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Removal of microplastics in unit processes used in water and wastewater treatment: a review

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Abstract: Many tons of micro- and nano-sized plastic particles enter the aquatic environment every year, due to increasing plastic production, with the consequent risk of microplastics contaminating our environment. Addressing this multifaceted threat requires innovative technologies that can efficiently remove microplastics from the environment. Therefore, there is an urgent need to study the efficiency of the removal of microplastics by different water and wastewater treatment technologies. After short overviewed the source, occurrence, and potential adverse impacts of microplastics to human health, we then identified promising technologies for microplastics removal, including physical, chemical, and biological approaches. A detailed analysis of the advantages and limitations of different techniques was provided. According to literature data, the performance of microplastics removal is as follows: membrane bioreactor (>99%) > activated sludge process (~98%) > rapid sand filtration (~97.1%) > dissolved air floatation (~95%) > electrocoagulation (>90%) > constructed wetlands (88%). Chemical treatment methods such as coagulation, magnetic separation, Fenton, photo-Fenton and photocatalytic degradation also show moderate to high efficiency of microplastics removal. Hybrid treatment such as the MBR-UF/RO system, coagulation followed by ozonation, adsorption, dissolved air flotation, filtration, and constructed wetlands based hybrid technologies have shown very promising results in the effective removal of microplastics. Lastly, research gaps in this area are identified, and suggestions for future perspectives are provided. We concluded this review with the current challenges and future research priorities, which will guide us through the path addressing microplastics contamination.

Introduction

Research on the occurrence and fate of microplastics (MPs) in the aquatic environment has been gaining momentum worldwide, especially in the last decade (Andrady 2011, Hidalgo-Ruz et al. 2012, Mrowiec 2017), and has considered the increase in plastic production and the consequent risk of MPs contaminating our environment. Global plastic production is over 380 million tons per year, while recycling rates are much less than the plastic waste generated, accounting for only 9% of all discarded plastic waste (Badola et al. 2022, Lv et al. 2019, Mrowiec 2018, Padervand et al. 2020). Plastics are used primarily in packaging production (40.5%), civil engineering and construction (20.4%), automotive needs (8.8%). Other uses of plastics include applications in the furniture industry, in the production of electrical and electronic equipment, medical applications and other (Plastics Europe 2022).

The potential impact on humans (via for example fish or sea salt ingestion) includes respiratory irritation, obesity, cardiovascular disease, asthma and cancer. The potential toxicity of microplastics arises from unreacted monomers, oligomers and chemical additives leaked from the plastic in the

long run (Thompson et al. 2009). Monomers and oligomers are both able to migrate from food packaging. As the concentration of the residuals reaches specific limits, they can be potentially absorbed by human bodies via different pathways. For instance, the presence of PS residuals in food materials is reported to cause serious health issues, while epoxy resins made of bisphenol A are absorbed by living tissues, then interfere with the rate of cell division (Thompson et al. 2009).

MPs can be discharged into the environment from the plastic industries, such as through leakages and accidents during transportation, wear of plastic items, use of personal care products such as toothpaste, dishwashing liquid and shower gel (Magni et al. 2019), and from synthetic textiles during the laundry process (Napper and Thompson 2016). MPs dumped into various surface water sources and found in municipal wastewater commonly originate from the aforementioned sources (Badola et al. 2022), and they are then discharged into wastewater treatment plants (WWTPs) (Durenkamp et al. 2016) through multiple pathways.

None of the current water and wastewater treatment technologies are designed to remove plastic particles because they have been developed to remove and neutralise

dissolved and suspended pollutant, and solid waste (Vuori and Ollikainen 2022, Mason et al. 2016). Most plastics are resistant to water, some absorb water slightly, and only a few dissolve. Extreme weather and coastal landfilling are ways for microplastics to enter the water environment. By analysing the matrices, these microplastics (including polystyrene, polyethylene and polypropylene) are identified from water, sediment and organisms, showing massive dispersions in the water environment (Kazour et al. 2019). Wastewater treatment technologies are commonly based on mechanical, biological and chemical processes, which incidentally also separate litter particles through filtration or attachment to precipitated nutrients and microbial flocs (Vuori and Ollikainen 2022, Talvitie et al. 2017a, Lv et al. 2019). The more effective water and wastewater treatment is in removing MPs, the more particles are separated into the sludge, increasing its potential for contamination (Lares et al. 2018; Lv et al. 2019). Therefore, it is urgent to study the efficiency of removal of MPs by different water and wastewater treatment technologies and understand their removal mechanism to reduce the amount of MPs entering the natural water system. However, there are few studies that summarise the removal mechanisms of MPs from critical water and wastewater treatment technologies.

Several review articles are available focused on MPs, especially on the occurrence of MPs in the marine environment, MPs analysis, and MPs remediation technologies. However, none of these review articles provides a systematic overview of MPs removal in unit processes of water and WWTPs. Hence there is an urgent need to study how microplastics interact with each and every unit process of WWTPs. This review provides a critical discussion on various techniques for microplastic particles' separation from aquatic environments. In addition to the separation methods conventionally utilised in the wastewater treatment process, more recent advanced separation techniques such as membrane bioreactors, magnetic-based separation, micromachines, and degradation-based separation, together with the advantages and limitations of each technology, are considered. The inclusion of both conventional and innovative WWTP process configurations in the study provides insights into which unit processes have the greatest potential to remove MPs and can be used in the future to achieve a reduction of MPs levels in the environment. In addition, we examined which MPs types (size and shape) were removed and which were left in the final effluent after treatments. The challenges and limitations of conventional techniques as well as the advantages of advanced techniques to separate small micron-size plastic particles from water are also presented and discussed. This review was expected to provide useful information to suggest improvement and highlight the further research areas of MPs removal technologies that can be employed in wastewater treatment.

Microplastics source and occurrence

Nowadays, 98% of MPs are retained by wastewater treatment plants (WWTPs) but MPs with a size smaller than 20 μm and "nanoplastics" NPs are not retained; therefore, WWTPs plants are considered to be one of the main sources of plastic pollution in wastewater effluents (Carr et al. 2016, Murphy et al. 2016, Malankowska et al. 2021).

Firstly, fibres lost from textiles during washing (Hernandez et al. 2017) and plastic beads used for exfoliation or purification in cosmetics and personal care products enter WWTPs through domestic discharge systems. The use of detergent appeared to affect the total mass of fibres released to the environment, yet the detergent type or overdosing of detergent did not significantly influence MPs release. Despite different release quantities, the overall microplastic fibre length profile remains similar regardless of wash condition or fabric structure, with the vast majority of fibres ranging between 100 and 800 μm in length irrespective of programme selected on the washing machine (Hernandez et al. 2017). This indicates that the fibre staple length and/or debris encapsulated inside the fabric from the yarn spinning could be directly responsible for releasing stray fibres.

Secondly, industrial plastics used in surface blasting, molding and many other processes are discharged into municipal wastewater collection systems before entering WWTPs (Gies et al. 2018, Long et al. 2019, Magnin et al. 2019).

The third factor responsible for MPs contamination in WWTPs is the wet sedimentation process. Fine plastic debris found in the atmosphere or in concrete and highway structures that results from the breakdown or abrasion of other plastics, such as packaging, textiles and tires, can enter wastewater through stormwater runoff (Kole et al. 2017, Long et al. 2019, Mintenig et al. 2017). Car tires release wear particles through mechanical abrasion. Wear and tear from tires significantly contributes to the flow of MPs or microplastics into the environment. The estimated per capita emission ranges from 0.23 to 4.7 kg/year, with a global average of 0.81 kg/year. The emissions from car tires (100%) are substantially higher than those of other sources of microplastics, e.g., airplane tires (2%), artificial turf (12–50%), brake wear (8%) and road markings (5%). Emissions and pathways depend on local factors like road type or sewage systems. The relative contribution of tire wear and tear to the total global amount of plastics ending up in our environment is estimated to be 5–10%.

Finally, WWTPs can receive MPs from landfill leachate, where due to harsh environmental conditions, landfilled plastic waste is fragmented into MPs, which are then transferred with leachate discharge to enter WWTPs (Zettler et al. 2013). He et al. (2019) investigated twelve leachate samples from four active and two closed municipal solid waste landfills. MPs were found in all the landfill leachate samples. In total, seventeen different types of plastics were identified in the leachate samples with calculated concentration ranging from 0.42 to 24.58 items/L. Polyethylene and polypropylene were the predominant polymer types. 99.36% of MPs were derived from the fragmentation of plastic waste buried in landfills. The size of 77.48% of the microplastics was between 100 and 1000 μm . The study shows that the generation, accumulation and release of MPs in landfills is a long-term process.

Along with wastewater, MPs can enter the environment via sewage sludge. Sewage sludge can contain from 20 to more than 180 particles of MPs per gram of dried sludge, depending on sludge management and testing methods (Lares et al. 2018, Talvitie et al. 2017a). Due to their relatively high phosphorus and nitrogen content, in many countries sludge is applied to agricultural land or used in landscaping (Nizzetto et al. 2016).

According to Horton et al. (2017), the amount of MPs in terrestrial environments can be 4 to 23 times higher than in the oceans. In addition, airborne MPs that have been emitted by the plastics industry and vehicles also enter WWTPs (Mintenig et al. 2017). WWTPs are therefore considered the main recipients of terrestrial MPs before they enter natural aquatic systems (Badola et al. 2022, Sun et al. 2019). It has been proven that untreated MPs are commonly discharged from WWTPs, enter water bodies, and eventually accumulate in the environment (Carr et al. 2016, Nocon et al. 2018, Moraczewska-Majkut et al. 2020, Wiśniowska et al. 2020).

MPs are thus commonly found in the atmosphere (Abbasi et al. 2019), soil (Guo et al. 2020), ocean (Wang et al. 2020a), freshwater (Han et al. 2020), and even in Arctic freshwater lake sediments (Gonzalez-Pleiter et al. 2020). MPs persist in the environment due to their slow degradation rates (Eerkes-Medrano et al. 2015). MPs always cause chronic toxicity due to their accumulation in organisms, and the prolonged exposure of humans and other organisms, although no evidence of their acute effects has been found (Prata et al. 2020, Chen et al. 2020). Additionally, they can affect the physiological activities of living communities by leaching contaminants from plastics (e.g., plasticisers, flame retardants) and by acting as a vector for persistent pollutants (Lee et al. 2020, Ahmed et al. 2021). They can also adsorb contaminants such as polycyclic aromatic hydrocarbons (Sørensen et al. 2020), heavy metals (Foshtomi et al. 2019, Pohl et al. 2022), bisphenol A (Murphy 2001), polybrominated diphenyl ethers (Singla et al. 2020), phthalates (Pohl et al. 2022), pharmaceuticals and personal care products (Ma et al. 2019a) due to their small volume (contaminant particle size is usually less than 5 μm) and large specific surface area.

Typically, the name microplastic refers to plastic particles between 100 nm and 5 mm in size (Saboor et al. 2022). Based on particle size, plastics are classified into different categories, including macroplastics (>25 mm), mesoplastics (5–15 mm), microplastics (<5 mm) and nanoplastics (<100 nm) (Badola et al. 2022, Saboor et al. 2022). Manufactured particles, such as microbeads, enter directly into wastewater and are counted as primary MPs. On the other hand, plastics that are formed during the process of breakdown from solid plastic waste into smaller particles are considered as secondary MPs (Ahmed et al. 2021, Saboor et al. 2022). The most common plastic materials found in effluents are polypropylene (PP), polyethylene (PE), polystyrene (PS), polyvinyl-chloride (PVC), polycarbonate (PC), polyamides (PA), polyester (PES) and polyethylene terephthalate (PET), depending on the type of products produced by the plant (Talvitie et al. 2017a,b). These are reversible thermoplastic polymers, highly recyclable materials that can be heated, cooled and shaped repeatedly (Talvitie et al. 2017a,b).

Sources, types and characteristics of MPs in aquatic ecosystems, including point sources such as WWTPs, need to be extensively studied. WWTPs act as a point source of MPs as the microplastics produced in/disposed of in household and industrial wastewater streams, and often in the stormwater drain system, make their way into WWTPs. This makes WWTPs important in the study of MPs. Across the globe, interest in studying microplastics in WWTPs is catching up and an overview of the key publications reported in the literature from various parts of the world is presented in Table 1.

Surface freshwater, including river, lake and reservoir water, and groundwater are the main raw sources for drinking water. Seawater is sometimes used, as freshwater sources are scarce. However, seawater desalination treatment requires high use of energy and is expensive (Bodzek 2019). These raw water sources are easily contaminated by agricultural and industrial activities, and animal farming discharge. MPs have been detected in different surface waters (Shen et al. 2020). The average abundance of MPs in freshwater environments ranges from several to millions of tons (Pivokonsky et al. 2018). These great differences are mainly influenced by the locations, natural conditions, human activities, etc. The number of MPs in the inland freshwaters of Wuhan in China ranged between 1660.0 ± 639.1 and 8925 ± 1591 particles/ m^3 ; here the major types were PE, terephthalate and PP (Wang et al. 2017). Low-density polyethylene has also been identified as the dominant type of MPs. Recently, it has been identified that even the Arctic Sea is a reservoir with some MPs contamination (Law and Thompson 2014). MPs have also been found in lakes and rivers and, due to the wind and river driven transport, the plastic litter reaches the coast and the ocean (Dubai and Liebezeit 2013, Wagner et al. 2014). Nowadays, 98% of MPs are retained by wastewater treatment plants (WWTPs) but MPs with a size smaller than 20 μm and NPs are not retained; therefore, WWTPs plants are considered to be one of the main sources of plastic pollution in wastewater effluents (Talvitie et al. 2017a,b, Carr et al. 2016, Murphy et al. 2016, Malankowska et al. 2021).

