# **Chapter 5 Microplastics in Wastewater Treatment Plants: Occurrence, Fate and Mitigation Strategies**



**Angel Joseph, Azmat Naseem, and Arya Vijayanandan**

**Abstract** Environmental contamination caused by microplastics (MPs) is an issue of grave concern which is pervasive in global water, air, and soil. Wastewater treatment plants (WWTPs) are recognized as the vital source of MPs into the environment. Hence understanding the fate and behavior of MPs in WWTPs is of utmost importance. Studies have also reported the presence of MPs in the treated effluent. Challenges in MPs removal in different treatment units need to be identified, and advancements of existing treatment technologies are to be explored for enhancing the removal of MPs in WWTPs. This chapter presents the occurrence and fate of MPs in different treatment processes such as biodegradation, adsorption, membrane processes, filtration, electrocoagulation, and advanced oxidation processes. The removal efficiency of MPs in different treatment units, the mechanism of removal, and the challenges involved in removing MPs in each treatment unit are discussed in detail. The efficiency of the existing treatment technologies in WWTPs are compared, and the modifications suggested by recent studies to improve the removal of MPs are presented in this chapter.

**Keywords** Microplastics · Occurrence · Mitigation · Advanced oxidation processes · Adsorption · Bioreactor · Electrocoagulation · Filtration · Wastewater treatment

# **5.1 Introduction**

Plastics are considered the modern marvel due to their benefits across all the fields ranging from health to food sectors saving countless lives (Golwala et al. [2021;](#page-14-0) Wang et al. [2021\)](#page-19-0). The property that makes the plastics suitable also renders their ubiquitous presence in the environment (Guo et al. [2020;](#page-15-0) Yang et al. [2021\)](#page-19-1). Depending on the usage pattern of plastic, the shelf life of the plastic items varies from 1 to 50 years or beyond before they are dumped as trash from where it can be recycled (9%),

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abandoned in the landfill (8%), utilized for energy recovery (12%), or dissipated in the environment (71%) (Dris et al. [2015a;](#page-14-1) Gregory [2009;](#page-15-1) Foerster [2018\)](#page-14-2). Microplastics (MPs) are of significant concern de to their persistence and capability to act as carriers for other toxic pollutants such as dioxins, aldrin, perfluorooctane sulfonic acid, triclosan, tonalite chlordane, hexachlorobenzene, etc. (Mammo et al. [2020\)](#page-17-0). Once MPs enter the food chain, it would result in bioaccumulation and will impart adverse health effects on living organisms and human beings as well (Miller et al. [2020\)](#page-17-1). It is imperative to highlight that MPs are not a single type of contaminant but a suite of contaminants and are considered among the emerging contaminants (Jain et al. [2021\)](#page-15-2). Owing to this serious concern, the European Parliament recently framed a resolution proposal (TA/2019/0071) emphasizing the urgent need for addressing MPs contamination related to wastewater treatment (Edo et al. [2020\)](#page-14-3). MPs are small fragments (≤5 mm), synthetic polymer fragments which have either been deliberately manufactured in smaller sizes known as primary MPs or developed due to the disintegration of bigger plastic known as secondary MPs (Frias and Nash [2019;](#page-14-4) Hartmann et al. [2019\)](#page-15-3).

MPs are reported to be extensively present in the soil (Guo et al. [2020\)](#page-15-0), oceans (Wang et al. [2020c\)](#page-19-2), atmosphere (Abbasi et al. [2019\)](#page-13-0), freshwater systems (Han et al. [2020\)](#page-15-4), and are also detected in the sediments (González-Pleiter et al. [2019\)](#page-14-5). It is essential to consider that WWTPs majorly act as the source for the entry of MPs into the environment as single treatment plant effluent can release up to  $10^{10}$  MPs per day (Mintenig et al. [2017\)](#page-17-2). These fragments of plastics, whether in larger or smaller forms, would cause serious adverse repercussions for biota, ecosystems, and the environment (Hartline et al. [2016;](#page-15-5) He et al. [2020;](#page-15-6) Magnusson and Norén [2014;](#page-17-3) Michielssen et al. [2016;](#page-17-4) Ou and Zeng [2018;](#page-17-5) Rummel et al. [2017;](#page-17-6) Wang et al. [2020b;](#page-19-3) Wright et al. [2020\)](#page-19-4). In the natural hydrological cycle, an appreciable proportion of MPs is detected in lakes, oceans, rivers, atmospheric precipitation, and wastewater and treated water (Alia et al. [2016;](#page-13-1) Andrady, [2011;](#page-13-2) Dris et al. [2015b\)](#page-14-6). In the case of the anthropogenic hydrological cycle, micro-sized litter finds its way in sewers (Tagg et al. [2015\)](#page-18-0), which are ultimately sent to the influent load of the WWTPs (Carr et al. [2016\)](#page-13-3).

