Review article

Strategies for mitigating challenges associated with trace organic compound removal by high-retention membrane bioreactors (HR-MBRs)

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Due to the limitations of conventional ultrafiltration/microfiltration-based membrane bioreactors (UF/MF-MBRs) in removing trace organic compounds (TrOCs), the concept of high-retention membrane bioreactors (HR-MBRs) was introduced. Despite the benefits, HR-MBRs still suffer several drawbacks. Therefore, this paper critically reviews the effectiveness and feasibility of the proposed strategies to alleviate fouling, salinity build-up and incomplete biodegradation of TrOCs during wastewater treatment by HR-MBRs. The severity of each challenge is compared amongst the various configurations together with the associated capital and operational expenditure to determine the most cost-effective set-up. Guidance is provided on strategies and/or lessons that could be adopted from well-established processes used at municipal scale. Chemical cleaning as mitigation for fouling degrades membranes leading to poor TrOCs removal, while pre-treatment and membrane surface modification increase operational expenditure (OpEX). However, there are other environmentally-friendly pretreatment and cleaning options which hold great potential for future application. These options such as advanced oxidation processes (AOPs) are critically discussed in this work. Further, in-depth discussion is made on the pros and cons of the various approaches (such as frequent sludge withdrawal, intermittent UF/MF filtration and using organic salts) to alleviate salt build-up. Finally, incomplete biodegradation of rejected TrOCs in the bioreactor transfers problems of toxic pollutants from wastewater treatment to sludge management. Herein mitigation strategies including using stronger biological agents and coupling HR-MBRs with other techniques are debated. Despite the challenges, HR-MBRs are a promising solution for clean water production from TrOCs impaired wastewater. Therefore, more research is needed to improve the performance of HR-MBRs.

Conventional and high-retention membrane bioreactors

Conventional membrane bioreactors (MBR) combine biological processes as well as membrane filtration, utilizing microfiltration (MF) and ultrafiltration (UF) membranes for wastewater treatment and reclamation. However, MF and UF membranes are ineffective in retaining Trace Organic Compounds (TrOCs), including pharmaceuticals, pesticides, hormones and antibiotics[1,2]. Therefore, high-retention membranes such as nanofiltration (NF), reverse osmosis (RO), forward osmosis (FO), and membrane distillation (MD) have been introduced in biological treatment to achieve high removal of TrOCs. The coupled process known as a high-retention membrane bioreactor (HR-MBR) has been the research focus in recent years. The high-retention processes ensure prolonged retention of the TrOCs in the bioreactor for further biodegradation.

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The most investigated high-retention membrane bioreactor (HR-MBR) processes to date are nanofiltration membrane bioreactor (NF-MBR)[3–5], reverse osmosis membrane bioreactor (RO-MBR)[6–8], membrane distillation bioreactor (MDBR)[9–11] as well as an osmotic membrane bioreactor (OMBR)[12–14]. Figure 1 shows the number of publications on the different configurations on HR-MBR. The graphs were drawn from the number of peer-reviewed scientific papers related to HR-MBR technologies in Scopus (abstract and citation database of peer-reviewed literature). RO-MBR remains the highly investigated HR-MBR process.

The main differences between conventional MBRs and high-retention membrane bioreactors (HR-MBRs) are the type of membranes used, driving force, solute rejection properties by the membranes and flux (Table 1), which have been reviewed by Luo et al.[15].

HR-MBRs were proposed to mitigate several challenges associated with MF/UF-MBRs, which originate from the inability of MF/UF-MBRs to remove slowly degradable small molecular weight compounds[16]. The poor removal of slowly biodegradable (recalcitrant) organics leads to the following consequences, as noted by Phattaranawik et al.[16]: (i) poor permeate quality that negatively affects polishing and disinfection steps, limiting direct reuse potential of the permeate[17]; (ii) since poorly biodegradable organics permeate through MF/UF-MBRs, there is inadequate time for microorganisms to acclimatize to achieve acceptable permeate quality and (iii) effective biodegradation of slowly biodegradable organic compounds require longer Hydraulic Retention Time (HRT), and this implies requirements of larger MF/UF-MBRs tanks which would increase capital and operational costs.

The removal of organic substances by MF/UF-MBRs depends mainly on the biological activity because MF/UF membranes have poor TrOCs removal. As noted before, there are several reasons for using HR-MBRs, including the better removal of dissolved solids, organic compounds, nutrients and pathogens[18–21] by facilitating the biodegradation of pollutants[22]. In this paper, we focus on the rejection of TrOCs by HR-MBRs, the challenges associated with HR-MBRs and their effects on TrOCs removal, and mitigation approaches and effectiveness.

HR-MBRs may be operated under aerobic or anaerobic conditions, although most of the studies to date have focused on aerobic HR-MBR. Further, the high-retention membranes may be submerged or applied as a side stream mode. Figure 2 shows a schematic representation of aerobic HR-MBRs. Anaerobic systems have a similar set-up without air bubbling.

In an NF/RO membrane bioreactor (NF/RO-MBR), hydraulic pressure serves as the driving force for the permeation of feed from the bioreactor (Fig. 2a, b). The membrane rejects low molecular weight TrOCs and salts in this process through various mechanisms discussed later. In OMBR (Fig. 2c, d), osmotic pressure is used to drive raw water from the bioreactor through FO membranes. An osmotic pressure develops across the membrane interface because of the high salt concentration in the draw solution. Low molecular weight salts such as NaCl and $MgCl_2$ are often used as draw solutes. Due to the dilution of the draw solution, an additional RO or MD process is used to produce desalinated water and reconcentrate the draw solution[15]. In MDBR (Fig. 2e, f), the MD membrane allows water vapor to permeate through the hydrophobic membrane. The driving force here is the temperature gradient between the feed and permeate side of the membrane. Hydrophobic MD membranes reject non-volatile TrOCs, while aeration stripping and biological degradation eliminate volatile organic compounds that would otherwise pass through the hydrophobic membrane[15].

Asif et al.[23] reviewed the fate of TrOCs during wastewater treatment by HR-MBRs. The authors discussed different removal mechanisms as well as influencing factors. However, the authors did not evaluate the different challenges in HR-MBR, their mitigation strategies, and their effectiveness. A 2014 review by Luo et al.[15] highlighted some challenges in HR-MBR, including salinity build-up, low permeate flux and membrane stability. However, a significant number of new relevant studies have made it necessary to present the current state of understanding on the topic.

Despite the potential, HR-MBRs have several challenges, including membrane fouling, salt build-up in the bioreactor and incomplete biodegradation (and therefore accumulation) of the rejected TrOCs. Further, although the removal of TrOCs is higher than in conventional MBRs, some TrOCs are still detected in the permeates of high-retention membranes. This review seeks to discuss, in detail, the major challenges associated with HR-MBRs and their effects on the overall performance of the removal of TrOCs, highlight the remediation strategies and assess their effectiveness and feasibility. A brief outline of conventional MBRs followed by HR-MBRs and their configurations are presented. Because most of the available studies only reported the overall TrOCs removal, our review focuses on that; however, the relative contribution of biological and membrane processes will be presented whenever available. The major challenges in HR-MBRs are identified, and their impacts on the gross performance of HR-MBRs in removing TrOCs are discussed. Mitigation strategies are provided for each problem, and their efficacy and feasibility are assessed. Based on the extensive literature review, this work provides knowledge gaps and recommendations for future research to mitigate challenges associated with TrOCs removal by HR-MBRs.

## Rejection of trace organic compounds (TrOCs) by HR-MBRs

In recent years, Trace Organic Compounds (TrOCs), including pharmaceuticals, hormones, personal care products, per- and poly-fluoroalkyl substances, disinfection by-products, flame retardants, and plasticisers have been detected in several water sources such as river streams, wastewater, tap water, groundwater, and marine water across the globe[24–28]. TrOCs may have unforeseen physiological consequences for humans and other organisms, and their toxicity level depends on the compound's class. Such negative consequences include liver damage, infant weight loss, respiratory problems, neurological disorders, immunodeficiency and reproduction disorders[29,30]. While some TrOCs degrade quickly and easily, other TrOCs are persistent[31]. The recalcitrant TrOCs resist biodegradation, sunlight photolysis, and other abiotic degradation processes, and thus the compounds, are expected to be in the biosphere permanently since most of the
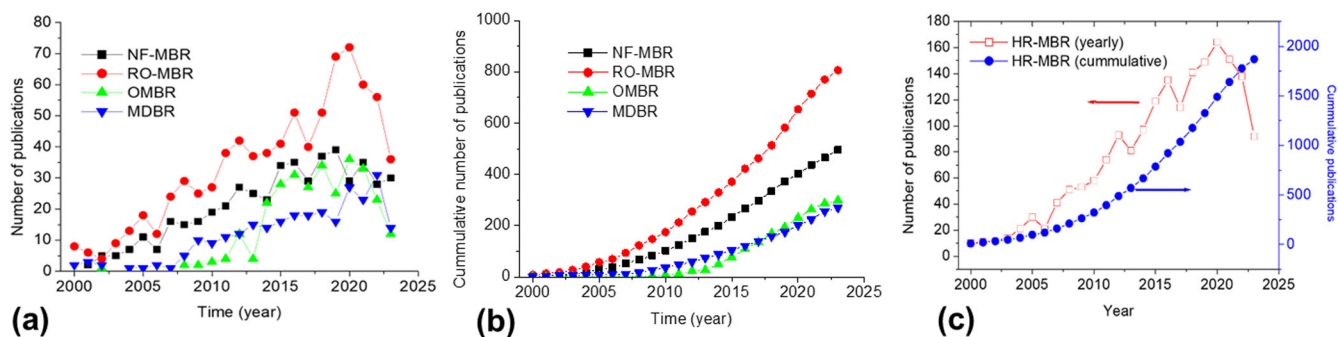
Fig. 1 | Publications on high-retention membrane bioreactors. Number of publications on the different HR-MBR configurations in Scopus (abstract and citation database of peer-reviewed literature): (**a**) number of yearly publications; (**b**) cumulative number of yearly publications; and (**c**) combined HR-MBR publications.