The concentration of chemical additives, like plasticisers, in plastic debris of remote and urban beaches is up to 35 ng/g on remote beaches and up to 700 ng/g on urban beaches for bisphenol A; between 0.1 and 400 ng/g on remote beaches and up to 9900 ng/g on urban beaches for polybrominated diphenyl ethers; and up to 3940 ng/g for phthalates (Hirai et al. 2011).

Removal of MPs from water and wastewater

All methods used to remove MPs can be classified into physical, chemical and biological methods, depending on the mode of treatment (Badola et al. 2022). Based on the available studies, it was observed that physical methods are studied most, followed by chemical and biological methods. The percentage of available studies on physical, chemical and biological methods was 45%, 31% and 24% respectively (Badola et al. 2022).

MPs are generally classified as persistent materials, but degrade more or less depending on their nature and chemical structure. If their half-life is less than the values specified in the REACH criteria (Table 2) they can be considered as degradable and do not pose an environmental risk (Padervand 2020).

Physical methods of MPs removal

Most of the studies that use physical principles such as adsorption, filtration, sedimentation, flotation etc. are classified as physical methods (Han et al. 2019, Ahmed et al. 2021, Sommer et al. 2018, Badola et al. 2022). Most of these methods have been verified in laboratory, pilot and even full-scale testing. Among various physical methods with high MPs removal efficiency are: biochar, magnetic polyoxometalate-based ionic liquid phase adsorbents, magnetic carbon nanotubes, electrocoagulation, rapid sand filter and dissolved air flotation, chitin and graphene oxide sponge, zirconium-based organometallic foam and others (Badola et al. 2022).

Table 1. Overview of the reported studies on microplastics in WWTPs around the globe

Country	Waste-waters	Unit processes	Type of MPs recovered	Size of MPs	Concentration of MPs in the influent	Removal of MPs in WWTP	References
Spain	Domestic wastewater	MBR, RSF	Fragments, Fibres, Microbeads, Films	210 μm – 6.3 mm	4.40 MsP/L	79%	Bayo et al. (2020)
United Kingdom	Domestic and industrial wastewater	Coarse screen, Grit chamber, Primary settling tank, ASP and clarification tank, Nitrification tank	Fragments, Fibres, Films	60 – 2,800 μm	8.1×10^8 MPs/day	96%	Blair et al. (2019)
South Korea	Domestic and industrial wastewater	Grit chamber, Primary settling tank, MBR, ASP, and settling tank, Coagulation tank, Membrane DF, RSF	Fragments, Fibres, Microbeads, Sheets	100 μm – 5 mm	8,400 MPs/L, 62,800 MPs/L, 11,680 MP/L	92–99%	Hidayaturrahman and Lee (2019)
China	Domestic wastewater and pre-treated industrial wastewater	Primary treatment, Secondary treatment, Seasonal tertiary treatment	Fragments, Fibres, Pellets, Granules	250 μm – 5 mm	1.57–13.69 MPs/L	79.3–97.8%	Long et al. (2019)
China	Domestic and industrial wastewater	Aerated grit chambers, OD, MBR, Secondary settling tank, UV disinfection chamber	Fragments, Fibres, Films, Foam	100 μm – 5 mm	4.0 MPs/L	97–99%	Lv et al. (2019)
Italy	Domestic wastewater	ASP and sedimentation tank, RSF and disinfection tank	Fragments, Films, Lines	0.01 – 5 mm	2.5 ± 0.3 MPs/L	84%	Magni et al. (2019)
China	Domestic and industrial wastewater	Primary aerated grit treatment tank, A/A/O, Denitrification, UF, Ozonation, UV tanks	Fibres, Particles	100 μm – 5 mm	12.03 ± 1.29 MPs/L	95%	Yang et al. (2019)
Canada	Domestic wastewater and storm water from combined sewers	Bar screen, Primary clarifier, Trickling filters, Solids contact tanks, Secondary clarifiers	Fragments, Fibres, Pellets	250 μm – 5 mm	31.1 ± 6.7 MPs/L	94%	Gies et al. (2018)
Finland	Domestic wastewater	MBR, ASP	Fragments, Fibres	250 μm – 5 mm	1.5×10^8 MPs/d	98.3%	Lares et al. (2018)
Denmark	Domestic and industrial wastewater	Coarse screen, Grit removal, Primary settling tanks, Aeration tank, Clarification tank	Fragments, Fibres, Microbeads, Films, Foam	100 μm – 5 mm	15.70 ± 5.23 MPs/L	98.41%	Murphy et al. (2016)
United States of America	Domestic wastewater and storm water	Bar screen, Grit chamber, Primary settling tank, ASP, TF and ASP, RSF, Anaerobic MBR	Fragments, Fibres, Microbeads	100 μm – 5 mm	133.0 ± 35.6 MPs/L	97–99%	Michielssen et al. (2016)

Sedimentation/clarification

Sedimentation is used to obtain concentrated suspensions of solids and to purify (clarify) liquids containing suspended solids (Bache and Gregory 2010, Ostrovsky et al. 2014). Sedimentation under the influence of gravity is used in the first stage of water and wastewater treatment, which includes the removal of granular suspensions (sand and gravel grains) in sand traps and the removal of fine suspensions and colloids in settling tanks (clarifiers). Preliminary clarification, used in wastewater treatment, is designed to ensure settling of solids prior to biological treatment (Riffat 2013). The sedimentation/clarification also allows for the removal of MPs from water systems. This method is not only used in primary wastewater treatment but also in secondary treatment.

Conley et al. (2019) reported the removal efficiency of MPs in three wastewater treatment plants with various treatment operations and service arrangements in the USA for one year. They found a high MPs removal efficiency of approximately 97.6% for the primary clarification. The size fractions included MPs particles larger than 418 μm , between 178–418 μm , and between 60–178 μm (Conley et al. 2019). Michielssen et al. (2016) observed that 84–88% of MPs with sizes ranging from 100–1000 μm were eliminated in wastewater treatment plants in the U.S. and other countries by primary sedimentation and clarification. In other studies, the sedimentation/clarification process, used before other treatment techniques (Bui et al. 2020, Ngo et al. 2019), showed removal efficiencies of 57–64% in wastewater from South Korea (Hidayaturrehman and Lee 2019). Murphy et al. (2016) also studied the performance of sedimentation and clarification at a municipal wastewater treatment plant in Glasgow, Scotland. The average MPs decreased from 15.7 MPs/L to 3.4 MPs/L with a removal efficiency of approximately 78%. Based on the results of Bayo et al. (2020a), approximately 74% of MPs were removed during sedimentation at a municipal wastewater treatment plant in Spain. Thus, pretreatment in WWTPs has the greatest impact on size distribution because it can effectively remove MPs of larger size.

The efficiency of MPs removal by sedimentation is affected by two key factors, including density and shape (Bui et al. 2020, Ngo et al. 2019). In the study of Lares et al. (2018), most of the MPs were eliminated (99%) in the initial stage at an input concentration of 57.6 MPs/L. The reason for the high efficiency obtained in this study may be due to the fact that more than 96% of the MPs were in the form of fibers. Based on the results of Hidayaturrehman and Lee (2019), the pretreatment stage retained more of the fibrous MPs (76–92%) than other types such as microspheres, sheets and irregular fragments. Long et al. (2019) argued that fragments and granules are two shapes of pollutants which are eliminated out of the flow most easily (respectively 91% and 83%) in WWTPs, whereas the removal rate of fibers is only around 79% at a WWTP in Xiamen, China. Hence, fibers are considered the most difficult shape of MPs to remove from the wastewater stream (Long et al. 2019). As a result, this shape of MPs pollutant is most dominant in WWTP final effluent (Long et al. 2019, Talvitie et al. 2017a, Ziajahromi et al. 2017). This result can be partly explained by the smoothness of the surface of each shape. For example, fibers and pellets are supposed to be smoother than other shapes, which means they have less resistance in

the wastewater environment to being captured by the treatment technologies in WWTPs (Anderson et al. 2018). In contrast, fragments are often angular, bifurcate and twisted, which not only increases the ability to be captured in solid flocs but also creates more chance for the colonization of microorganisms, by increasing the degree of sedimentation or degradation.

The major drawback of sedimentation in the removal of MPs is that pollutants are not completely removed, they only sink or are trapped in sludge which allows for a high risk of the MPs reverting back into the wastewater due to the turbulent environment. In addition, MPs which are dumped as sewage sludge in a landfill will return to WWTP through leachate or enter the natural water environment by stormwater runoff. Therefore, an alternative sustainable treatment process for MPs removal is needed.

Flotation

Flotation is a physicochemical method of separating solids ground in water, in which hydrophobic impurities along with air bubbles are brought to the surface of the suspension to form foam. Dissolved air flotation (DAF) is the most widely used method for separating low-density solids, oil and fibrous materials, including MPs, from soil or sediment in dense liquids (Han et al. 2019). In this method, air is dissolved at a high pressure in water that results in the formation of bubbles. These bubbles attach solid particles on their surfaces (including MPs), which are later removed by skimmers (Bui et al. 2020). Recently, DAF offered high efficiency for MPs removal. In a WWTP in Hameenlinna, Southern Finland, the detected MPs removal rate of a dissolved air flotation facility was 95% (Talvitie et al. 2017b). However, the influent concentrations of MPs in this study were relatively low (2 ± 0.07 MPs/L). There have been no studies to evaluate the effectiveness of DAF in removing MPs under different conditions such as density, size, shape and composition of MPs. Coppock et al. (2017) proposed a portable density flotation to separate MPs with particle sizes ranging from 100 μm –10 mm from sediments with an average efficiency of 95.8%. Zinc chloride, with a density of 1.5 g/cm^3 , was used as an effective flocculating agent, allowing fine sediment to settle, while allowing dense polymers to float.

Unlike sedimentation, air flotation technology removes contaminants by trapping low-density MPs (such as PE, PP), and the medium-density plastics (such as PS and PA), which can float alone or with air bubbles and cannot be captured by using the sedimentation technique. However, compared to the sedimentation process, this method is expensive to operate and maintain (Talvitie et al. 2017b). Another factor that should be taken into account is the natural buoyancy of contaminants in the wastewater environment that can be changed by the adsorption of chemical compounds onto the surface of MPs particles. Similar to the sedimentation process, the morphology of MPs is an important factor affecting their removal in air flotation. Again, fragments and granules are the two contaminant shapes removed in WWTPs (Xiamen, China) at 91% and 83% respectively, while the fiber removal rate is only about 79% (Long et al. 2019). Therefore, fibers are considered the most difficult MPs shape to remove from wastewater. The use of polyaluminium chloride increases the process of flocculation.

Classical filtration

Filtration is a basic and effective method that is commonly used to remove MPs from water and wastewater (Ahmed et al. 2021). Filters with different pore structures and pore size and different materials are used in the filtration process, among which metal-based filters (stainless steel), glass fiber, and polymer-based filters such as polycarbonate, nitrocellulose and nylon are the most frequent (Wang and Wang 2018). The most common pore size in filters is 0.45–1 μm . Some filter materials have a curvy and deep pore structure, such as stainless steel and nylon filters, while others have narrow and straight circular pores, such as polycarbonate filters (Saboor et al. 2022). Due to the microscopic size of the particles or contaminants in the liquid, filter cartridges become clogged. To solve this problem, iron salt is added to the coagulation/flocculation of the solid fraction and a pre-filtration step is performed using a larger pore size filter (Crawford and Quinn 2017).

Straining through sieves is also used to separate MPs, which leads to the sorting of particles into different size ranges depending on the choice of sieve mesh size (Ahmed et al. 2021). The most commonly used screening system for separating MPs from water and sediment samples is multistage screening, which uses a series of sieves with different mesh sizes (Crawford and Quinn 2017). Olivatto et al. (2019) studied the separation of MPs found in samples from Guanabara Bay, Brazil using sieving and manual sorting. MPs were isolated by wet sieving using two meshes, including a mesh size of 355 μm at the bottom and 4.75 mm at the top. Gimiliani et al. (2020) presented an effective separation and quantifying method comprising sieving with 2.0, 1.0, 0.5 and 0.25 mm mesh sizes, sediment collection, drying and microscopic evaluation of samples retained on each sieve. Zhang et al. (2020a) used a fine sieve (mesh size 2.5–10 mm) to remove larger solids (> 2.5 mm) during wastewater pretreatment, because sieves with larger (50–100 mm) and medium mesh sizes (10–40 mm) were unable to retain MPs. For the membrane bioreactor (MBR), sieves are used (mesh size 0.2–2 mm) as an alternative to sedimentation to prevent membrane fouling. Generally, due to the irregular shapes of the MPs, some larger MPs may still pass through this type of sieve.

Disc filter (DF) is a promising technology to decrease the concentration of MPs in wastewater treatment effluent. Several studies have shown that DF is used in WWTPs as a tertiary treatment at full scale in many countries (Talvitie et al. 2017b, Hidayatullahman and Lee 2019). In a study of the Viikinmaki WWTP in Helsinki (Talvitie et al. 2017a), MPs were retained by the filter, gradually forming a sludge cake on the filter surface. High-pressure backwashing was used to remove it. The pore size of the filter, which is a woven material usually made of PP, polyester or PA, is generally 10–40 μm . Hydraulic retention time, water flow rate, membrane fouling and pore size are the main parameters affecting efficiency of the DF process (Talvitie et al. 2017b). The removal rates obtained for different types of MPs were between 40–98.5%, and the concentration of MPs was reduced from 0.5 to 0.3 MP/L using a 10 μm filter and from 2.0 to 0.03 MP/L using a 20 μm filter. Hidayatullahman and Lee (2019) reported an overall MPs removal efficiency of 99.1% at a WWTP in South Korea with a DF (pore size 10 μm), while the MPs removal efficiency at the DF stage was 79.4%. Simon et al. (2019) used a DF with 13 polyester mesh discs with a pore

size of 18 μm , which removed 89.7% of MPs with a pore size >10 μm . However, the effectiveness of this method was diminished by large size plastics that accumulated on the filters and blocked the pores (Simon et al. 2019).