Previous studies focused on the source, composition, and concentration of MPs have reported that the MPs in the influent of WWTPs are majorly composed of small-sized fibers discharged from laundries (Baldwin et al. [2011\)](#page-13-4), microbeads used in personal grooming products (Ezel et al. [2016\)](#page-14-7). Pollutants such as heavy metals (Yazdani Foshtomi et al. [2019\)](#page-19-5), polycyclic aromatic hydrocarbons (Sørensen et al. [2020\)](#page-18-1), pharmaceutical compounds and cosmetic products (Liu et al. [2019a;](#page-16-0) Ma et al. [2019\)](#page-16-1), and polybrominated biphenyl ethers (Singla et al. [2020\)](#page-18-2) gets adsorbed from surrounding media to MPs due to their large surface area and low volume (Thompson et al. [2004\)](#page-18-3). High concentrations of micro-sized plastic litter in water affect the efficiency of water purification processes like filtration units that get choked or malfunctioned due to clogging (Chesters et al. [2013;](#page-14-8) Guo et al. [2012\)](#page-15-7). There is also a possibility that MPs experience shear forces generated by pumping or mixing, which further disintegrates MPs into much smaller fragments leading to the formation of nano plastics released in the water matrix (Lee et al. [2019\)](#page-16-2).

The research focusing on the sources, occurrence, and fate of MPs in WWTPs has gained a lot of attention in recent years (Bakaraki Turan et al. [2021;](#page-13-5) Edo et al. [2020;](#page-14-3) Habib et al. [2020;](#page-15-8) Liu et al. [2021;](#page-16-3) Ngo et al. [2019;](#page-17-7) Raju et al. [2018;](#page-17-8) Sun et al. [2019;](#page-18-4)

Xu et al. [2021\)](#page-19-6). Various technologies are being adopted for the removal of MPs at different treatment stages. A detailed review of the occurrence and the fate of MPs in WWTPs are presented in this chapter. In addition, this article aims to provide a comprehensive review of the mechanism of removal of MPs in specific treatment units for a better understanding of research gaps that need to be addressed in the future. Occurrence of MPs in wastewater treatment plants.

Contamination of MPs in municipal sewage is reported in several parts of the world (Iyare et al. [2020\)](#page-15-9). WWTPs are considered the prime source of MPs in the water bodies (Tang et al. [2020a,](#page-18-5) [2020b\)](#page-18-6) MPs are mainly introduced into wastewater through stormwater discharge and outlets from residential structures and multiple production units (Salmi et al. [2021\)](#page-18-7). The release of MPs can occur throughout the life cycle of the plastic products from the production and formulation stage to transport, utilization, and final disposal (Wang et al. [2021\)](#page-19-0). These MPs are being transported to different environmental matrices through runoff, sewage, landfills, industries, and weathering (Lambert et al. [2014\)](#page-16-4).

The origin and composition of MPs found in WWTPs are challenging to analyze due to their smaller particle size and wide range of unknown sources (Kaliszewicz et al. [2020;](#page-15-10) Ziajahromi et al. [2016,](#page-19-7) [2017\)](#page-19-8). Primary MPs detected in WWTPs include tiny beads that are used in several cosmetic products such as scrubs, face wash, toothpaste, etc. (Chang [2015;](#page-14-9) Fendall and Sewell [2009\)](#page-14-10). The other sources can be plastic pellets, fishing gear, and building paints which yield small-sized MPs (0.5–6 mm) released into water systems (Kärrman et al. [2016\)](#page-16-5). However, the most considerable contribution is from the automobile sector. The tires release 50,000–135,000 tons of MPs of range  $> 0.01$  mm, and the smallest recorded micro fragments of size  $> 5 \mu m$ originate from fertilizers and detergents (Viveknand et al. [2021\)](#page-19-9). A common source of MPs in the WWTPs is microbeads (Kärrman et al. [2016\)](#page-16-5), and it is estimated that nearly 94,500 microbeads are discharged on single wash from cosmetic products (Napper et al. [2015\)](#page-17-9).