Table 1 | Differences between conventional MBR and HR-MBR

Characteristic	Configuration			
	MF/UF-MBR	NF/RO-MBR	OMBR	MDBR
Membrane type	MF/UF (hydrophilic)	NF/RO	FO	Hydrophobic MF (porous)
Driving force	Hydraulic pressure (low)	Hydraulic pressure (high)	Osmotic pressure	Vapor pressure
Salt rejection (%)	Poor	40–99.9[241]	~100[242]	100[16]
Total organic carbon (TOC) in permeate (mg/L)	3–10[243]	1–4[241]	<3[186,244]	<0.8[16]
Water flux (L/m²/h)	10–30[243]	<2.5[241,245]	<10[186,244]	1.2–15[10,16]
Capital cost (US$)	7990–38,790[231]	100,000 – 293,000,000[246]	-	375,000[233]
Operational cost	0.4–0.58 (US$/m³)[231]	0.2–1.2 (US$/m³)[246]	-	375,700 US$/year[233]
Energy consumption	-	0.16–4.0 (kWh/m³)[246]	-	22 MWh/d[233]
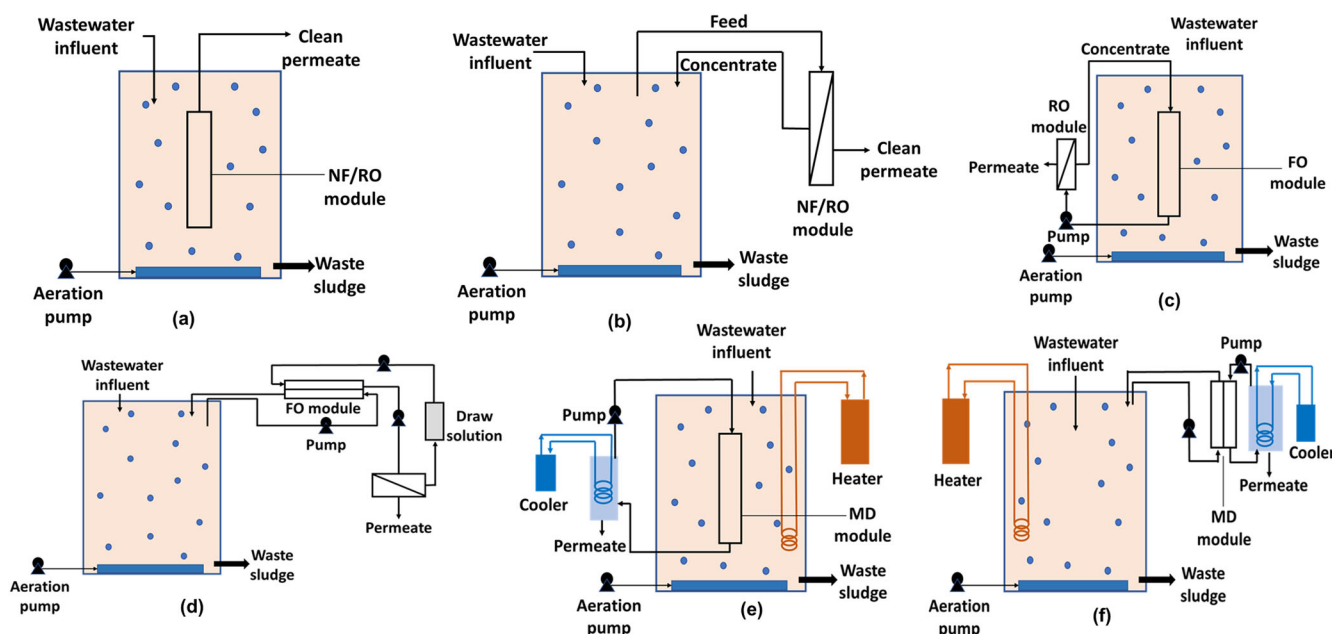
Fig. 2 | Configurations of high-retention membrane bioreactors. Representation of aerated submerged and side stream HR-MBRs: (a) submerged NF-MBR, (b) side stream NF-MBR, (c) submerged OMBR, (d) side stream OMBR, (e) submerged MDBR and (f) side stream MDBR system.

water treatment process cannot remove them[25]. Similarly, conventional MBR processes utilizing MF and UF membranes fail to remove TrOCs from wastewater[1,2], hence the introduction of HR-MBRs, which have better TrOCs removal efficiency than the individual biological and membrane filtration processes[32] and conventional MBRs[17]. For example, TrOCs have been detected in the draw solution of MF-MBRs at concentrations of 3.8–100.4 ng/L[33].

In HR-MBR, TrOCs are removed in the biological reactor as well as in the membrane filtration step and this makes properties of the bioreactor, the high-retention membranes, and the solute properties important parameters. TrOCs removal in the bioreactor is through biodegradation by microbes[34] and adsorption of TrOCs onto the sludge[35], subsequently retained by the high-retention membranes. Adsorption and biodegradation are influenced by hydraulic retention time (HRT), sludge retention time (SRT), dissolved oxygen (DO), pH, addition of sorbent materials and solids' concentrations, amongst others. In addition, the physicochemical properties of TrOCs such as charge, molecular structure, hydrophobicity, and molecular interaction force amongst others also play a role in adsorption and biodegradation.

For example, Song et al.[36] found that the removal of ketoprofen, primidone, ibuprofen, diclofenac, carbamazepine, gemfibrozil, simazine, atrazine, and diuron in the bioreactor was poor (<30%) and this was attributed to the presence of electron-withdrawing groups, such as chloro,

amide, and nitro in their molecular structures[37]. Further, these compounds were relatively hydrophilic with Log D values lower than 3.2 at neutral pH, thus they poorly adsorbed to the sludge phase. Therefore, their removal mechanism was mainly due to biodegradation. Compounds with low hydrophobicity such as caffeine, sulfamethoxazole, trimethoprim, and amitriptyline were effectively removed in the biological process because of their high intrinsic biodegradability due to the presence of electron donation groups (amine and hydroxyl) in their molecular structure[38]. Their removal in the biological reactor could reach 70%[36]. Contrary, highly hydrophobic compounds such as linuron, clozapine, diazinon, triclosan, and triclocarban highly absorb on the sludge and therefore are biodegraded highly. Degradation rates and adsorption of TrOCs in the bioreactor have been recently reviewed by Gu et al.[39], where both processes vary for the different TrOCs and are influenced by the properties of TrOCs as well as the bioreactor. In another review, Semblante et al.[40] highlighted the role and mechanisms of sorption, biodegradation, and abiotic transformation of TrOCs in conventional wastewater treatment. In the membrane filtration step, the TrOCs are removed through size exclusion[41,42], electrostatic interactions[43,44] and non-electrostatic interactions[42,45], which include hydrophobic interactions, van der Waals interactions and hydrogen bonding[46]. To achieve high TrOCs removal, the properties of both the high-retention membranes and TrOCs are important. In addition, the chemistry of the feed water and operational parameters also play a role.

The removal of TrOCs by HR-MBRs was recently reviewed by Asif et al.[23], who focused on the retention mechanisms and influencing factors. Other publications on the removal of TrOCs by MBRs and HR-MBRs, together with their removal mechanisms, include the works of Zhang et al.[32], Lim et al.[47], Luo et al.[48] and Criscuoli et al.[49]. Using antibiotics as model TrOCs, Haffiez et al.[50] focused on specific bioreactor parameters that affect the biodegradation of organic waste. In another work, Mahlangu et al.[51] reviewed the removal of TrOCs by high-retention membrane processes and focused on the removal mechanisms, challenges and opportunities for further improvement.

TrOCs removal by MDBR

The removal of TrOCs by membrane distillation bioreactors (MDBR) is controlled by interactions between the membrane and TrOCs; and this makes the characteristics of TrOCs and membrane important. The organics are retained through charge exclusion as well as exclusion based on the volatility and hydrophobicity of the compounds. MD membranes have pore sizes larger than that of NF/RO membranes and this makes TrOCs removal by sieving irrelevant[52]. In addition, the membrane porosity play no role in the retention of TrOCs[53]. Therefore, volatility and adsorption (due to the hydrophobicity of the membrane) are the major TrOCs removal mechanisms. Due to the negative surface charge of MD membranes, negatively charged TrOCs are highly rejected than positive compounds through charge repulsions[54]. The volatility of TrOCs is the main determining factor for their removal by MD because the mass transfer occurs in the vapor phase. Thus, non-volatile compounds are completely rejected, while volatile TrOCs permeate the membrane leading to poor removal[55]. Regarding hydrophobicity, it has been reported that hydrophilic TrOCs with low volatility are rejected more than hydrophobic TrOCs with high volatility[56]. The degradation of TrOCs due to the high feed temperatures (thermal degradation) in MD is another feasible TrOCs removal mechanism. However, this depends on the characteristics of TrOCs and operating conditions[57].

TrOCs removal by NF/RO-MBR

According to the solution-diffusion model, the removal of TrOCs by NF/RO membranes is mainly through size exclusion and adsorption where the solutes first adsorb onto the membrane surface, partition into the membrane, and diffuse through the membrane to the permeate side[58]. Regarding size exclusion, TrOCs that are larger than the membrane molecular weight cut-off (MWCO) are well rejected by the membrane. The size of the TrOCs determines their diffusion through the membrane, and this parameter has been represented by molecular size such as molecular weight, Stokes diameter, molecular width and length, minimum projection area and van der Waals volume[59,60]. When the TrOCs size is relatively smaller than the pore size of the membrane, adsorption becomes the important mechanism to achieve removal. In this case, TrOCs with high affinity for the membrane are adsorbed onto the membrane surface and partition to the permeate side[61]. Membrane-TrOCs affinity interactions include electrostatic, polar, hydrophobic, hydrogen bonding and π–π interactions[62]. Therefore, adsorption is affected by the TrOCs charge, polarity, hydrophobicity, and functional groups. Surface carboxylic functional groups give NF/RO membranes a negative surface charge. Therefore, negatively charged TrOCs are well rejected due to electrostatic repulsions, while positively charged TrOCs are attracted to the membrane surface resulting in poor retention[63]. TrOCs with high dipole moment are aligned such that their dipole charge is opposite to that of the membrane surface charge, and this promotes their adsorption and lower removal[64,65]. Hydrophilic TrOCs are rejected more than hydrophobic TrOCs due to the high affinity of hydrophobic TrOCs to adsorb onto the membrane surface[66,67]. Finally, hydrogen bonding and π–π interactions promote the adsorption of TrOCs onto the membrane surface leading to poor rejection[44,68].

