using any sacrificial agents. Realizing plastic degradation to pure CO₂ via photooxidative C—C bond cleavage over the photocatalyst was the initial stage. The resulting CO₂ was further reduced into C₂ fuels in the subsequent stage using the same photocatalyst and the photoinduced C—C coupling process. For instance, single-unit-cell thick Nb₂O₅ layers photodegraded PE 100 % into CO₂ within 40 h, and the generated CO₂ was then further photo reduced to CH₃COOH.

In another experiment, H₂O was converted into H₂ using photoreduction (Xu et al., 2022). At the same time, non-recyclable polymers like PE plastic bags, PP plastic boxes, and PET plastic bottles were photodegraded into CO₂, which was then eventually photo reduced into CO. They also reported that Co-Ga₂O₃ nanosheets with H₂O could effectively photoconvert commercial PE plastic bags into syngas (H₂ + CO). By this method, H₂ and CO production rates were ~647.8 and ~158.3 μmol g^{−1} h^{−1}, respectively. To increase the effectiveness of non-recyclable plastics degrading in the future, it is required to design photocatalysts with high CO₂ reduction activity.

5.5.2.6. Primary sedimentation/grit chamber. Generally, the first stage of WWTP includes the grit chamber and primary sedimentation (Bui et al., 2020). The basic principle and associated units like coarse screens, fine screens, aerated grit chambers, and subsequent stages are shown in SF 6. For the sludge sample, the treatment method using Fenton's reagent was described in the 'Advanced oxidation process (AOP)' section. To remove wastes like wipes and cigarette butts, wastewater was passed through coarse and fine screens. Fine screens were static wedge-wire plates with a 6-mm spacing between their wires, while coarse screens were standard bar screens with 20-mm openings between the bars. Wastewater moved towards the aerated grit chamber and provided coarse air bubbles. In sedimentation tanks, wastewater containing microorganisms (biosolids) and inert materials like MPs moved and settled (Bilgin et al., 2020).

MPs were mainly eliminated during the aeration process at the back of the grit chamber through sedimentation and skimming (Ngo et al., 2019). Liu et al. (2019c) experimented with the primary sedimentation technique, where the abundances of MPs in influent and effluent were 79.9 items/l and 47.4 items/l, respectively, and the removal rate of MPs was 41 %. The process could achieve a high MPs removal rate (66 % in Ziajahromi et al., 2017b; 57 %–64 % in Hidayaturrehman and Lee, 2019). Indeed, Murphy et al. (2016) investigated the performance of the process and found that the average MPs reduced from 15.7 items/l to 3.4 items/l with a removal efficiency of ~78 %. The result was in line with the investigation by Bayo et al. (2020), who reported that the process could remove nearly 74 % of MPs from WWTPs.

Interestingly, high MPs elimination rate (92 %) was observed in most of the major WWTPs in Canada, where particles were fiber types (Gies et al., 2018). Till now, the highest removal MPs rate was 99 %, where the input MPs concentration was 57.6 items/l, and the experiment was carried out by Lares et al. (2018). More than 96 % of MPs in the study were in fiber form, which could be the possible reason for the high removal efficiency. According to Hidayaturrehman and Lee (2019), the initial treatment stage retained more fiber types MPs (76 %–92 %) compared to other MP forms like sheet, microbead, and fragment. The removal of MPs in the pretreatment stage was often highly successful, and the subsequent steps could

eliminate the remaining MPs. However, it is necessary to consider more advanced technologies in subsequent or tertiary treatment stages to remove MPs.

5.5.2.7. Adsorption removal and TiO_2 -based photocatalytic treatments treatment. A novel removal approach was suggested by Tiwari et al. (2020) to remove nano-scale plastic materials from aquatic environments through synthesized Zn—Al layered double hydroxides (LDHs). For the synthesis of Zn—Al LDG, 2 M NaOH was used to coprecipitate the solution of Zn^{2+} (0.75 M ZnCl_2) and Al^{3+} (0.25 M $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) at pH 8. The precipitate was filtered, dried at 70 °C for 1 day, and stored after aging. A series of batch sorption experiments were conducted to get a clear insight into the interaction mechanism and applicability of Zn—Al LDH for removing NPDs from the aquatic system. The interaction between LDHs and nanoscale plastic particles was dominated by physical adsorption, leading to the observation of super reusability. Through this process, the removal efficiency of NPDs was achieved 100 % in the freshwater environment at pH 4; however, removal performance would quickly decrease by adding competing ions to the solution.

In another novel technique, Sun et al. (2020) demonstrated that MPs were adsorbed on a sponge made of chitin and graphene oxide (Chitin-GO). Chitin-GO sponges were synthesized following the freezing-thawing method (Duan et al., 2013). The sponges were placed in deionized water for 1 h, and the water absorption rate was measured. Such a sponge effectively adsorbed various MPs from water with a pH range of 6 to 8. They observed that the efficiency rate was over 70 % for amine-modified and carboxylate-modified PS particles. In addition, some driving forces like hydrogen bond interactions, electrostatic interactions, and π - π interactions were influential factors in adsorption treatments.