Secondary MPs directly discharged into the sewers include tiny fiber litter (of size  $12-16 \mu m$ ) from clothing, and it is estimated that every wash of a single garment using a household machine would generate > 1900 fibers (Browne et al. [2011\)](#page-13-6). The formation of secondary MPs is due to regular disintegration of larger plastics under nominal atmospheric conditions due to exposure to the sunlight or by mechanical disintegration during pumping and mixing in WWTPs (Eriksen et al. [2014\)](#page-14-11). Larger plastic items are broken down by different mechanisms such as hydrolysis, photo-oxidation, mechanical breakage caused by turbulent water or sand abrasion, or microorganisms-assisted bio-assimilation (Gewert et al. [2015;](#page-14-12) Niaounakis [2017\)](#page-17-10). Determining particle shape, size, amount, and surface chemistry of MPs is challenging, especially in the case of secondary MPs (Andrady [2017\)](#page-13-7). In a recent study, the samples obtained from the WWTPs reported MPs' presence that can be categorized into six shapes: pellets, granules, fibers, films, fragments, and foams (Yang et al. [2021\)](#page-19-1). Fibers and fragments are most abundantly found in WWTPs, and these fibers possessing a greater length to width ratio, are difficult to remove in the treatment units (Sol et al. [2020\)](#page-18-8).

Physical, chemical, and biological treatment techniques could be responsible for changing the configuration of primary MPs to secondary MPs (González-Pleiter et al. [2019\)](#page-14-5), rendering WWTPs the key contributor of secondary MPs in the terrestrial as well as in aquatic media (Liu et al. [2021\)](#page-16-3). Depending upon the level of treatment, WWTPs have a high propensity of removing MPs; however, they are critical contributors to contamination as water remains the prime carrier for the diffusion of MPs in the environment (Bayo et al. [2020;](#page-13-8) Bretas Alvim et al. [2020;](#page-13-9) Edo et al. [2020;](#page-14-3) Sol et al. [2020;](#page-18-8) Ziajahromi et al. [2016\)](#page-19-7). At various treatment levels involved in WWTPs, such as preliminary followed by primary, secondary, and terminating at the tertiary, approximately 87–99% of plastic litter is removed from the sewage (Viveknand et al. [2021\)](#page-19-9). A schematic representation of the sources and pathway of MPs during in WWTPs is shown in Fig. [5.1.](#page-3-0)

Despite the high rates of removal of MPs in WWTP, still, a large proportion of this debris-loaded wastewater is regularly released to the water streams (Carr et al. [2016\)](#page-13-3). A Scotland-based treatment plant was reported to release about  $6.5 \times 107$  MPs daily into its neighborhood (Murphy et al. [2016\)](#page-17-11), and another WWTP in Italy was found to discharge around  $1.6 \times 10^8$  MPs per day (Magni et al. [2019\)](#page-16-6). Murphy et al. [2016](#page-17-11) reported that the influent loading of MPs in a WWTP in England was close to 15.70  $(\pm 5.20)$  particles/L and was lowered to 0.25 ( $\pm$  0.04) particles/L in the discharged effluent. A study conducted in a WWTP in China reported that the MPs with mass < 10 kg were released into the water system per day, and the proportion of fragments is abundant (Lv et al.  $2019$ ). Gies et al.  $(2018)$  estimated the dominance of different types of MPs such as fibers (65.6%), fragments (28.1%), pellets (5.4%), granules  $(0.45\%)$ , foam  $(0.22\%)$ , and sheets  $(0.20\%)$  in samples collected from a WWTP in Canada.



<span id="page-3-0"></span>**Fig. 5.1** Different sources, occurrence and pathways of MPs in WWTPs (figure modified from Zhang et al.[\(2020\)](#page-19-10))

# **5.2 Treatment Methods and the Mechanism Involved in Removing MPs from WWTPs**

Existing WWTPs are not carefully designed for removing MPs from wastewater. Literature suggests that a significant quantity of MPs was released to the environment in each treatment option adopted in the WWTPs (Joo et al. [2021\)](#page-15-11). Depending upon the treatment methods and physicochemical characteristics of the polymer, such as particle density, size, charge, hydrophobicity, etc., variable removal efficiencies can be expected in different treatment technologies (Bond et al. [2018\)](#page-13-10). The efficacy of any removal treatment method is highly dependent on the quality of influent and the characteristics of MPs in it.