In general, filters with smaller pore size show higher MPs removal efficiency. It is significant that DF can effectively remove all shapes and size fractions, but some of the smallest (20–100 μm) pass through the filter (Talvitie et al. 2017b). The literature review suggests that DF offers a relatively low efficiency in removing MPs, which can be increased by using coagulation processes prior to DF.

Rapid sand filtration (RSF) is a popular water treatment technology for both waterworks and wastewater treatment plants. This technology provides fast and effective contaminant removal with low operating and maintenance costs. In wastewater treatment plants, RSF is typically used as a third stage of treatment (Ngo et al. 2019). RSF are constructed with sand layers consisting of 1 mm gravel with a grain size of 35 mm, 0.5 m quartz with a grain size of 0.1–0.5 mm, and anthracite is often used. This method is effective for MPs >20 μm in size and is able to effectively remove all types of MPs (Badola et al. 2022). In the presence of three layers (anthracite, quartz sand and gravel), RSF captures suspended solids by physical adsorption or mechanical straining. Due to hydrophilic interactions, MPs particles are adhered to the surface of sand grains or adsorbed on silica grains, which reduces the performance over time (Enfrin et al. 2019). The adsorption of MPs is difficult to reverse due to the presence of functional groups, such as hydroxyl groups on the surface of MPs resulting in stronger interactions with RSF (Cai et al. 2018). In a pilot study, the RSF process was applied as a third stage of wastewater treatment in WWTP Turku, Finland (Talvitie et al. 2017b). 97% removal of MPs (0.7–0.02 MP/L) of all shapes and sizes, even the smallest fractions (20–100 μm), was obtained. Hidayatullahman and Lee (2019) showed that the removal efficiency of MPs at the RSF stage was 73.8% and the total removal efficiency was 98.9% when RSF was applied as the third stage of wastewater treatment in combination with coagulation. Therefore, RSF was considered a suitable technology for MPs removal at the low concentration range. These studies showed that the MPs removal efficiency of the RSF process is lower than that of the MBR process, while it is higher than that of dissolved air flotation, granular activated carbon (GAC) adsorption and membrane filtration, demonstrating its potential as an effective process for MPs removal in WWTPs (Badola et al. 2022). Therefore, the RSF process can be very attractive when combined with other treatment processes such as coagulation.

Membrane filtration

Membrane technology is one of the possible methods to remove plastic litter from water mainly because membrane-based operations have the potential to replace energy-intensive conventional technologies due to their low energy consumption, operation flexibility and simplicity, good stability, easy control and scale-up. Membrane separation processes differ based on the separation mechanism and size of the separated particles (Bodzek 2019). Pressure driven membrane processes are by far the most widely applied membrane processes in water and wastewater treatment. There are four main types of these processes. These are microfiltration (MF), ultrafiltration (UF),

nanofiltration (NF) and reverse osmosis (RO) (Ezugbe and Rathilal 2020, Bodzek 2019). The main difference exhibited by these processes is their membrane pore size. Diffusion membrane processes (forward osmosis (FO) and membrane distillation (MD)) and processes using ion exchange membranes (electrodialysis, electrodeionisation) are also considered (Ezugbe and Rathilal 2020). Membrane material, pore size, thickness and surface characteristics affect the performance of the membrane process. In membrane filtration, the major drawback is the fouling phenomenon which occurs by adsorption of particles on the surface and inside the pores of the membrane. Fouling decreases membrane filtration performance which results in higher energy cost, operation time and maintenance (Malankowska et al. 2021). Enfrin et al. (2019) revealed that MPs could interact with the membrane surface because of their intrinsic physicochemical properties such as hydrophobicity, surface charge and roughness. Nevertheless, membrane technology is highly efficient in the removal of low-molecular weight contaminants such as small MPs (<100 μm) and NPs.

Ultrafiltration (UF) and microfiltration (MF) are increasingly used for the treatment of high quality drinking water in an economic manner thanks to low energy consumption, high separation efficiency and compact plant size (Bodzek 2019, Bodzek et al. 2019). It is a low-pressure membrane process (1–10 bar) that uses symmetric/asymmetric membranes having a pore size between 0.05–10 μm for MF and 1–100 nm for UF. UF/MF membranes can reject particulates and macromolecules such as proteins, fatty acids, bacteria, protozoa, viruses and suspended solids. Therefore, MF/UF are used to replace existing classical processes (sedimentation, flocculation, coagulation and sand filtration and chlorination) used in water and wastewater treatment. However, these technologies are not specifically designed for the removal of MPs that remain in the final treated water (Mason et al. 2016, Bodzek 2019, Talvitie et al. 2017b). In many cases, MF/UF are integrated with processes used in water and wastewater treatment, such as sedimentation, classical filtration, flotation, biological and advanced oxidation processes and used for pre-filtration in reverse-osmosis plants to protect the osmotic membranes against fouling (Bodzek 2019). UF coupled with the coagulation step is one of the main water treatment technologies in current water plants.

A variation on composite membranes are **dynamic membranes (DM)**, which are obtained by passing a solution containing membrane-forming components through porous supports (Ersahin et al. 2012). Immersion of the porous supports in a suitable colloidal suspension of the membrane-forming material and drying is also used. Porous carbon electrode tubes, hard poly(vinyl chloride), sintered metal powders and ceramic tubes are used as porous materials. Organic polyelectrolytes and hydrated metal oxides in colloidal form are most commonly used as film-forming components. In contrast to MF and UF, the DM filtration process exhibits a lower pressure, which means reduced energy consumption (Li et al. 2018). Low filtrate resistance and easy cleaning of DM are mentioned as main advantages (Li et al. 2018).

DM technology is more effective at removing low density (poorly settling) contaminants and undegradable MPs due to the rapidly forming secondary membrane (DM layer) with microparticles (Li et al. 2018). DM was applied to remove

MPs from synthetic wastewater in gravity mode using a DM laboratory filtration kit (Li et al. 2018). It achieved the removal of about 90% of MPs from synthetic wastewater using a 90 μm support mesh. After 20 min of MPs filtration, the turbidity of the effluent was reduced to <1 NTU (Nephelometric Turbidity Unit), which confirmed the rapid formation of DM resulting in better MPs removal efficiency. Overall, DM technology showed excellent performance to remove microcontaminants including MPs during wastewater treatment, and mitigated the disadvantages of membrane fouling in UF/MF. The combination of DM technology with coagulation or activated sludge process can be highly effective to remove micro-contaminants and MPs in wastewater treatment (Li et al. 2018). Further research is needed to unravel the mechanism of DM layer formation.

Reverse Osmosis (RO) is actually used in municipal and industrial water treatment systems to purify water using RO or NF membranes (pore size > 2 nm) by removing salts, heavy metals and other organic impurities. RO is currently applied in food and beverage production, biopharmaceutical manufacturing, power generation, production of high purity water, and desalination of brackish waters and seawater, as well as in the recovery of water from industrial and municipal wastewater (Antony et al. 2011).

In terms of MPs removal in wastewater, RO has been implemented at a WWTP in Sydney, Australia after tertiary treatment (Ziajahromi et al. 2017). They characterized and quantified the microplastic in samples coming from a WWTP that produced a highly treated effluent, including screening and sedimentation, biological treatment, flocculation, disinfection/de-chlorination processes, UF, and finally RO process. Results indicate the presence of MPs fibers in the samples after the RO process. A removal efficiency of MPs, bigger than 25 μm , of only 90.45% was obtained (Ziajahromi et al. 2017). The result is significantly lower than that of MBR which is 99.9% with smaller MPs (20 μm). After 4 treatment stages including primary, secondary and tertiary treatment processes and RO, the WWTP still releases ten million pieces of plastic debris per day to the natural aquatic environment (Ziajahromi et al. 2017).

Wang et al. (2020b) studied the occurrence of phthalate esters and MPs in the effluent simultaneously of four wastewater treatment plants and the receiving water bodies in winter and spring. MPs were mostly in the form of granules and fragments with size <0.01 mm in the four WWTP effluents (276–1030 MPs/L) and receiving water bodies (103–4458 MPs/L). The main techniques were clarification, filtration and reverse osmosis with removal rates of 42.7–69.2%, 25.3–59.3% and 22.6–51.0%, respectively. The total removal rates of phthalate esters and microplastics in the four WWTPs were 47.7–81.6% and 63.5–95.4%, respectively. The results revealed that the surrounding environment considerably affected the amount of phthalate esters and microplastics in surface waters.

Overall, membrane treatment technology is not specially designed to remove MPs efficiently, due to common issues of membrane fouling and decreasing water flux. More research should be devoted to minimize membrane abrasion and fouling in membrane-based treatment technology. However, membrane treatment technology can be attractive if it is combined with a biological process such as MBR or chemical process such as coagulation.

Magnetic separation process

Magnetic separation is the most reliable for the separation of MPs/NPs from sediment or water samples under magnetic force, although it is not suitable for MPs removal in WWTP. This method is particularly effective for small-sized MPs, because of their large surface area to volume ratio which enhances the binding affinity of MPs with Fe nanoparticles. Grbic et al. (2019) studied the performance of magnetic removal of MPs from seawater, freshwater and sediment. Fe nanoparticles were coated with hexadecyltrimethoxysilane (HDTMS) to create the hydrophobic characteristics for allowing binding with MPs, which helps to isolate the MPs from water under a magnetic field (Fig. 1). 92% and 93% removals of small-sized (< 20 µm) PE and PS and large-sized (>1 mm) MPs from seawater were obtained. The recovery rate of medium-sized (200 µm–1 mm) MPs was 84% and 78% from freshwater and sediment. Therefore, MPs recovery by magnetic extraction process is particularly suitable for drinking water treatment (Grbic et al. 2019).

Shi et al. (2022a) studied magnetic nano-Fe₃O₄ for MPs removal. The results showed optimal magnetization of MPs via surface absorption. At 1.3 g/L nano-Fe₃O₄ and 150 min treatments, the average removal rate of four common types of MPs including PE, PP, PS and PET in sizes of approximately 200–900 µm was 86.87%, 85.05%, 86.11% and 62.83%, respectively. The removal rate varied among polymer- and different-sized MPs, and was positively related to the density of nano-Fe₃O₄ adsorbed on MPs' surfaces. In addition, the removal rate of MPs in artificial seawater was relatively high in comparison to pure water. Furthermore, the established approach was effectively applied to remove MPs in environmental water bodies including river water, domestic sewage and natural seawater, with a removal rate of higher than 80%. Pramanik et al. (2021) also studied removal efficiency of NPs/MPs by two types of ferrofluid used and found an average removal of 43% for magnetite and 55% for cobalt ferrite. All three plastics tested, i.e., PE, PVC and polyester, had similar removal efficiency by nano-ferrofluid particles, meaning that this removal technique does not rely on the plastic component type. Altogether, this study provided a novel and simple approach to remove MPs in water, and shows potential application.

Shi et al. (2022b) prepared magnetic sepiolite, which was used to remove PE with 98.4% efficiency. SEM and XRD analysis showed that the magnetic sepiolite was deposited (wrapped,

embedded or adsorbed) on the PE surface so effectively that the mixtures could be separated from the aqueous solution in a suitable magnetic field as strong magnetic materials. The PE removal efficiency after using the recycled magnetic sepiolite from the magnetic tube five times was still above 90%.

Tang et al. (2021) synthesized magnetic carbon nanotubes (M-CNTs) for the first time as adsorbents to remove MPs. M-CNTs were effectively adsorbed on PE, PET and PA and all the MPs/M-CNTs composites were readily separated from aqueous solutions by magnetic force. When the 5 g/L of M-CNTs was added, target MPs (5 g/L) were completely removed within 300 min. The maximum adsorption capacities of PE, PET and PA were 1650, 1400 and 1100 mg-M-CNTs/g, respectively. Furthermore, the adsorbed M-CNTs can be recycled via thermal treatment (600°C) and these M-CNTs were featured with the same magnetic properties and comparable MPs removal capacity as the original ones. After being used four times, M-CNTs were still able to remove ~80% of total MPs in the testing solution. The observed effectual removal of MPs from prepared solutions and wastewater highlights M-CNTs as promising techniques for the control of MPs pollution.

Adsorption

Biochar (BC) and activated carbon (AC) are extensively used as adsorbents to treat water containing MPs and NPs (Ahmed et al. 2016, Sommer et al. 2018). Adsorbent surface area and porosity are two major properties for effective removal of MPs (Siipola et al. 2020). BC used for removal of MPs are made from various substances like corn, hardwood, pine, spruce bark, etc., alone or in combination (Siipola et al. 2020, Wang et al. 2020c). Siipola et al. (2020) reported that activated BC was the most suitable adsorbent for MPs removal, even with relatively low surface area (200–600 m²/g). Despite the small surface area (187 m²/g) with macro-scale porosity, spruce bark BC resulted in better performance for MPs retention than pine bark ACs with surface area 556–603 m²/g. Activated BC effectively retained large size MPs particles, whereas 10 µm spherical microbeads did not adsorb as efficiently. Hence, meso- and macro-porosity can be very beneficial for the removal of MPs. The BC surface roughness may influence the retention of large MPs particles most likely through physical attachment. They also found that PE particles and fleece fibers were 100% retained, although the mechanism of MPs adsorption is yet to be identified (Siipola et al. 2020). A comparison study between simple sand filter and BC filter showed above 95% removal efficiency for fine

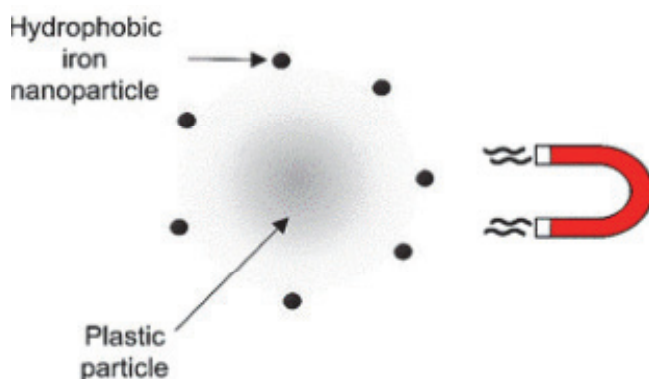


Fig. 1. Concept of MPs removal in magnetic field (Saboor et al. 2022, Grbic et al. 2019)

(approx. 10 μm) size PS MPs spheres (microbeads) while the sand filter showed an efficiency of 60%–80%. This indicates that BC filters are a better option for the removal of MPs as compared to sand filters (Wang et al. 2020c). Both BC and AC may act as a filter when packed in a column for MPs removal (Zhang et al. 2020a). Therefore, adsorption with AC or BC via a filtration setup is an economical process to remove MPs.