TrOCs removal by OMBR

In FO, the transport of water through the membrane is in opposite direction to reverse salt flux (from the draw solution). As a result, reverse salt flux prevents diffusion of TrOCs from the feed side to the draw side leading to higher rejection than in NF/RO[69]. The removal of non-ionic TrOCs is largely dictated by size exclusion but some deviations have been reported[70]. Charge interactions also play a role where repulsive membrane-TrOCs interactions results in higher removal. However, at higher draw solute concentration, the effects of charge interactions are canceled because to the suppression of the double layer surrounding a charged solute[71]. This effect is caused by increase in ionic strength in the feed side due to reverse salt flux at high ionic strength[69] For uncharged TrOCs, their interaction with the membrane is not influenced by ionic strength; therefore, size exclusion becomes the important removal mechanism[72]. Other important TrOCs removal mechanisms in FO are adsorption or dipolar interactions where TrOCs with high dipole moment (e.g., carbamazepine with 3.6 Debye) orientate towards the membrane pore resulting in poor removal[73]. The removal of TrOCs by MDBR, NR-MBR, OMBR and RO-MBR is presented on Tables 2–5, respectively.

Figure 3 presents a summary of TrOCs removal efficacy by HR-MBR processes. The data is drawn from Tables 2–5 which present detailed information on studies that have reported on the removal efficiency of TrOCs by HR-MBRs. The chemical structures of the TrOCs are presented in Fig. 4. TrOCs removal is based on the removal rates reported by different studies, and the TrOCs are ranked according to biodegradation and/or sorption in the bioreactor (from lowest to highest). Most of the TrOCs are poorly removed in the bioreactor, however, they are well retained by the various membrane processes.

Fouling of high-retention membranes
Classification of fouling types and fouling mechanisms

Fouling remains a major challenge in conventional MBRs and HR-MBRs, whereas, in the former, it is primarily attributed to the deposition of macromolecules that are biopolymeric as well as colloidal and particulate in nature. In the latter, fouling is even more complex due to the additional effects of elevated salinity (a challenge discussed in a separate section). Membrane fouling is classified based on the type of foulants accumulated on the membrane surface. Accordingly, the major fouling types are organic fouling, colloidal fouling, inorganic fouling, biofouling and combined fouling (i.e., fouling by more than one type of foulant). Colloidal, organic, and inorganic foulants in the bioreactor result in colloidal fouling, organic fouling and scaling, respectively; biofilm formation on the high-retention membranes results in biofouling that degrades membrane performance[18–20,74].

Organic fouling is a term used to describe membrane fouling by natural organic matter (NOM) and polysaccharides omnipresent in raw water. Organic fouling has been widely reported, and the flux decline has been ascribed to increased hydraulic resistance exerted by the fouling layer. Organic fouling is exacerbated by the presence of divalent cations (e.g., calcium), which result in the formation of compact foulant-divalent cation complexes (Fig. 5a) that resemble an egg-box[75,76]. Although wastewater has lower salt content compared to seawater and brackish water, the high rejection properties of membranes used in HR-MBR result in salinity build-up in the feed. Therefore, the accumulation of divalent cations may exacerbate organic fouling during wastewater treatment by HR-MBRs.

Inorganic fouling is described as precipitation and crystallization or scaling of rejected salts (e.g., magnesium sulfate ($MgSO_4$), calcium carbonate ($CaCO_3$), and calcium sulfate ($CaSO_4$) dehydrate or gypsum) on the membrane surface leading to flux decline[77]. Due to the high-retention of sparingly soluble salts, their concentration on the membrane surface increases over time, and this raises the osmotic pressure gradient across the membrane leading to flux loss[78]. This concept, called concentration polarization, has been dubbed the major cause of flux decline in inorganic fouling.

Colloidal fouling refers to membrane fouling by colloids such as silica and aluminum oxide. Interactions between the colloids and membranes control the deposition of colloidal particles on the membrane surface. Colloidal fouling results in flux decline due to hydraulic resistance and cake-enhanced concentration polarization (CECP—a phenomenon where the

Table 2 | Removal of Trace Organic Compounds (TrOCs) from wastewater by membrane distillation bioreactors (MDBR)

Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates[a]	Ref
An anaerobic osmotic MBR was coupled with MD (OMBR-MD) to treat municipal sewage containing TrOCs. Synthetic wastewater simulating municipal wastewater was prepared and spiked with 7 TrOCs at 2 µg/L per compound. The MD process was used to reconcentrate the FO draw solution.	Biological removal by sorption for 17α-ethinylestradiol, betamethasone, fenofibrate, fluconazole, ketoprofen, loratadine and prednisone was about 30%, 31%, 20%, 25%, −18%, 5% and 33%, respectively. Removal by biodegradation was 60%, 50%, 44%, 10%, 55%, 60% and 22% for 17α-ethinylestradiol, betamethasone, fenofibrate, fluconazole, ketoprofen, loratadine and prednisone, respectively.	There was more than 99.99 removal of all TrOCs by the MD process (PTFE membrane) except for betamethasone which was removed by 96.5% after 22 days of MD operation.	[247]
A direct contact membrane distillation unit was integrated with an anaerobic membrane bioreactor for the removal of TrOCs and energy recovery. Synthetic wastewater simulating high-strength domestic wastewater was spiked with 26 TrOCs. The feed was prepared from 4000 mg/L glucose, 750 mg/L peptone, 2250 mg/L sodium acetate, 175 mg/L potassium dihydrogen phosphate, 175 mg/L magnesium chloride, and 175 mg/L urea.	Caffeine (75%), sulfamethoxazole (83%), ketoprofen (25%), trimethoprim (95%), paracetamol (55%), naproxen (35%), primidone (15%), ibuprofen (16%), triamterene (45%), carazolol (55%), tris(2-carboxyethyl)phosphine (30%), diclofenac (15%), carbamazepine (18%), gemfibrozil (22%), simazine (25%), amitriptyline (90%), atrazine (20%), diuron (22%), linuron (62%), clozapine (75%), phenylphenol (50%), bisphenol A (15%), diazinon (90%), triclosan (65%) and triclocarban (70%).	Integration of MD (PTFE membrane) improved removal achieving 80% to complete removal of all TrOCs except for diclofenac (75%), atrazine (74%) and phenylphenol (78%). The MDBR was continuously operated for 30 days.	[36]
A direct contact membrane distillation system was used to treat the effluent of a continuously operated MBR. The MBR was fed with simulated acid mine drainage- and sewage-contaminated groundwater spiked with 12 TrOCs (5 µg/L each) and four metal salts (10 mg/L each).	Acetaminophen (100%), bezafibrate (100%), diclofenac (53%), sulfamethoxazole (80%), amitriptyline (95%), carbamazepine (55%), primidone (82%), triclosan (75%), trimethoprim (93%), atrazine (75%), linuron (75%) and pentachlorophenol (62%).	Degradation in the bioreactor ranged from 52–100%, and biodegradation depended on the electron-withdrawing properties of the TrOCs. The MD process (PTFE membrane) achieved about 85% to complete retention of all TrOCs.	[248]
The removal of 5 TrOCs by enzymatic MBR and MD from synthetic wastewater was investigated where the feed constituted 5 TrOCs at a concentration range 920–1130 µg/L. A photolysis step was added to enhance degradation.	Sulfamethoxazole (40%), diclofenac (75%), bisphenol A (90%), oxybenzone (45%), and carbamazepine (60%).	The MD process (PTFE membrane) achieved greater than 99% TrOCs retention. The retained TrOCs were degraded by the ultraviolet (UV) photolysis process (27–88% degradation).	[9]
A membrane distillation process was used for the removal of TrOCs from synthetic wastewater prepared from 100 mg/L glucose, 100 mg/L peptone, 17.5 mg/L KH$_2$PO$_4$, 17.5 mg/L MgSO$_4$, 10 mg/L FeSO$_4$, 225 mg/L CH$_3$COONa and 35 mg/L urea. 29 TrOCs were added at 5 µg/L each.	Enterolactone (92%), primidone (98%), ketoprofen (90%), formononetin (92%), naproxen (45%), gemfibrozil (88%), metronidazole (80%), diclofenac (0%), fenoprop (44%), estriol (95%), ibuprofen (98%), clofibric acid (38%), 17α-ethinylestradiol (90%), oxybenzone (98%), carbamazepine (35%), estrone (99%), 17β-estradiol (99%), 17β-estrodiol-17-acetate (99%), bisphenol A (98%), octocrylene (97%), ametryn (55%), amitriptyline (95%), pentachlorophenol (88%), atrazine (0%), propoxur (30%), triclosan (98%), benzophenone (85%), 4-tert-butyphenol (95%) and 4-tert-octylphenol (95%).	The subsequent MD step removed more than 95–100% of all 29 TrOCs, with most organics completely removed by the PTFE membrane.	[249]
The efficiency of the MDBR system in removing TrOCs was examined. The feed was simulated domestic wastewater spiked with 25 TrOCs at 5 µg/L each.	Clofibric acid (30%), salicylic acid (95%), ketoprofen (98%), fenoprop (33%), naproxen (98%), ibuprofen (85%), primidone (62%), diclofenac (18%), gemfibrozil (65%), propoxur (83%), carbamazepine (5%), pentachlorophenol (78%), estriol (98%), atrazine (25%), ametryn (85%), benzophenone (78%), amitriptyline (98%), 4-Tert-butyphenol (95%), oxybenzone (99%), estrone (98%), 17α-ethinylestradiol (97%), 17β-estradiol (99%), triclosan (50%), 17β-estrodiol-17- acetate (99%) and octocrylene (90%).	The MD process (PTFE membrane) achieved 95% to complete TrOCs removal efficiency. The coupled processes removed TrOCs through biodegradation, sludge adsorption, and rejection.	[10]
A persulfate-assisted direct contact membrane distillation process was evaluated for effectiveness in treating secondary effluent spiked with 12 TrOCs. The MD feed was synthetic wastewater prepared from 400 mg/L glucose, 100 mg/L peptones, 35 mg/L urea, 17.5 mg/L monopotassium phosphate, 17.5 mg/L magnesium sulfate, 10 mg/L ferrous sulfates, and 225 mg/L sodium acetate.	Diclofenac (25%), pentachlorophenol (58%), carbamazepine (63%), triclosan (75%), sulfamethoxazole (78%), primidone (82%), atrazine (82%), linuron (85%), amitriptyline (90%), trimethoprim (95%), bezafibrate (100%) and acetaminophen (100%).	The MD process (PTFE membrane) completely removed all TrOCs and produced high-quality effluent.	[11]
A hybrid anaerobic osmotic membrane bioreactor–membrane distillation system was used for the removal of TrOCs from municipal sewage spiked with 7 TrOCs at 2 µg/L per compound.	17α-ethinylestradiol (90%), betamethasone (94%), fenofibrate (99%), fluconazole (92%), ketoprofen (97%), loratadine (99%) and prednisone (97%).	The inclusion of MD (PTFE membrane) resulted in all TrOCs retained by more than 97%, with fenofibrate and loratadine reaching greater than 99.9% removal.	[250]
An osmotic membrane bioreactor−membrane distillation hybrid system was used for the treatment of synthetic wastewater, simulating medium-strength wastewater. The feed was spiked with 30 TrOCs, with each compound added at 5 µg/L.	Clofibric acid (80%), salicylic acid (99%), ketoprofen (98%), fenoprop (80%), naproxen (95%), metronidazole (95%), ibuprofen (99%), primidone (65%), diclofenac (72%), gemifibrozil (90%), propoxur (92%), enterolactone (45%), carbamazepine (95%), pentachlorophenol (98%), N, N-Diethyl-meta-toluamide (98%), atrazine, estriol (90%), ametryn (80%), amitriptyline (95%), benzophenone (98%), 4-tert-butylphenol (99%), oxybenzone (99%), estrone	Except for enterolactone (85%), estriol (90%) and bisphenol A (90%), the poorly degraded TrOCs were removed by MD (PTFE membrane) in the range of 99% to complete removal.	[189]

