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Microplastics Fouling and Interaction with Polymeric Membranes: a Review

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Abstract

The emergence and accumulation of microplastics (MPs) in various aquatic environments have recently raised significant concerns. Wastewater treatment plants (WWTPs) have been identified as one of the major sources of MPs discharge to the environment, implying a substantial need to improve advanced techniques for more efficient removal of MPs. Polymeric membranes have been proven effective in MPs removal. However, fouling is the main drawback of membrane processes and MPs can foul the membranes due to their small size and specific surface properties. Hence, it is important to investigate the impacts of MPs on membrane fouling to develop efficient membrane-based techniques for MPs removal. Although membrane technologies have a high potential for MPs removal, the interaction of MPs with membranes and their fouling effects have not been critically reviewed. The purpose of this paper is to provide a state-of-the-art review of MPs interaction with membranes and facilitate a better understanding of the relevant limitations and prospects of the membrane technologies. The first section of this paper is dedicated to a review of recent studies on MPs occurrence in WWTPs aiming to determine the most frequent MPs. This is followed by a summary of recent studies on MPs removal using membranes and discussions on the impact of MPs on membrane fouling and other probable issues (abrasion, concentration polarisation, biofouling, etc.). Finally, some recommendations for further research in this area are highlighted. This study serves as a valuable reference for future research on the development of anti-fouling membranes considering these new emerging contaminates.

Keywords: Microplastics, Microfibers, Membrane fouling, Wastewater treatment plants

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Contents

Nomenclature:

Dissolved air flotation	DAF
Disc filter	DF
Fourier-transform infrared spectroscopy	FTIR
Membrane bioreactor	MBR
Microplastics	MPs
Nanoplastics	NPs
Oxidation ditch	OD
Polyamide	PA
Polyacrylamide	PAM
Polyethylene	PE
Polyester	PEST
Polyethylene terephthalate	PET
Polypropylene	PP
Polystyrene	PS
polysulfone	PSF
Polyvinyl chloride	PVC
Reverse osmosis	RO
Forward osmosis	FO
Rapid sand filtration	RSF
Styrene acrylonitrile	SAN
Ultrafiltration	UF
Wastewater treatment plants	WWTPs

1. Introduction

Plastics have become convenience products due to their unusual but practical useful properties (e.g., lightweight, high strength, durability, and low price). Hence, plastic production has intensively increased over the past decades with reports showing a production increase of over 240 folds in less than 100 years, from 1.5 million tons in 1951 to around 359 million tons in 2018 (Can-Güven, 2020; Gunaalan et al., 2020). This has inevitably led to the accumulation of plastic wastes and their fragments, particularly microplastics (MPs), both on land and in water bodies such as oceans (Rocha-Santos and Duarte, 2015; Y.T. Zhang et al., 2020). MPs are defined as plastic particles with a characteristic size smaller than 5 mm (Wright et al., 2013), which can further be classified based on their shape [fragment, fiber, pellet, bead, line, film, foam] and type [e.g., polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polyamide (PA)]. Owing to both the comparatively high buoyancy and durability of MPs, they can remain in aquatic environments for extended periods which can eventually lead to multiple potential environmental and health implications (Klein et al., 2015).

Plastics/MPs can be a significant source of contamination in the oceans. They commonly contain a wide range of additives classified as functional additives, colorants, fillers, and reinforcements; most of which are not chemically bonded to the structure of MPs. Consequently, such additives are readily leached from MPs into the host water (Gunaalan et al., 2020). In addition, MPs have the potential to adsorb toxic chemical substances (e.g., heavy metals and active pharmaceutical compounds) because of their hydrophobicity and high specific surface area (Bakir et al., 2014; F. Yu et al., 2020; H. Yu et al., 2020). A recent study reported MPs as the carrier of a huge content of toxic metals to aquatic environments, where they have the potential to be ingested by marine organisms (Sarkar et al., 2021). Hence, environmental hazard analysis of MPs as well as developing elimination and control strategies to combat the MPs release to the environment have become a focal point of research in recent years.

Recent studies have shown that wastewater treatment plants (WWTPs) are one of the main MPs pathways to aquatic environments, accounting for over 25 % of total MPs release into the oceans (Boucher and Friot, 2017; Eggen et al., 2014; Hidayaturrahman and Lee, 2019; Magni et al., 2019). Although WWTPs generally contain multiple separation stages that can act as individual or integrated barriers against MPs, no associated studies have shown complete MPs removal from wastewater (Franco et al., 2020; Raju et al., 2020; N. Tang et al., 2020). The remaining of even a small percentage of MPs in the WWTPs effluents leads to a high level of contamination in the aquatic environments given the large volume of treated wastewater that is being discharged into them daily. Murphy et al. (2016) studied MPs discharge from a large secondary WWTPs in Scotland and reported that even with the MPs removal rate of 98.41 %, still over 23 Billion MPs are being annually discharged into the oceans through WWTPs alone (Murphy et al., 2016). Therefore, there is a need for additional/modified tertiary treatment procedures in WWTPs to successfully eliminate MPs from WWTPs effluents (Kalčíková et al., 2017; Park et al., 2020; Talvitie et al., 2017).

Current tertiary treatment technologies in WWTPs have not been designed to capture MPs. Intensive research and developmental studies are hence required to enable WWTPs for higher and more efficient MPs removal (Barcelo and Pico, 2020). Among the relevant tertiary treatment techniques, membrane filtration has significant advantages because of its wide market availability and easy retrofitting with minimal cost. The latter can reduce the need for extensive feasibility analysis compared to other treatment techniques, making membrane technology an attractive separation process for targeted MPs removal. MPs removal efficiency by membrane-based technologies including membrane bioreactors (MBR), disc filters (DF), and reverse osmosis (RO) membranes have been investigated recently (Bayo et al., 2020; Lares et al., 2018; Lv et al., 2019). Talvitie et al. (2017) and Lv et al. (2019) have done comparative studies on MPs removal using common removal techniques including membrane-based processes and proved that higher MPs separation efficiency can be achieved by membrane-based processes compared to the other common treatment techniques used in WWTPs [e.g. rapid sand filtration (RSF), dissolved air flotation (DAF) and oxidation ditch (OD)]. They attributed this to the small pore size of the relevant membranes making them capable to efficiently remove MPs through size exclusion.

However, MPs impact on membrane fouling and concentration polarisation propensity can be a significant challenge for the industrial application of membrane processes. Fouling, that is the deposition of particles on the membrane surface or inside its pores, is generally considered as the main drawback for membrane application (Chai et al., 2020; Shen et al., 2020). Fouling can impose significant economical and operational issues if not controlled properly; therefore numerous studies have been carried out on mitigating membrane fouling (Huang et al., 2021; Long et al., 2021; Teng et al., 2021; Xiao et al., 2021). Recent studies have proven the contribution of MPs to an increase of membrane fouling as well as membrane fouling mechanism alterations (Enfrin et al., 2020; Im et al., 2021; Li et al., 2020; Maliwan et al., 2021); a few studies have also explored MPs fouling mitigation by surface modification, integration of coagulation, and integration of gas scouring in membrane processes (Enfrin et al., 2021c, 2021a; Li et al., 2021; Ma et al., 2019b, 2019a). However, no pervious study has comprehensively reviewed the impact of MPs fouling on polymeric membranes and related MPs-membranes interactions. Hence, critically reviewing the relevant literature that can provide collective insights into these matters is highly beneficial for developing efficient membrane techniques with both minimal MP fouling and optimised membrane MP remediation when required.

Generally, the efficiency of using membrane-based techniques for MPs separation can be affected by various parameters (Siegrist and Joss, 2012; P. Wang et al., 2020). For instance, the concentration of MPs and other contaminants present in water, MPs characteristics, membrane characteristics, membrane fouling, and interaction of MPs with membranes can all contribute to the MPs removal rate. Herein, the main objective is to critically review the available literature on MPs removal by membranes and their fouling effects. The study starts with a summary of MPs occurrence in WWTPs to investigate the characteristics of frequent MPs and the impacts of MPs characteristics on their removal rate. This is followed by reviewing different membrane processes for MPs separation. Interaction of MPs and membrane will be then discussed in more detail aiming to investigate MPs fouling that is followed by reviewing studies that have investigated the fouling mitigation of MPs. Then, other potential interactions of membrane and MPs including biofouling, secondary pollution, abrasion impact of MPs on polymeric membranes, and possible mitigation pathways/options are discussed. Potential membrane technologies for MPs removal that have not been employed are also suggested. Lastly, the prospects of the membrane/membrane integrated techniques to target MPs removal will be discussed. The outcomes will open new avenues for further research on the material/technical development of membrane processes and mitigating MPs fouling impact on membranes.

2. MPs occurrences in WWTPs

2.1. Characteristics of the most frequent MPs in WWTPs effluents

MPs have differing types, shapes, and sizes that influence their occurrence and removal. Therefore, identifying the most frequent MPs properties benefits the design of advanced treatment processes. Fig. 1 shows the morphology of the MPs detected in a WWTP in China

(L. Zhang et al., 2021). The occurrence and characteristics of MPs in WWTPs have been reviewed in a number of studies (Gatidou et al., 2019; Sun et al., 2019). However, due to the large number of studies being conducted over the past two years on MPs occurrence in different WWTPs around the world, a collective overview of the recent findings has been presented here to identify their contribution to advanced treatment technologies, in particular membrane separation techniques.

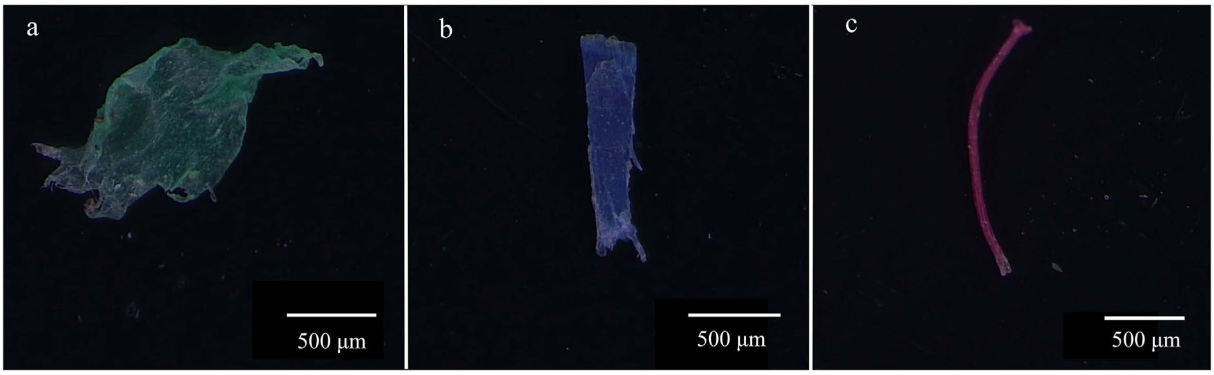

Fig. 1. Images of different shapes of MPs in wastewater samples in China. Reproduced from (L. Zhang et al., 2021) with permission from Elsevier.