Primary treatments involving coarse screening remove most of the MPs, while grit sedimentation, screening, degreasing, and primary sedimentation contribute to further removal. The biological processes followed by sedimentation in secondary treatment accounts for approximately 20% removal efficiency (Xu et al. [2021\)](#page-19-6). About 69–79% of MPs introduced into WWTPs are eliminated at the primary screening stage with grit removal methods (Ziajahromi et al. [2021\)](#page-19-11). Low-weight floating MPs could bypass the grease skimming method (Sun et al. [2019\)](#page-18-4). Primary sedimentation could remove particles with a spherical shape having a diameter  $> 27-149 \,\mu m$  (Iyare et al. [2020\)](#page-15-9). In combination with the grit and grease removal units, the primary settling unit potentially improves the efficacy of removing MPs. A recent study reported that treatment units such as sedimentation and skimming units have the potential to remove almost 72% of MPs present in sewage influent (Xu et al. [2021\)](#page-19-6).

Another study reported higher removal efficiency for MPs having size (106–  $300 \,\mu$ m) than MPs with range ( $>300 \,\mu$ m) (Lee and Kim, [2018\)](#page-16-8). Smaller MPs can be trapped easily in the grit and grease removal stage and adhere to the biofilm or flocs. A study conducted by Liu et al. (2019) suggested that MPs removal is closely associated with turbidity removal from primary sedimentation as well as secondary treatment. In WWTPs, the major fraction of microbeads ends up in the sludge, with the remaining microbeads released into receiving streams (Mason et al. [2016;](#page-17-12) Murphy et al. [2016\)](#page-17-11). Many of the available treatment techniques eliminate MPs by entrapping this litter in sludge; however, dedicated techniques for removing MPs are lacking. There is still the persistence of MPs in the final effluent discharged via WWTPs, signifying the inability of existing treatment methods in completely eliminating MPs in wastewater (Ziajahromi et al. [2017\)](#page-19-8). Secondary and tertiary treatments in WWTPs remove 88– 94% of MPs loaded in the sewage influent, and about 72% removal efficacy was reported during preliminary and primary treatment (Iyare et al. [2020\)](#page-15-9). The following section discusses the detailed removal mechanisms of MPs in specific treatment units in WWTPs.

#### *5.2.1 Biological Processes*

Bioreactors are adopted as a secondary treatment option for the removal of MPs in WWTPs. The MPs are hardly degraded within a short period. The extracellular polymer substances secreted by the microorganisms facilitate the initial trapping of MPs (Zhang et al. [2020\)](#page-19-10). The degradation of MPs by the microbes followed by the formation of sludge aggregates causes the effective removal of MPs in bioreactors. The accumulated MPs in the sludge were then removed in a secondary clarifier (Jeong et al. [2016\)](#page-15-12). Recent studies have reported the biological degradation of MPs by various organisms (Yuan et al. [2020;](#page-19-12) Miri et al. [2021\)](#page-17-13). Polymer digestion by the microbes present in the sludge is the plausible mechanism of MPs removal (Miri et al. [2022\)](#page-17-14).

Biodegradation of MPs takes place in four successive steps: colonization, biofragmentation, assimilation, and mineralization (Dussud and Ghiglione [2014\)](#page-14-14). The biodegradation of MPs initiates with the formation of microbial colonies over the surface of the polymer (Oberbeckmann et al. [2015\)](#page-17-15). This colonization causes macromolecular modifications on the MPs by physical (biofilms forcing cracks and pits on the polymer surface) and chemical (the secretion of acids by the microbial colonies weakens the polymer structure) pathways (Yoshida et al. [2016\)](#page-19-13). Moreover, the enzymes secreted on the MPs aids in disintegrating the polymer to a low molecular weight oligomer during bio-fragmentation (Skariyachan et al. [2018\)](#page-18-9). The bio-fragmentation stage is continued with subsequent assimilation of fragmented oligomers by the microbial colony. This stage is called assimilation, and in the final stage of mineralization, bacteria consume all useful energy out of oligomers and release oxidized waste into the surrounding environment through multiple biochemical pathways (Hou et al. [2021;](#page-15-13) Miri et al. [2022\)](#page-17-14). However, in most of the biological treatment processes, degradation of MPs is minimum; rather removal occurs due to adsorption and entrapment (Zhang et al. [2020\)](#page-19-10).

The activated sludge process (ASP) has been effective in mitigating wastewater MPs after preliminary treatments (Bretas Alvim et al. [2021\)](#page-13-11). The mineralization of MPs in ASP involves two processes (a) sorption and (b) degradation (Fig. [5.2\)](#page-5-0). Most

<span id="page-5-0"></span>

of the secondary treatment facilities in WWTPs combine biological processes with a clarification unit for the effective removal of biomass which also aids in the removal of MPs (Tang et al. [2020a,](#page-18-5) [2020b\)](#page-18-6). The biological flocs promote growth on MPs' surface, which causes accumulation of MPs debris in the system, and subsequently, MPs may get settled in the clarifier (Hurley et al. [2018\)](#page-15-14). Experimental results from a lab-scale SBR detected the presence of smaller fragments in the sludge and larger MPs in the effluent with a removal efficiency of 52% (Kalčíková et al. [2017\)](#page-15-15).