In another investigation, Batool and Valiyaveetil (2020) employed CaCO_3 co-precipitation to remove plastics. A modified nanoprecipitation technique was used to create poly(methyl methacrylate) (PMMA) and poly(vinyl acetate) (PVAc) nanoparticles with the fluorescent dye, perylene-3,4,9,10-tetracarboxylic tetrabutylester (PTE) (Fessi et al., 1989). PTE was synthesized according to a previous method demonstrated by Dubey et al., 2016, and PTE stock solution with various polymers was prepared in acetone (50 ml). The complete characterization of the particles was reported using UV-Vis spectroscopy, dynamic light scattering, and SEM. Through this process, the effectiveness rate was 99 % within 30 min. The removal effectiveness of synthetic polymers was significantly influenced by their negative surface charges suggesting that MPs were physically trapped on CaCO_3 surfaces. Surprisingly, researchers achieved an expectable recovery through an adsorption method, although the physical interaction caused particles to adsorb in an unselective manner. However, it is challenging to extract MPs from other suspended contaminants.

Researchers have explored a design regarding the degradative potential of TiO_2 -based micro- and nanodevices for the photocatalytic treatment of MPs (Sekino et al., 2012). Photo-active micromotors have drawn much attention due to their significant potential in water purification, the clean-up of environmental contamination, and medical applications (Zhang et al., 2018a; Eskandarloo et al., 2017). According to Wang et al. (2019c), effective photocatalytic removal of PE MPs from wastewater was achieved using an Au-optimized TiO_2 -based micromotor. Initially, TiO_2 particles (700 nm) were synthesized through titanium (IV) isopropoxide (TTIP) hydrolysis and condensation reaction process (Wang et al., 2018b). At the final stage of the synthesis method, the magnetic Au@Ni@TiO_2 (or Au@Fe@TiO_2) particles were released by adopting ultrasonication. The environmental MPs were extracted following a chemical and enzymatic protocol (Velev et al., 2009), and residues were analyzed through Raman microscopy. The micromotor was propelled by electron-hole production processes that started water and hydrogen peroxide photochemical reactions. However, the major drawbacks of the system were the lack of selectivity and the ability of micromotors to recognize. Therefore, the system was not potentially applicable in real wastewater due to the requirement of low concentration of H_2O_2 to promote the phoretic interactions, which allowed the micromotor to move.

A protein-based N- TiO_2 photocatalyst has recently been reported by Ariza-Tarazona et al. (2019) to degrade PE MPs in both solid and liquid phases. An N- TiO_2 was synthesized according to a method proposed by Zeng et al. (2015). The conventional sol-gel process was utilized to synthesize a second N- TiO_2 material that would be used as a control. A film was produced from the nitrogen-based precursor solution, and subsequent treatment was conducted to crystallize the anatase TiO_2 phase. X-ray diffraction was used to characterize the crystalline phase in N- TiO_2 materials. All photocatalytic tests were carried out in a closed reaction chamber at an ambient temperature for 20 h. In their findings, photocatalytic treatment for the pollutants caused a mass loss of 1.1 % with a constant rate of $12.2 \times 10^{-4}/\text{h}$ in the solid phase after 20 h of treatment. These values were reported to be 6.4 % and $38.2 \times 10^{-4}/\text{h}$ in the aqueous phase. The study also said that interaction between the photocatalyst surface and MPs and the semiconductor's surface area significantly impacted MPs removal efficiency. The structural changes during the mineralization process had also been well-supported by the SEM imageries and FTIR spectra. To avoid the arrest of photocatalysis, environmental factors, the interaction of MPs with N- TiO_2 , and the N- TiO_2 surface area should be carefully controlled and monitored.

5.5.2.8. Magnetic filtration. Magnetic fields are helpful in isolating MPs grafting with magnetic seed substances providing many facilities, including separation ability and less waste sludge (De Vicente et al., 2011). The process consisted of three major steps: forming magnetic seeds, improving the aggregation induced by magnetic seeding, and improving removal efficacy (Wan et al., 2011). Grbic et al. (2019) suggested a magnetic filtration process to remove MPs from different environmental compartments. According to their study, hydrophobization of Fe nanoparticles accelerated the MPs sorption. This was performed through surface modification along with a silane to activate Fe nanoparticles with hydrophobic hydrocarbon tails. In their experiments, hydroxyl groups in the native oxide layer of Fe reacted with the methoxysilane group of hexadecyltrimethoxysilane (HDTMS) to produce siloxane bonds. Notably, HDTMS were employed as magnetic seeds that could modify hydrophobic iron nanoparticles. Hydrophobic iron nanoparticles adhered to MPs surfaces resulting in hydrophobic interaction that allowed MP magnetic isolation (Fig. 12a). Using this method, researchers were capable of removing 92 % of 10–20 μm MPs (PS and PE beads), 81 % of 0.2–1 mm MPs (PUR, PP, PS, PVC, and PE), 93 % of >1 mm MPs (PET, PS, PE, PVC, PP, and PUR), and 78 % of the 0.2–1 mm MPs (PVC, PE, PUR, PP, and PS) from seawater, sediment, and freshwater, respectively (Zhang et al., 2021a).

Tang et al. (2021) reported that the use of magnetic carbon nanotubes (M-CNTs) was attractive due to the notable interaction ability between M-CNTs and MPs. At first, M-CNTs were produced from the suspension of carbon nanotubes and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ solution. Prior to MPs removal tests, various amounts of M-CNTs and MPs were mixed, and MPs were separated from the aquatic solutions by magnetic force after a sharp shake (150 rpm). In this method, some features such as π - π electron conjugation, electrostatic interaction, hydrophobic interaction, complication, and hydrogen-bond interaction were identical to adsorb and eliminated MPs.