Fig. 2 presents the characteristic summary of the most frequent MPs found in WWTPs effluents reviewing studies on the detection of MPs in over 170 WWTPs globally. The results indicate that PE, PP, polyester (PEST)/ polyethylene terephthalate (PET), and PS have been the most common polymer types; and fiber has been the most dominant shape of MPs present in WWTPs effluent. Fibers are mainly made of PEST, and more specifically, PET (Ramírez-Álvarez et al., 2020; Zambrano et al., 2019). Fibers found in WWTPs are mostly originating from the domestic washing machine discharges as each cycle of laundry can discharge up to 1900 fibers into the sewage system (Browne et al., 2011; Carney Almroth et al., 2018; Dris et al., 2015; Lares et al., 2018; Mason et al., 2016; Michielssen et al., 2016; Sun et al., 2019; Talvitie et al., 2015; Ziajahromi et al., 2017b). In fact, fibers shed during synthetic textile washing account for over 35 % of fiber MPs released into the oceans (Bakaraki Turan et al., 2021). Development of 'smarter' textiles or devising more efficient filters in household washing machines can decrease MPs discharge into the sewage system (Carney Almroth et al., 2018; De Falco et al., 2020, 2018; Napper and Thompson, 2016). Hence, improving the quantitative knowledge of MPs origin in WWTPs can alleviate MPs release by controlling them at the source. The study of how to control MPs release into the wastewater from their original source is beyond the scope of this review.

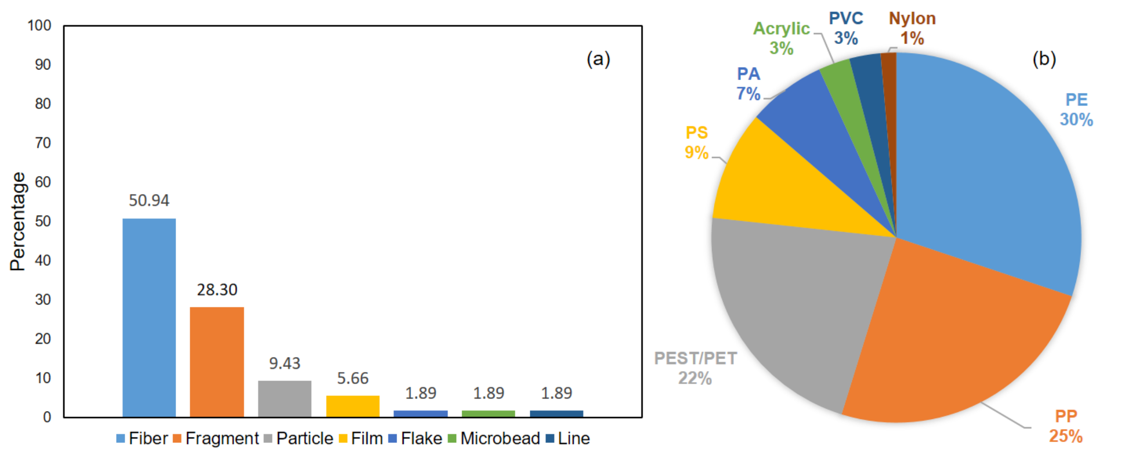

Fig. 2. Frequent MPs that have been observed in over 170 WWTPs: (a) shape, and (b) type.

The main issue with fibers is the difficulty of their complete removal even after advanced treatment processes as fibers can longitudinally pass through the gaps present in the treatment facilities or filter/membrane pores (Ziajahromi et al., 2017b). The presence of fibers is a major issue for membranes and mesh filters that separate MPs through size exclusion (Poerio et al., 2019). Hence, developing advanced technologies or modifying the existing ones to remove microfibers more efficiently is critical for successful MPs management in WWTPs. For membranes, this can be achieved through devising surface modification techniques such that the membranes repel the microfibers (through electrostatic repulsion, hydrophilic-hydrophobic interactions, and alike). More details are included in Section 3. Table 1 is a review of frequent MPs characterisations in WWTPs and their removal rate.

Table 1. MPs occurrence in different WWTPs around the world.

Country (Number of sites)	**Identified size rang (µm)**	**Removal rate %**	**Influent concentration (MPs/L)**	**Effluent concentration (MPs/L)**	**Most frequent type**	**Most frequent shape**	**Most frequent size (µm)**	**Reference**
Finland (1 site)	20- >300	Fibers: 97 Particles: 98	textile fibers: 180 synthetic particles: 430	textile fibers: 4.9 synthetic particles: 8.6	-	Synthetic particle, textile fiber	-	(Talvitie et al., 2015)
France (1 site)	100-5000	83-95	260-320	14-50	-	Fiber	<1000	(Dris et al., 2015)
US (7 sites)	45-400	99.9	-	0.0008	PE	Particle	-	(Carr et al., 2016)
US (17 sites)	>125	-	-	0.05	-	Fiber and fragment	125-355 µm: 57 % >355 µm : 43 %	(Mason et al., 2016)
Scotland (1 site)	>65	98.41	15.70	0.25	PEST, PA, PP	Flake, fiber	598	(Murphy et al., 2016)
Australia (3 sites)	25-500	90	1.5	0.28	PE, PET	Fibers, irregular shaped particles	25-100	(Ziajahromi et al., 2017b)
Germany (12 sites)	20-5000	97	-	MPs particles > 500 µm: 0-0.005 MPs particles < 500 µm: 0.01-9 Fibers: 0.09-1	PE	-	50-100	(Mintenig et al., 2017)
Canada (1 site)	>1	99	31.1	0.5	-	Fiber , fragment	-	(Gies et al., 2018)
Finland (1 site)	>250	98.3	57.6	1	PEST, PE	Fiber, particle	<1000	(Lares et al., 2018)
Denmark (10 sites)	10-500	99.3	7216	54	PE, PEST	Fragment, fiber	<91	(Simon et al., 2018)

China (1 site)	>25	MBR: 82.1 OD: 53.6	0.28	MBR: 0.05 OD: 0.13	PP, PE, PS, PET	Fragments, fiber	>500	(Lv et al., 2019)
South Korea (3 sites)	>1.2	99	13,813.3	132	-	Microbead, fragment	-	(Hidayaturrahman and Lee, 2019)
Italy (1 site)	63-5000	84	2.5	0.4	PEST, PA	Line, film	50-100	(Magni et al., 2019)
China (1 site)	50-5000	95.16	12.03	0.59	PET, PS, PP	Fiber	1110.72 ± 862.95	(L. Yang et al., 2019)
UK (1 site)	60-2800	96	3-10	-	PP	Fiber, films	-	(Blair et al., 2019)
China (1 site)	> 47	64.4	79.9	28.4	PA, PE, PP	Fiber and fragment	66.5	(X. Liu et al., 2019)
China (7 sites)	> 43	79.3-97.8	1.57-13.69	0.20-1.73	PP, PE, PS	Granules, fibers, fragments	>125	(Long et al., 2019)
France (1 site)	>20	98.83	244	2.84	PE	Fragments, fiber	20-80	(Kazour et al., 2019)
United States (3 sites)	> 43	85.2-97.6	70-250	1-30	-	Fiber	<418	(Conley et al., 2019)
Germany (1 site)	> 10	-	-	wet weather days: 5.9 dry weather days: 3	PET, PP, PE, PS	Particles, fiber	<100	(Wolff et al., 2019)
China (2 sites)	>100	60.4-86.9	1.01-2.06	0.27-0.4	PE, PP	Fiber	>1000	(Ruan et al., 2019)
Spain (12 sites)	25- 5000	93.7	-	10.7	PE, PP, PEST, acrylic	Fragments	25-104	(Edo et al., 2020)
Turkey (3 sites)	>26	55-97	2.8	1.6	PE, PP, acrylic, PS	Fiber	>500	(Akarsu et al., 2020)
Spain (1 site)	-	90.3	3.20	0.31	PE	Fragments and fiber	400-600	(Bayo et al., 2020b)
South Korea (50 sites)	Influent: >45	98.7-99.99	10-470	0.004-0.51	PP, PE, PET	Fragment	-	(Park et al., 2020)

	Effluent: >100							
China (2 sites)	>149	66.1-62.7	23.3-80.5	7.9-30.3	PVC, PE, PP	Fiber, fragment	<1000	(N. Tang et al., 2020)
Australia (1 site)	>1.5	76.61	11.80	2.76	PEST, PP, PA, PE	Fiber, fragment	<125	(Raju et al., 2020)
Spain (1 site)	>150	74.8	11.1	2.8	PE, PP, PEST	Fiber	500-2000	(Bretas Alvim et al., 2020)
Mexico (3 sites)	>250	-	-	0.49	Nylon, PET	Fiber	250-500	(Ramírez-Álvarez et al., 2020)
China (9 sites)	> 13	35-98	18-890	6-26	PE, PP, PS	Fragment and film	<500	(F. Wang et al., 2020)
Canada (3 sites)	>125	-	-	13.3	PET	Fiber	-	(Grbić et al., 2020)
Spain (5 municipal and 2 industrial wastewaters)	>100	78-97	264-1567	39-131	PVC, High density PE, Poly Ethyl Methacrylate ,PP, PS, PE	Fibers and fragments	<1000	(Franco et al., 2020)
China (1 site)	>20	75.7	126	30	PEST, PA	Fiber	20-100	(Jiang et al., 2020)
Thailand (3 sites)	>300	84	12.2	2	PEST, PE, polyacrylate, PP	Fiber	-	(Hongprasith et al., 2020)
Australia (3 sites)	>25	98	81.66	0.7	PET, PE, PP	Fiber	-	(Ziajahromi et al., 2021)
China (4 sites)	>25	89.2–93.6	0.70–8.72	0.07–0.78	PP, PE	Fiber	>500	(L. Zhang et al., 2021)
Spain (1 municipal and 1 industrial wastewaters)	>100	> 90	645.03-1567.49	16.40-131.35	PVC, HDPE, PE, Ethylene acrylic acid	Fiber	100-355	(Franco et al., 2021)
China (1 site)	>68	96	44.07	1.97	PET, PP, PE	Fiber	450-900	(Y. Zhang et al., 2021)
Iran (1 site)	>25	90	9.2	0.84	-	Fiber	25-125	(Sarkar et al., 2021)