In recent years, granular activated carbon (GAC) filtration was employed to treat some emerging contaminants in an aqueous environment (Östman et al. 2019). Wang et al. (2020d) evaluated the MPs removal capability of the GAC filtration system in a drinking water treatment plant. The MPs removal capacity (PE PP and PAM) of this technology was up to 60.9%, less effective than other conventional technologies such as coagulation/flocculation, sand filtration, RSF and ozonation. In the GAC process, contaminants are removed by a combination of biodegradation and physical adsorption. However, so far the mechanism to remove MPs in GAC is still unclear. Therefore, the GAC filtration process can be an effective technology for MPs removal at low MPs concentration ranges.

Comparison among physical processes

Physical treatment methods can be applied to remove a wide range of MPs from water, with their average removal

efficiencies summarized in Table 2. The wide range of MPs removal rates is due to the different process conditions and the different sizes of particles removed. A wide range of MPs can be removed through filtration processes such as GAC filtration, RSF and DF. DAF is also attractive for removing MPs efficiently with the flocculation process. Membrane treatment such as UF, RO and DM technology can be more effective hybrid systems, i.e., MBR, coagulation – UF. Among all other membrane treatments, DM technology is cost-effective and highly efficient for removing MPs from synthetic wastewater but still insufficient to remove large scale MPs from wastewater. On the other hand, density separation and magnetic separation are more efficient at removing MPs from sample water. The adsorption process is suitable to adsorb MPs from water but this process was not studied sufficiently. Moreover, among the physical treatment technologies, the quantitative analysis revealed that filter-based methods showed better MPs removal efficiency than others. MPs removal through physical methods followed the order: filtration process > flotation process > adsorption process > membrane process > magnetic and density separation process. Furthermore, a more detailed characterization of MPs in different treatment technologies is needed to select the most suitable methodologies for the efficient removal of MPs from WWTP effluents.

Table 2. Advantages, limitations and effectiveness of physical treatment technologies in MPs removal

Process type	Efficiency	Advantage	Disadvantage	References
Adsorption (AC and BC)	100%	Sufficient surface area and suitable porosity effectively retained large size MPs.	10 μm spherical MPs did not adsorb as efficiently.	Siipola et al. 2020
UF	41,7%	PE particles can be completely bound by the UF membrane.	Fouling	Ma et al. 2019b, Ziajahromi et al. 2017
RO	25%	MPs >25 μm was completely removed.	Fouling	Ziajahromi et al. 2017
Dynamic membranes	>90%	Less energy consumption and trans-membrane pressure, low filtration resistance, low cost.	Membrane fouling. Not effective for large scale water treatment.	Li et al. 2018
Density Separation	high	Can remove low density MPs. Reliable and practical method.	Heavy salts are very expensive, and some are hazardous.	Murphy et al. 2016
Grit/primary Sedimentation	78.34%	Low-cost process. Effective for large MPs	Need to secondary and tertiary treatment to remove small MPs.	Liu et al. 2019a, Yang et al. 2019, Murphy et al. 2016
GAC filtration	99,9%	Remove small size MPs with biological activity	Clogging is the main problem	Wang et al. 2019a, Zhang et al. 2020b
RSF	97,2%	Low operational and maintenance cost.	Fouling take place; backwash is needed. MPs are broken into smaller particles.	Enfrin et al. 2019, Talvitie et al. 2017b, Hidayaturrehman and Lee 2019, Michielssen et al. 2016
Disc filters	98.5%	Sludge cake formation. Float MPs are especially removed.	Backwash needed due to membrane fouling.	Hidayaturrehman and Lee 2019, Talvitie et al. 2017b
Flotation	95%		Disadvantage – removes contaminants by trapping low-density MPs (such as PE, PP), and the medium-density plastics (such as PS, and PA)	Talvitie et al. 2017b
Magnetic separation	78–93%	Efficient for smaller MPs. Better for drinking water treatment.	MPs recovery from sediment is lower.	Grbic et al. 2019

Chemical methods of MPs removal

Chemical methods are also used in the treatment of water and wastewater containing MPs, either alone or in combination with others to enhance the effectiveness of physical processes (e.g. sedimentation, membrane processes). Several methods, like ozonation, advanced oxidation processes (e.g. photocatalysis) and coagulation are most commonly used for plastic removal/degradation (Ahmed et al. 2021).

Coagulation/flocculation

As MPs particles are tiny in size (diameter < 5 mm), it is highly challenging to separate them through filtration processes continuously. Difficulties like filtration surfaces fouling make them often inefficient and discontinuous, hence, pretreatment by coagulation and flocculation will improve filtration efficiency. Many wastewater and drinking water treatment plants worldwide use coagulation/flocculation processes to form enlarged contaminant particles that are easier to separate (Shirasaki et al. 2016). Among the various coagulating agents to coagulate and agglomerate MPs particles, Fe- and Al-based salts and flocculants (e.g. polyacrylamide – PAM) are commonly used to bind fine particles through adsorption-complexation (Chorghé et al. 2017).

It is obvious that the formation of MPs flocs closely depends on the MPs concentration. Indeed, with a certain amount of coagulant, it would be difficult to form flocs in water with a lower MPs concentration, resulting in a lower MPs removal efficiency. Hidayaturrahman and Lee (2019) studied MPs removal by coagulation using polyaluminium chloride (PAC) with different initial MPs dosages (4200 MPs/L, 5840 MPs/L and 31,400 MPs/L). The results showed that the removal efficiencies of MPs amounted to 53.8%, 47.1% and 81.6% respectively. Rezania et al. (2018) also found that the ability to remove MPs positively correlated with the coagulant dose. For PE with small size (< 0.5 mm), MPs removal efficiency increased from 8.3% up to 36.9% when the aluminium coagulant dosage increased from 13.5 mg/L Al to 405 mg/L Al. However, as the flocculant dosage was increased, the removal rate of MPs would tend to decrease. This is explained by the fact that MPs' zeta potential decreased as the coagulant dosage increased excessively, resulting in difficulty in forming MPs flocs.

The efficiency of the coagulation process depends on the type of coagulant used. For example, in the study of Ma et al. (2019b,c), both aluminium and ferric-based coagulants were simultaneously examined with the presence of PE MPs, which have been commonly detected in various wastewater. As a result, the performance of Al coagulant was more efficient than that of ferric-based coagulant in PE removal. The smaller the PE particle size, the higher the removal efficiency. The corresponding removal efficiencies of small PE particles ($d < 0.5$ mm) were only 8.24% and 12.65% in the presence of 0.5 mM and 5 mM $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ at pH 7.0, respectively, while in the presence of $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ (15 mM), the removal efficiency was 36.89%. However, the removal efficiency for larger particles ($0.5 < d < 1$ mm) was only 20.61%, and 11.73% for particles of size $1 < d < 2$ mm and 4.51% for particles $2 < d < 5$ mm (Ma et al. 2019b,c). Zhou et al. (2021) compared PACl and FeCl_3 at drinking water treatment plants, and found that the former was more effective than the latter at removing PS and PE MPs (e.g. ~78% removal of PS by PACl (90 mg/L) vs. ~64% removal of PS by FeCl_3 (90 mg/L) and ~30% removal of PE by PACl (90 mg/L) vs. 17% removal of PE by FeCl_3 (90 mg/L)).

The efficiency of the coagulation process for MPs also depended on the pH values of the water solution. Ma et al. (2019b) investigated PE removal by coagulation process with $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ (5 mmol/L or 135 mg/L Al) at pH 6, 7 and 8. For low coagulant dosage (0.5 mM $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$) in Fig. 2a, the removal efficiency of PE was barely influenced, even for the small particle size. The corresponding removal efficiencies for small PE particles $d < 0.5$ mm were $8.17 \pm 1.12\%$, $8.28 \pm 1.06\%$ and $7.67 \pm 0.98\%$ at pH levels of 6.0, 7.0 and 8.0, respectively. However, the removal efficiency of PE decreased with increasing pH solution with a high coagulant dosage (5 mM $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$), especially for the small particle size (Fig. 2b). The removal efficiencies regarding the smaller PE particles ($d < 0.5$ mm) decreased from $27.52\% \pm 0.94\%$, $25.83 \pm 2.91\%$ and $22.15 \pm 1.72\%$, at pH levels of 6.0, 7.0 and 8.0, respectively, whereas the corresponding removal efficiencies for the larger PE particles ($2 < d < 5$ mm) only decreased from $5.34 \pm 1.13\%$, $4.27 \pm 1.91\%$ and 2.73 ± 1.89 , respectively.

Ma et al. (2019c) also tested the effects of pH condition (pH 6, 7 and 8) on the performance of the coagulation using the

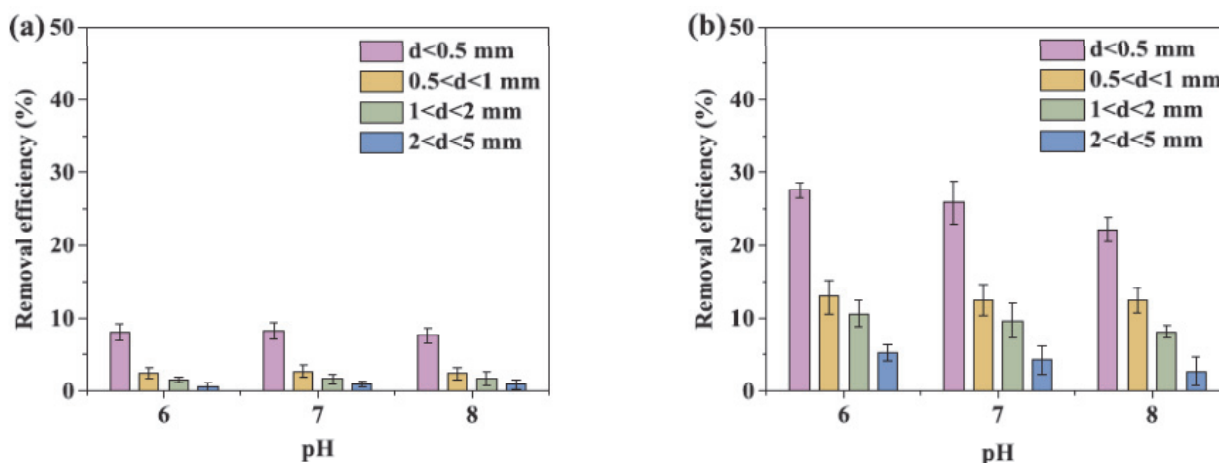


Fig. 2. Removal of PE with different particle sizes under different pH conditions: (a) 0.5 mM $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$; (b) 5 mM $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$. Other experimental conditions: weight of PE particles: 0.1 g (Ma et al. 2019b)

$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (2 mmol/L) with 0.92–0.97 g MPs/m³. However, the removal efficiency of PE was little influenced by ionic strength. The removal efficiency of PE increased under the high dosage of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (2 mmol/L), and the corresponding removal efficiency also increased with increasing solution pH. For 0.2 mmol/L $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, the removal efficiency for small-particle-size PE ($d < 0.5 \mu\text{m}$) was 6.03%, 6.71% and 6.67% at pH 6.0, 7.0 and 8.0, respectively. For 2.0 mmol/L $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, however, the removal efficiency for small-particle-size PE ($d < 0.5 \mu\text{m}$) was 11.56%, 13.27% and 17.23% at pH 6.0, 7.0 and 8.0, respectively.

In many cases, low MPs removal efficiency was observed in the coagulation process using conventional dosage. The solution is to use flocculation/flocculants, which is the final element of the coagulation process. In the flocculation, chemical bonds are formed between micelles, easy-to-remove sludge or suspended solids which combine and separate an easily removable sludge or slurry. Several studies indicated that polyacrylamide (PAM) was effective in enhancing the efficiency of coagulation (Ma et al. 2019b,c). The results indicated that the removal efficiency increased from 26% (without anionic PAM) to 61% (with anionic PAM of 15 mg/L). However, there was 45% of PE eliminated by adding 15 mg/L cationic PAM and 61% with the same dosage of anionic PAM. The findings showed that anionic PAM was more effective than cationic PAM in PE MPs removal. Anionic surfactants such as sodium dodecyl sulphate also facilitate the coagulation of MPs since negative charges induced by surfactant adsorption are neutralized in the presence of $\text{Al}^{3+}/\text{Fe}^{3+}$ ions resulting in the precipitation of microplastic particles. The coagulation removal efficiency was not affected by increasing sodium dodecyl sulphate surfactant (Xia et al. 2020).