On the other hand, Rhein et al. (2019) reported that magnetic seeded filtration (MSF) represented two major steps, hetero-agglomeration, and magnetic isolation, in MPs removal from the aquatic environment. This method used magnetite (Fe_3O_4) as magnetic seeds, and MPs were diffused using ultrasonic techniques. The ferromagnetic matrix was submerged in agglomeration cells to trap the magnetite substances and generated hetero-agglomerates following the suspension transportation into cells (Fig. 12b). The position of both the agglomeration cell and the separation matrix determined a significant difference in the two stages: for separation, the entire cell was submerged in a magnetic field, and the separation matrix was manually lowered into the suspension to cause magnetic separation. The efficiency rate of MPs extraction in this method was 95 %, where ultimate surface charge acted as a vital factor. Low or opposite charged surface potentials were considered to increase or decrease electrostatic repulsion. Moreover, magnetic seed level and ionic strength were crucial elements

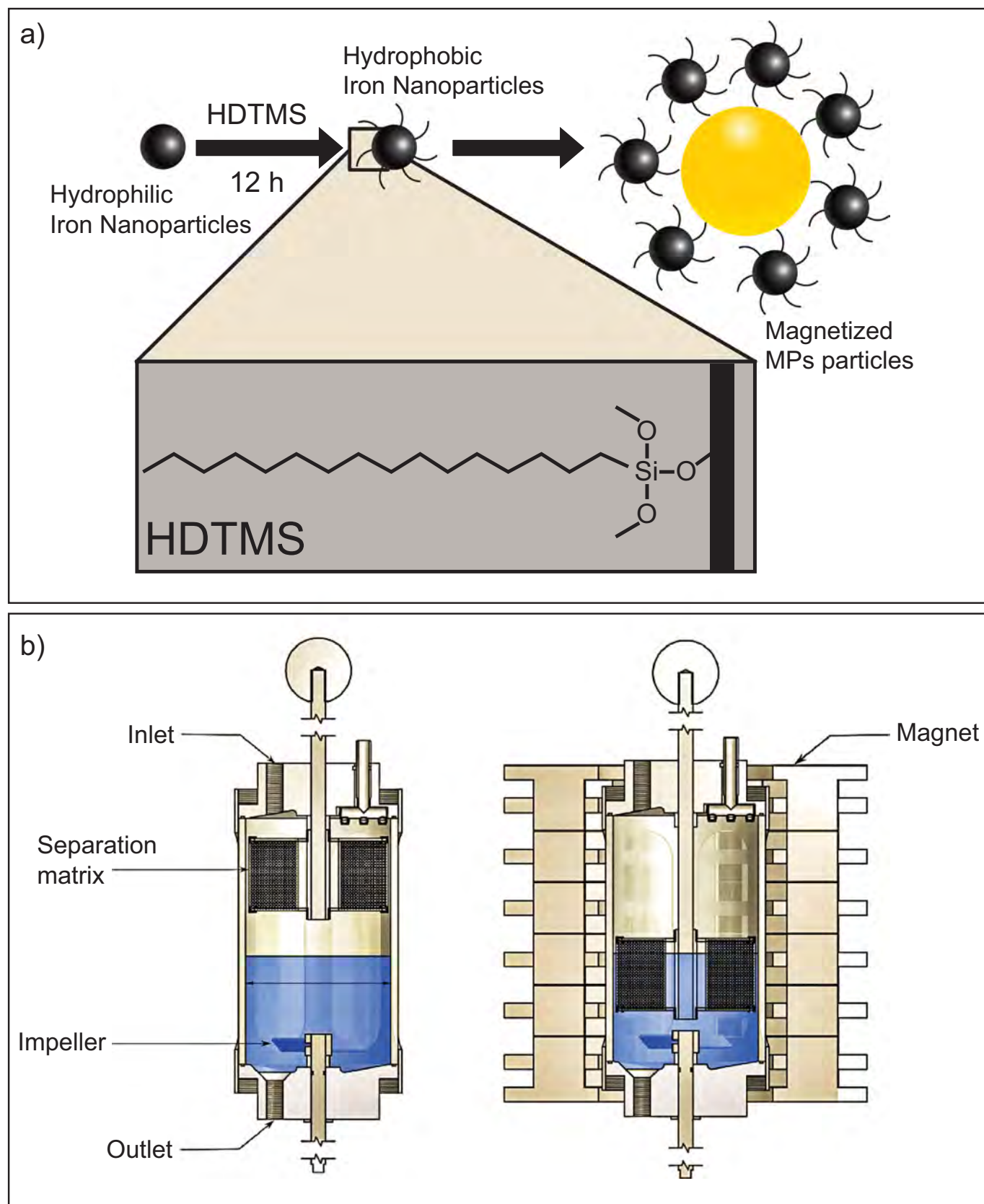


Fig. 12. a) Magnetic filtration device for hetero-agglomeration and MPs separation through a magnetic field. From Zhang et al., 2021b; b) Compartments of magnetic isolation device. From Rhein et al., 2019.

in increasing MPs separation efficiency when electrostatic repulsion occurred in agglomeration partners. The authors recommended magnetic extraction for the MPs removal; however, some potential drawbacks in applying the magnetic removal strategy. For instance, the aggregation behaviour is dominated by the surface properties of magnetic seeds, MPs, and other lipophobic/lipophilic materials (Zhang et al., 2021b). Nevertheless, little research deals with how different elements in the aquatic environment interact, let alone the extensive and various surface reactions of weathering MPs (Jiang et al., 2020). Moreover, this method is more sensitive to MPs' shape and size. For example, the protocol may not be an ideal and sensible methodology for commercial spherical MPs with defined sizes and without MPs films, fragments, and fiber possessing with large size fractions (Hu et al., 2014; Wang et al., 2014). Concerning this, more advanced technologies are required.