Table 2 (continued) | Removal of Trace Organic Compounds (TrOCs) from wastewater by membrane distillation bioreactors (MDBR)

Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates[a]	Ref
	(98%), bisphenol A (90%), 17α-ethynylestradiol (98%), 17β-estradiol (99%), triclosan (99%), β-estradiol-17-acetate (99%), 4-tert-octylphenol and octocrylene (98%)		
A membrane distillation process was coupled to a forward osmosis system to treat raw sewage with a pH of 7.1, electrical conductivity of 1075 µS/cm, and total organic carbon of 72 mg/L. 12 TrOCs were added to the feed to make a final concentration of 5 µg/L each. Granular activated carbon or ultraviolet (UV) treatment was introduced in the feed of the FO to prevent membrane fouling.	Pentachlorophenol, triclosan, linuron, atrazine, amitriptyline, diclofenac, carbamazepine, sulfamethoxazole, caffeine, trimethoprim, primidone, bezafibrate	The hybrid OMBR-MD process utilizing a PTFE membrane removed pentachlorophenol (94%), triclosan (98%), linuron (94%), atrazine (96%), amitriptyline (98%), diclofenac (98%), carbamazepine (99%), sulfamethoxazole (98%), caffeine (98%), trimethoprim (99%), primidone (98%) and bezafibrate (100%). Granular activated carbon adsorption or UV-oxidation prevented contaminant accumulation in the draw solution and led to over 99.5% rejection of TrOCs.	251

[a]Some TrOCs removal rates were estimated from rejection graphs presented in the referenced articles.

PTFE polytetrafluoroethylene, *CTA* cellulose triacetate.

fouling layer prevents back-diffusion of salts from the vicinity of the membrane surface to the feed leading to accelerated salt build-up on the membrane surface and subsequently a rise in osmotic pressure gradient and decline in driving force (Fig. 5b))[79–82]. The cake layer exacerbates concentration polarization effects on flux.

Biofouling occurs through five major steps (Fig. 6), namely: (a) conditioning of the membrane surface, (b) attachment of microbes on the membrane surface, (c) production of extracellular polymeric substances (EPS), which help adhere cells and facilitate attachment of cells onto the membrane surfaces, (d) cell growth and multiplication, and (e) detachment of microbes[83].

A review by Meng et al.[84] discusses the biofouling of low-pressure membranes by polysaccharides and proteins in conventional MBRs and fouling mitigation strategies. However, fouling in low-pressure membrane filtration may differ from that of high-pressure filtration processes due to the lack of hydraulic compaction of the fouling layer[85]; thus, the work of Meng and co-workers may be relevant to biofouling in OMBR and MDBR but not NF/RO-MBR. Further, the work of Meng and co-workers was centered on biofouling. Wastewater constitutes a mixture of foulants which have different fouling mechanisms and effects. Therefore, this work further elaborates on the different fouling types. However, the work of Meng et al. is still important in understanding biofouling in HR-MBRs when biofoulants predominate in the feed.

Membrane fouling is controlled by membrane properties, foulant properties, and operating conditions discussed by Tang et al.[86]. Foulant characteristics include foulant type, size and charge[82,87]; membrane properties comprise surface roughness[22,88], charge, hydrophilicity and functional groups, while feed composition and operational parameters involve the concentration of foulant[82,89], solution pH[90], ionic strength[90,91], presence and concentration of divalent cations[82,87,90–92], initial permeate flux[90], cross-flow velocity[87,90,93], feed temperature[89,93] as well as module design and spacers[94,95]. The roles of the various factors have largely been investigated using synthetic water. However, there are studies based on real wastewater samples. However, the findings from fouling based on synthetic water are still relevant and provide insights into membrane fouling mechanisms during wastewater treatment by HR-MBRs. In MD, flux decline is mainly due to pore wetting by the contacting solutions, increase in salt concentration in the feed solution and cake layer formation[36], while in NF and RO, fouling is mainly due to pore blocking and concentration polarization by rejected salts which lower the driving force. In FO, the reduction in flux is predominantly due to increased hydraulic resistance by the formed cake layer and reverse salt flux which drops the concentration gradient between the feed and draws the side of the FO membrane. Fouling in FO is more reversible than that in high-pressure-driven processes due to the negligible compression of the cake layer[85]. Figure 7 shows the four classic filtration models (complete pore

blocking, standard blocking, intermediate blocking, and cake filtration) widely used to explain flux decline in membrane fouling[96,97].

Fouling degrades the membrane and leads to a shorter lifespan. Previous studies have reported that when organic and colloidal foulants co-exist in the feed, fouling is mainly dominated by organic fouling, regardless of the concentration ratio between the different fouling types[82]. Table 6 summarizes the major fouling or flux decline mechanisms in membrane filtration.

Variation in fouling mechanisms and fouling propensity

From Fig. 2, it can be noted that NF/RO-MBR are pressure-driven processes while OMBR and MDBR are not. Previous studies have reported on variations in the fouling mechanisms and propensity of the different processes. From the different configurations, the fouling propensity to some extent is controlled by membrane surface properties, operating pressure, temperature as well as water transport mechanisms. In MD, the occurrence of large pores containing air and vapor affect fouling due to the effects of the liquid-air interface on the adhesion of foulants and crystal nucleation[98,99]. Contrary, FO and RO membranes have sub-nanometer pores which are smaller than most foulants. Therefore, foulants do not accumulate inside the membrane pores like in MD[99]. Although some studies have claimed that MD and FO are less susceptible to fouling than RO due to their low operating pressure[100,101], there are some findings that have demonstrated that the effect of pressure on fouling is negligible[102]. Temperature plays a role in membrane fouling because it affects solubility, crystallization kinetics and concentration polarization of inorganic salts. For organic fouling, high temperatures may affect fouling through denaturation proteins or depolymerization of polysaccharides[103]. Hydraulic-osmotic pressure difference in FO and RO transports water across the membrane interface. On the other hand, water permeation is driven by vapor pressure difference between the feed and permeate side. In FO and RO, fouling lowers flux through cake-enhanced concentration polarization (CECP). Further, it causes hydraulic drag which reduces the hydraulic pressure. For MD fouling, both CECP and to a lesser extent hydraulic drag reduce vapor pressure. In addition, MD is also prone to cake-enhanced temperature polarization due to the temperature difference in the permeate and feed side of the membrane[104].

A study by Siddiqui et al.[105] compared organic fouling propensity in FO and RO mode using sodium alginate as model foulant. The authors found that FO was more prone to fouling than RO due to the reduction in the intensity of internal concentration polarization (ICP) and increase in the effective osmotic driving force during fouling in FO mode. However, their findings were contrary to common claim in the literature where conclusions on fouling are based only on the flux profiles. The researchers also found that the specific fouling resistance for FO was greater than that of RO and this was due to the contribution of reverse solute diffusion from the draw

Table 3 | Removal of Trace Organic Compounds (TrOCs) from wastewater by nanofiltration membrane bioreactors (NF-MBR)

Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates	Ref
Nanofiltration membranes were used to treat MBR effluent targeting the removal of 12 TrOCs detected in the MBR feed (municipal wastewater).	Diclofenac (8%), erythromycin (−44%), clarithromycin (74%), azithromycin (53%), methiocarb (100%), imidacloprid (−18%), thiamethoxam (1%), clothianidin (88%), acetamiprid (39%), oxadiazon (33%) and triallate (99%).	The poorly biodegraded TrOCs were removed by the membranes and the removal by NF90 was greater than 99.9% for azithromycin, clarithromycin, and diclofenac while the removal by NF270 membrane was 80%, 75% and 91% for azithromycin, clarithromycin, and diclofenac, respectively.	[3]
An enzymatic membrane bioreactor using laccase as the biological agent was equipped with a nanofiltration membrane for the degradation and removal of 29 TrOCs. Synthetic wastewater was spiked with TrOCs at 5 µg/L per organic compound.	Clofibric acid (20%), metronidazole (65%), fenoprop (5%), ketoprofen (5%), naproxen (10%), primidone (15%), ibuprofen (35%), propoxur (30%), diclofenac (35%), carbamazepine (5%), gemfibrozil (20%), amitriptyline (60%), N, N-diethyl-meta- toluamide (DEET) (40%), atrazine (40%), ametryn (15%), benzophenone (90%), octocrylene (95%), salicylic acid (95%), estriol (70%), enterolactone (15%), pentachlorophenol (35%), 4-tert-butylphenol (98%), estrone (78%), bisphenol A (70%), 17α–ethinylestradiol (95%), 17β–estradiol (85%), 17β-estradiol-17- acetate (95%), 4-tert-octylphenol (95%), and triclosan (95%).	The NF90 membrane attained 95–99% retention of all TrOCs from the biological process except for diclofenac and carbamazepine which were removed by 90%.	[4]
Nanofiltration was integrated into a laccase and persulfate-mediated oxidation process that was used for the degradation of TrOCs in MBR treating synthetic wastewater.5 TrOCs were spiked into the feed to achieve 500 µg/L per organic.	Bisphenol A (60%), diclofenac (15%), sulfamethoxazole (7%), carbamazepine (10%) and oxybenzone (99%).	The NF90 membrane removed the poorly degraded TrOCs and achieved complete removal of all TrOCs except for sulfamethoxazole (95%).	[252]
Nanofiltration membranes were used to treat MBR effluent targeting the removal of TrOCs. The MBR was fed with synthetic wastewater simulating municipal sewage. The feed was spiked with 40 TrOCs at initial concentration of 2 µg/L for each compound. The simulated wastewater contained glucose (400 mg/L), peptone (75 mg/L), KH_2PO_4 (17.5 mg/L), $MgSO_4$ (17.5 mg/L), $FeSO_4$ (10 mg/L), and sodium acetate (225 mg/L).	Paracetamol (90%), N, N-Diethyl-meta-toluamide (5%), caffeine (50%), ibuprofen (93%), t-octylphenol (90%), atrazine (3%), meprobamate (10%), primidone (5%), nonylphenol (10%), bisphenol-A (99%), naproxen (35%), carbamazepine (10%), linuron (15%), gemfibrozil (20%), dilantin (0%), triamterene (25%), sulfamethoxazole (90%), ketoprofen (65%), atenolol (95%), estrone (98%), 17β-estradiol (100%), amitriptyline (98%), androstenedione (100%), estriol (100%), testosterone (100%), triclosan (90%), trimethoprim (20%), etiocholanolone (100%), androsterone (100%), diclofenac (20%), 17α-ethynylestradiol (90%), triclocarban (98%), clozapine (70%), omeprazole (60%), hydroxyzine (90%), enalapril (92%), risperidone (90%), simvastatin (95%), simvastatin-hydroxy acid (50%) and verapamil (70%).	The poorly degraded compounds were removed by the NF membranes (NF270 and NF90), and removal rates were between 80% and 100%.	[253]
Domestic wastewater treated by MBR was used directly as an influent for a reverse osmosis process for the removal of TrOCs. The MBR effluent had the following characteristics: 56 mS/cm electrical conductivity, chemical oxygen demand of less than 20 mg/L, 17 β-estradiol less than 5 ng/l and testosterone of 120 ng/L.	17β-Estradiol (13%) and testosterone (55%).	The poorly degraded TrOCs in the MBR effluent was removed by the NF270 and XLE membranes which achieved greater than 95% removal.	[254]
The removal of TrOCs by MBR coupled to a nanofiltration process was investigated. The feed for the MBR was urban wastewater, and NF further treated the MBR effluent.	N-nitrosodimethylamine and its precursors, namely azithromycin, citalopram, erythromycin, clarithromycin, ranitidine, venlafaxine and o-desmethylvenlafaxine. The bioreactor reduced N-nitrosodimethylamine precursors above 94% under aerobic conditions; however, the removal was lower (72%) under anaerobic conditions. Reduction in degradation was also noted for azithromycin (68–59%), citalopram (31–17%), venlafaxine (35–15%) and erythromycin (61–16%).	The NF process (NF90 membrane) achieved more than 90% removal of all compounds.	[255]
A pilot plant MBR was coupled to NF/RO and used to treat raw municipal wastewater over 100 days. The removal of 10 TrOCs which occurred at the concentration range of 0.06–59.50 µg/L, was monitored. The MBR influent had chemical oxygen demand of 2204 mg/L.	Acetaminophen (92%), ibuprofen (97%), caffeine (98–99%), nicotine (94%), carbamazepine (4%), diclofenac (67%), triclosan (46–84%), 4-octylphenol (42–78%), 4-tert-octylphenol and bisphenol A (31–73%)	The NF90 membrane removed 59 to greater than 99% of TrOCs. Acetaminophen, ibuprofen, and caffeine we rejected by over 99%; carbamazepine, diclofenac and 4-tert-octylpheno were rejected by 81–87% while 4-octylphenol (78%) and triclosan (79%) had lower than 80% removal.	[256]
An integrated MBR-NF/RO system was used to remove pharmaceuticals and personal care products from municipal wastewater. The TrOCs were present in the feed at the 7.12–18.4 ng/L concentration range.	Caffeine (87%), atenolol (87%), metoprolol (45%), amoxicillin (75%), trimethoprim (60%), sulfadimidine (75%), sulfamethoxazole (58%), norfloxacin (50%), ofloxacin (60%), ciprofloxacin (70%), lomefloxacin (65%), enrofloxacin (50%), oxytetracycline (65%), tetracycline (70%), chlortetracycline (65%), doxycycline (70%), clarithromycin (80%), erythromycin-H_2O (70%), roxithromycin (75%), azithromycin (78%), carbamazepine (35%), benzhabeite (85%), bisphenol A (83%), nonylphenol (80%), estrone (85%), 17β-estradiol (78%) and estriol (93%).	The poorly degraded TrOCs were completely removed by NF membrane (NF2540) except for trimethoprim (95%), sulfadimidine (98%), sulfamethoxazole (95%), carbamazepine (98%), bisphenol A (99%), nonylphenol (99%) and 17β-estradiol (95%).	[257]

Table 3 (continued) | Removal of Trace Organic Compounds (TrOCs) from wastewater by nanofiltration membrane bioreactors (NF-MBR)

Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates	Ref
The complementary performance of combined MBR with UV-oxidation or NF/RO for the removal of TrOCs from synthetic wastewater simulating medium-strength wastewater was investigated. The feed had a total organic carbon, total nitrogen, and chemical oxygen demand of ~180, 25, and 600 mg/L, respectively. 22 TrOCs were added into the feed at a concentration of 5 µg/L per compound.	Salicylic acid (97%), metronidazole (40%), fenoprop (18%), ketoprofen (65%), acetaminophen (85%), naproxen (45%), primidone (88%), ibuprofen (95%), diclofenac (10%), carbamazepine (30%), gemifibrozil (98%), estriol (98%), pentachlorophenol (60%), 4-tert-butylphenol (90%), estrone (95%), bisphenol A (93%), 17α-estradiol-17-acetate (90%), 4-tert-octylphenol (95%) and 4-n-nonyphenol (90%). Compounds with Log D greater than 3.2 were degraded more.	The NF270 membrane attained about over 95% to complete removal of the TrOCs except for metronidazole (75%), carbamazepine (85%), pentachlorophenol (90%) and bisphenol A (85%).	[124]
Three NF membranes were evaluated for their efficiency in treating MBR effluent, targeting the removal of 11 pharmaceuticals and personal care products in municipal wastewater. The MBR effluent had chemical oxygen demand of 5.9 mg/L and electrical conductivity of 258 µS/cm.	Acetaminophen (18%), atenolol (30%), carbamazepine (−17%), clopidogrel (70%), diclofenac (50%), dilantin (1%), ibuprofen (25%), iopromide (44%), glimepiride (14%), naproxen (55%) and sulfamethoxazole (61%).	The membranes achieved the following removal: acetaminophen (10% NE40, 18% NE70 and 30% NE90), atenolol (25% NE40, 60% NE70 and 62% NE90), carbamazepine (40% NE40, 70 NE70 and 85% NE90), clopidogrel (35% NE40, 80% NE70 and 81% NE90), diclofenac (85% NE40, 100% NE70 and 100% NE90), dilantin (38% NE40, 65% NE70 and 75% NE90), ibuprofen (30% NE40, 50% NE70 and 90% NE90), iopromide (30% NE40, 55% NE70 and 65% NE90), glimepiride (45% NE40, 70% NE70 and 80% NE90), naproxen (40% NE40, 100% NE70 and 100% NE90) and sulfamethoxazole (30% NE40, 35% NE70 and 70% NE90). The order NE90 > NE70 > NE40 showing the role of molecular weight cut-off on solute retention.	[258]

solution. Further, there was no evidence of hydraulic pressure compressing the fouling layer and there was greater foulant accumulation in FO than in RO. In another study, the fouling propensity between RO, FO and MD was compared under identical hydrodynamic conditions using calcium sulfate and sodium alginate as model representatives of inorganic and organic foulants, respectively[106]. There was more flux decline in MD due to scaling after 36 h, while FO resisted scaling and no flux decline was observed. Contrary, there was lesser flux decline in MD due to organic fouling (14%) compared to FO and RO (46–47%). This was ascribed to the high operating temperature raising the diffusion coefficient of foulants, augmenting transport away from the membrane and elevating the critical flux. The results presented in this review indicate that the different process may resists fouling by one foulants while getting fouled by another. Therefore, the extent of fouling depends on the foulant type as well as filtration process.

Tables 7–10 present studies that have reported the fouling of high-retention membranes (MDBR, NF-MBR, OMBR, and RO-MBR, respectively) during wastewater treatment by HR-MBRs and the findings indicate that the type of fouling is the same for the different HR-MBR processes and mainly depends on the composition of the feed. For instance, organic, colloidal, scaling and biological fouling has been reported for all the configurations. However, there are noticeable differences in the flux decline mechanisms and implications of fouling to the process. For example, flux decline in MDBR is associated with membrane wetting as well as thermal and mass transfer resistance. For OMBR, flux decline is ascribed to internal concentration polarization of salts from the draw side. Due to the high pressure applied in NF-MBR and RO-MBR, the flux decline is due to additional hydraulic resistance imparted by the compacted cake layer (for compressible foulants). In addition, cake-enhanced concentration polarization is one major contributor to reduction in flux during wastewater treatment using NF-MBR and RO-MBR configurations.

Fouling has negative effects on the typical design/performance values in the HR-MBR application. Besides the decline in flux, fouling in NF/RO leads to increase in the specific energy consumption in kWh per m³ (kWh/m³) of permeate production due to additional energy required to drive permeation through the fouled membrane. The increase in energy consumption is also due to the resistance to fluid flow through the membrane (because of cake resistance), the friction losses in the retentate and permeate channels of membrane modules and the non-conventional operation of high pressure pumps and energy recovery devices[107]. For FO and MD,

permeation is due to concentration gradient as well as temperature gradient, respectively. Therefore, the effects of fouling on specific energy consumption are less severe in the vapor phase separation process (MD) compared to the liquid phase separation processes (NF and RO). Furthermore, fouling in MD results in immediate failure in membrane separation. Fouling may promote membrane pore wetting which subsequently leads to direct permeation of feed water into the distillate stream and remarkably undermine pollutant removal rate[108].

Influence of fouling on TrOCs removal by high-retention membranes

In HR-MBR, the feed from the biological process will impact the membrane filtration step mainly because wastewater contains a blend of microorganisms, organic foulants, colloids and salts. Additionally, the presence of foulants in the feed (wastewater) may either decrease or improve the biodegradation of TrOCs by the microbes in the reactor. For instance, 1 mg/L of humic acid was found to promote the degradation of bisphenol A and oxybenzone (phenolic compounds) while inhibiting the biodegradation of sulfamethoxazole, carbamazepine, and diclofenac (non-phenolic compounds) in direct contact membrane distillation (DCMD) coupled with photolysis[109]. Further, fouling can influence the performance of high-retention membranes where the removal of TrOCs can either be enhanced, remain unchanged or even decrease (Fig. 8).