As inferred from Table 1, a broad range of MPs concentrations are reported in influent (0.2-1567 MPs/L) and effluent (0.0008-131 MPs/L) of WWTPs. Likewise, various size ranges of MPs have been reported dominant in the WWTPs studied in the literature. For instance, N. Tang et al. (2020) have outlined the dominant size of the identified MPs in both their respective WWTP influent and effluent to be between 0.3 mm to 1 mm. The study only sampled MPs larger than 149 μm, and hence, the particles smaller than this size were not detected and consequently not considered for the analysis. Similarly, other studies with smaller MPs measurement ranges have reported smaller dominant sizes of MPs in their samples (Edo et al., 2020; Raju et al., 2020; Wolff et al., 2019). For instance, Wolff et al. (2019) identified MPs >10 μm in a WWTP in Germany, reporting that 95 % of MPs in the effluent of the secondary treatment stage were between 10 and 100 μm. Raju et al. (2020) captured MPs >1.5 μm in a WWTP in Australia and reported the highest portion of MPs in the influent and effluent to be in the range of 1.5-125 μm. Mintenig et al. (2017) studied 12 different WWTPs with measured MPs larger than 20 μm and reported the dominant MPs size in the treated wastewater to be in the range of 50-100 μm. Likewise, Talvitie et al. (2017) sampled MPs > 20 μm and found 70 % of the particles in the effluent being in the size range of 20-100 μm.

Park et al. (2020) investigated MPs occurrence in 50 WWTPs in South Korea and presented MPs size distribution down to 45 in influents and down to 100 μm in effluents. Fig. 3 shows the results of the study. In this figure, ΔN is the number of MPs in a size group, Δdp is the difference in the median length between the two adjacent size groups and d_p is MPs size. The results suggested the abundance of smaller MPs relative to the larger ones. The result suggested the abundance of smaller MPs relative to the larger ones which is in line with the results presented in Table 1.

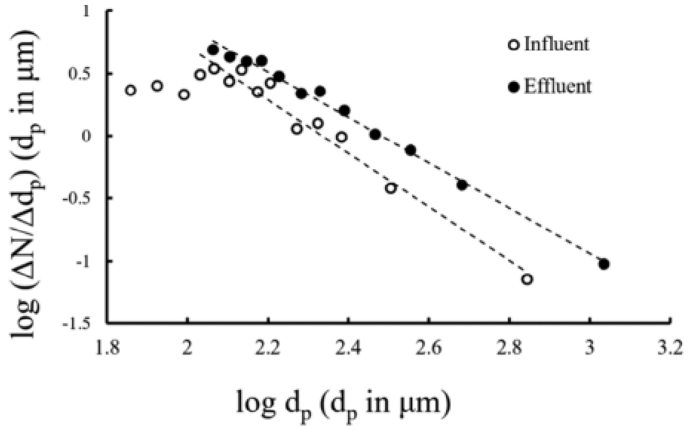

Fig. 3. MPs size distribution in influents and effluents of 50 WWTPs in South Korea. Reproduced from (Park et al., 2020) with permission from ACS Publications.

The discrepancy observed for the dominant concentrations and size ranges of MPs detected in WWTPs can be ascribed to the varied detection techniques and different minimum cut-off sizes used in the literature. It is however interesting to note that in most of the studies, the reported dominant size ranges of MPs were close to the minimum cut-off size being measured. Thus, there is a high possibility that MPs smaller than the minimum detection limit/sampling cut-off size were not detected by the relevant studies. Although most studies have detected MPs larger than 20-25 microns, there are a few studies that detected MPs smaller than 20 microns (down to 1 micron) in WWTPs (Gies et al., 2018; Hidayaturrahman and Lee, 2019; Raju et al., 2020; F. Wang et al., 2020; Wolff et al., 2019). Furthermore, while the presence of nanoplastics (NPs) has been overlooked by other researchers, there is a high possibility for the presence of NPs in different steps of WWTPs. Hence, sampling of small MPs and NPs needs to be considered in order to make a more robust conclusion about the most occurring size ranges of MPs and NPs in WWTPs.

MPs detection techniques are generally classified as visual methods, spectroscopic methods, and chromatographic methods. Visual methods are mainly used to determine the physical properties of MPs (e.g., size, shape, and colour) and can be also used to directly count the MPs (Rocha-Santos and Duarte, 2015; Shim et al., 2017). This method, however, needs to be coupled with other complementary techniques such as spectroscopic methods (e.g., Fourier-transform infrared spectroscopy (FTIR) and Raman spectroscopy) for a more meaningful analysis of the MPs properties (J. Li et al., 2018; Prata et al., 2019). FTIR and Raman spectroscopy produce chemical spectrums inferring the chemical bonds and composition of MPs (Bergmann et al., 2015; Hidalgo-Ruz et al., 2012; Prata et al., 2019; Ziajahromi et al., 2017a). Pyrolysis gas chromatography, mass spectrometry, and liquid chromatography are the other techniques used for MPs detection. Products of MPs thermal degradation are used for the determination of their composition via the mass spectrometer. Liquid chromatography is based on the different solubility of MPs that can verify the polymer type. Different analytical techniques are used to determine the physicochemical properties of MPs in WWTPs. However, no standard techniques have been developed for MPs detection and hence varied individual or integrated methods have been employed in the literature (Goedecke et al., 2020; Picó and Barceló, 2020; Primpke et al., 2020). Developing the current identification techniques and establishing some relevant standards that can be followed by industry will benefit future attempts to study the frequency and relevant properties of MPs in WWTPs.

2.2. Fate and removal of MPs in WWTPs

Municipal WWTPs usually consist of preliminary treatment (e.g., coarse screening and grit removal), primary treatment (e.g., screening and grit chamber and sedimentation), secondary treatment (e.g., activated sludge processes, bio-filters, and oxidation ditches), tertiary treatment (e.g., activates carbon, sand filtration, and membrane technologies) and disinfection (e.g., injection of a chlorine solution, ozone, and ultraviolet irradiation). A schematic illustration of the major steps of municipal WWTPs is depicted in Fig. 4. Generally, the applied treatment techniques in a plant determine its MPs removal efficiency (Gatidou et al., 2019; Kalčíková et al., 2017). Hence, different studies have been conducted to ascertain the contribution of each stage towards MPs removal. For instance, Michielssen et al., (2016) have examined the fate of MPs at each treatment step (preliminary, primary, and secondary) within two WWTPs. Their results indicated that the combined preliminary (screening and grit removal) and primary (gravity separation and surface skimming on primary clarifiers) treatment processes could remove nearly 86 % of MPs. The secondary treatment step (activated sludge and trickling filters) had limited contribution to the MPs removal leading to an overall MPs removal of 91 %. Murphy et al., (2016) have reported that 78.34 % of MPs were captured in preliminary (grit and grease) and primary treatment processes, with a further 20.1 % removal rate in the secondary treatment stage.

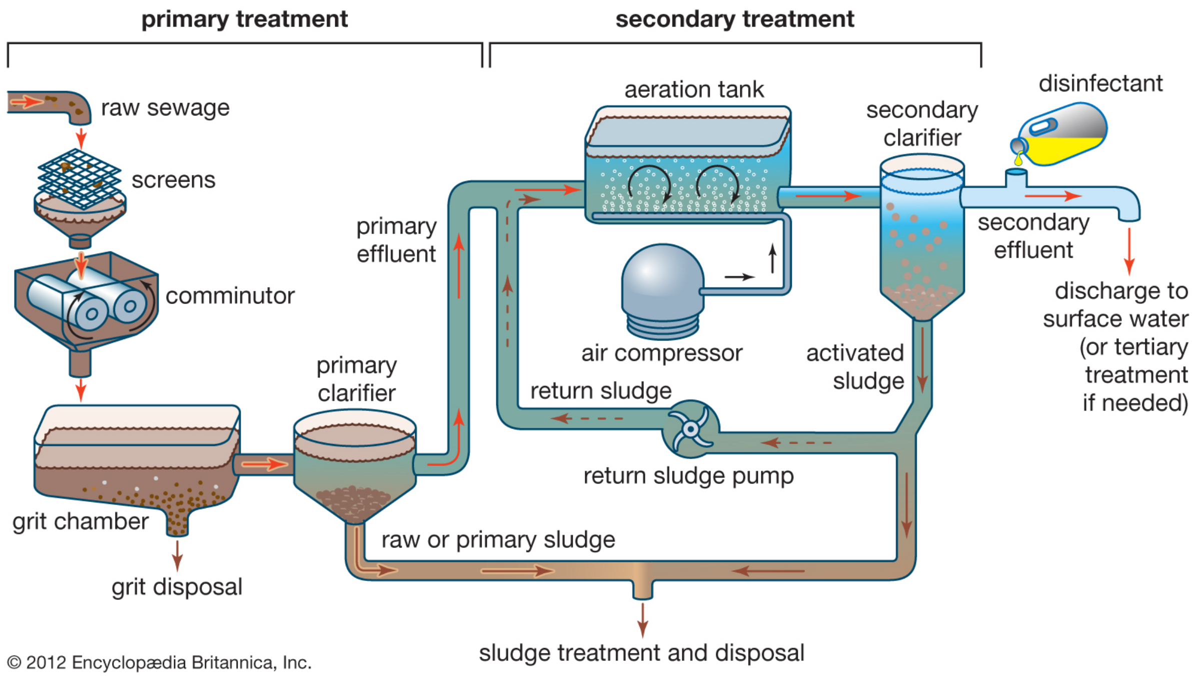

Fig. 4. Schematic illustration of the major treatment steps in municipal WWTPs. By courtesy of Encyclopædia Britannica, Inc., copyright 2015; used with permission (Wastewater treatment - Primary treatment | Britannica).