Most recently, Wang et al. (2020c) assessed the influence of MPs' particle size and polymer type on the efficiency of the coagulation process coupled with sedimentation. The results show that larger particles will have a higher removal efficiency. Specifically, 100% of large particles ($> 10 \mu\text{m}$) and 45%–75% of small particles ($5\text{--}10 \mu\text{m}$) were removed by coagulation. Compared to filament and pellet forms, fibers can be eliminated highest (51–61%) because fibrous MPs were easier to attach to flocs. The results also indicated that PET was removed the most (59–69%) compared with PP, PS and PAM. This finding has also been reported by Katrivesis et al. (2019) and Lares et al. (2018).

Coagulation coupled with UF is one of the main water treatment technologies in current water plants proving a significant removal of organic matter. A specific schematic diagram of MPs removal during coagulation and UF processes of MPs is shown in Fig. 3 (Ma et al. 2019b).

However, the worrying levels of MPs in freshwaters make an in-depth investigation of the behavior of MPs during the removal by coagulation and UF processes mandatory, also

considering that they are water treatment technologies used in the production of drinking water (Bodzek 2019, Ma et al. 2019b,c). Ma et al. (2019b,c) studied the removal behavior of PE in drinking water treatment by UF and coagulation using $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ coagulants. The density of PE (0.92–0.97 g/cm³) is very close to that of water, making it difficult to remove by sedimentation or flotation. In the UF process, PE particles were completely removed because of the small pore diameter of the UF membrane, and slight membrane fouling was induced after coagulation at a conventional dosage, especially for the large PE particle size. The larger the PE particles, the more heterogeneous the Al-based floc layer was, which led to less membrane fouling. With increasing coagulant dosage, membrane fouling was gradually aggravated owing to the thick cake layer formed. However, this behavior may not be a general rule but may depend on various parameters related to the membrane process as well as the properties of the plastic (chemical composition, size and shape). Based on this systematic investigation, the removal behaviors of MPs exhibited during coagulation and UF processes have application potential for drinking water treatment.

Overall, coagulation or agglomeration of MPs can be an effective step in the MPs treatment process to enhance the efficacy of water and wastewater treatment. Moreover, it can play a significant role in overcoming the fouling problems of the membrane-based treatments. The efficacy of MPs removal closely depended on pH value, size, shape and components of MPs, dosage and type of coagulant and flocculant aids. It is essential that future study focuses on finding the best coagulants/flocculant aids and their optimum conditions for MPs removal.

Electrocoagulation (EC)

EC is an advanced technology of the chemical coagulation process, which is comparatively cost-effective, energy-efficient, and capable of being automated with the help of electrodes. Environmentally friendly EC has also been used to remove PE MPs in a stirred batch reactor (Perren et al. 2018). EC is a technique in which coagulant is generated in situ by means of oxidation by an anode usually made of aluminium or iron. A typical EC reactor comprises several electrolytic cells, each containing a cathode and an anode, which can be made of the same or different materials (Moussa et al. 2017). Due to the electrical current supply (electrons flow) aluminium or iron are transferred from an anode material to a solution in the form of Al^{3+} or Fe^{2+} . Simultaneously, the evolution of hydrogen gas and release of hydroxide anions occur at a cathode. Hydroxide anions move towards an anode and form ionic pairs with metals' cations. Those pairs form polymeric aluminium or iron hydroxides, i.e., compounds responsible for coagulation. These coagulants break up the emulsion or colloids and make changes in the stabilization of the surface charges of suspended

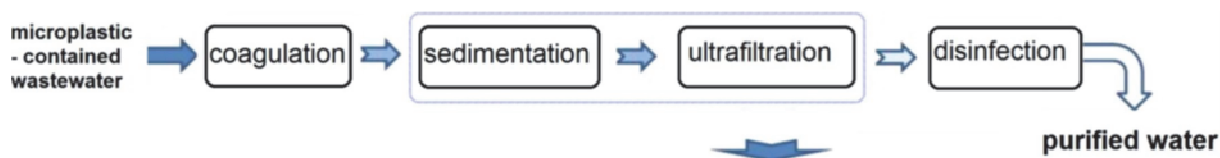


Fig. 2. Scheme of the coagulation UF process for removal of MPs (Ma et al. 2019b)

MPs, which permits them to become close enough to each other and thus attached via Van der Waals forces (Akbal and Camcı 2011).

EC has been reported to be effective over pH 3–10 (Padervand et al. 2020), which makes it more attractive as an effective method for MPs removal from many types of wastewater and their effluents without adding other chemicals. Also, alteration in current density rarely affects the efficacy of MPs removal, which is above 90% (Perren et al. 2018). In the presence of 0.2 g/L of NaCl and pH 7.5, 99.24% of microbeads were removed using this method (Padervand et al. 2020). Recently, Akarsu and Deniz (2020) obtained up to 98% MPs removal from laundry wastewater using a Fe/Al electrode in this process within only 60 min. Despite having such cost-effective and efficient performance in MPs treatments, the EC process has some operational drawbacks such as requirements for continuous replacement of sacrificial anodes, cathodic passivation and high cost of power supply. The development of more viable anodes and future research on operational modifications to avoid the cathodic passivation are required to overcome these limitations.

Oxidation

Several studies have reported MPs removal by oxidizing agents (e.g. ozone, hydrogen peroxides, oxidizing acids) as well as some advanced oxidation processes (AOP). Chemical oxidation aims to mineralize the polymers and convert them into CO₂, water and other substances. In some cases, radiation from different sources (e.g. UV-vis radiation, solar energy), electric current and ultrasound are used to improve the efficiencies of these oxidation processes (Miao et al. 2020). Advanced oxidation processes (AOPs) have gained much attention in recent years, by mineralizing the targeted substances by producing highly reactive radicals (e.g. •OH) (Klavarioti et al. 2009). Although many AOPs have been developed and implemented for wastewater treatment purposes (Feng et al. 2011), very few of them are used for MPs treatment. Ozonation, photo-Fenton, electro-Fenton methods and photocatalytic oxidations are the most widely used and efficient methods.

Ozonation. Ozone as one of the most potent oxidants can react with various polymeric substances, with unsaturated bonds as well as the aromatic rings of the polymers (Ahmed et al. 2017). Some studies have confirmed its effective effect on polymer degradation (Chen et al. 2018), via highly reactive secondary forms of oxidants (e.g. hydroxyl radicals). This process is applied either as a direct treatment method for MPs removal or used to improve the efficacy of some conventional biological methods. There is some evidence of significant changes in PE, PP and PET polymers exposed to ozone. For instance, ozonation may facilitate polymer degradation by increasing polymer surface tension, boosting the polymer surface's adhesion properties, reducing hydrophobicity and increasing solubility, reducing intrinsic viscosity, and decreasing melting points of the polymers and modifying mechanical properties. Chen et al. (2018) reported a high polymer degradation rate (>90%) at 35–45°C by exposure to ozone. Hidayaturrehman and Lee (2019) obtained the highest MPs (particle size 1–5 µm) removal efficiency in ozonation method (89.9%) compared to other advanced treatment methods such as membrane disc-filter (79.4%) and

rapid sand filtration (73.8%). Moreover, the enhancement of 17.2–22.2% removal efficacy was obtained by integrating the GAC-filtration method with ozonation. There is evidence of enhancing microbial mineralization and removal efficacy of MPs with ozonation. In a laboratory-based ozone investigation, mineralization of β-14C PS films by *Penicillium variabile* was found to be increased significantly from 0.01% to 0.15% (Tian et al. 2017). These cases proved that ozonation could be used as a useful tertiary treatment step in wastewater treatment. The main challenge with ozonation is the high production cost of ozone and environmental issues (Ahmed et al. 2017).

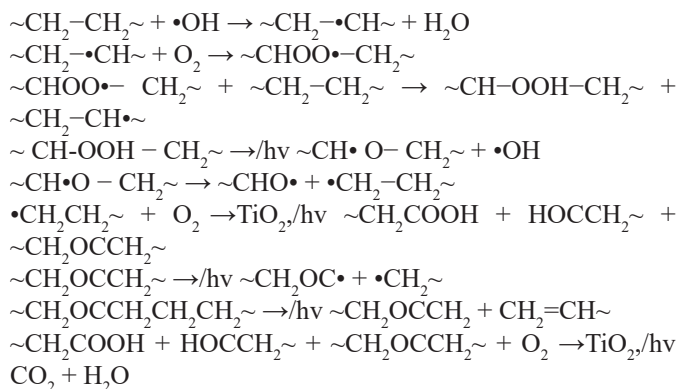
Fenton process. Fenton process has been one of the most widely used AOPs for wastewater treatment. In this process, highly reactive hydroxyl radicals are generated from the reaction of hydrogen peroxides (H₂O₂) and Fe²⁺-containing heterogeneous catalysts, which further oxidize the targeted organic impurities and other contaminants to CO₂, water and mineral products. The Fenton process enhances the oxidation power of H₂O₂ with the help of an iron-catalyst. As iron is a non-toxic and abundant element in the environment, this method has become popular. MPs are rarely affected by the Fenton process. Tagg et al. (2017) examined the influence of Fenton's reagent on PE, PP and PVC MPs, and observed no significant changes in any of the polymers even at three different doses of H₂O₂ and FeSO₄·H₂O. An average of 25.5% removal efficacy was obtained within 24 h through this modified Fenton process. A significant effect of pH was observed, as an enhancement of 1.69–3.89% removal efficacy was attained by adding sodium pyrophosphate as a chelating agent (at pH 7.95).

To overcome the limitations of the classical Fenton process, new methods have been developed in which the main oxidant (H₂O₂) is generated electrochemically (de Luna et al. 2012) or uses UV radiation to generate •OH radicals from H₂O₂ molecules in the presence of an Fe(II) catalyst (Ahmed et al. 2017). Miao et al. (2020) applied the electro-Fenton method to the degradation of PVC MPs and obtained 75% dichlorination efficiency and 56% mass loss efficiency within a 6 h experiment at 100°C. A TiO₂/graphite cathode was used to achieve this high efficiency. Since this method is effective for PVC MPs, it may also be a potential method for other chlorinated MPs species such as 2,4-dichlorophenol, PS, PP and PE (Miao et al. 2020). While the traditional Fenton process was not able to induce significant changes in MPs solely due to the •OH radicals produced, UV irradiation increased the rate of oxidative degradation. Feng et al. (2011) found more than 99% mineralization of crosslinked sulphonated PS foams in just 250 min by a photochemically assisted Fenton process. Overall, more investigation is required for the effective implementation of the Fenton process in MPs treatment.

Photocatalysis. The photocatalysis process initiates with the excitation of the corresponding photocatalyst through the absorption of an appropriate amount of energy from a definite light source (Bodzek et al. 2021). Photo-excitation results in the generation of photogenerated electron and hole pairs eCB⁻/hVB⁺ (CB = conduction band, VB = valence band). Valence band holes hVB⁺ react with adsorbed molecules of water and hydroxyl groups on the surface of heterogeneous photocatalyst, producing hydroxyl radicals (•OH). At the same time, electrons eCB⁻ reduce O₂ in the solution to form superoxide anion (•O₂⁻), which undergoes reaction with water,

affording hydroperoxyl radicals ($\bullet\text{HOO}$). The highly reactive radicals then oxidize various organic contaminants, including polymers effectively (Ali et al. 2016, Padervand et al. 2020, Ouyang et al. 2021). It has been proposed as an energy-efficient, durable and cost-effective process for polymer degradation (Tofa et al. 2019).

Different mechanisms of MPs photocatalysis have been proposed, including the hydroxyl radicals promoting degradation process (Liang et al. 2013). Various nanostructured semiconductors are used as photocatalysts to generate the desired reactive species, of which metal oxide nanomaterials having semiconducting properties with a particular bandgap (ZnO , TiO_2) are most appropriate (Bodzek et al. 2021). ZnO nanoparticles are considered as one of the most promising photocatalysts due to their appropriate bandgap for catalysis (3.37 eV), high redox potential, non-toxicity, excellent electron mobility and flexibility in sizes and shapes to be formed (Qi et al. 2017). Photocatalytic degradation of low-density PE based MPs was investigated through heterogeneous rod-like zinc oxide nano-catalysts (Tofa et al. 2019). From the optical images, morphological changes including the appearance of wrinkles, brittleness, cracks and spots on photo-exposed surfaces of the microplastics were observed. Also, the results revealed variations in the elasticity properties of the sample exposed to photocatalytic conditions in comparison with non-irradiated wastewater, and this is directly in correlation to the changes in the strength of chemical bonds. Liang et al. (2013) obtained FTIR data which confirmed the presence of newly formed functional groups such as carbonyl and vinyl during the photocatalytic treatment. They proposed the following mechanism for the mineralization of microplastics in wastewater:



Protein-based N-TiO_2 photocatalysts were also reported to hold the potential to degrade MPs in both aqueous and solid phases. Ariza-Tarazona et al. (2019) obtained 6.40% mass loss of high-density polyethylene MPs within 18 h, while irradiating it with visible light radiations in the presence of N-TiO_2 photocatalysts. The catalyst surface area, as well as the extent and nature of interactions between the MPs and catalyst surface, influenced the removal efficacy significantly (Ariza-Tarazona et al. 2019).

Photo-active micromotors have gained massive attention during recent years due to their extensive capability for environmental contaminants remediation and water purification (Eskandarloo et al. 2017, Zhang et al. 2018). Micromotors are very small particles that can move themselves in a specific direction autonomously when placed in a chemical solution (Hermanová and Pumera 2022). Recently, several studies have been conducted on the degradation capability and mechanism of TiO_2 -based nano-devices and micromotors in photocatalysis of MPs. MPs have been treated in the photocatalytic process by using Au-decorated TiO_2 -micromotors to make this process more efficient (Wang et al. 2019b). The micromotor propulsion is supplied by photochemical reactions in water and hydrogen peroxide initiated by electron-hole generation processes (Fig. 4).