The effects of fouling on the retention of TrOCs are controlled by the properties of the organics. Hajibabania et al.[110] studied the effects of fouling on the removal of organic compounds by NF membranes and found that the consequences were dependent on the properties of the compounds. The removal of ionic compounds including diclofenac (85–90%), naproxen (90–92%) and ibuprofen (92–94%) was not affected by membrane fouling and this was ascribed to electrostatic repulsions between the ionic solutes and the negatively charged membrane surface which prevented adsorption of the compounds on the fouling layer[111]. Contrary, fouling generally decreased the removal of nonionic hydrophobic TrOCs. This was due to the formation of loose cake layers (because of the vast range of molecular weight of organic matter) where the hydrophobic compounds (e.g., risperidone (95–65%) and fluoxetine (96–80%)) gradually increased in concentration leading to a reduction in their retention. However, the extent of decrease in the rejection of hydrophobic compounds was less severe for highly hydrophobic compounds due to their adsorption onto the foulants in the feed.

Table 4 | Removal of Trace Organic Compounds (TrOCs) from wastewater by osmotic membrane bioreactors (OMBR)

Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates	Ref
An anaerobic osmotic MBR was coupled with membrane distillation to treat synthetic wastewater simulating municipal wastewater that was spiked with 7 TrOCs at 2 μg/L per compound. The feed had chemical oxygen demand of 404 mg/L.	Biological removal by sorption for 17α-ethinylestradiol, betamethasone, fenofibrate, fluconazole, ketoprofen, loratadine and prednisone was about 30%, 31%, 20%, 25%, −18%, 5% and 33%, respectively. Removal by biodegradation was 60%, 50%, 44%, 10%, 55%, 60% and 22% for 17α-ethinylestradiol, betamethasone, fenofibrate, fluconazole, ketoprofen, loratadine and prednisone, respectively.	The FO process (cellulose triacetate (CTA) membrane) removed 99% 17α-ethinylestradiol, 96% betamethasone, 97% fenofibrate, 99% fluconazole, 96% ketoprofen, 98% loratadine and 99% prednisone.	[247]
A fertilizer-drawn forward osmosis process was used for the removal of TrOCs from synthetic wastewater initially treated through a biological process. The model, TrOCs, were added at concentrations of 10 ug/L each, and the chemical oxygen demand was 800 mg/L.	Caffeine, atenolol, and atrazine	TrOCs removal by the FO process (CTA membrane) was influenced by draw solute type, strength, and membrane orientation. High removal rates (96%–99%) were achieved for all TrOCs from the different draw solutions, and the removal was in the following order: atenolol > atrazine, > carbamazepine.	[259]
Two forward osmosis membranes were compared for their efficiency in removing TrOCs from synthetic wastewater after a biological process. The synthetic feed contained 100 mg/L glucose, 100 mg/L peptone, 17.5 mg/L KH₂PO₄, 17.5 mg/L MgSO₄, 10 mg/L FeSO₄, 225 mg/L CH₃COONa, 35 mg/L urea and 30 TrOCs at 5 μg/L per organic compound.	Clofibric acid (50%), salicylic acid (99%), ketoprofen (98%), fenoprop (78%), naproxen (98%), metronidazole (99%), ibuprofen (99%), primidone (30%), diclofenac (78%), gemfibrozil (98%), propoxur (60%), formononetin (98%), enterolactone (40%), carbamazepine (40%), pentachlorophenol (90%), DEET (95%), estriol (98%), atrazine (35%), ametryn (70%), amitriptyline (98%), benzophenone (99%), 4-tert-butylphenol (99%), oxybenzone (99%), estrone (99%), bisphenol A (98%), 17α-ethynylestradiol (99%), 17β-estradiol (98%), triclosan (99%), β-estradiol-17-acetate (99%), and octocrylene (90%)	The FO membrane (CTA membrane) achieved high removal of the poorly degraded compounds, where the cellulose triacetate membrane achieved more than 98% removal of all compounds besides clofibric acid (95%), fenoprop (90%), metronidazole (90%), primidone (88%), carbamazepine (78%), atrazine (50%), ametryn (85%) and octocrylene (90%). The thin film composite (TFC) membrane performed better compared to its cellulose triacetate counterpart and removed greater than 98% of all TrOCs except for atrazine (90%).	[13]
An osmotic membrane bioreactor was evaluated for the removal of 3 TrOCs from simulated sewage wastewater with the following characteristics: 350 mg/L chemical oxygen demand, 16 mg/L ammonium nitrogen, 28 mg/L total nitrogen and 3.5 mg/L phosphate. Potassium chloride, sodium chloride and sodium acetate were used as draw solutes.	Caffeine (92–100%), atenolol (94–100%), and atrazine (14–50%) under anoxic conditions. Biological removal under oxic conditions was 92–100% for caffeine, 30–90% for atrazine and 90–100% for atenolol.	The FO process (CTA membrane) achieved the following removals when using different draw solutes: 98–100% removal for sodium acetate, 92–100% removal for sodium chloride and 94–100% from potassium chloride. Caffeine was removed more than atrazine and atenolol in all draw solutes used.	[14]
The role of draw solution in removing TrOCs in an osmotic MBR was investigated. The feed was prepared from deionized water spiked with TrOCs at 2 μg/L per compound. Draw solutions were prepared using sodium chloride, sodium acetate, magnesium acetate, ethylenediaminetetraacetic acid, disodium salt, and magnesium chloride.	17α-ethinylestradiol, betamethasone, fenofibrate, fluconazole, ketoprofen, loratadine and prednisone	The CTA FO membrane had the following removal rates for the different draw solutions: 70–99% for sodium chloride; 68–94% for magnesium chloride; 76–98% for sodium acetate; 62–95% for magnesium acetate, and 62–99% for ethylenediaminetetraacetic acid disodium salt.	[260]
An osmotic MBR was used for the removal of antibiotics from municipal wastewater. The TrOCs were present in raw water at the 4.1–716.9 ng/L concentration range, while the chemical oxygen demand was 459.8 mg/L.	Oxacillin (55%), cefalexin (25%), amoxicillin (35%), ampicillin (50%), ciprofloxacin (18%), lomefloxacin (21%), norfloxacin (30%), enrofloxacin (30%), ofloxacin (18%), azithromycin (75%), erythromycin (85%), roxithromycin (70%), clarithromycin (70%), oxytetracycline (25%), tetracycline (18%), chlortetracycline (30%), sulfadiazine (42%), sulfathiazole (30%), sulfamethazine (15%), and trimethoprim (30%).	The CTA FO membrane achieved greater than 90% rejection of the antibiotics except for oxacillin (88%), amoxicillin (83%), ampicillin (85%), chlortetracycline (82%), sulfadiazine (75%), and sulfathiazole (80%).	[33]
The removal of cytostatic drugs using anaerobic OMBR was investigated using synthetic domestic water with total organic carbon, ammonia nitrogen, total nitrogen, and total phosphorus concentrations of 127.5 ± 12.7, 15.3 ± 1.0, 40.3 ± 1.2 and 5.3 ± 0.5 mg/L, respectively. The feed was spiked with 8 TrOCs on days 84 (100 ng/L) and 87 (100 μg/L).	Removal by adsorption was 50% for cyclophosphamide, 100% for azathioprine, 0% for doxorubicin, 100% for epirubicin, 100% for flutamide, 100% for mitotane, and 100% for tamoxifen. Removal by biodegradation was 98% for methotrexate, 70% for mitotane, 16% for azathioprine and 20% for flutamide.	The rejection of the TrOCs by the polyamide thin film composite FO membrane ranged from 99–100% for all compounds except for azathioprine (98%) and cyclophosphamide (96%).	[261]
An osmotic anaerobic MBR was coupled to membrane distillation for the removal of TrOCs in domestic sewage. The feed was spiked with 7 TrOCs at a concentration of 2 μg/L per compound. The bioreactor was monitored for 54 days.	17α-Ethynyl-estradiol, ketoprofen, betamethasone, fenofibrate, fluconazole, loratadine and prednisone	The PTFE FO membrane removed greater than 99.54% betamethasone, 99.05% ketoprofen, 95.77% 17α-ethinylestradiol, 100% fenofibrate, 95.88% fluconazole, more than 99.91% loratadine and over 98.51% prednisone after 47 days of operation.	[262]
An osmotic MBR was used to treat municipal wastewater targeting the removal of 20 TrOCs over 35 days. The TrOCs were detected in the raw municipal wastewater.	Sucralose (15%), fluoxetine (55%), tris(2-chloroethyl)phosphate (58%), sulfamethoxazole (63%), tris(1,3-dichloro isopropyl)phosphate (65%), tris(1-chloro-2-propyl) phosphate (70%), bisphenol A (70%), diclofenac (70%), diphenhydramine (90%), N, N-diethyl-meta- toluamide (95%), atenolol (96%), acesulfame (98%), naproxen (98%), trimethoprim (98%), ibuprofen (98%), triclocarban (99%), oxybenzone (99%), caffeine (99%), propylparaben (99%), and acetaminophen (99%).	The FO process (CTA membrane) achieved 98–100% removal of all TrOCs except for acetaminophen (70%), acesulfame (88%), N, N-diethyl-meta- toluamide (28%) and bisphenol A (43%).	[113]