The introduction of biofilters to remove MPs is considered a very simple and innovative concept. Liu et al. [\(2020\)](#page-16-9) tested the biofilter consisting of different layers of stone wool as an advanced refining step for treated wastewater. The secondary effluent entered the filter through the top portion, and treated effluent was discharged from the bottom of the filter (Liu et al. [2020\)](#page-16-9). The study reported that the filter could reduce MPs by 79–89% in effluents of the wastewater treatment plant. However, it is also observed that the biofilters are more recommended for the removal of larger plastic fragments, and hence a complete MPs removal could not be achieved by this method (Kuoppamäki et al. [2021\)](#page-16-10). Maximum removal occurred at the top of the biofilter with the lower layers allowing limited surplus treatment efficacy (Thuptimdang et al. [2021\)](#page-18-10). Large-sized MPs were retained by biofilter with no particles greater than 100  $\mu$ m from the filter in the final effluent (Kuoppamäki et al. [2021\)](#page-16-10).

Lee and Kim [\(2018\)](#page-16-8) compared the removal efficiencies of 3 different configurations of the activated sludge process for MPs removal: sequencing batch reactor (SBR), anaerobic–anoxic–oxic  $(A^2O)$ , and media process  $(A^2O)$  basin with filled carrier). It was observed that all processes attained removal efficiencies > 98%, and SBR reported the highest rate of 99.2%. Jiang et al. 2020 reported that the anoxic– oxic process (A/O) removed approximately 17% of MPs in wastewater. MPs show higher retention potential in most of the biological processes, and the treatment of MPs fraction with the sludge is of great concern (Liu et al. [2019c\)](#page-16-11).

A significant portion of the MPs is reported to be accumulated in the waste activated sludge (WAS). WAS produced in the specific treatment units is further treated with an anaerobic digester (AD) (Wei et al. [2019\)](#page-19-14). The treatment of WAS enriched with MPs has been challenging in recent times. The size and composition of the MPs are the predominant factors that determine the efficacy of anaerobic digestion (Chen et al. [2021\)](#page-14-15). The composition of WAS enriched with larger-sized fractions of MPs has shown a reduction in methane production than in smaller fragments (Wei et al. [2019;](#page-19-14) Zhang et al. [2020\)](#page-19-10). It has been reported that the presence of MPs in the WAS reduces the efficiency of the anaerobic digestion as the MPs also act as a carrier for the other antibiotics and other toxic pollutants (Wei et al. [2019\)](#page-19-14). A recent study has investigated the co-effect of MPs and antibiotics in the anaerobic digestion and observed adsorption of antibiotics to MPs due to its hydrophobicity and larger specific area (Wang et al. [2020a\)](#page-19-15). This causes a reduction in the anaerobic efficiency and the prolonged residence of antibiotics in the anaerobic digester, which causes inhibition in antibiotic degradation (Zhu et al. [2018\)](#page-19-16). Higher content of MPs in WAS causes inhibition of the hydrolysis stage of anaerobic digestion that results in the increased formation of dissolved organic matter (Chen et al. [2021\)](#page-14-15). This further causes the inhibition of the acidogenesis stage that leads to lower methane

production (Lin et al. [2020\)](#page-16-12). In general, biodegradation of MPs in anaerobic digestion processes is difficult, and understanding the fate of MPs in the system is essential. The leaching of monomers during anaerobic digestion is another matter of concern, and the mechanisms involved in the anaerobic digestion of MPs are still not clearly understood.