The performance of micromotors in removing MPs was tested on commercially supplied PS MPs, primary MPs isolated from personal care products, and MPs collected from the Baltic Sea and the Warnow River (Hermanová and Pumera 2022). As this method is very new in MPs treatment compared to other conventional and advanced treatment methods, future research is essential to obtain more effective and efficient advanced photocatalysts, so that the method can be successfully applied to MPs treatment in real wastewater.

Comparison of chemical treatment technologies

Overall, the application of chemical treatment methods significantly enhances the MPs removal efficacy of WWTPs. An overview of each of the methods, their advantages, obtained efficiencies and drawbacks is represented in Table 4. Photocatalytic degradation is a potential strategy but very few WWTPs have implemented this method so far due to their miscellaneous drawbacks (Table 3). The average efficiencies of MPs removal obtained with chemical methods followed the order: photo-Fenton process > electro-coagulation >

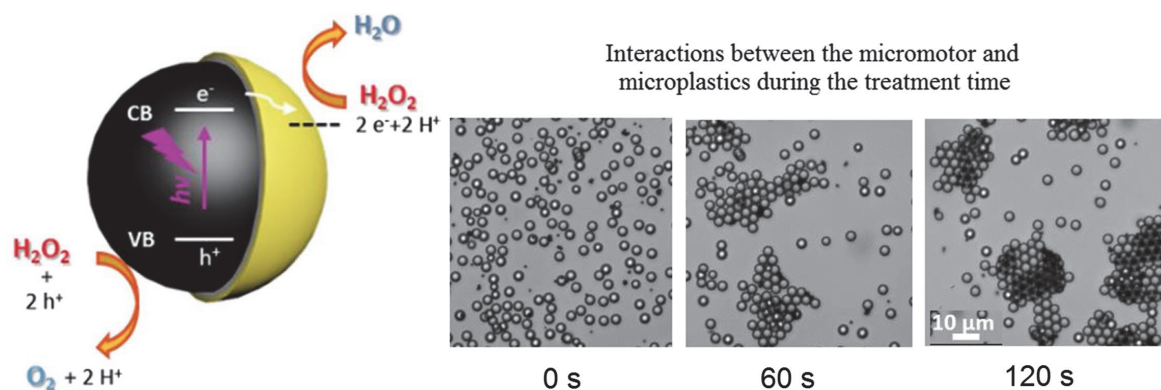


Fig. 4. Photocatalytic removal of PS MPs in water by an Au-Ni- TiO_2 micromotor (Wang et al. 2019b). The images represent the interactions leading to photodegradation of the MPs on the surface of the micromoto

ozonation > electro-Fenton process > solgel agglomeration > coagulation > modified Fenton process. Unfortunately, none of these treatment strategies can remove MPs from contaminated sludge and wastewater when implemented alone without any other physical or biological treatment strategies. Moreover, by-products, as well as some secondary sludge produced in some methods such as coagulation and EC, require further treatment.

AOPs are widely studied and applied for treating different recalcitrant pollutants in the environment. Because ROS produced in AOPs could effectively degrade many pollutants, several AOPs have been studied to remove MPs. Homogeneous and heterogeneous AOPs, including UV photolysis, UV/H₂O₂, O₃, UV/visible light-induced photocatalysis, heat activated PS and PMS, and plasma, could effectively decompose various types of MPs with different sizes. However, the decomposition mostly occurred on the surface of MPs even though several studies reported complete removal of MPs (Kim et al. 2022).

One of the disadvantages of chemical methods of removing MPs from the aquatic environment is the possibility of fragmentation of MPs during oxidation and mineralization (Gerritse et al. 2020). Fragmentation of plastics is thought to be initiated by polymer chain backbone weathering through exposure to sunlight (UV), oxidants, hydrolysis and physical shearing, for example, through currents, waves, or friction with sand. The oxidation and shortening of polymer chains and leaching of plasticizers makes plastic materials brittle and stimulates the formation of surface cracks and fragmentation. As a result, micro- and nanometer sized plastic particles may be released from the surface of larger fragments. In time this

can result in the generation of numerous micro- and nanoplastic particles from a single plastic object.

Biological methods of MPs removal

Microorganisms in MPs removal

Biological methods use organisms to address the contamination of MPs present in the environment by degrading them. Several organisms have been tested for their potential to degrade MPs present in water and wastewater, among them mostly microorganisms show potential for MPs degradation (Harrison et al. 2011). As shown in Fig. 5, microorganisms can break down complex plastic polymers to simpler monomer forms. Aerobic degradation results in CO₂ and water as products while anaerobically it forms CO₂, water, methane and H₂S (Chandra and Enespa 2020, Badola et al. 2022). Several microorganisms have been successfully tested in this process, first of all fungi, and bacteria. A list of such microorganisms is mentioned in Table 4 (Badola et al. 2022).

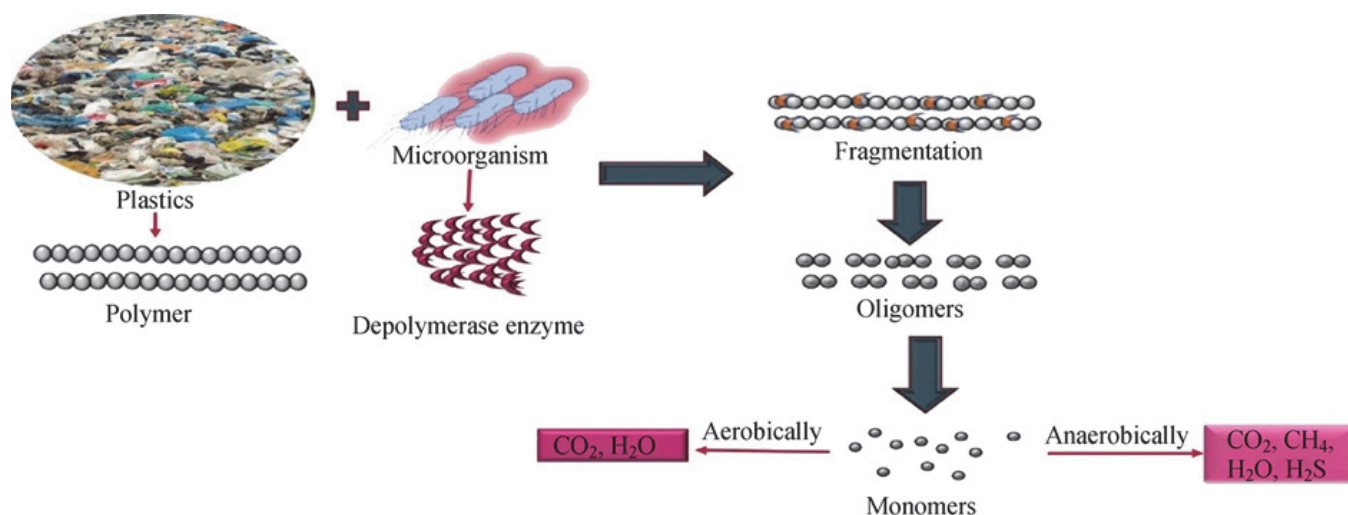
In addition to microorganisms, some other organisms also proved to adsorb MPs in water. Research works have been conducted on organisms like Red Sea giant clam (*Tridacna maxima*) (Arossa et al. 2019), Antarctic krill (*Euphausia superba*) (Dawson et al. 2018), some Corals and microalgae (Corona et al. 2020, Cunha et al. 2020) for assessing their capacity to adsorb MPs, but their reported efficiency was very low. It is important to mention that the accumulation of MPs in the tissues of mussels is unfavorable due to the high consumption of seafood in some parts of the world. In fish, MPs accumulate in the gills and the digestive system. In a study

Table 3. Advantages and challenges of chemical treatment methods for MPs removal

Process type	Efficiency	Advantages	Disadvantages	References
Ozonisation	89.9%	<ul style="list-style-type: none"> Efficient tertiary treatment. Facilitate treatment by modifying properties 	<ul style="list-style-type: none"> Difficulties in ozone production. High operational cost. 	Ahmed et al. 2017
Modified Fenton process	25.49%	<ul style="list-style-type: none"> Reagents availability. Cost-effective process. 	<ul style="list-style-type: none"> Lower efficacy. Applied for specific type MPs. Optimal and cost-efficient reagents 	Tagg et al. 2017
Electro-Fenton process	75%	<ul style="list-style-type: none"> Eco-friendly process. Highly efficient method. Lower reagent costs. Lower sludge production. 	<ul style="list-style-type: none"> Excessive cost requirements. Required modifications. Required investigation for application on different MPs. 	Miao et al. 2020
Photo-Fenton process	>99%	<ul style="list-style-type: none"> Highly efficient method. No requirements of excessive catalysts or reagents. 	<ul style="list-style-type: none"> pH should be maintained in an optimum level. More investigation is required for practical uses. 	Ahmed et al. 2017, Feng et al. 2011
Photocatalytic degradation	Possible complete mineralization	<ul style="list-style-type: none"> No additional chemicals. Ecofriendly process. Efficient mineralization of particles with the help of solar energy. 	<ul style="list-style-type: none"> Lower efficacy. Generation by-products. High energy-consuming process Require photo-reactor and difficult to recover. 	Ariza-Tarazona et al. 2019, Tofa et al. 2019, Wang et al. 2019b
Coagulation	61%	<ul style="list-style-type: none"> Controllable operational conditions. Adequate for small MPs. 	<ul style="list-style-type: none"> Inappropriate for large-sized MPs. Uses of additional chemicals 	Hidayaturrahman and Lee 2019
EC	>90%	<ul style="list-style-type: none"> Minimal sludge. Adequate for smallest MPs. Cost-effective. No secondary contaminants 	<ul style="list-style-type: none"> Sacrificial anodes are required to be replaced repeatedly. Cathode passivation is observed. Electricity is required. 	Perren et al. 2018

Table 4. List of microorganisms used to remove MPs

Microorganism	Type of microorganism	Type of plastic	Efficiency	Reference
<i>Bacillus subtilis</i>	Bacteria	Polyethylene	9.26%	Vimala and Mathew 2016
<i>Serratia marcescens marcescens</i>	Bacteria	Low Density PE	–	Odusanya et al. 2013
<i>Rhodococcus ruber</i>	Bacteria	Polyethylene	8%	Orr et al. 2004
<i>Chaetomium globosum</i>	Fungus	Polyurethane	–	Oprea and Doroftei 2011
<i>Bacillus sphericus</i> Alt; <i>Bacillus cereus</i> BF20	Bacteria	Low Density Polyethylene	Weight loss 2.5–10%	Sudhakar et al. 2008
<i>Zalerion maritimum</i>	Fungus	PE granules	–	Paço et al. 2017
<i>Alcanivorax borkumensis</i> Low-	Bacteria	Low Density PE film	Weight loss 3,5%	Delacuvellerie et al. 2019
Cyanobacteria: <i>Phormidium lucidum</i> and <i>Oscillatoria subbrevis</i>	Bacteria	Low Density PE	–	Sarmah and Rout 2019
<i>Exiguobacterium</i> sp. YT2	Bacteria	Polystyrene film	7.5%	Yang et al. 2015
<i>Paenibacillus urinalis</i> NA26; <i>Bacillus</i> sp. NB6; <i>Pseudomonas aeruginosa</i> NB26	Bacteria	Polystyrene film	–	Atiq et al. 2010
<i>Bacillus cereus</i> <i>Bacillus gottheilii</i>	Bacteria	PE, PS, PET and PP	Weight loss: 0.0019/day 0.0016/day	Auta et al. 2017
<i>Bacillus</i> sp. Strain 27; <i>Rhodococcus</i> sp. Strain 36	Bacteria	Polypropylene	4–6.4%	Auta et al. 2018
<i>Aneurinibacillus aneurinilyticus</i> ; <i>Brevibacillus agri</i> ; <i>Brevibacillus</i> sp.; <i>Brevibacillus brevis</i>	Bacteria	polypropylene film and granules	22.8–27.0%	Skariyachan et al. 2018
<i>Stenotrophomonas panacihumi</i>	Bacteria	PP film	–	Jeon and Kim 2016
<i>Pseudomonas citronellolis</i>	Bacteria	PVC film	13%	Giacomucci et al. 2019
<i>Mycobacterium</i> sp. NK0301	Bacteria	PVC film	–	Nakamiya et al. 2005
<i>Aspergillus</i> sp. S45	Fungus	Polyester film	15–20%	Osman et al. 2018
<i>Penicillium</i> sp.	Fungus	polyester/ polyether film	8.9%	Magnin et al. 2019
<i>Acinetobacter gernerii</i>	Bacteria		–	Howard et al. 2012
<i>Bacillus muralis</i>	Bacteria	PET	–	Narciso-Ortiz et al. 2020
<i>Zalerion maritimum</i>	Fungus	Polyethylene	43%	Paço et al. 2017
<i>Rhodococcus ruber</i>	Bacteria	Polyethylene	8%	Orr et al. 2004

**Fig. 5.** Degradation of plastic particles under the influence of microorganisms (Badola et al. 2022, Chandra and Enespa, 2020)

by Corona et al. (2020), MPs removal efficiency of mushroom coral collected from the reef of the island of Magoodhoo, Faafu Atoll, Republic of Maldives showed efficiency of 97% for the size of 200–1000 μm . PE MPs fragmentation and size alteration ingested by Antarctic Krill (*Euphausia superba*), a planktonic crustacean, were studied by a group of environmentalists in Australia (Dawson et al. 2018). The experiments confirmed that smaller microplastics are much more easily fragmented under environmental conditions, and the physical size decreased from $\sim 31 \mu\text{m}$ for the MPs to less than $1 \mu\text{m}$ for the fragmentation products (Dawson et al. 2018).