5.5.2.9. Oil film extraction (OFE). This method is based on the hydrophobicity property of MPs and is not affected by the density of MPs (Patil et al., 2022). A cost-effective OFE method was introduced by Crichton et al. (2017) using canola oil which was based on oleophilic characteristics. Here, the MPs were allowed to contact the oil, and the mixture was left to settle until the oil layer was separated completely on top of the water layer. They found that the removal rate was 96 % through this process. Following this, Mani et al. (2019) used castor oil to eliminate MPs from sediment and aquatic system, and the average elimination rate of MPs was 95 %. However, according to Lares et al. (2019), solid samples could impede extraction funnels during isolation. When hydrophilic materials were discharged as underflow, the extraction funnel limited the large size of recovered MPs (Kim et al., 2022b). The use of an olive oil-based approach was one of the potential MP extraction methods from sediment containing PET, PU, PVC, PC, and PE, and the recovery rate was over 90 % (Scopetani et al., 2020).

This extraction method requires extra digestion with high biogenic levels before oil and environmental matrix shaking. Digestion before extraction is effective as the oxidized digestion process can impede OFE and cause hydrophilic surface on MPs (Truc and Lee, 2019). Moreover, remaining oil traces can interfere with oil film extraction, requiring different cleaning methodologies with ethyl alcohol and hexane.

5.5.2.10. Froth flotation. Froth flotation has been widely employed in the recycling industry to extract minerals based on the strength of interactions of specific particles as well as the adhesion of particles to the air (hydrophobic particles) or the water (hydrophilic particles) (Junhao et al., 2021). In this method, the plastics (hydrophobic materials) attached to the bubble surface subsequently carry them upward as well as the air-liquid interface (Nguyen et al., 2019) (SF 7). Three plastic flotation methods were common: reagent adsorption, gamma flotation, and surface modification (Fraunholz, 2004). Apart from the other two methods, surface modification was time-consuming and was affected by unrealistic plastic floatability, which led to complexity in understanding surface modification and its actual employment (Jiang et al., 2020).

Bolto and Xie (2019) discussed the implementation of functionalized polymers in treating sewage disposals that might effectively remove grease, surfactant, oil, and algae. Some other studies reported that nano-sized PS particles with hydrophobic properties could deposit on the air bubble and water interface to concentrate hydrophobic matrices (Dong et al., 2016; Yang et al., 2013). This method was supported by Enfrin et al. (2019) and Sun et al. (2019) as an MPs-targeted treatment technique to reduce the effect on treating units, enhance WWTP efficacy, and decrease MPs emission from wastage.

In addition, this method could use several wetting agents such as saponin, sodium lignin sulfonate, tannic acid, methyl cellulose, and calcium lignin sulfonate. In a Denver cell with 3 dm³ capacity and a 600 rpm rotational speed, flotation tests were conducted by Pita and Castilho (2017). The operation was carried out with freshwater, and tannic acid wetting reagents, and the pH remained at the range of 7–7.2. It is mentionable that plastic recovery was decreased along with increasing tannic acid. Contact

angles were determined carefully using a direct method as flotation performance increased according to the increase of contact angles. The study revealed that plastic removal ranged from 82 to 98 % when tannic acid was used at <1 mg/l.

Talvitie et al. (2017) designed an advanced MPs extraction technique from WWTPs based on dissolved air flotation (DAF). Water was pumped to a flotation tank at 1-atm after being saturated with air under high pressure, producing dispersed water. Dispersed water provided released air bubbles that adhered to the suspended solids and caused them to float to the surface, where they were removed by skimming. A flocculation chemical, Polyaluminium Chloride (PAX), was added to the wastewater at a dosage of 40 mg/l prior to flotation to improve flocculation, resulting in an MP removal rate of 0.95 %.

Huang et al. (2017) treated MPs with microwave-assisted potassium permanganate modification (MPPM) to gain surface modification. After surface modification, the particles were taken out of the potassium permanganate solution and employed in the flotation studies after being rinsed with tap water and soaked for a predetermined period. During the flotation trials, terpeneol was introduced to the flotation column to act as a frother. At the end of the flotation test, the recovery rates of PVC/PMMA and submerged polycarbonate (PC) were 98.95 % and 89.61 %, respectively.