# *5.2.2 Filtration*

Rapid sand filtration (RSF) is the commonly adopted low-cost treatment method for the removal of MPs. The removal of MPs has been enhanced by incorporating RSF units as the last stage option in tertiary treatment (Padervand et al. [2020\)](#page-17-16). After the systematic investigation of WWTPs in Finland, it was observed that RSF could remove 97% of MPs present in the effluent from the secondary treatment stage (Talvitie et al. [2017\)](#page-18-11). Although higher removal efficiencies are being reported, the main demerit of such systems is that it causes fragmentation of MPs. Hence the magnitude as well the surface properties of the MPs are of great importance in this context, as these factors determine the extent of MPs and filter media interaction (Bhattacharjee [2016\)](#page-13-12). A recent study in South Korea reported that RSF and coagulation units could be coupled together to achieve improved removal efficiency (Hidayaturrahman and Lee [2019\)](#page-15-17). Investigation on the transport behavior of smaller fragments of MPs (0.02  $\mu$ m and 2  $\mu$ m) through sand and biochar medium has been evaluated in a recent study (Tong et al. [2020\)](#page-18-12). The study also concluded that improvement in the surface roughness of the filter media has contributed to enhanced interaction between the polymer and caused better deposition of MPs in the filter media. In addition, the development of composite filter media such as biochar/Fe<sub>3</sub>O<sub>4</sub>-biochar having magnetic attraction can act as a permeable barrier and has great potential to immobilize the MPs fragments (Faisal et al. [2018\)](#page-14-16). A study focused on the elucidation of the mechanism behind the immobilization of MPs (Fig. [5.3\)](#page-8-0) in biochar media concluded that stuck, trapped, and entangled are the three factors that control MPs fate in biochar media (Wang et al. [2020d\)](#page-19-17). The development of such cost-effective filter media can improve the overall performance of tertiary treatment systems.

#### *5.2.3 Adsorption*

Several studies have targeted the development of adsorbent media that can be used suitably to adsorb the MPs present in wastewater (Li et al. [2019;](#page-16-13) Nolte et al. [2017\)](#page-17-17). In a recent study, the application of graphene oxide and chitin for the adsorption of MPs had been explored and identified to be an effective method (Sun et al. [2020\)](#page-18-13). The adsorption of MPs to graphene-supported adsorbents is primarily due to the pie bond, H-bond, and electrostatic attraction. Three-dimensional graphene-based adsorbents can be used for the separation of MPs. The adherence and high sorption



<span id="page-8-0"></span>**Fig. 5.3** Mechanisms involved in the removal of MPs using filtration (figure modified from Wang et al.[\(2020d\)](#page-19-17))

of MPs to the surface of the marine microalgae and seaweed have been reported in a recent study (Sundbæk et al. [2018\)](#page-18-14). The alginate released from the seaweed surfaces enhances the sorption capacity, and about 94% of MPs have been removed in that manner (Padervand et al. [2020\)](#page-17-16). The effect of surface charges on MPs and microalgae in the adsorption process has been analyzed in a study conducted by Nolte et al. [\(2017\)](#page-17-17). The study also concluded that the rate of adsorption of positively charged polystyrene is comparatively higher than the negatively charged MPs. Anionic polysaccharides present in the algal chemical structure enhanced the adsorption of MPs on the surface of algae (Dey et al. [2021\)](#page-14-17). Hence the surface charge of the MPs is an important parameter that determines the rate of adsorption. A recent study shows effective MPs removal using granular activated carbon (GAC) adsorption with thermal regeneration. This study proposes to retrofit the existing STP with GAC combined with thermal regeneration as the process ensures more than 92% removal of MPs ranging from 20 to 50  $\mu$ m (Kim & Park [2021\)](#page-16-14).

## *5.2.4 Membrane Technologies*

The application of membranes is highly effective for the treatment of MPs present in wastewater. Such membrane filtration units are reported to remove MPs with higher efficiency and maintain stable effluent quality and ease of treatment (Poerio et al. [2019\)](#page-17-18). It is reported that the influent load and particle concentration are the critical parameters to be considered in the filtration of MPs (Li et al. [2018\)](#page-16-15). The design of polymer coatings with elongated mesh is also found to be an effective method for the removal of MPs (Mohana et al. [2021\)](#page-17-19). Size exclusion, electrostatic interaction, hydrophobic interaction, and biofilm formation are the basic mechanisms by which MPs are usually removed in a membrane technique (Joo et al. [2021\)](#page-15-11).

Coupling the biological reactors with membrane units is another effective option for removing polymeric debris and MPs present in industrial wastewater (Talvitie et al. [2017\)](#page-18-11). A simple schematic representing the removal of MPs while passing through the membrane modules (ultrafiltration unit) is presented in Fig. [5.4.](#page-9-0) The effective interaction between MPs and the ultrafiltration unit results in the complete removal of MPs (Enfrin et al. [2020\)](#page-14-18). A comparison study was conducted by Lv et al. [2019](#page-16-7) between MBR and oxidation ditch. The results revealed that MBR has exceptional efficiency over the oxidation ditch in the treatment of MPs. However, the limiting factors such as membrane congestion and control of biofilm reduce the efficiency of MBR in treating MPs (Lares et al. [2018;](#page-16-16) Nicolella et al. [2000\)](#page-17-20). Studies have reported that smaller plastic fragments are very difficult to be removed in the tertiary treatment units and the treatment options such as biological filters and advanced separation systems are not effective as membrane bioreactors which ensures almost 99.9% removal efficiency (Mason et al. [2016;](#page-17-12) Talvitie et al. [2017\)](#page-18-11). However, optimization of different parameters, including membrane surface charge, fouling phenomena, transmembrane pressure, membrane material, pore size, and hydraulic retention time are important for the effective removal of MPs.