Traditionally, plastics were considered as non-biodegradable items but now these are known to be degraded and metabolized by different organisms, especially by microbes. The abundance of microorganisms in the environment and their potential in attacking MPs seems to be one of the most effective solutions to MPs. Moreover, several enzymes that are capable of hydrolyzing the different plastics have been identified (Wei and Zimmermann 2017). Another recent biological technique which works on the mechanism of microbial ‘trap and release’, was engineered for MPs removal (Liu et al. 2021). In this method, MPs are efficiently trapped and aggregated in sticky exopolymeric substances produced by engineered bacterium, *Pseudomonas aeruginosa* biofilms and then the trapped MPs can be dispersed or released by biofilm dispersal mechanism for downstream resource recovery or recycling. This ‘trap-and-release’ bio-aggregation method works for every type and size of plastic material. Further, it does not depend on the concentration of MPs. The increased total mass will help simple and easier removal by filtration or sedimentation in tanks.

Biological wastewater treatment processes

Secondary treatment aims to treat the wastewater emanating from primary treatment and eliminate the residual organics and suspended solids. The secondary treatment in WWTPs combines biological treatment processes and clarification processes (Sun et al. 2019, Tang et al. 2020). In this stage, aerobic or anaerobic biological treatment methods are employed to remove dissolved and colloidal biodegradable organic matter. In addition, it uses an alternating system anaerobic, anoxic & oxic (A^2O) for biological nutrient removal. Activated sludge (AS) and biological beds (BF) (effluent filters/biofilters), membrane bioreactors (MBR) and hydrotreatment plants (constructed wetlands) are most commonly and widely used technologies for secondary treatment of wastewater and the most effective methods for MPs removal (Talvitie et al. 2017b). Figure 6 shows the standard secondary treatment processes in which MPs removal efficiencies are tested. The MPs removal efficiencies of the standard secondary treatment processes are summarized in Table 5.

As shown in Figure 6, the **activated sludge (AS) system** removes MPs mainly by entrapment in sediment flocs, degradation due to ingestion by protozoa or metazoans, and microorganisms and formation of sludge aggregates (Jeong et al. 2016). Sludge containing microplastics was removed during the subsequent degradation secondary settling process (Jeong et al. 2016). MPs removal in this process occurs via adsorption or aggregation. Microorganisms secrete extracellular polymeric substances (EPS) to absorb the available contaminants as well

as MPs and then degrade them to produce desired products. Sometimes, microorganisms take up MPs by mistake due to visual similarity with their nutrients and then discard them after agglomeration into flocs due to their inability to decompose or convert them into harmless substances (Ahmed et al. 2021). In addition, chemicals such as ferrous sulphate or other coagulants used during secondary treatment can have a positive effect on MPs removal because they can cause suspended solids to aggregate into flocs (Murphy et al. 2016). Lares et al. (2018) demonstrated very high removal efficiency (98%) of MPs in a classical AS process. Similarly, Murphy et al. (2016) and Edo et al. (2020) also proved that this technology can remove up to 92.6% and 93.7% of MPs, respectively. Another study by Hidayaturrahman and Lee (2019) found that MPs removal efficiency ranged from 42% to 77%, and Bayo et al. (2020a,b) in a municipal wastewater treatment plant (Spain) also found MPs removal of about 62%. A study of municipal wastewater treatment systems in Italy revealed that about 64% of MPs were removed after using a grid chamber and AS system (Magni et al. 2019). Other researchers reported 2–55% removal of MPs in biological treatment processes (Lv et al. 2019, Yang et al. 2019). Ziajahromi et al. (2017) found a removal rate of MPs from activated sludge of 66.7%, while in an A^2O process only 28.1% was removed in a Wuhan wastewater treatment plant, China (Liu et al. 2019b) and 54.47% in a Beijing wastewater treatment plant in China (Yang et al. 2019). The removal efficiency of MPs in the A^2O process is relatively low due to the sludge return. Furthermore, the degradation of MPs in A^2O is quite slow. In this context, the conventional classical activated sludge method is more advantageous for MPs removal in wastewater treatment plants. Overall, the efficiency of MPs removal by AS is not stable and varies over a relatively wide range.

The AS process shows variation for different sizes and shapes of MPs (Zhang et al. 2020a). For example, Liu et al. (2019a) found that most of the MPs removed in the AS process were $< 300 \mu\text{m}$ in size, whereas other researchers obtained the highest removal efficacy for 1–5 mm sized particles (Lares et al. 2018). In addition, during the secondary treatment, more MPs fragments are removed than fibers. This was supported by the studies showing that the relative abundance of MPs fragments decreased while that of fibers increased after the secondary treatment (Talvitie et al. 2015, 2017a, Ziajahromi et al. 2017). The average fiber concentration was 25 times higher than other MPs fragments (Talvitie et al. 2015). One possible reason is that the easily settled or skimmed fibers had already been largely removed during the pretreatment, whereas those remaining might have some properties, such as neutral buoyancy, which was resistant to being further removed. In terms of sizes, large MPs particles can be further removed during the secondary treatment, resulting in a relatively low amount in the secondary effluent. Studies showed that MPs with a size larger than 500 μm were almost absent from the secondary effluent (Mintenig et al. 2017, Ziajahromi et al. 2017). Talvitie et al. (2017a) found that microparticles $> 300 \mu\text{m}$ only account for 8% after secondary treatment. Equally important is the relationship between MPs’ size and their form in WWTP effluent. An assessment was made and it was found that among MPs with a size of $> 500 \mu\text{m}$, PE (mean 59%) and PP (mean 16%) dominated

(Nocoń et al. 2018, Moraczewska-Majkut et al. 2021). In contrast, Dris et al. (2015) found that microplastics within the size range of 500–1000 nm still accounted for 43% after secondary treatment. The reason for this high proportion was unclear. It might be related to specific microplastic removal efficiency achieved by various secondary treatment processes with different operational conditions. Variations in MPs removal are due to (i) the change of microbes, (ii) the nature of the MPs in the wastewater (size, shape, surface structure), and (iii) abiotic factors (e.g. temperature, pH) (Ahmed et al. 2021). The other influencing factors that could affect the MPs removal rate by the activated sludge process are the retention time (Carr et al. 2016) and nutrient level in wastewater (Rummel et al. 2017). The longer the contact time, the higher are the chances of surface biofilm coating on the plastic debris that modifies the surface, size and relative densities of the contaminants (Carr et al. 2016). Such changes may make a significant impact on the neutrally buoyant MPs to increase the likelihood of eliminating them by skimming or settling processes, which then improves the removal rate of the wastewater treatment technology.

Constructed wetlands (CWs) are familiar and natural technology for wastewater treatment with a comparatively lower cost than other biological treatment methods. Studies have been conducted recently to investigate the feasibility of MPs removal from wastewater using CWs (Liu et al. 2019b, Ziajahromi et al. 2020). Vegetated wetlands are the prime locus for detaching, storing, transforming and finally releasing MPs particles (Helcoski et al. 2020). A few studies have been conducted on the contribution or performance of vegetated wetlands, including natural and CWs in MPs removal from polluted water (Helcoski et al. 2020, Wang et al. 2020d). Wang et al. (2020d) showed the effective role of macro-invertebrates (e.g. snails, bristle worms, beetles) in MPs distribution throughout the wetlands. They claimed that macro-invertebrates in the wetlands ingest a non-negligible amount of MPs. Over 90% removal efficacy was achieved in both horizontal and vertical flow type CWs, which is comparable with other conventional tertiary treatment methods of WWTPs, such as biological filtration (84%), dissolved air flotation (95%), DF (40–98.5%), MBR (99.9%) and sand filters (97.1%). The time for MPs degradation is correlated

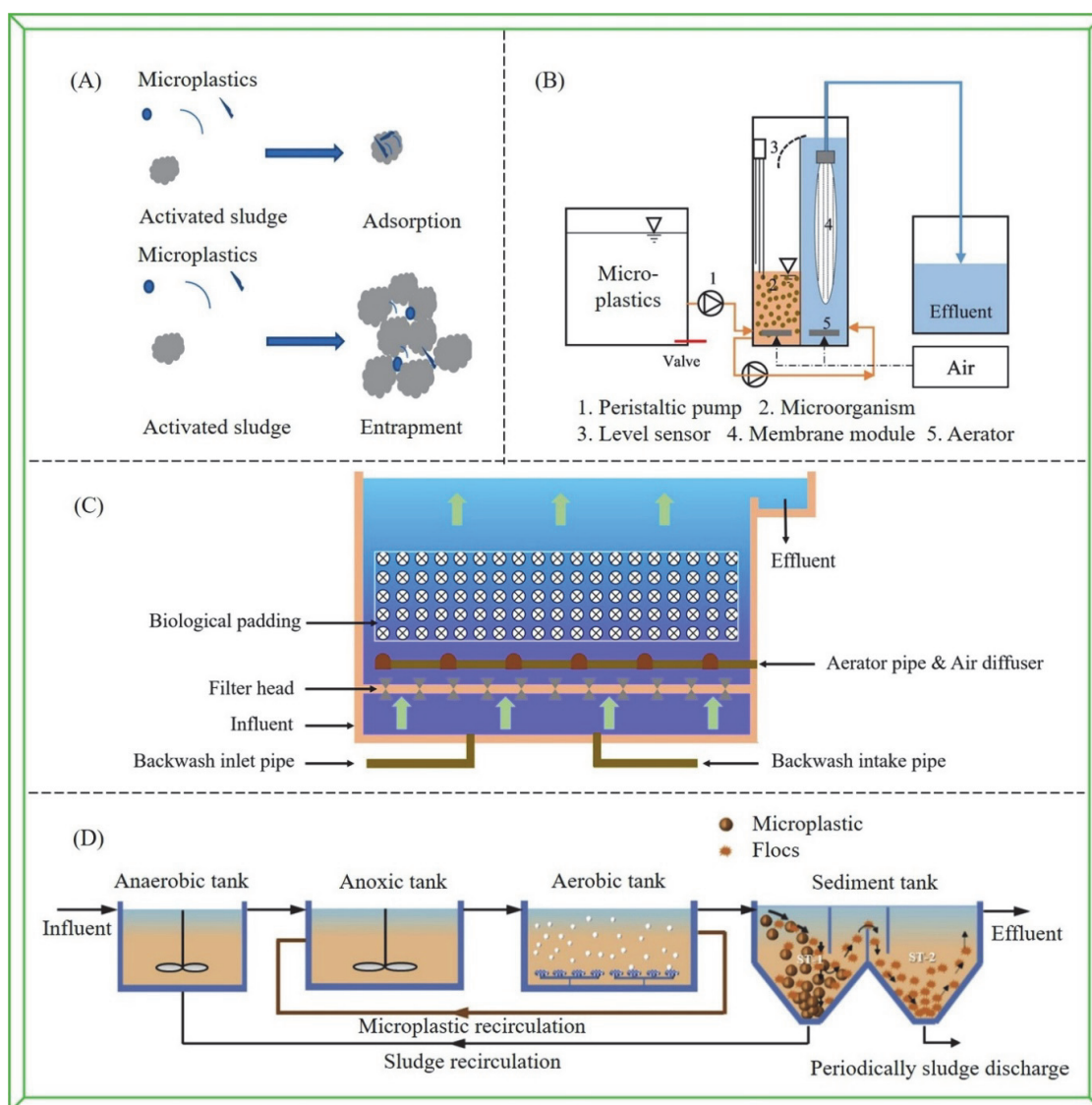


Fig. 6. The schematics of the bioreactor systems in microplastics removal. (A) Activated sludge process (Zhang et al. 2020a), (B) MBR (Li et al. 2020), (C) Biofilter, (D) A²O (Liu et al. 2020)

between habitats differing in the density and stem cover of wetland vegetation and the type and form of MPs. 98% MPs removal efficacy was obtained through the whole WWTP when CWs were used in its tertiary treatment steps. Therefore, CWs can be an efficient, environmentally friendly and cost-effective tertiary treatment process to significantly reduce MPs from wastewater. Moreover, the efficacy can be enhanced through integrating different features of different types of CWs (e.g. surface flow CWs, subsurface flow vertical type, subsurface flow horizontal type CWs). Therefore, further combined applications of such different CWs are strongly recommended for MPs removal from wastewater.

Biofilter technology integrates physical and biological purification processes, and biofilm filtration and adsorption were the main mechanisms for MPs removal. The microbe film growing on the surface of the inert filter material is in contact with the MPs and increases the contact area between MPs and microorganisms. Excess microbes and retained microplastics are easily removed by backwashing in the ascendant water flow (Rocher et al. 2012). Biological bed/biofilter technology is often used after a bioreactor system. MPs entering the biofilter are smaller in size and lower in density, which increases the difficulty of MPs removal. However, biofilter technology still shows the highest MPs removal efficiency

Due to incomplete pollutant removal, undecomposed MPs present in sewage sludge readily infiltrate terrestrial ecosystems and re-disperse throughout the environment. Effective removal of MPs can be ensured by an additional element of the WWTP, such as a filtration process (Moraczewska-Majkut et al. 2021). Filtration devices (e.g. membrane filtration) should be placed at the post-treatment stage, i.e., after the secondary sedimentation tank. This could prevent the increasing amounts of microplastics in the effluent. The fate and processing of these undecomposed MPs in the sludge phase have rarely been discussed in the literature. Therefore, further research on this topic is urgently needed.