Zhang et al. (2021a) used coagulative colloidal gas aphyrons (CCGAs) that were facilitated by cetyltrimethyl ammonium bromide (CTAB) and anhydrous aluminum chloride to reduce PMMA and PS at the micron scale from aquatic systems. Due to the synergistic effects of flotation and coagulation, it was a practical approach for MPs removal despite the lack of specialized apparatus and significant reagent consumption (Zhang et al., 2021b). In the beginning, PACl was synthesized by gradually titrating the NaHCO₃ solution into the vigorously agitated AlCl₃ solution. Then, the cationic surfactant of CTAB and the coagulant of polyaluminum chloride (PACl), which served as the frother and bubble modifier, were combined to create the bubble formation solution. Using this method, Imhof et al. (2012) reduced 55 % of MPs from sediment, and the observed density separation method was a better option. Meanwhile, Nguyen et al. (2019) noted that unpredictable factors could make MPs extraction difficult. In addition, some researchers observed an unstable efficacy of this method that warranted future research.

5.5.2.11. Density separation (DS). DS method is one of the most popular effective techniques for MPs separation from the soil (Junhao et al., 2021). Liu et al. (2018) reported that soil was the first treated material through ultrasonic as each particle in the bulk sample sank or floated due to their relative density (Zhang et al., 2018b). Although NaCl was widely used in suspension solutions, the NaCl solution (1.2 g/cm³) could not extract relatively high-density plastics such as PET (1.38 g/cm³) and PVC (1.35 g/cm³). (Ruggero et al., 2020). In this regard, researchers suggested employing a saturated solution of sodium iodide (NaI), zinc chloride (ZnCl₂), calcium chloride (CaCl₂), sodium bromide (NaBr), and zinc bromide (ZnBr₂) (Han et al., 2019). The use of a suspension medium depended on availability, price, recovery rate, and environmental effect. For instance, using NaI and ZnCl₂ solutions was not environmentally convenient. In this procedure, a 1.7 g cm⁻³ density ZnCl₂ solution was prepared by dissolving 72.07 g of ZnCl₂ in 46.84 mL of distilled water. This process had six stages, where suspension medium was added into a beaker with soil. The supernatant containing MPs was drawn into another beaker and passed into the filtration procedure after stirring and sedimentation (Fig. 13). MPsmight adhere to the beaker wall during emptying or transferring between the filtration process and the beaker; therefore, transparent and smooth glassware was necessary for the procedure.

Coppock et al. developed Munich Plastic Sediment Separator (MPSS) that could separate MPs from the solution medium and directly transfer them to the filter. Firstly, ZnCl₂, (NaCl), and (NaI) solutions were made by dissolving the salts in ultrapure water to reach densities between 1.2 and 1.8 g cm⁻³. A compact extraction unit was designed to be adequately mixed in a single step and rapidly cleaned to remain free from cross-