According to Table 1, the MPs removal rate in the WWTPs is in the very broad range of 35-99.9 %. Considering that a large volume of treated wastewater is discharged into the environment daily, there is a need to develop novel tertiary treatment units or modify the existing ones to target MPs capturing and removal from wastewater. There is controversy as to whether the MPs shape and size contribute to their overall separation efficiency. Franco et al. (2020) claimed the shape and size are not significant properties by studying MPs removal in five municipal and two industrial WWTPs. However, other studies in different WWTPs have observed that the shape and size of MPs did contribute to their removal rate (Hidayaturrahman and Lee, 2019; Michielssen et al., 2016; Talvitie et al., 2015). For instance, Talvitie et al. (2015) studied the MPs removal in a WWTP in Finland and reported that primary sedimentation had the highest efficiency for fibers removal whilst secondary sedimentation mainly removed particles. Hence, MPs shapes contributed to their removal rate in each step. In another study, it was found that microbeads were completely removed whilst fibers, fragments, and paint chips could not be completely removed after secondary treatment processes (Michielssen et al., 2016). Hidayaturrahman and Lee (2019) have also investigated the fate and removal of different shapes of MPs, reporting that the microbead removal rate in each stage of the WWTP was lower than that of irregular MPs like fibers and fragments. This can be ascribed to microbeads having smooth surfaces making them difficult to be adsorbed on other materials such as natural organic matter or suspended solids. Different operation conditions, equipment types, and process configurations used in WWTPs can explain the controversy over the influence of MPs shape on their separation efficiency. A more sensible example in this regard is the separation of fibers which are prone to pass through membranes longitudinally (Bayo et al., 2020a). Therefore, fiber removal might be lower than microbeads or fragments removal in WWTPs that employ membrane technologies.

Furthermore, the size ranges of MPs have been reported to contribute to their separation efficiency in WWTPs in different ways. This can be attributed to the MPs size contribution to their flotation rate, movement across streams, capacity to adsorb contaminants, and flocculation or sedimentation (Enfrin et al., 2020; Rios Mendoza et al., 2018). Magni et al. (2019) studied three size ranges of MPs (0.1-0.01 mm, 0.5-0.1 mm, and 5-0.5 mm) and observed that the removal rates decreased with the particle size (65 %, 77 %, and 94 % respectively). In contrast, Long et al. (2019) reported that smaller MPs tend to get aggregated and settled into the sludge faster so their removal rate in WWTPs is higher than their larger counterparts. The higher abundance of smaller MPs in the WWTPs effluents inferred from Table 1, might indicate the higher removal efficiency of larger MPs. However, this is also affected by the WWTP facilities as some mechanical treatment processes such as automatic screens tend to break MPs leading to an increase in the number of smaller MPs in the effluent.

It can be concluded that the morphology and size of MPs influence their removal efficiency in wastewater treatment processes. This shows the importance of considering the shape and size of MPs when designing the relevant treatment technologies to improve MPs separation efficiency. In order to design a treatment technique for MPs removal, or to ameliorate the existing treatment technologies, MPs characteristics and their concentration in WWTPs should be more specifically considered.

3. A review of membrane processes for MPs removal

Membrane separation is one of the most common approaches used in several stages of WWTPs and can be a great platform to enhance MPs removal. This is due to the development of new membrane materials capable of removing pollutants at low concentrations (micropollutants) and small sizes (MPs and nanoparticles also being predominantly present as micropollutants in WWTPs) whilst remaining economically feasible (Wu, 2019). Membrane separation works based on three different fundamental mechanisms: adsorption, electrostatic repulsion (between charged solutes and charged membrane surface), and size exclusion or sieving (Sanguanpak et al., 2019). In the following sections, various membrane processes are reviewed and discussed in terms of their MPs removal rates highlighting future research needs in this area.

3.1. Membrane bioreactor

Membrane bioreactor (MBR) is a common process in WWTPs that combines activated sludge treatment with membranes. MBR performance includes membrane sieving effect and pollutant biodegradation by microorganisms. The employed membranes inside MBR are generally ultrafiltration (UF) or microfiltration (MF) types configured as plate, hollow fibre, or tubular units (Chong et al., 2012). MBR processes have a substantial micropollutant removal capability due to its dual treatment mechanism: biodegradation and membrane filtration (Besha et al., 2017; Luo et al., 2014; Ma et al., 2018; Masiá et al., 2020; Ngo et al., 2019). Besides, the small pore size of MBR filters (commonly in the range of 0.1-0.5 μm) makes them suitable candidates for MPs removal (Chong et al., 2012; Goswami et al., 2018; Ngo et al., 2019).

Recently, a few studies evaluated the retention of MPs in different steps of their studied WWTPs and compared their efficiency to MBR. Talvitie et al., (2017) explored MBR, disc filter (DF), rapid sand filtration (RSF), and dissolved air flotation (DAF) in different WWTPs in Finland to examine their efficiency to remove MPs ranging between 20 and 300 μm. The MBR filtration tank contained 20 flat sheet ultrafiltration (UF) membrane cartridges. The MBR technique showed the highest MPs removal rate amongst the studied treatment techniques. The MPs removal rates were 99.9 % by MBR, 97 % by RSF, 95 % by DAF, and 40-98.5 % by DF. The influent of the MBR contained mainly fiber and spherical shape MPs whilst its effluent only contained fibers. The polymer types in the MBR influent were PEST, PP, PE, and polyacrylate whilst the MBR effluent only contained PEST MPs.

Michielssen et al. (2016) compared the removal of MPs ranging between 20 µm and 4.75 mm in MBR, granular sand filtration and activated sludge systems. The MBR membrane showed a slightly higher MPs removal rate compared to the granular sand filtration and activated sludge techniques (99.4 %, 97.2 %, and 95.6 % MPs removal rates respectively). In terms of the separation efficiency by the MPs configuration, microbeads showed complete removal in all three techniques. Fibers and fragments were respectively the most prominent MPs types that remained in the WWTPs effluents. However, a slight improvement was observed by MBR which resulted in a 10 folds decrease (from 15 to 1.25 billion particles) in the daily discharge of MPs to the environment. Lares et al. (2018) also confirmed higher MPs (> 250 µm) removal efficiency of MBR (99.4 %) compared to activated sludge (98.3 %). This was investigated in a pilot-scale MBR that was employed at the Mikkeli WWTP in Finland where PEST fibers (mainly PET) smaller than 1 mm were the most prominent MPs in all stages of both processes.

Leslie et al., (2017) reported a slight improvement of MPs removal by integration of MBR in a WWTP. Typical MPs in both influent and effluent of the WWTP were fiber shaped and smaller than 300 µm. The effluent of the WWTP without MBR contained an average of 58 MPs/L whilst that of the process including the MBR resulted in a concentration of 51 MPs/L in the effluent. The comparatively low improvement of removal rate by the MBR was ascribed to MBR design, MPs characteristics, and operational pressure. The MBR systems used in that study were not designed to metabolise plastic materials which contributed to their low MPs removal efficiency. Moreover, the majority of MPs in the influent of the MBR were fibers that may pass longitudinally through the membranes, particularly at the high operational pressures (1-4 bar) applied in the MBR system.

Lv et al. (2019) compared an MBR system with an oxidation ditch (OD) system for MPs removal. OD is a secondary wastewater process that removes pollutants with a combined system of biological treatment and aeration (Shammas and Wang, 2009). The influent wastewater had a concentration of 0.28 MPs/L containing PET, PS, PE, and PP, where the dominant shapes were fragments and fibers. The MPs had a broad particle size distribution of 25 μm to above 500 μm. The membrane used in the MBR system was a PVDF hollow fiber membrane cartridge with a pore size of 0.1 μm. Although most of the studies have reported their MPs removal rates based on MPs number, this study reported the removal rate of the MPs based on both mass and number. MBR and OD respectively captured 99.5 % and 97 % of MPs on the basis of MPs mass, accounting for 82.1 % and 53.6 % based on MPs number. The superior removal rate of the MBR system was correlated to its microfiltration membrane with smaller pore sizes than the characterised MPs and the relevant size exclusion process. According to the results obtained, MPs removal rate on the basis of MPs number was lower as the equipment broke down MPs during the removal process and increased the number of smaller MPs in the effluent. However, the OD process broke down MPs to a higher grade than the MBR.

Likewise, Pizzichetti et al. (2021) reported the presence of MPs in the permeate of membranes with smaller particle sizes than those in the feed stream. This also confirmed MPs disintegration through membrane filtration owing to mechanical stress of hydraulic pressure which can be detrimental to the efficient application of membranes. These findings illustrate the significant role of a treatment process in increasing the number of small MPs. Small MPs can be hazardous because their high specific surface areas making them capable to adsorb more toxic chemicals as well as being more prone to be digested by marine organisms than the larger plastics (Adam et al., 2019; Choi et al., 2020; Cole et al., 2020). Therefore, investigation of this obstacle is essential to avoid secondary pollution of MPs and NPs associated with membrane application. It can be suggested that optimisation of hydraulic pressure could prevent MPs breakup. This however needs to be further investigated and the correlations to be clarified.

A comparison study (Bayo et al., 2020a) of MBR and RSF technique implemented a parallel MBR-RSF process in a WWTP in Spain where the MBR system included 10 flat-sheet membrane units. The results confirmed a higher MPs (> 200 µm) removal efficiency for the MBR as compared to RSF. Removal rates based on MPs shape were also investigated in this study. Both systems removed particles more efficiently than fibers. Again, this was attributed to fibers escaping the units through the longitudinal passage from the gaps and filter pores. The majority of MPs in the influent and the effluent of both systems were fibers smaller than 1 mm. The influent of both systems consisted of 14 different polymers with the majority being PE, Acrylate, and PP whilst the effluent of RSF contained PE, Nylon, and Polyvinyl. The effluent of the MBR only included melamine (MUF) which did not exist in the influent that shows MUF being a secondary pollutant. The source of MUF was not mentioned, however, since MUF is used for membrane modification, it was most likely shed from the membrane itself during the filtration process. Therefore, although MBR processes seem potential applicants for MPs removal, their membranes need to be optimised to minimise the MBR secondary pollution effect on wastewater through fiber shedding.