A case study has showcased the capability of removing MPs in effluent at a WWTP using membrane process in Mikkeli, Finland (Hermabessiere et al. [2017\)](#page-15-18). Dynamic membrane (DM) is considered an appealing membrane-based method for wastewater that works on the principle of cake layer-based formation (Poerio et al. [2019\)](#page-17-18). In wastewater treatment, DM performs the function of a secondary membrane filter; however, compared to MF/UF, membrane performance is weakened due to dense fouling and thick layers (Baresel et al. [2019\)](#page-13-13). The reverse osmosis (RO) process can also be effectively implemented in WWTPs to remove MPs. Tertiary treatment involving ultrafiltration and RO are reported to be efficient for removing MPs (Ziajahromi et al. [2017\)](#page-19-8).



<span id="page-9-0"></span>**Fig. 5.4** Removal of MPs using ultrafiltration unit (figure modified from Enfrin et al. [2020\)](#page-14-18)

#### *5.2.5 Electrocoagulation*

The electrocoagulation processes are considered to be an effective method for the removal of MPs as the coagulants produced from the metal electrodes are more likely to encounter the MPs present in wastewater with several advantages, including lower sludge production and minimal operating cost (Akarsu & Deniz, [2021;](#page-13-14) Elkhatib et al. [2021\)](#page-14-19). The performance of electrocoagulation processes has been evaluated using a bench-scale electrocoagulation unit (Fig. [5.5\)](#page-10-0) and found to be most effective in removing MPs at neutral pH as the rate of coagulant formation is higher at neutral pH (Perren et al. [2018\)](#page-17-21).

In a recent study conducted by Ariza-Tarazona et al. [\(2019\)](#page-13-15), the efficiency of iron and aluminum salt coagulants in removing polyethylene has been compared and concluded that aluminum-based coagulants had shown better removal efficiency than iron salt. This study also noted a reduction in the removal efficiency with the rise in pH, and the smaller plastic fragments below 0.5 mm are found to be hardly removed at a higher pH range. The study also inferred that an enhancing coagulation agent such as polyacrylamide had improved the removal efficiency of smaller MPs. Electrocoagulation processes have been successfully employed for the removal of microbeads using aluminum as the electrode material. Perren et al. [\(2018\)](#page-17-21) investigated different parameters, including the effect of pH and electrode to electrode distance that affects the MPs removal rate. Higher removal efficiency (more than 90%) was reported over the lower pH values (pH 3). Kim et al. [\(2021\)](#page-16-14) reported that about 90% of MPs were converted to separable flocs in the electrocoagulation unit, which would later be removed in any tertiary treatment unit.



<span id="page-10-0"></span>**Fig. 5.5** Electrocoagulation unit adopted for the removal of MPs (figure modified from Perren et al. [2018\)](#page-17-21)

### *5.2.6 Advanced Oxidation Processes*

The applicability of the advanced oxidation process (AOP) has been explored as a tertiary treatment option for removing MPs. Recently, the combined influence of Fenton and heat-activated persulfate methods in the long-term degradation of polyethylene and polystyrene MPs have been studied (Liu et al. [2019c\)](#page-16-11). The influence of the average size and oxygen to carbon ratio  $(O/C)$  ratio of MPs in the surface properties and the adsorption capacity was analyzed in this study, and concluded that these parameters have significantly changed the oxidation rate ofMPs. The alterations in the surface of MPs in an advanced oxidation process are usually evaluated for the O/C ratio (Liu et al. [2019b\)](#page-16-17). The morphological and structural variations of polyethylene MPs during the dark and UV light degradation have been evaluated in a recent study (Da Costa et al. 2018). It is observed that salinity increased the rate of degradation of MPs compared to UV irradiation (Suhrhoff et al. [2016\)](#page-18-15). Hence the salinity of the surrounding media plays an important factor in the photodegradation of MPs as salts enhance the formation of oxidized sites (Vasile and Pascu [2005\)](#page-18-16).