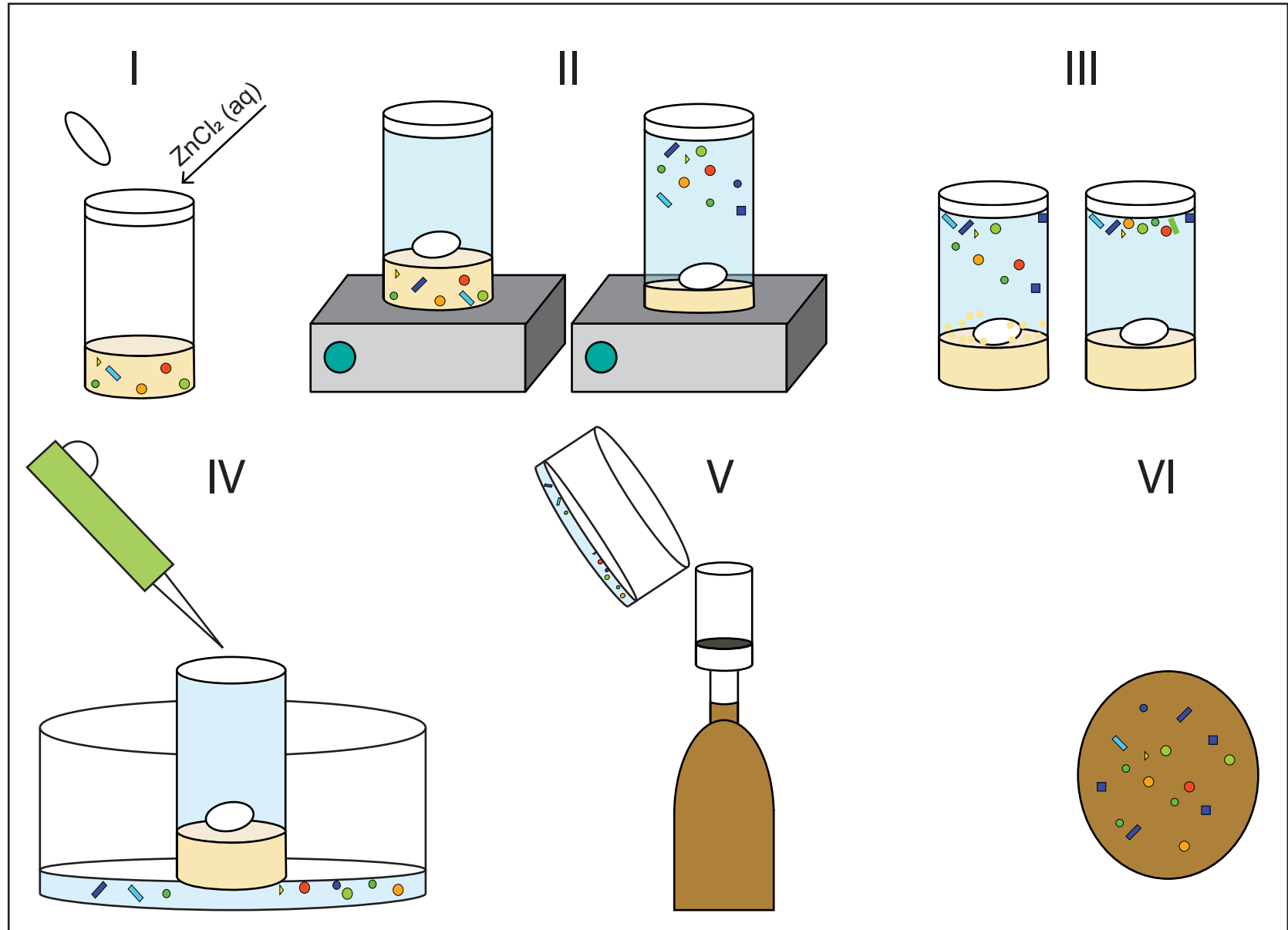


Fig. 13. The basic scheme of density separation method I) the addition of ZnCl_2 solution and agitator along with a density $>1.6 \text{ g/cm}^3$, II) Three times of stirring suspension III) allowing sediment to settle down and MPs to float, IV) the addition of extra ZnCl_2 slowly, V) filtration and rinsed with deionized water after treatment, VI) collection and MPs free from ZnCl_2 . From Konechnaya et al., 2020.

contamination. Moreover, sediment-microplastic isolation (SMI) was constructed to achieve free movements of particles preventing MPs from being trapped in the unit. By this method, the MPs' recovery rate was 92.5–98 %. In an experiment, conducted by Imhof et al. (2012), the technique avoided excessive MP loss without emptying repetition and separation, and the recovery rate range was from 95 % to 100 %.

After being motivated, Zobkov and Esiukova (2017) followed the original configuration of MPSS and procedures proposed by Imhof et al. (2012). They utilized artificial reference particles (ARPs) to monitor the MPs extraction efficiency. They observed that MPs' separation efficiency drastically dropped to 13 %–39 % due to the complexity of extracting organic matters from MPs. Meanwhile, Mahat (2017) followed the Bauta Microplastic-sediment separator (BMSS) made from glass and metal, where a suspension medium mixture of CaCl_2 and ZnCl_2 was used. BMSS had three major components: the sediment chamber, the glass column, and the separation chamber. The separation chamber could be reversed to obtain the light-density sample for filtration. Four methods were optimized for their procedure for separation, filtration, and chemical digestion, providing an 82–100 % efficiency rate.

Claessens et al. (2013) improved a novel device based on “Barnett's fluidized sand bath” that used gas and liquid flow upward to separate lighter particles in the soil from heavier ones before suspending them in NaI solution. In their experiments, the water flow rate was regulated to maximize extraction effectiveness and reduce sand contamination of the sample. Their device was made of PVC; therefore, it could not separately identify other commonly available PVC (Liu et al., 2019b). Both methods could process large sample numbers, and the extraction rate was >90 % for >35 μm MPs. However, it was unclear whether they were as effective for <35 μm MPs. In addition, these techniques were only effective in loose soil, and it was unclear if they could separate MPs in cohesive soil.

Later on, Nuelle et al. (2014) developed a two-step technique based on the air-induced overflow method and fluidization in NaCl solution. Various solvents (H_2O_2 , NaOH, and HCl) were tested to dissolve biogenic materials of both animal and plant origin. Some specific plastic pellets and raw plastic materials were used to be resistance against H_2O_2 , NaOH, and HCl. The basic concept was to use a combination of sediment fluidization in a lower-density salt followed by MP flotation in a higher-density salt. Using a saturated NaCl solution, the air-induced overflow (AIO) method was created to pre-extract sediments, and the range of recovery rate was 68–99 %. It is clear that the removal rate of MPs significantly depends on the practice employed, the surrounding media, and the physical and chemical structures of MPs. The summarization of different MPs' removal methods is presented in ST 1, which will help to decide to adopt the practices to remove MPs effectively. In addition, some adopted MPs' removal techniques in different countries are also summarized in Table 5. For instance, USA employed filtration and chemical digestion using 0.43 μm mesh size for fiber, PS, cellulose, and nylon, and the average removal efficiency rate was 89.4 % (Conley et al., 2019).

6. Recommendations and future work directions

There is a growing body of literature on the toxicity of MPs in various aquatic organisms and MPs removal from the aquatic environment. However, more studies are required to fill the existing research gaps to better understand MPs' toxicity with other additives and removal strategies. The present study identified some potential research gaps and suggestions concerning these issues:

- Due to the complexity of the MP pollution issues, hybrid solutions such as an amalgam of several policy frameworks are needed to bring about the expected improvements.
- Although MP toxicity has been studied in laboratory experiments, the shape, size, and concentration of investigated polymers in a controlled setting are very different from those of MPs in the natural aquatic environment. More studies are required considering the actual situation in the future.