Furthermore, as MBR is a hybrid process, the impact of MPs on the MBR biological treatment step needs to be also considered. Multiple studies reported that MPs and the chemicals attached to them may be toxic to the microbial culture leading to an inhibitory effect on biological treatment (Cluzard et al., 2015; Li et al., 2020b; Wei et al., 2019; X. Zhang et al., 2020; Zhao et al., 2020). For instance, MPs can inhibit denitrification by changing the microbial process which leads to the accumulation of ammonium in water (Dai et al., 2020; Seeley et al., 2020; L. Tang et al., 2020). Y. T. Zhang et al. (2020) evaluated the long term effect of PVC MPs at the concentration of 15-150 MPs/L on biological wastewater treatment processes and concluded that the microbial populations and relative abundances of methanogens and acidogens were reduced due to the presence of MPs. These observations were subsequently correlated to the toxic leachate and excess oxidative stress of MPs.

Li et al., (2020a) studied the influence of MPs on the microbial community of an MBR system by adding pure/pristine PVC MPs to synthetic wastewater. The exposure of the MBR system to MPs did not show any significant effect on the microbial community. Maliwan et al. (2021) reported a slight impact of MPs on the microbial diversity and predominant microbes of a MBR system. However, this might not be the case in reality where MPs entering the water treatment facilities may carry toxic pollutants that can damage the microbial community. Moreover, aged and fresh MPs have different adsorption capacities and interaction mechanisms (G. Liu et al., 2019) which may also change their impact on MBR microbial community. Hence, the impact of MPs on MBR biological organisms should be further studied and strategies to minimise its impact developed.

As MPs are persistent pollutants, they may not be degraded effectively in the biological process of current MBR (Hahladakis et al., 2018). Hence, proper sludge treatment should be considered in order to avoid secondary pollution upon sludge disposal. Furthermore, considering that the partial MPs biodegradation depends on the MPs chemical and physical properties (Kumar et al., 2020; Tokiwa et al., 2009), identifying the characteristics of present MPs and tailoring the biological treatment of MBR to their treatment may increase the MBR MPs removal efficiency.

3.2. Disc filter

Disc filters (DF) is used in some WWTPs as a final polishing step for particle removal through size exclusion or cake layer formation on its surface (Simon et al., 2019). A DF includes a number of woven filter meshes typically made of PP, PA, or PEST with pore sizes of 10-40 μm, which are located in a closed tank. A few recent studies have evaluated the efficiency of DF in MPs removal and showed the overall high efficiency of DF (Hidayaturrahman and Lee, 2019; Mintenig et al., 2017; Simon et al., 2019; Talvitie et al., 2017).

In a study conducted at the Oldenburg WWTP in Germany, a system of 12 rolling filters was used as a tertiary treatment process for MPs (>20 μm) removal. The filters were made of PA pile fabrics and were connected to a PEST support that contained a small amount of PE. The filters (pore size of 10-15 μm) were located after the primary and secondary treatment steps and were back-washed several times per day. The system showed excellent performance and successfully eliminated 100 % of MPs > 500 μm, 93 % of MPs < 500 μm, and 98 % of synthetic fibers from the treated wastewater. There were six different types of MPs in the influent of filter (PE, polyvinyl alcohol, PP, styrene acrylonitrile (SAN), PET, and paint) whilst the filtration effluent contained small quantities of PE, PA, and SAN. However, this technique imposed secondary contamination in the form of PA fibers, possibly originated from the PA filter itself (Mintenig et al., 2017).

Talvitie et al., (2017) surveyed the efficiency of DF process as a tertiary treatment step in the Viikinmaki WWTP. The DF was a pilot-scale system including two discs, each with 24 filter panels. Physical retention in the filters and the formation of sludge cake inside the filter panels were the basis for the removal process. Two DF systems with different pore sizes of 10 μm (DF 10) and 20 μm (DF 20) respectively were examined separately for the removal of MPs in the size range of 20-300 μm. Iron-based coagulant and a cationic polymer were also used to improve particle removal. Fibers and fragments were the most prominent MPs in the influent of both DF. The influent of DF 10 consisted of PEST, PE, PS, PVA, PA, and acrylamide, whilst the effluent only contained PEST and PE giving 40 % (0.5 to 0.3 MPs/L) MPs removal rate by DF 10 alone. Conversely, the removal rate of DF 20 was 98.5 % (2 to 0.03 MPs/L) where the influent contained PEST, PE, PP, PU, PVC, and the effluent only consisted of PES MPs. DF 20 showed a higher MPs removal rate compared to DF 10 despite the smaller pore size of the DF 10. This was attributed to excessive use of polymer during the process of DF 10 which resulted in the blockage of the filter by sticky polymer flocs that subsequently needed accelerating backwash. This in return decreased the removal efficiency of the DF 10 giving some MPs the opportunity to pass through the filter during the high-pressure backwash operation. Furthermore, the influent concentrations and polymer types were not identical in the two systems (DF 10 and DF 20).

In a recent study, a DF was implemented at the final stage of a WWTP including primary treatment processes (grit chamber and primary settling), secondary treatment processes (bioreactor and secondary settling), and a coagulation tank using Al-based coagulants (average dosage of 30.5 mg/L). The study analysed and compared MPs removal by DF and an ozonation system. A removal rate of 79.4 % (from 1444 to 297 MPs/L) was recorded which was smaller than that of MPs removal efficiency of ozonation process (89.9 %). The DF pore clogging made it necessary to operate more frequent backwashes. This in turn contributed to the low removal rate of the DF due to MPs leaching to the effluent during the high-pressure backwash operation (Hidayaturrahman and Lee, 2019). The study shows that the efficiency of DF is limited because the requirement for multiple backwash operations can increase the MPs leakage to the effluent and decrease their removal efficiency.

Simon et al. (2019) also investigated the performance of DF for MPs removal using a DF including 13 PEST discs in a WWTP. The filters had a pore size of around 18 μm and were used to investigate the removal of MPs with a size > 10 μm as the final treatment stage at a WWTP in Denmark. This system removed 89.7 % of MPs based on the number of particles and 75.6 % of MPs based on their mass similar to the case study of (Lv et al., 2019) that also observed better number based removal efficiencies. MPs smaller than those present in the influent were not detected in the effluent. Hence, the authors concluded that the DF process does not cause MPs degradation. However, surprisingly, there were particles larger than the mesh pore size of the filters present in the effluent showing the possibility of large particles bypassing the filters. This could have been induced because of the flexible nature of the filter cloth, damage to the filter by continuous mechanical stresses, and/or distortions due to the high-pressure backflush cycles. The latter has also been reported by (Hidayaturrahman and Lee, 2019) and (Talvitie et al., 2017). Similar to the case study conducted in Germany (Mintenig et al., 2017), secondary contamination of MPs was also identified in the effluent and was correlated to the shedding of the fibers from the DF. Despite this secondary contamination, this study showed a high removal efficiency of MPs by DF confirming their potential prospects for MPs management. The number and mass of the MPs distribution before and after filtration were investigated. PE and PEST were the dominant MPs based on the total number of MPs in the influent whilst PVC was the most frequent MPs (65 %) based on the total MPs mass (although being less frequent (only 6 %) based on the MPs number). This can be correlated to the comparatively higher density of PVC particles. After filtration, PS was the most abundant MPs based on both number and mass. Different relative fractions of all polymer types before and after filtration indicates that MPs type influences their removal rate by membrane processes. It can be attributed to the different surface and chemical properties (i.e., surface charge, roughness, hydrophilicity, surface functional groups) and varied morphology/shape of the different types of MPs; This claim however needs further investigation through experimental analysis.

Overall, DF demonstrated high efficiency in MPs removal, but the secondary contamination induced by the leakage of fibers from the DF filters can be a significant disadvantage for their application. To make DF technology a suitable approach for MPs removal, the structure of the filters needs to be reinforced to make them more stable. DF can also be integrated with other treatment approaches to target the exclusion of secondary contaminants from the effluent. Further studies on DF application for MPs removal can help develop more robust, reliable, and efficient treatment strategies integrated with DF technology.

3.3. Reverse osmosis

Reverse osmosis (RO) is one of the most important and widely recognised technologies in the water industry. The most commonly applied commercial RO membranes used in water and wastewater treatment systems are made of PA thin film composites (Asadollahi et al., 2017; Zargar et al., 2016b), where the module is either hollow fiber or spiral wound (Shenvi et al., 2015). Although RO membranes are generally capable of achieving high removal rates for micropollutants (Albergamo et al., 2019; Kimura et al., 2003), only one study (Ziajahromi et al., 2017b) have investigated the application of a UF-RO combined system for MPs removal.

Ziajahromi et al. (2017b) investigated the transport and fate of MPs over 25 µm. The system was implemented as an advanced treatment unit at the final treatment stage of a WWTP. The influent of the UF and RO contained different types of MPs including PE, PET, PS, and PP within a dominant size range of 100-190 µm. The study showed a decrease in the concentration of MPs from 2.2 MPs/L to 0.28 and 0.21 upon UF and RO filtration steps respectively; thus over 90 % MPs removal was achieved. However, despite the small cut-off size of RO membranes, PET fibers were detected in the effluent. This was attributed to the occurrence of pores with larger sizes in the membrane, the membrane materials, and other local gross membrane imperfections or small gaps between pipework.

Table 2 presents a summary of the previous studies about membrane processes for MPs removal in WWTPs, detailing membrane pore sizes, removal rates, MPs size range, and concentrations before and after the relevant filtration processes. Overall, most of the employed membrane processes (MBR, DF, and RO) could efficiently remove MPs. However, it is not completely feasible to compare their efficiency owing to the different initial types, concentrations, and size ranges of MPs in each report and the lack of sufficient studies on RO process and MPs. It can be concluded that membrane processes have a high potential to be employed for MPs removal; however, identifying the best membrane process warrants further experimental research being conducted in this area.

Table 2. MPs removal rate of membrane processes.