The disintegration of MPs by the photocatalytic method has been recently explored and reported as a potential treatment option for the mineralization of polymer (Tofa et al. [2019\)](#page-18-17). The light source is chosen depending upon the type of photocatalyst and the amount of energy required for the extraction of electrons from the valence band of the photocatalyst. Many of the recent studies have effectively utilized photocatalysis for the effective degradation of MPs. The applicability of  $TiO<sub>2</sub>$  (Ariza-Tarazona et al. [2020;](#page-13-16) Luo et al. [2021\)](#page-16-18) and ZnO (Uheida et al. [2021\)](#page-18-18) photocatalysts for the photocatalytic degradation of MPs have been majorly explored in this field. Liang et al. [\(2013\)](#page-16-19) reported improved photocatalytic degradation of low-density polyethylene by grafting hydrophilic polyacrylamide on the  $TiO<sub>2</sub>$  photocatalyst.

The apparent conditions of the degradation for polypropylene (PP), polystyrene (PS), and polyethylene (PE) are cracks/flakes and granular oxidation. However, the most vulnerable MPs that can change the configuration to a smaller range or even nano range are the ones that develop cracks and flakes (Cai et al. [2018\)](#page-13-17). These fissures are referred to as stress concentrators and fractures, formed at the brittle or weak spots of MPs (Cooper, [2012\)](#page-14-20). The application of UV irradiation initiates the splitting of C-H bonds and C–C bonds, which results in the formation of peroxy free radicals (Cai et al. [2018;](#page-13-17) Gewert et al. [2015\)](#page-14-12). Furthermore, application of UV irradiation activates hydroxyl, carbonyl, hydroperoxide, and chromophore groups in the surface of MPs that causes the formation of free radicals and marks the beginning of chain reactions that ultimately leads to the complete mineralization of MPs. The removal efficiency reported with AOP is comparatively lower; however, these methods could achieve maximum mineralization of MPs (Lam et al. 2020).

The pros and cons of different technologies need to be evaluated for selecting the suitable treatment scheme for removing MPs. High energy demand, membrane fouling, sludge accumulation, microbial aggregation on the membrane surface are some of the inherent demerits encountered with conventional treatments and

membrane processes that can be avoided by the extended application of photocatalytic processes with efficient utilization of solar energy. Filtration systems are a comparatively low-cost option for MPs mitigation. However, the lower removal rate of MPs has been reported with filtration units, and this might be due to the longitudinal movement of MPs fragments through the filter media (Sun et al. [2019\)](#page-18-4). Hence additional final stage treatment units should be supported with filtration systems in WWTPs to remove smaller fragments of MPs. In most cases, during the backwashing of the filters, the solids removed may again be sent back to the beginning stage, adding the loading rate of MPs in the WWTPs.

The retention of a significant fraction of MPs in the sewage sludge limits the visibility of biological processes in the treatment of MPs. However, research focusing on the leaching of monomers from MPs during the biodegradation and studies focusing on the transformation of MPs in specific units of WWTPs are scanty. The interactions of MPs with the surface of the membrane are not clearly understood, and further studies need to be conducted in this area. The effect of other pollutants and the operating parameters on the fate of MPs in different treatment units should be investigated, and the underlying mechanisms of removal need to be substantiated. Future studies should also focus on the reaction intermediates generated during photocatalysis and other AOPs during the treatment of MPs.

#### **5.3 Conclusions**

Plastic polymers are inevitable in human lives due to their continued demand in all sectors. Though it seems a considerable removal, still WWTPs are majorly contributing to MPs contamination to the environment. The MPs-loaded effluent released from the WWTPs ultimately merges with waterbodies. Being an emerging contaminant, MPs serve as carriers of metals and organic contaminants. Bioaccumulation of MPs is inevitable considering the cumulative emission of MPs into the water bodies. Therefore, maximizing the removal of MPs in WWTPs would eliminate a possible pathway of MPs into the environment. However, there are uncertainties in comparing the removal efficiencies of MPs in WWTPs due to the multiple possible removal mechanisms and lack of standardized sampling/identification methods. Proper standardization and the periodic assessment of treatment units are required for a greater degree of evaluation of the fate of the MPs in WWTPs. Different technologies are being explored for the removal of MPs, such as adsorption, filtration, membrane technologies, electrocoagulation, and advanced oxidation processes such as photocatalysis. The coupling of different secondary and tertiary schemes would significantly reduce the MPs concentration in the treated effluent. Membrane filtration is reported to be a promising technology; however, further research is required to address the challenges related to membrane fouling issues. Ample attention should be given to the development of treatment methods that focus on the complete mineralization of the MPs to curb the emission, and further studies are needed in this direction.

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