- There is insufficient data for freshwater species. Freshwater species are severely suffering from MP toxicity compared with marine species.
- MPs could transport some hazardous compounds, and it is essential to identify the level of the compounds that exert toxicity along with MPs to aquatic species.
- Effective waste disposal techniques are to be developed to improve the aquatic environment to reduce plastic pollution (Hartmann et al., 2019).
- Most of the studies were carried out at a particular stage of organisms. More research, especially on different life cycle stages and cross-species response, is still needed to improve our understanding of the toxicity of MPs in aquatic organisms.
- It is essential to check for MPs in other organs, like the eviscerated flesh in fish. Moreover, the neurotoxicity of MPs in different aquatic organisms can be under consideration.
- There is a significant research gap regarding the toxicity of MPs additives in zooplankton.
- Advanced management approaches should be developed, including recycling, reusing, reducing, and recovering plastic waste, as a significant research gap exists.
- It is crucial to investigate the biological transport and metabolism of MPs in aquatic organisms and the interaction between biomolecules and MPs to understand the harmful mechanisms of MPs at the cellular and molecular levels.
- Developing and standardizing appropriate analytical methods for identifying and removing NPs and plastic additives is necessary.
- There is a considerable research gap regarding the potential Trojan Horse impacts of MPs on the biological response of aquatic organisms.
- To cover a wide range of MPs in the aquatic environment, developing more reliable technologies for MP detection and quantitative analysis is crucial.
- More research is required to improve the performance of MPs removal in membrane-based treatment by reducing membrane abrasion and fouling.
- Adaptation of dynamic membrane technology in membrane bioreactors should be encouraged to enhance MPs removal efficiency.
- It is vital to do more studies regarding the MPs removal from the sludge phase through biological treatment methods.
- The use of solar energy should be actively investigated for commercial-scale photocatalysis treatment plants in addition to UV irradiation.
- New and cathodic materials should be created for effective MP removal in the Fenton method.
- More potent and effective photo-catalysts should be synthesized to remove MPs through photocatalytic degradation.
- More bio-inspired materials and their affordable synthesis pathways should be looked for sol-gel agglomeration approach
- It is necessary to improve more active anodes for the electrocoagulation method.
- Hybrid treatments should be specifically developed for the removal of MPs.

7. Conclusions

Wide applications, increased production, and improper disposal of plastics have resulted in serious global environmental pollution. As a consequence, MPs significantly affect aquatic organisms due to their bioaccumulation properties. MPs can carry other legacy and emerging contaminants, such as heavy metals, PAHs, DDT, PFAS, etc. The adsorption of different contaminants on MPs depends on the physical and chemical properties. It is notable that the MPs toxicity varied from species, concentration level, and associated chemical additives. In fishes, the accumulation of MPs along with additives changed feeding behaviour, enzyme function, and growth. Moreover, neurotoxicity and oxidative stress were also observed in fish. ROS formation and growth retardation were common impacts in microalgae that were pervasively observed. Potential effects on zooplankton

Table 5
Different MPs removal treatment processes associated with other information of various WWTPs around the world.