Method	Membrane Pore size	Removal rate (%)	Initial MPs concentration (MPs/L)	MPs Concentration after removal (MPs/L)	MPs size range (µm)	Reference
MBR	0.2 µm	99.4	133.0	0.5	20- 4750	(Michielssen et al., 2016)
	0.08 µm	12	58	51	10-5000	(Leslie et al., 2017)
	0.4 µm	99.9	6.9	0.005	20-300	(Talvitie et al., 2017)
	0.4 µm	99.4	57.6	0.4	> 250	(Lares et al., 2018)
	0.1 µm	82.1	0.28	0.05	>25	(Lv et al., 2019)
	-	79	4.40	0.92	> 200	(Bayo et al., 2020a)
	0.1 µm	100	10	0	< 5	(Li et al., 2020a)
DF	10 µm	40	0.5	0.3	20-300	(Talvitie et al., 2017)
	20 µm	98.5	2	0.03	20-300	(Talvitie et al., 2017)
	10-15 µm	MPs > 500 µm: 100 MPs < 500 µm: 93 synthetic fibers: 98	MPs > 500 µm: 0.05 MPs < 500 µm: 0.2 synthetic fibers: 0.9	MPs > 500 µm: 0 MPs < 500 µm: 0.01 synthetic fibers: 0.02	20-5000	(Mintenig et al., 2017)

	10 µm	79.4	1444	297	> 1.2	(Hidayaturrahman and Lee, 2019)
	18 µm	89.7	29	3	10-500	(Simon et al., 2019)
UF-RO	-	90	2.2	0.21	25-500	(Ziajahromi et al., 2017b)

4. Effects of MPs on membranes fouling, performance and durability

4.1. MPs impact on membrane fouling

Membrane lifetime and permeate flux are significantly affected by concentration polarisation and fouling (Asadollahi et al., 2017; Li et al., 2019; Z. Yang et al., 2019). Concentration polarisation occurs because of an increase in the concentration of solutes near the membrane surface, which creates a boundary layer near the membrane walls (Bassyouni et al., 2019; Zargar et al., 2020, 2015). Concentration polarisation triggers the surface fouling and as per their interdependency, modification techniques usually address both of them simultaneously (Guha et al., 2017). The size range of pollutants (e.g., MPs), the membranes pore size and surface properties, the support layer structure, and the module configuration (e.g., number and thickness of the spacer sheets, number of fibers and membrane sheets in modules and alike) are the major factors affecting membrane fouling. Particles larger than the membrane pore size can cause pore blocking or cake layer formation, while particles smaller than the membrane pore size can cause internal and irreversible fouling (Enfrin et al., 2019). For instance, Abdelrasoul et al. (2013) studied latex particles (0.2-200 µm) removal by polysulfone membranes (MWCO 60,000 kDa) and observed two different fouling mechanisms based on the latex particles size. Particles smaller than 10 µm (smaller than membrane pore size) caused irreversible fouling by blocking the membrane pores whilst particles larger than 10 µm created reversible fouling by deposition of a porous cake layer on the membrane surface because of physical adsorption.

Generally, pore clogging directly relates to membrane pore size, concentration of particles, surface characteristics of both foulants and membrane, and water flow rate (Enfrin et al., 2020; Lin et al., 2014). Another important factor in pore clogging is the proximity of the membrane pore size and the contaminants particle size where similar sizes facilitate the membrane pore blockage. Therefore, the proposed membrane pore size for MPs removal should be adjusted according to MPs size ranges in the effluent of a WWTP if practical. Considering the limited number of studies targeting the analysis of small MPs down to 1 μm and NPs in WWTPs, further studies need to be conducted to determine the most frequent MPs and NPs size ranges in the targeted WWTP effluents to design the tertiary membrane processes accordingly.

Enfrin et al. (2020) have surveyed the fouling of a polysulfone (PSF) membrane (pore size of 31 nm) used for PE MPs and NPs removal. The presence of MPs caused water flux reduction owing to the interactions between NPs/MPs and the membrane pores and surface. The hydrophobic characteristics of both the membrane and particles caused an increase in fouling, while their negative charge facilitated the fouling mitigation. The membrane surface morphology before and after integration of MPs in the filtration process is shown in Fig. 5 which confirms the cake layer formation after MPs filtration. Overall, the low performance of the membrane cleaning and the potentially irreversible fouling by NPs/MPs confirm the importance of further investigations correlating membrane fouling and MPs contamination. The results imply that the current treatment options need further development to deal with new emerging NPs/MPs pollutants.

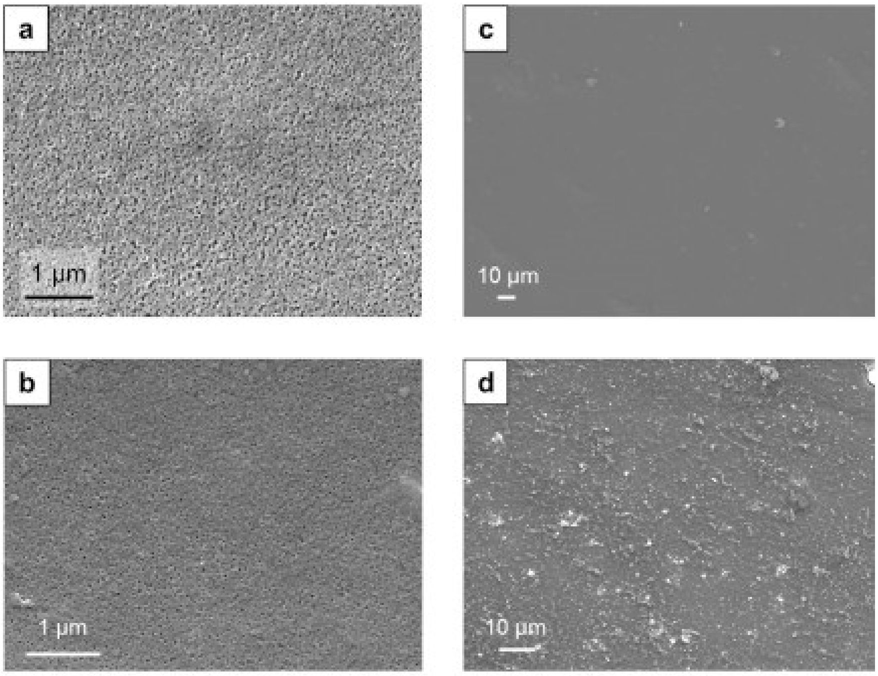

Fig. 5. Membrane surface a) before filtration, b) and c) after pure water filtration and d) after polluted water with NPs/MPs filtration. Reproduced from (Enfrin et al., 2020) with permission from Elsevier.

Li et al. (2020a) explored MBR membrane fouling in the presence of PVC with a concentration of 10 MPs/L and reported higher membrane fouling when MPs were present. PVC addition into the influent of MBR decreased the removal efficiency of organic matter (from 80 to 50 %) and ammonia (from 95 to 40 %). Considering membrane pore size (0.1 µm) and PVC particle size (< 5 µm), MBR removed almost all the MPs. The presence of MPs increased both reversible and irreversible fouling of membrane where the increase of irreversible fouling was attributed to the entrance of some tiny MPs into the membrane pores (The smallest size of MPs was not mentioned in the study). Interestingly, Maliwan et al. (2021) reported less fouling in their MBR system when MPs were present. The study investigated MBR fouling in the presence of polyester (fiber), PA (fiber), PP (fragment), and PE (fragment) with a size range of 0.3-5.6 mm and concentrations of 7 MPs/L, 15 MPs/L and 75 MPs/L. The decrease of fouling was correlated to the scouring effect of MPs. This inconsistency between the two recent studies is due to the different size ranges of MPs that were used. In the former study, MPs less than 5 µm were used that could cause pore clogging of MBR filters, however, in the latter study MPs particles larger than 300 µm were investigated that could act as a scouring material. There is a high possibility that NPs and MPs smaller than 25 µm are present in WWTPs that have not been considered in the previous reports. Therefore, the occurring MPs and NPs in WWTPs may potentially cause pore clogging and increase irreversible fouling in MBR systems.

4.2. Fouling mitigation

Membrane fouling can be alleviated by the application of efficient pre-treatment processes or membrane modification approaches. For instance, a combination of coagulation and membrane filtration or application of UF membranes before RO systems can mitigate membrane fouling (Jiang et al., 2017; Wolf et al., 2005). Membrane characteristics namely hydrophilicity, surface charge, and morphology play a significant role in the extent of fouling (Kumar and Ismail, 2015). For instance, the formation of an ultrathin water layer on the surface of hydrophilic membranes hinders hydrophobic particles' adhesion to the membrane due to hydrophilic-hydrophobic repulsion (P. Wang et al., 2020). Similarly, repulsive forces between the charged foulants and the charged membrane surface hinder the formation of a fouling layer on the membrane surface (Kumar and Ismail, 2015). Hence, increasing the hydrophilicity of membranes or surface charge alteration relevant to the most frequently occurring foulants in the system can greatly contribute to decreasing the fouling propensity of the developed membranes. In terms of morphology, narrower pore size distribution, larger porosity, and a smoother surface can result in less fouling (Enfrin et al., 2019; Kumar and Ismail, 2015).

Membrane surface modification (e.g., through the integration of hydrophilic additives on/into the membranes or plasma treatment/polymerization), creates a smoother and more hydrophilic surface followed by an improvement in the permeability and antifouling behaviour of the membrane (Azari and Zou, 2013; Changani et al., 2020; Nguyen et al., 2013; Orooji et al., 2018; Shenvi et al., 2015; Suhaimi et al., 2020). Recently, inorganic fillers such as titania, zeolite, silica, alumina, and carbon-based materials have been proposed as additives in the membrane matrix to decrease fouling (Chai et al., 2020; Masjoudi et al., 2021; Shen et al., 2020; Zargar et al., 2017a, 2017b, 2016). Taking into account that most MPs are hydrophobic and negatively charged (Enfrin et al., 2020), modifying a membrane to be both hydrophilic and negatively charged can decrease MPs fouling on the membrane. The MPs fouling mitigation strategies need to be investigated through detailed experimental analysis.