Table 5 | Removal of Trace Organic Compounds (TrOCs) from wastewater by reverse osmosis membrane bioreactors (RO-MBR)
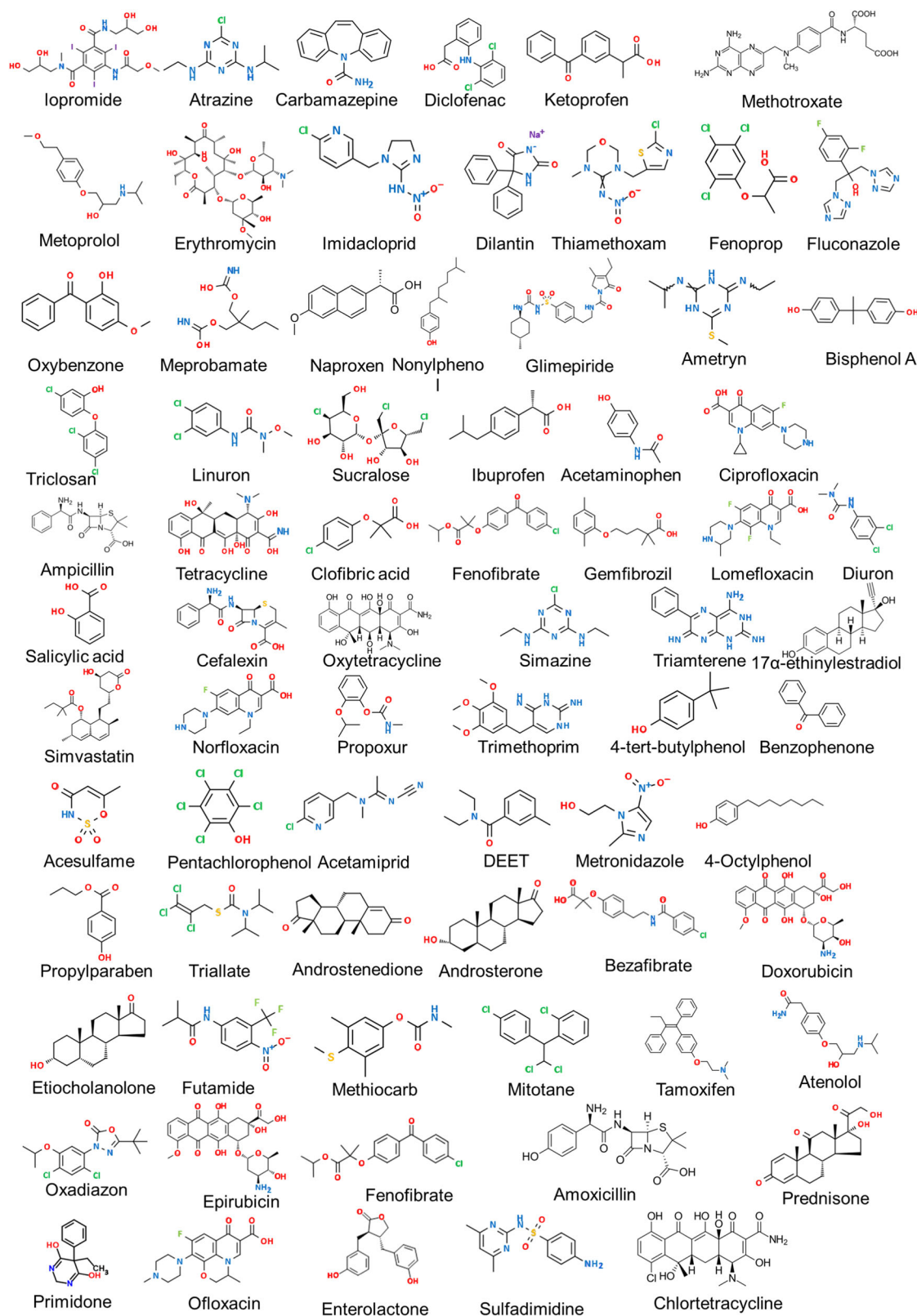
Description and feed type	TrOCs (Biological removal rates)	Membrane removal rates	Ref
A combined membrane bioreactor and reverse osmosis (MBR–RO) system was assessed for its efficacy in treating municipal wastewater. The removal of trihalomethanes was monitored. Total trihalomethanes in the MBR effluent were quantified at 182–689 mg/L.	Trichloromethane, bromodichloromethane, chlorodibromomethane, tribromomethane, monochloroacetic acid, monobromoacetic acid, dichloroacetic acid, trichloroacetic acid, bromochloroacetic acid, and dibromoacetic acid	High physical removal of trihalomethanes (over 83%) by the RO membranes (X20 and LFC1) treating MBR effluent was achieved.	[19]
A reverse osmosis process was used to treat MBR effluent targeting the removal of 12 TrOCs detected in the MBR feed (municipal wastewater).	Diclofenac (8%), erythromycin (−44%), clarithromycin (74%), azithromycin (53%), methiocarb (100%), imidacloprid (−18%), thiamethoxam (1%), clothianidin (88%), acetamiprid (39%), oxadiazon (33%) and triallate (99%).	All TrOCs in the MBR effluent were removed by the XLE membrane achieving greater than 99–100% removal.	[3]
An integrated MBR-RO system was used for the removal of 20 TrOCs in municipal wastewater. The organic compounds were present in the feed to the highest concentration of 2.90 μg/L.	Codeine (88%), hydrocodone (92%), carbamazepine (50%), diazepam (66%), lorazepam (45%), famotidine (83%), ranitidine (90%), azithromycin (75%), clarithromycin (83%), erythromycin (78%), sulfamethoxazole (65%), ofloxacin (0%), metronidazole (95%), atenolol (85%), metoprolol (67%), nadolol (88%), propranolol (60%), sotalol (65%), salbutamol (75%), and clopidogrel (65%).	Integrating the RO process (crosslinked aromatic polyamide RO membrane) improved the rejection of the TrOCs and achieved complete retention for all organics.	[8]
Reverse osmosis membranes were used to treat MBR effluent targeting the removal of TrOCs. The MBR was fed with synthetic wastewater spiked with 40 TrOCs at an initial concentration of 2 μg/L per compound. The simulated wastewater constituted of glucose (400 mg/L), peptone (75 mg/L), KH$_2$PO$_4$ (17.5 mg/L), MgSO$_4$ (17.5 mg/L), FeSO$_4$ (10 mg/L), and sodium acetate (225 mg/L).	Paracetamol (90%), N, N-Diethyl-meta-toluamide (5%), caffeine (50%), ibuprofen (93%), t-octylphenol (90%), atrazine (3%), meprobamate (10%), primidone (5%), nonylphenol (10%), bisphenol-A (99%), naproxen (35%), carbamazepine (10%), linuron (15%), gemfibrozil (20%), dilantin (0%), triamterene (25%), sulfamethoxazole (90%), ketoprofen (65%), atenolol (95%), estrone (98%), 17β-estradiol (100%), amitriptyline (98%), androstenedione (100%), estriol (100%), testosterone (100%), triclosan (90%), trimethoprim (20%), etiocholanolone (100%), androsterone (100%), diclofenac (20%), 17α-ethynylestradiol (90%), triclocarban (98%), clozapine (70%), omeprazole (60%), hydroxyzine (90%), enalapril (92%), risperidone (90%), simvastatin (95%), simvastatin-hydroxy acid (50%) and verapamil (70%).	The BW30 RO membrane achieved 99–100% removal of all compounds except for DEET (96%), caffeine (98%), meprobamate (98%), linuron (97%), carbamazepine (96%), triamterene (96%), trimethoprim (92%) and diclofenac (90%). On the other hand, the ESPA2 RO membrane achieved complete removal of all compounds except for DEET (93%), t-octylphenol (97%), bisphenol A (97%), linuron (95%), trimethoprim (98%) and diclofenac (90%).	[253]
A reverse osmosis process was used to treat the effluent of OMBR. The FO process treated synthetic wastewater from a bioreactor. The feed was spiked with 31 TrOCs (5 μg/L per compound) to simulate medium-strength municipal sewage.	Clofibric acid (30%), salicylic acid (95%), ketoprofen (98%), fenoprop (33%), naproxen (98%), ibuprofen (85%), primidone (62%), diclofenac (18%), gemfibrozil (65%), propoxur (83%), carbamazepine (5%), pentachlorophenol (78%), estriol (98%), atrazine (25%), ametryn (85%), benzophenone (78%), amitriptyline (98%), 4-Tert-butyphenol (95%), oxybenzone (99%), estrone (98%), 17α-ethinylestradiol (97%), 17β-estradiol (99%), triclosan (50%), 17β-estrodiol-17-acetate (99%) and octocrylene (90%).	The RO (ESPA2 membrane) treating FO draw achieved the following removals: 99–100% removal from sodium chloride, 98–100% removal from sodium acetate and 97–100% from ethylenediaminetetraacetic acid disodium salt.	[12]
A membrane bioreactor coupled to reverse osmosis was evaluated for the removal of estrogens (E1 to E3). The bioreactor was fed with secondary effluent wastewater with 391 mg/L chemical oxygen demand and total estrogens concentration of up to 182 ug/L. The study was conducted in three phases where different membranes were used.	Estrogens (E1 to E3)	In the first phase, 87.7% removal of estrogens was achieved by the LFC1 membrane. This improved to 88.5% in the second phase, which used the same membrane. In the third phase, estrogens were not detected in the RO permeate, showing complete removal by the ESPA1 membrane.	[182]
A reverse osmosis system was used to reconcentrate the draw solution of FO treating municipal wastewater. The removal of 20 TrOCs detected in the FO draw was investigated in the process.	Sucralose (15%), fluoxetine (55%), tris(2-chloroethyl) phosphate (58%), sulfamethoxazole (63%), tris(1,3-dichloro isopropyl)phosphate (65%), tris(1-chloro-2-propyl) phosphate (70%), bisphenol A (70%), diclofenac (70%), diphenhydramine (90%), N, N-diethyl-meta- toluamide (95%), atenolol (96%), acesulfame (98%), naproxen (98%), trimethoprim (98%), ibuprofen (98%), triclocarban (99%), oxybenzone (99%), caffeine (99%), propylparaben (99%), and acetaminophen (99%).	The RO membrane (SW30) removed 100% TrOCs except for N, N-diethyl-meta-toluamide (75%), bisphenol A (65%) and tris(2-chloroethyl)phosphate (95%).	[113]

Fouling greatly reduced the removal of nonionic hydrophilic compounds (caffeine (65–50%), sulfamethoxazole (90–30%) and trimethoprim (98–46%)) due to cake-enhanced concentration polarization which was promoted by the formation of porous fouling layers[112].

In another study investigating the removal of TrOCs using a unique ultrafiltration-osmotic MBR, the removal of nonionic and hydrophobic TrOCs by FO membrane was lower compared to the charged compounds.

Contrary, the RO membrane rejected almost all nonionic compounds, while hydrophobic nonionic compounds were poorly rejected[113]. The removal of positively charged and nonionic TrOCs by fouled membranes has been reported to be higher than that of the virgin membrane and this has been ascribed to the fouling layer (organic, colloidal and biological) preventing partitioning of the compounds through the membrane (pore blocking) and adsorption of TrOCs onto the organic fouling layer. Therefore, the presence

Fig. 3 | Removal of trace organic compounds by HR-MBRs. Summary of trace organic compounds (TrOCs) removal by biological and membrane processes.

of foulants in the feed and on the membrane surface can enhance the removal of TrOCs by providing additional sites for adsorption thereby prolonging their time for biodegradation.