WWTP Country	Treatment method	Sample volume/ MPs concentration	Used mesh size	Spectroscopy tools	Polymer size	Polymer type/composition	MPs influent (MPs/l)	MPs effluent (MPs/l)	Efficiency rate (%)	References
Thailand	Pilot-scale ultrafiltration, Primary, secondary, and tertiary	20 L	0.1 μ m	FTIR	0.05–5 mm	Fiber, PET	16.55 \pm 9.92, 77 \pm 7.21	3.52 \pm 1.43, 10.67 \pm 3.51	96.97	Tadsuwan and Babel (2022)
UK	Secondary and tertiary	301–10,380 items/g	178	μ FTIR	25–178 μ m	PET, PP, and PE	955–17,214	2–54	99.8 %	Horton et al. (2021)
Turkey	Primary, secondary, and tertiary	10 L	26 μ m, 6 mm	FTIR	<500–3000 μ m	PP, PS, PE, Acetate, Cellulose, fiber	1.5, 2.6 and 3.1	0.6, 0.7 and 1.6	55–97	Akarsu et al. (2020)
Thailand	Screening, grift chamber, secondary treatment	5–10 L	0.33 and 4.75 mm	FTIR	0.3–5 mm	Fiber (32–57 %), PE, PS, PP, and polyacrylate	12.2	2	–	Hongprasith et al. (2020)
Spain	Primary and secondary treatment	0.47–13 L	0.45 μ m	FTIR	400–600 μ m	PP, PET, LDPE, HDPE, PMMA, PEP, PS, NYL, PUR, BPI, RBB, MCR, AC	15.70	0.25	90.3 %	Bayo et al. (2020)
USA	Filtration, chemical digestion, counting, and characterization	Influent: \sim 0.5 L, Effluent: \sim 1.5–15.5 L	0.43 μ m	μ FTIR	60–418 μ m, >418 μ m	Fiber, PS, cellulose, nylon	2.5	4.8	89.4 (average)	Conley et al. (2019)
China	Grit chamber, primary and secondary sedimentation	30 L	5 mm and 50 μ m	μ FTIR	681.46 \pm 528.73 μ m	Microfiber, PP, PET, and PS	12.03 \pm 1.29	0.59 \pm 0.22	95.16	Yang et al. (2019)
Hong Kong	Chemical-induced primary treatment/Secondary	100–200 L	100 μ m	ATR FTIR	0.1–0.5 mm	Fiber, hexabromocyclododecane, polybrominated diphenyl ethers	1.01–2.06	0.27–0.4	60.4–86.9	Ruan et al. (2019)
Italy	Tertiary, screening, grit and grease, biological and sedimentation	30 L	63 μ m	μ FTIR	0.5–0.1 mm	acrylonitrile-butadiene, PE, ethylene-propylene, PES, PS, PA, PUR, and PP	2.5 \pm 0.3	0.4 \pm 0.1	84	Magni et al. (2019)
Korea	Tertiary	Influent: 10 L, Effluent: 100 L	100, 200 μ m	ATR FTIR	106–300 μ m, >300 μ m	Fiber and fragment	60.16	0.86	98.5 (average)	Lee and Kim (2018)
Denmark	Secondary, filtration, ultrasonic treatment	1 L	Influent: 500 μ m, 1 mm and 2 mm, effluent: 10 μ m	Focal plane array FTIR	10–500 μ m	Acrylate, PE, PP, PE-PP, PUR, PVC, EVA, PA, PS, PVA, SAN, Pest, and VAC-acrylic	7216 (median)	54 (median)	99.3	Simon et al. (2018)
Finland	Screening, grit separation, primary clarification, biological treatment, and sedimentation	Effluent: 4–30 L	260 and 500 μ m	FTIR	<250 μ m	Fiber, fragment, PES, PA, PE, PP, and PET	57.6	1	98.3	Lares et al. (2018)
Australia	Primary, secondary, tertiary, and reverse osmosis (in-situ)	3 and 200 L	25–500 μ m	FTIR	60–500 μ m, >500 μ m	PET and PS	2.2	Primary: 0.28, Secondary: 0.48 and tertiary: 1.54 reverse osmosis: 0.21	90	Ziajahromi et al. (2017a, 2017b)
Germany	Filtration	0.39–1 m ³	10 μ m	Focal plane array μ FTIR	20 μ m	PE, PS, PA, PEST and styrene acrylonitrile (SAN), PUR, PVC	73 (average)	0 to 9 \times 10 ³	97	Mintenig et al. (2017)
Netherland	Filtering	1 L	0.2 μ m	FTIR	10–300 μ m and 300–5000 μ m	Fiber, foil, sphere	73 (average)	65 (average)	72	Leslie et al. (2017)
Scotland	Secondary	10 L	65 μ m	FTIR	0.598 mm	PET, PA, PS, PES, PVA, PVC, PVE, PUR, PS acrylic, PV acrylate, polyaryl ether, alkyd, and acrylic	15.70	0.25	98.41	Murphy et al. (2016)
USA	Filtering, tertiary treatment	2 \times 10 ⁵ L (average)	45, 180, and 400 μ m	FTIR	10–300 μ m, 100–600 μ m	Fragment, PE, and PP	1	373 items/ 4.23 \times 10 ⁵ L	99.9	Carr et al. (2016)
USA	Secondary and tertiary	Influent: 1–2 L, Effluent: 34–38 L	4.75, 0.85, 0.3, 0.106, and 0.02 mm	Stereomicroscope	0.58 mm	Fiber, fragment, paint chip, and microbead	133	Secondary: 5.9 SAL/L, Tertiary: 2.6 SAL/L, MBR: 0.5 SAL/L	95–99 %	Michielssen et al. (2016)
Finland	Filtering, primary and secondary sedimentation	0.3 L	20, 100, and 200 μ m	Microscope	–	Synthetic particle and fiber	Synthetic substance: 430, fiber: 180	Synthetic substance: 8.6, fiber: 4.9	Synthetic substance: 98, fiber: 97.3	Talvitie et al. (2015)
France	Screening, grit and oil removal, settling, biological treatment	Automatic sampler	1.6 μ m	Stereomicroscope, FTIR	100–5000 μ m	Synthetic fiber	260–320 \times 10 ³ items/m ³	14–50 \times 10 ³ items/m ³	88.97	Driss et al. (2015)

included growth retardation, increased mortality, changes in eating habits, lipid build-up, and lower reproductive function. Additionally, MPs and additives have toxicological effects on polychaetes, including neurotoxicity, cytoskeleton instability, decreased feeding rate, growth, survival, and capacity to burrow, weight loss, and elevated mRNA transcription rates. Some strategies like control of MPs' sources, limiting MPs production, and enforcing legislation against the enormous use of MPs can be adopted to minimize MPs' concentration in the aquatic environment. Notably, certain fungi, bacteria, and algae can degrade plastic polymers effectively, and the biodegradation method is eco-friendly; however, this method is slow in degradation kinetics. Therefore, engineered separation strategies have been widely employed. Among eleven advanced treatment techniques, coagulation and filtration, electrocoagulation, oil film extraction, advanced oxidation process (AOPs), adsorption removal technique, and density filtration method are commonly used due to their high removal efficiency rates. Nevertheless, more studies regarding MPs toxicity in aquatic organisms and more efficient and comprehensive scale removal technologies are required in future research.

Ethics approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

All of the authors consented to publish this manuscript.

Code availability

Not applicable.

CRediT authorship contribution statement

A. S. Shafiuddin Ahmed: Conceptualization, Data extractions, Investigation, Supervision, Writing- original draft, Writing – review & editing.

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Data availability

No data was used for the research described in the article.

Declaration of competing interest

The authors declare that they have no conflict of interest during this study which could have appeared to influence the work reported in this manuscript.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.162414>.

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