In a combined system of membrane and coagulation, particles can be trapped by coagulants, creating larger particles leading to less irreversible fouling and pore clogging. Besides, a hybrid system of membrane and coagulation can also improve the removal efficiency of MPs that are smaller than the membrane pore size owing to the particles being trapped and hence effectively removed by coagulants. Furthermore, common MPs such as PE and PP might agglomerate in water due to their hydrophobicity so developing combined systems of membrane and coagulation process has a high potential to address the MPs contamination issue.

Ma et al. (2019a) studied the removal and fouling of PE MPs (<0.5-5 mm) using a combination of Fe ($FeCl_3.6H_2O$) coagulation and a PVDF membrane with a pore size of 30 nm. The schematic illustration of the applied process is shown in Fig. 6. The integrated system could completely remove the PE particles to below detectable limits due to the small pore size of the UF membrane which completely removed the escaped particles from the coagulation step through size exclusion. Interestingly, membrane fouling at the presence of PE particles was less than that of the Fe-based flocs alone. This corresponds to the fact that PE particles caused greater porosity of the cake layer resulting from the gaps created between the adsorbed flocs on the membrane surface. This resulted in an easier overall passage through the membrane. Therefore, the presence of MPs may even mitigate membrane fouling by other pollutants in WWTPs owing to their contribution to form a sparse cake layer. However, this is again related to the integrated MPs sizes in this study that was about 0.5 mm to 5 mm. The presence of smaller MPs and NPs could cause pore clogging and intensive membrane fouling.

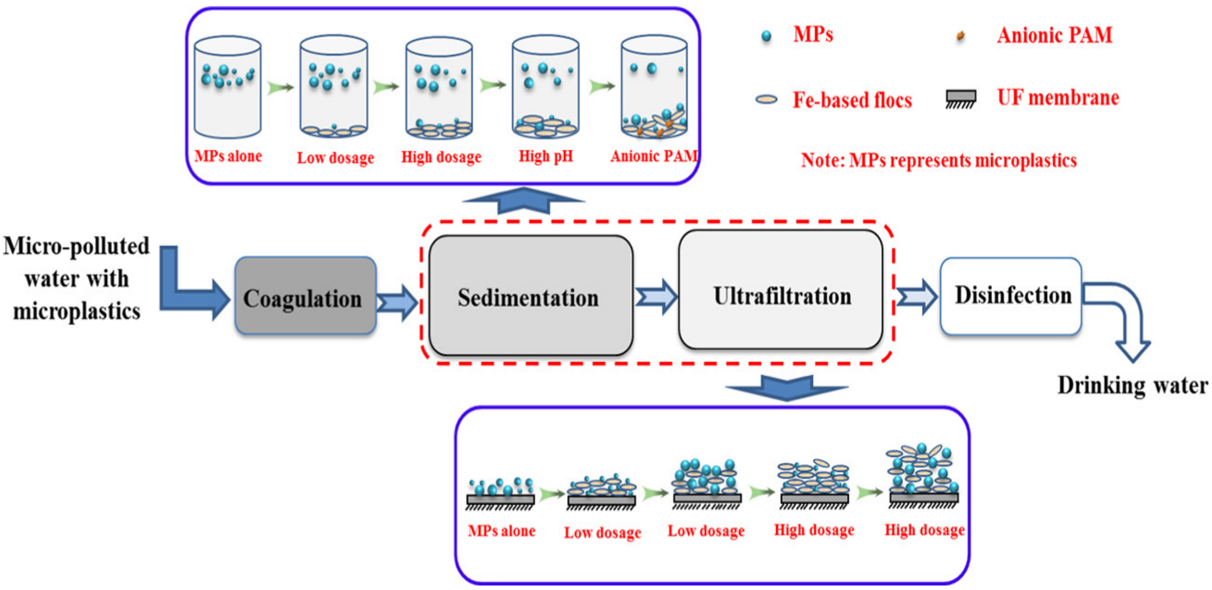

Fig. 6. Coagulation and ultrafiltration process for MPs removal. Reproduced from (Ma et al., 2019a) with permission from Elsevier.

The same group (Ma et al., 2019b) studied the combined system with Al- based salts for MPs removal and investigated MPs fouling impact on membrane. It was observed that the average floc size of Al-based coagulants was smaller than that of Fe-based coagulants. This resulted in the larger specific surface area of Al-based flocs for the same dosage of both coagulants to trap MPs. Hence, the Al-based coagulants performed better than the Fe-based variants in removing PE MPs. Coagulation could contribute to the fouling of membranes resulting in a slightly higher fouling due to the thick cake layer formation by coagulants. Both of the noted studies were conducted using only one kind of MPs (PE) with a size about 0.5 mm to 5 mm. The presence of other MPs with different shapes, and sizes (especially smaller MPs and NPs) in wastewater can make the fouling impact more complicated and hence, warrants further investigations.

J. Li et al. (2021) investigated MPs fouling by integration of coagulation. The study worked on fouling of round shape PS particles with the size of 0.1, 1, 10, and 18 μm and concentrations of 0.01 mg/L to 1 mg/L. The filtration process was conducted by employing a PVDF hollow fiber membrane module with a mean pore size of 0.03 μm and using AL-based salt for coagulation. Effect of concentration and size of MPs on membrane fouling were also studied. Higher concentration caused more severe fouling while the effect of MPs size did not follow a clear trend. The fouling rate of MPs based on their size was as: 1 μm> 0.1 μm > 10 μm > 18 μm. Generally, larger particles create a more porous filter cake with lower resistance, but interestingly, 1 μm- sized particles caused higher fouling compared to 0.1 μm particles. This can be correlated to the different structures of filter cake formed by these two different sizes. SEM analysis confirmed that MPs of 1 μm size formed a less porous layer. Therefore, 1 μm was defined as a critical size for membrane fouling. The critical size of MPs to minimise fouling should be however further investigated using different types of MPs to make a more robust conclusion. Furthermore, it was observed that integration of coagulation reduced the transmembrane pressure by 85 % which confirms alleviated MPs fouling by forming a looser and more porous cake layer.

Enfrin et al. (2021c) studied plasma surface modification through grafting carboxylic acid, amine, and siloxane functional group on the PSF UF membrane. Three modified membranes: hydrophilic-positive (amine functional group), hydrophilic-negative (carboxylic acid functional group) and hydrophobic (siloxane functional group) were used to filter MPs/NPs solutions with a concentration of 10 mg/L. The adsorbed MPs/NPs on the hydrophilic membrane surface decreased more than 60 % compared to the pristine PSF membrane while the hydrophobic surface modification had no impact on MPs/NPs accumulation on the membrane surface compared to pristine PSF membrane. Furthermore, both hydrophilic membranes (negative and positive charge) showed less water flux decline (less than 10 %) compared to pristine and hydrophobic membrane (about 40 %) after 6 hours of filtration. These results demonstrated that repulsive polar forces between MPs/NPs and membranes can prevent fouling.

Enfrin et al., (2021b) also studied the impact of physical cleaning by gas scouring on fouling mitigation of MPs/NPs on pristine and surface modified (mentioned above) UF PSF membranes. The integration of gas scouring caused less water flux decline of pristine and hydrophobic membrane (less than 23 %) compared to the membranes whiteout gas scouring (about 40 %) whilst the integration of gas scouring did not impact fouling tendency of hydrophilic membranes. However, even with the integration of gas scouring, the pristine and hydrophobic membranes had higher water flux decline compared to hydrophilic membranes. This demonstrated that hydrophilic surface modification causes superior fouling mitigation compared to gas scouring integration.

MPs fouling mitigation by coagulants was confirmed by (Li et al., 2021), while (Ma et al., 2019b) and (Ma et al., 2019a) reported higher fouling by integration of coagulation. This is correlated to the different MPs size ranges that were employed in the studies owing to scouring impact of larger MPs. Nevertheless, as MPs may cause irreversible fouling, integration of coagulation could mitigate irreversible fouling by formation of larger particles preventing pore clogging. This however has not been studied as of yet. Overall, based on the relevant studies reviewed, hydrophilic surface modification causes superior fouling mitigation compared to both integration of gas scouring and coagulation. This is due to the repulsive forces between hydrophobic MPs/NPs and hydrophilic membranes that helps to mitigate MPs fouling.

4.3. MPs contribution to biofouling

Biofouling is also an inevitable phenomenon in membrane application which occurs by deposition or growth of microorganisms on the membrane surface and/or within membrane pores, specifically in RO and MBR processes (Aslam et al., 2018; Mehrabi et al., 2020; Vrouwenvelder et al., 2009). Biofouling causes an increase in pressure drop and/or a decrease in flux, thereby increases the operational costs and decreases membrane lifetime (Creber et al., 2010; Mahmoudi et al., 2020; Valladares Linares et al., 2016; Vrouwenvelder et al., 2008) Biofouling is the most complicated fouling type in membrane processes due to the fast reproduction of microorganisms (Bucs et al., 2018). It is also difficult to prevent, control, or remove biofouling because even with high removal of microorganisms in pre-treatment, there are still enough microbial cells remaining to grow on the membrane, and even after intensive chemical cleaning treatment, biofilms/microorganisms can re-emerge (Firouzjaei et al., 2020; Fridjonsson et al., 2015; Miller et al., 2012).

MPs can be a potential carrier of microorganisms owing to their hydrophobic and nonpolar surfaces. Besides, leached additives of MPs can act as potential nutrient sources. Attachment of microorganisms (i.e., bacteria, algae, protozoans, and fungi) to MPs surfaces can form biofilm within minutes to hours (Miao et al., 2019; Rummel et al., 2017). The extent to which this occurs depends on the surface chemistry and the structure of the MPs (e.g., polymer type, adsorbed and leaching chemicals, size, and age) as well as ambient conditions (e.g., temperature, salinity, pressure, the availabilities of light and oxygen, and the presence of other pollutants) (Harrison et al., 2018). Maliwan et al. (2021) investigated MBR biofouling layer in the presence of MPs and reported that MPs presence changed the structural composition of the biofouling cakes and caused a higher extent of biofouling. Overall, MPs may increase and change membrane biofouling cake layer due to the accumulation of microbial communities on their surface during WWTPs processes. However, general conclusions about the MPs effect on membrane biofouling are difficult to be drawn because little information is available in this area.