Membrane bioreactor (MBR)

Membrane bioreactor (MBR) is a term generally used to define wastewater treatment processes where a perm-selective membrane, e.g., generally MF or UF, is integrated

with a biological process promoted by biological catalysts (bacteria, enzymes) (Poerio et al. 2019, Xiao et al. 2019). The removal mechanism is dual in nature, i.e., biodegradation and membrane filtration. MBR configurations, both aerobic and anaerobic MBRs, can be divided into two classes as side-stream MBR (sMBR) (membrane module outside bioreactor) and submerged in bioreactor (iMBR) (Fig. 7) (Poerio et al. 2019, Xiao et al. 2019).

iMBR offers a lower cleaning frequency, and lower energy consumption, but, otherwise, sMBR can handle higher MLSS (Mixed liquor suspended solids) concentration than iMBR. For this reason, it is easier to carry out maintenance operations and module replacements and cleaning since the system is more compacted. During this process, the UF/MF membrane directly separates solids from mixed liquid in biological reactors. The MBR method can eliminate the secondary clarifier and stop any biological solids loss within the effluent and permit a really high concentration of biomass. It provides large flux and fine filter precision. Furthermore, the versatility of this technology permits an easy integration with other processes (e.g. pervaporation, reverse osmosis), perfectly in line with green chemistry principles, within the logic of process intensification, which offers numerous new opportunities in terms of competitiveness, product quality improvement, process or product novelty and environmental friendliness (Judd 2016). Nowadays, MBR is deemed to be one of the most powerful technologies for efficient municipal and industrial wastewater treatment around the world (Poerio et al. 2019, Xiao et al. 2019). This technology provides significant improvement, with respect to the traditional methods of wastewater treatment, such as high effluent quality, small footprint, complete separation of hydraulic retention time (HRT), and solids retention time (STR), easy scale-up, etc. Regarding fouling control, various methods have been developed with this technology. They comprise, for example: intermittent permeation or relaxation, membrane backwashing, air backwashing and using specified proprietary antifouling products.

The combination of pressure-driven membrane techniques with a biological process (MBR) could enhance the rate of MPs removal from primary effluent. In MPs treatment, the role

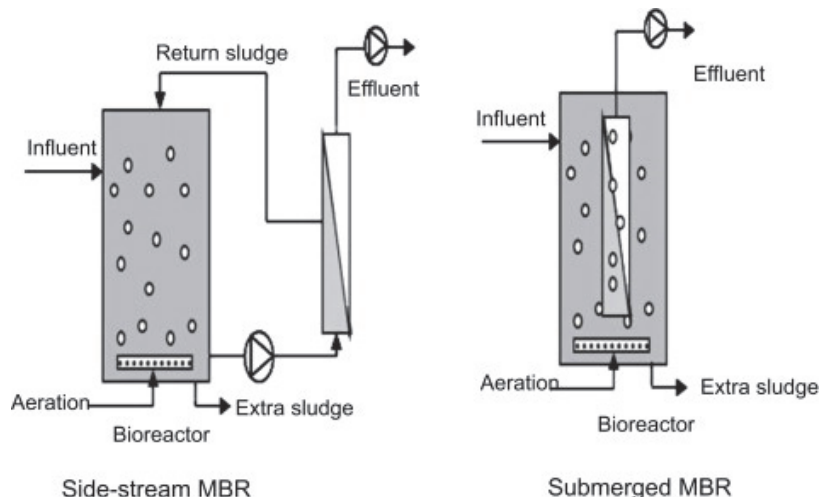


Fig. 7. Membrane bioreactor (MBR) configurations (Poerio et al. 2019, Xiao et al. 2019)

of MBR is to reduce solution complexity by the biodegradation of the organic matter; this will permit the removal of MPs and their further treatment. Thanks to the membrane process, the MPs is concentrated in the retentate stream.

In the work (Talvitie et al. 2017b), the performance of MBR was compared with other final-stage wastewater treatment technologies (disc-filter, rapid sand filtration, and dissolved air flotation) for MPs removal (Table 5). The MBR removed 99.9% of MPs during the treatment (from 6.9 to 0.005 MP/L), rapid sand filter 97% (from 0.7 to 0.02 MP/L), dissolved air flotation 95% (from 2.0 to 0.1 MP/L) and disc-filter 40–98.5% (from 0.5–2.0 to 0.03–0.3 MP/L) of the MPs during the treatment. The study shows that with advanced final-stage wastewater treatment technologies, WWTPs can substantially reduce the MPs pollution discharged from wastewater treatment plants into aquatic environments. An MBR containing 20 submerged flat sheet UF membranes with 0.4 µm pore size and 8 m² surface area showed a significant improvement in MPs removal (99%), higher quality of final effluent, and a great potentiality in decreasing the number of process stages, replacing the conventional secondary clarifiers in conventional AS. MPs removal from 6.9±1.0 item/L to 0.005±0.004 item/L was achieved (Talvitie et al. 2017a). The studies showed that MBR allowed for the highest reduction of MPs in the final effluent, demonstrating that the membrane-based technology is the most efficient.

Similarly, Lares et al. (2018) obtained 99.4% MPs removal, which indicated that the MPs removal rate of MBR is consistent and significant. MBR is characterized by a high removal capacity for all size fractions (especially the smallest size, 20–100 µm) and all shapes of MPs from wastewater compared to other advanced treatment (Talvitie et al. 2017b). However, compared with other treatment technologies, the performance of MBR seems to be not influenced by the shape, size and composition of MPs. Most recently, Li et al. (2020) studied the effectiveness of PVC gel removal (particle size < 5 µm) by the MBR with a 0.1-µm submerged membrane and a 0.1-m² surface area. Under operating conditions of 2.5-hour HRT, temperatures around 19.1°C and pH 7.5, the results showed that virtually no MPs were detected in the permeate of the MBR system. Baresel et al. (2019) investigated an MBR combined with a 0.3 m² granular activated carbon biofilter for the removal of MPs from real wastewater from Henriksda WWTP (Stockholm) with a 10-hour HTR. A UF system was applied downstream of the biological reactor. A 100% removal efficiency of MPs in MBR treated wastewater was obtained.

It has been observed that MPs of smaller sizes, especially fibers, cannot be completely removed by MBR due to their high length-to-width ratio (Ngo et al. 2019, Freeman et al. 2020). MPs, therefore, remain in the sludge after filtration, which needs to be treated again as solid waste, resulting in

a possible increasing treatment cost. Other major limitations of MBR technology in wastewater treatment are the control of biofilm thickness, fouling and liquid distribution, which determine the effectiveness of the method (Poerio et al. 2019, Bui et al. 2020). The effect of MPs on membrane fouling and the degradation and/or transformation of MPs in MBR should be studied in future research. Many studies have concluded that MBR is highly effective and relatively stable in removing MPs. Therefore, MBR may be the most effective technology so far among the common wastewater treatment technologies for the elimination of MPs from wastewater.

A very promising discovery, which could in the future be associated with MBR technology, is the isolation of a novel bacterium (*Idonella sakaiensis*) able to use PET as its major energy and carbon source (Yoshida et al. 2016). This bacterium produces two different enzymes when in contact with PET, which can efficiently convert PET into the less dangerous monomers (terephthalic acid and ethylene glycol). Dawson et al. (2018) recently reported the size reduction of MPs (from 31.5 µm to less than 1 µm) when exposed to Antarctic Krill (*Euphausia superba*). The enzymes can be easily integrated with the MBR, so in the future it will probably be possible to degrade the MPs in the enzymatic membrane reactor, as already demonstrated for PET degradation (Barth et al. 2015).

Comparison of different biological processes

Biological treatment methods can be applied to remove MPs to a significant extent from different environmental conditions; the MBR process and CWs showed the best efficacy among all of them. Conventional AC also achieved a similar removal percentage, but only in limited studies. It is difficult to declare any exact removal percentage for microbial treatment processes because they always fluctuate according to the microorganisms involved. On the other hand, aerobic digestion and AD can be applied efficiently only for biodegradable MPs particles. A comparative overview of the biological methods, their advantages and drawbacks are summarized in Table 6. The removal of MPs via biological methods decreased in the order: MBR > CWs > activated sludge > microbe processes. The MBR process and CWs have potential in leading biological methods of MPs removal.

MPs' fragmentation and size alteration through ingestion by various microorganisms, e.g., zooplankton, marine fungi and bacterial strains, has been confirmed by a group of environmentalists (Dawson et al. 2018). But the mechanism of fragmentation and type of interactions between MPs and zooplankton in which biota-facilitated degradation occurs, still remains unclear. However, the experiments confirmed that smaller MPs are much more easily fragmented under environmental conditions. Dawson et al. (2018) hypothesize that fragmented MPs have increased potential for interaction

Table 5. MPs concentrations before and after treatment with different technologies (Talvitie et al. 2017a,b Padervand et al. 2020)

Method	Effluent type	Before (MP/L)	After (MP/L)	Removal (%)
Disc filter 10 µm	After 2nd stage treatment	0,5	0,3	40,0
Disc filter 20 µm	After 2nd stage treatment	2,0	0,03	98,5
Rapid sand filter	After 2nd stage treatment	0,7	0,02	97,1
Dissolved air flotation	After 2nd stage treatment	2,0	0,1	95,0
Membrane bioreactor	After 1st stage treatment	6,9	0,005	99,9

at the molecular level, as seen in other MPs studies, and this warrants significant attention to nanoparticle toxicology in the discussions surrounding global plastic pollution. WWTPs generally provide a high MPs removal rate, with a large proportion of MPs trapped during WWTPs processes (Xu et al. 2021). Therefore, a majority of the remaining MPs are transferred to WWTP sludge, which is often directly applied in green construction and as an agricultural fertilizer, resulting in sludge being a significant route for the release of MPs into soils and aquatic environments.

Concluding remarks

The occurrence and impact of plastic particles in water bodies is increasing worldwide. According to the literature, millions of tons of micro- and nano-sized plastic particles enter the aquatic environment every year. Studies on microplastic hazards and separation have been growing over the past decade. With the current research on MPs particles, a number of methods have been developed and evaluated to help fill this research gap in the future. WWTPs are a significant source of MPs in addition to domestic and industrial sources. MPs have been found to act as an important vector for various pollutants such as heavy metals, additive mixtures, surfactants, antibiotics, pesticides and pharmaceuticals. Various methods

of purification of MPs are discussed in terms of their effectiveness, and their advantages and limitations. Filtration is considered to be the most effective physical method for the removal of MPs, although further work is still needed for its implementation in large-scale municipal wastewater treatment. CWs and MBR technologies are the most efficient among biological treatment methods. In chemical treatment, EC, coagulation, photo- and electro-Fenton methods show promising results in MPs removal. Hybrid treatment such as the MBR-UF/RO system, coagulation followed by ozonation, GAC, DAF, RS, filtration, and CWs based hybrid technologies have shown very promising results in the effective removal of MPs.

An efficient method for MPs treatment and a policy that can be implemented strictly across the globe is urgently warranted to control MPs in the environment. Most research regarding the removal of MPs is conducted in-vitro under controlled conditions, and there is a high likelihood of a reduction in efficiency under natural conditions. When these methods are performed in real case scenarios, such as for treatment of wastewater, which is a mixture of contaminants, efficiency could alter and show different results for different treatment methods. Although wastewater treatment plants have shown good efficiency, there is an urgent need to create and add specific MPs removal units to water treatment plants.

Table 6. Different types of MPs removed in biological wastewater treatment process

Treatment process	Efficiency (%)	Type of microplastic removed	References
Submerged MBR (KUBOTA)	100.0		Talvitie et al. (2017a)
Submerged MBR	100.0		Li et al. (2020)
MBR	99.9	20–100 µm MPs	Talvitie et al. (2017b)
MBR	99.4%	PES, PE, PA and PP	Lares et al., 2018
MBR	99	PVC Fragments, fibres	Lv et al. (2019)
BAF biological aerated filter	99	PE100–300 µm	Talvitie et al. (2017b)
Anaerobic submerged MBR	99.4		Lares et al. (2018)
AS	98.3%	Various types	Lares et al., 2018
MBR	97.6	PES fibres and PE fragments	Lares et al. (2018)
OD oxidation ditch	97	Fragments, fibres	
AS USA	95.9	SAL	Michielssen et al. 2016
ASP	93.8	Microbeads	Michielssen et al. (2016)
A ² O	93.7	PE, PP, PE and acrylic fibres	Edo et al. 2020
AS and clarification	92	Fragments, fibres	Blair et al. (2019)
TF and AS	89.8	Microbeads	
MBR, AS, and settling tank	83.1–91.9	Fragments	Hidayaturrehman and Lee (2019)
MBR	79.01	Fibres, PP, PS	Bayo et al. (2020a,b)
AS South Korea	75–91.9	Primary and secondary MPs	Hidayaturrehman and Lee (2019)
A/A/O	71.67±11.58	Not mentioned	Yang et al. (2019)
AS	66.7	Polystyrene	Ziajahromi et al. (2017)
AS, sedimentation	64	Fibres	Magni et al. (2019)
A ² O	54.4	–	Yang et al. (2019)
AS Slovenia	52	PE <100µm	Kalčíková et al. (2017)
A ² O	28.1	PET, PE, PES, PAN, PAA	Liu et al. (2019a,b)
Anoxic tank, aeration basin, clarifier	2.4	Smaller microplastics	Alavian Petroody et al. (2020)

Although significant progress has been made in MPs research in terms of their analysis, interactions with other contaminants, toxicological effects, and removal by different treatment technologies, there are still many gaps. Future research directions on MPs are suggested as follows:

- Membrane-based treatment, to minimize membrane fouling and increase MPs removal.
- Mechanism of MPs degradation and/or transformation in MBR.
- Isolation and amplification of a number of MPs degrading microbes for their targeted applications.
- MPs removal from the sludge phase produced in biological treatment methods.
- Further development of CWs for application in MPs removal.
- Synthesizing new cathode materials for efficient removal of MPs in Fenton processes.
- Utilization of solar energy for commercial-scale photocatalysis treatment plants for MPs removal.
- More bio-inspired materials and their cost-efficient synthesis routes for the sol-gel agglomeration method should be sought.
- Development of more viable anodes for the EC method.
- Hybrid treatments are needed to be specially designed to remove MPs.
- Development of new modelling techniques to evaluate the transport route of MPs in the soil, sediments and water.

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