4.4. Other potential interaction of MPs and membranes

Furthermore, there have been some claims regards the deteriorating impact of MPs on the membrane structure (Enfrin et al., 2019). Particulate contaminants like MPs can damage the membrane surface by enlarging surface pores, creating new pores, and decreasing membrane thickness through excessive direct contact (Wang et al., 2020). Abrasion increases the permeate flux whilst decreasing the rejection, which may additionally compromise the integrity of a membrane (Arimi et al., 2016). The irregular shape of many MPs can damage membranes because MPs edges can erode membrane surfaces, especially in cross-flow systems and high-pressure operations like RO process (Enfrin et al., 2019). A recent study (Pizzichetti et al., 2021) evaluated MPs abrasion on 3 different microfiltration membranes: polycarbonate (PC), cellulose acetate (CA), and polytetrafluoroethylene (PTFE) with a pore size of 5 µm in a dead-end filtration system. PA and PS with an initial concentration of 100 mg/l in a size range of 20-300 µm were filtrated separately through different membranes. PS particles induced more abrasion compared to PA filtration because of their higher shape irregularity; Besides, abrasion of CA membrane was more significant compared to the PTFE and PC owing to lower hardness.

Moreover, the characteristics of the fouling layer itself can play an important role in the removal of pollutants during the separation processes. This is because of the formation of hydrogen bonds, hydrophobic adsorption, and electrostatic repulsion between particles and the fouling layer (P. Wang et al., 2020). For instance, the presence of pharmaceutically active compounds, a type of micropollutants, decreases organic foulant accumulation on membrane surfaces due to the hydrogen bonding and hydrophobic interactions between pharmaceutical compounds and organic foulants (C. Li et al., 2018). Hence, MPs in a fouling layer may increase or decrease the removal rate and fouling of other pollutants present in wastewater. Furthermore, since most MPs are negatively charged, their presence in the fouling cake layer repels positively charged particles.

Im et al. (2020) investigated the removal rate of perfluorinated pollutants by a forward osmosis (FO) process with the co-existence of PVC MPs with a concentration of 0.5 g/L and reported that the presence of MPs increased the rejection rates for some of the perfluorinated pollutants owing to their adsorption on MPs. However, the employed concentration of MPs in this study was far higher than the real concentration in WWTP effluents Furthermore, the study reported that MPs presence decreased water permeability of membranes due to acceleration of concentration polarisation by organic foulants and MPs. However, the impact of MPs needs to be separately investigated as the presence of organic foulants itself increases concentration polarisation. Nevertheless, MPs' contribution to the formation of fouling layers and their interaction with other pollutants has yet to be extensively explored through targeted research. Research development in this field can significantly contribute to the identification and further modification of membranes to minimise fouling and maximise membranes filtration efficiency.

Besides, the possibility of secondary pollution of MPs via membranes has been identified in different research studies (Bayo et al., 2020a; Hidayaturrahman and Lee, 2019; Mintenig et al., 2017; Talvitie et al., 2017). This can be correlated to physical and chemical cleaning processes, mechanical stress, and aging of membranes. Physical and chemical cleaning of membranes include backwashing with high pressure and using chemical agents that contribute to membrane embrittlement and degradation in long term. Therefore, it can cause secondary pollution by releasing MPs (Ding et al., 2021, 2020; Huang et al., 2020; C. Wang et al., 2020). Further research is essential to investigate this matter and modify the employed membrane structures accordingly.

It also deserves to discuss how to control and dispose the microplatics in rejected water of membrane treatment. This is required to validate the feasibility and optimise the MPs removal by membrane processes. Recently, sustainable biological degradation and photocatalysis treatment technologies have been employed to remediate MPs pollution (Ariza-Tarazona et al., 2020; Jiang et al., 2021; Othman et al., 2021). While employing these methods for treating the whole volume of wastewater requires high cost and retention time, a hybrid system of membrane and degradation can be an economical and at the same time a sustainable choice for solving the problem of MPs presence in wastewater. Magnetic extraction has also been employed recently for MPs removal (Grbic et al., 2019). This can be another option for the separation of MPs from rejected water as exposing the rejected water to a magnetic field can be a more feasible approach than that to the whole volume of wastewater. In conclusion, efficient removal and degradation of MPs require a combination of technologies to implement an optimised, economical and sustainable treatment in WWTPs.

5. Potential membrane technologies for MPs removal

Dynamic membrane and forward osmosis membrane are two other options for MPs removal which have not been studied for this purpose as of yet. However, they can be suggested as promising technologies for MPs removal due to their lower fouling and abrasion impact resulting from the low hydraulic pressure in the process (Hartanto et al., 2016; Hartanto et al., 2019). A dynamic membrane (DM) process is a combination of porous support material (e.g., cloth or mesh) with a fouling layer formed during wastewater filtration (Ersahin et al., 2012). Filtering a high initial flux forms the DM layer within 0.3-24 h (Hu et al., 2016). A broad range of pore sizes from 10 to 500 μm have been employed in various studies as the mesh structure (Saleem et al., 2017). The DM layer can form a new layer after being fouled and washed; hence, there is no need to physically replace the membrane (Fan and Huang, 2002). This is a significant advantage for DM process compared to the other conventional membrane techniques.

Li et al., (2018) used a dynamic membrane (DM) with a 90 μm mesh in a dead-end filtration unit under gravity-driven mode for treating synthetic wastewater containing diatomite micro-particles (in the size range of 1.65 μm to 516 μm). In this study, an excellent micro-particle removal efficiency (99.5 %) was observed which was attributed to the ability of the DM to remove low-density particles. Overall, the DM technique has a great potential to develop into a suitable MPs removal approach given its relatively low cost and energy consumption, and the simplicity of its cleaning and maintenance requirements; however, more research studies investigating the removal and fouling of MPs by DMs are required to make more robust conclusions regarding its efficiency.

Although, co-existence of MPs to investigate other pollutants removal in FO system has been studied recently (Im et al., 2021), the removal and fouling of MPs were not discussed. FO could stand as a promising technology for this application due to its noticeable advantages. First, fouling on membrane in FO system is less severe compared to other membrane technologies due to low or no hydraulic pressure. Secondly, FO requires far less energy than the other filtration processes that is again correlated to low or no hydraulic pressure. Lack of hydraulic pressure can also benefit mitigating/preventing breaking down of MPs and abrasion effect. Finally, FO can be proposed to be used in WWTPs effluents using seawater as draw solution that could decrease required energy for draw solution recovery. This technology can prevent the entrance of MPs as well as other micropollutants that threaten the aquatic environment. In a project in Denmark, a pilot FO system for treating MBR effluent was employed for micropollutant removal. The study reported a high rejection rate of up to 99 % for 35 different micropollutants (R. Li et al., 2021). The mentioned project also considered

MPs removal, but no data has been published yet from this group regarding the MPs removal ("The hunt for a potent solution for micropollutants - Aquaporin," n.d.).

6. Conclusions

This study has comprehensively reviewed research on the assessment of MPs presence in WWTPs and their relevant correlation to membrane fouling as well as its associated issues and prospects. Since WWTPs cannot completely remove MPs, advanced treatment is required to reduce MPs entry into the aquatic environments. Varied advanced treatment strategies have been studied for MPs removal, and among them, membrane processes have shown good potential to be employed as effective MPs removal technologies. However, fouling, abrasion and other membrane limitations regarding MPs removal need to be considered to develop more efficient membrane-based techniques. This review has provided a substantial insight on MPs and membrane interactions and MPs fouling and can pave the way for developing more efficient membranes/membrane-based technologies and modifying the existing ones to address MPs concerns in wastewater.

7. Perspectives

Although previous studies have led to valuable achievements to date, there are still considerable knowledge gaps in this field. This comprehensive review has identified the following topics that warrant further research.

- Existing wastewater treatment processes are not designed for MPs removal as they are essentially emerging contaminants. Future studies could focus on optimisation and modification of existing wastewater treatment units towards targeting MPs removal and minimising their emission into aquatic environments. Forward osmosis system is a promising technique with great potentials to address MPs pollution in WWTPs effluents for retention of MPs with low cost and energy. However, extensive economic feasibility studies are required in this respect.

- Most studies have not considered NPs which probably might have higher concentrations in WWTPs effluents and be more hazardous to aquatic environment. It is strongly suggested that future research give a special focus to the investigation of NPs presence and removal from WWTPs as well as studying their membrane fouling effects. The possibility of pore clogging and irreversible fouling of membrane by NPs, in MBR treatment which its applied membranes have larger pore sizes, are also suggested to be studied.
- Fiber MPs account for a very significant and persistent portion of MPs in WWTP effluent; fibers removal in WWTPs has shown some complexity due to the longitudinal penetration of fibers into small gaps or membrane pores. Accordingly, it is necessary to develop membrane technologies that are capable of efficient fiber retention or integrate further tertiary treatment approaches downstream of the WWTPs to capture the 'leaked' microfibers. Hybrid treatment of membrane and coagulation could be a potential solution to trap fibers passing the membrane longitudinally. Besides, as fibers are negatively charged, developing negatively charged membranes can repel fiber and prevent their passage through the membrane. It should be noted that most of the commercial membranes are negatively charged so this can be studied employing commercial membranes. The impact of both suggestions on the removal rate of other pollutants and their economic efficiencies need to be also considered.
- Despite the high removal rate of membrane methods, secondary contaminations via degradation of membrane filters have been observed. The extent of this contamination source still needs to be established and warrants further investigation.

- MPs break up due to the pressure applied on membranes in the pressure-driven membrane processes should be further studied. Optimisation of applied pressure and velocity may possibly mitigate this problem.
- The impacts of MPs characteristics such as their morphology, type, and size on fouling of different membrane technologies should be further explored. This can benefit developing techniques with less severe fouling to frequently occurring MPs in WWTPs effluents.
- In all studies, pristine MPs were employed for fouling investigation, whereas in WWTPs, aged MPs and their affiliated chemicals may have different physicochemical characteristics that can alter their impacts on fouling. The occurring MPs in WWTPs effluents can be collected and used for a more realistic investigation of membrane fouling.
- The presence of MPs in WWTPs can interfere with the treatment procedures targeting other pollutants. Experimental studies investigating the impacts of MPs on the treatment/ removal of other contaminants by membranes are highly beneficial to design more robust and durable treatment processes.

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Declaration of interests

None.

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