In this work, the effect of fouling on TrOCs removal is centered on the membrane filtration step because foulants from the biological process accumulate on the membrane and change the membrane surface properties. However, it is acknowledged that biological activity is affected by the presence of foulants, and the performance of the biological process will have a role in the removal of TrOCs by membranes. For instance, poor degradation of TrOCs in the bioreactor will imply high TrOCs concentration in the feed of

Fig. 4 | Trace organic compounds. Chemical structures of TrOCs investigated in this work (extracted from chemspider.com).

high-retention membranes, and this can lead to poor removal due to concentration polarization. The role of the concentration of TrOCs in the feed on their removal by high-retention membranes has been investigated, and both improvements and decline in rejection have been reported for different membranes[114], while some researchers did not observe any correlation between the retention of TrOCs and their concentration in the feed[115]. Reduction in TrOCs removal at high concentrations has been ascribed to charge shielding effects, a concept whereby elevated TrOCs concentration increases adsorption on the membrane surface and raises the TrOCs concentration in the electric double layer. This shields the membrane's surface
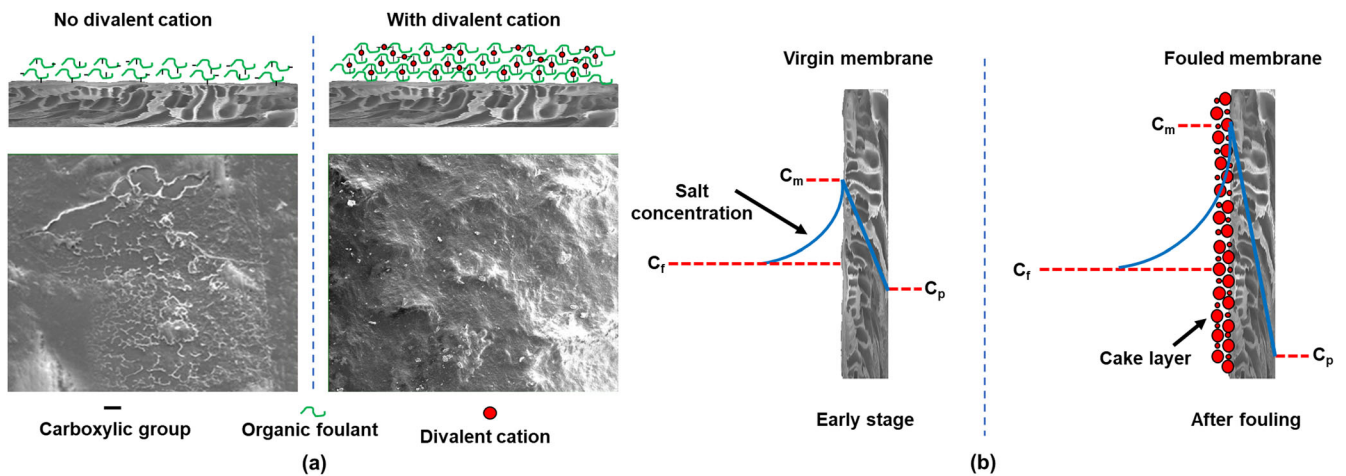
Fig. 5 | Organic fouling of membranes. a Organic fouling in the presence and absence of divalent cations and **b** salt concentration polarization profile in virgin and fouled membranes, adapted from Ju and Hong[79]. C_f is the concentration in the feed, C_m is the concentration on the membrane surface, and C_p is the concentration in the permeate.
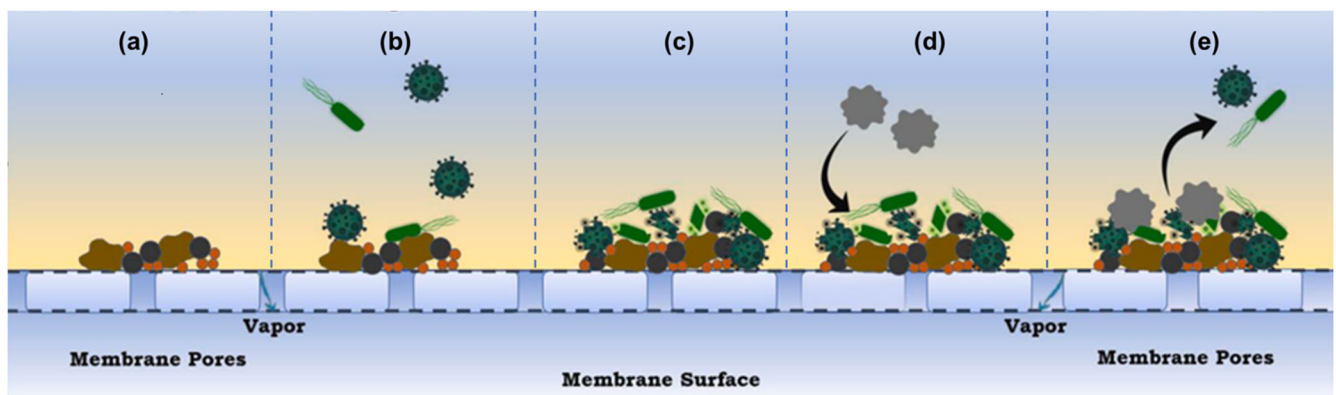
Fig. 6 | Biofouling in membrane distillation. Stages of biofilm formation on membrane surfaces during waste and wastewater treatment by MD[338]: **a** formation of conditioning film, **b** attachment of microorganisms, **c** excretion of extracellular polymeric substances (EPS), **d** biofilm formation, and **e** detachment of microorganisms. Similar fouling mechanisms occur in FO, NF, and RO processes.

Fig. 7 | Membrane fouling stages. Membrane fouling and fouling models showing different stages of organic, colloidal, inorganic (scaling) and biofouling.
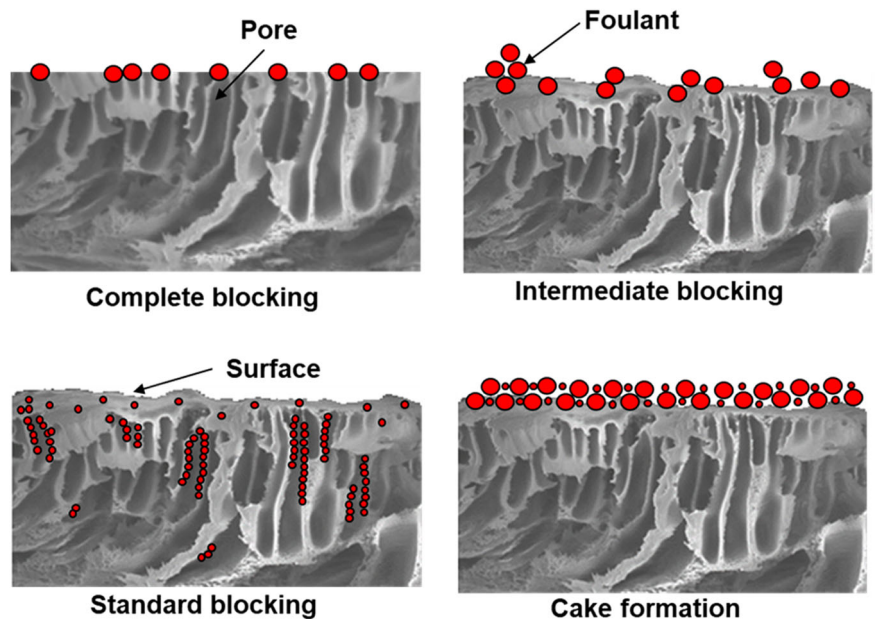
Table 6 | Major fouling and flux decline mechanisms for the different fouling types

Fouling type	Major fouling and flux decline mechanisms
Organic fouling	○ Cake build-up leading to an increase in hydraulic resistance[21,40] ○ Calcium complexation leads to the formation of tight organic fouling layers[82] ○ Cake-enhanced concentration polarization decreases driving force[21,40] ○ Pore blocking restricts water passage[263–267]
Colloidal fouling and scaling	○ Cake-enhanced concentration polarization effects that increase osmotic pressure gradient across the membrane interface[40,41] ○ Increase in hydraulic resistance due to the formation of compressed fouling layers[263] ○ Surface crystallization or scaling that exacerbates concentration polarization[41,50]
Biofouling	○ Biofilm growth on the membrane surface leads to an increase in biofilm hydraulic resistance[39,40] ○ Enhanced concentration polarization near the membrane surface resulting in biofilm-enhanced osmotic pressure[268]
Combined fouling	○ Synergistic effects from the different foulants[82,269,270]

Table 7 | Membrane fouling studies during wastewater treatment by membrane distillation bioreactors (MDBR)

Feed type	Membrane	Impact of membrane flux[a]	Ref.
Synthetic wastewater with 0.67 g/L chemical oxygen demand and 0.04 g/L total nitrogen. The feed was prepared from 4.27 g/L glucose, 0.85 g/L meat extract, 1.07 g/L peptone, 0.19 g/L KH_2PO_4, 0.19 g/L $MgSO_4$, 0.16 g/L $FeCl_3$ and 3.2 g/L CH_3COONa.	Hydrophobic polyvinylidene fluoride membrane	About 45% flux decline due to biofouling after 23 days of MDBR operation. The major fouling constituent was extracellular polymeric substances. Accelerated flux decline was ascribed to thermal and mass transfer resistance.	271
Synthetic wastewater was prepared from 1500 mg/L glucose, 20 mg/L ammonium sulfate, 10 mg/L potassium phosphate, and 50 mg/L urea, leading to the 1632 mg/L chemical oxygen demand of the synthetic wastewater.	Hydrophobic polyvinylidene fluoride membrane	16–50% flux decline after 7 days of operation in an anaerobic MDBR. Microbial by-products and colloidal particles were the major foulants. Crystallization of calcium chloride, magnesium sulfate, manganese chloride, zinc sulfate, ferric chloride, cupric sulfate, and cobalt chloride also occurred and contributed to fouling.	272
Synthetic wastewater is prepared from glucose, peptone, $NaHCO_3$, urea, KH_2PO_4, NH_4Cl and trace elements to simulate sewage.	Hydrophobic polytetrafluoroethylene membrane	Crystallization fouling by salts including $CaCO_3$, $MgCO_3$, $CaSO_4$, $Ca_3(PO_4)_2$ and $Mg_3(PO_4)_2$ occurred after 3 days of operation, leading to a 50% flux decline.	273
Synthetic wastewater simulating high-strength domestic wastewater. The feed comprised 4000 mg/L glucose, 750 mg/L peptone, 2250 mg/L sodium acetate, 175 mg/L potassium dihydrogen phosphate, 175 mg/L magnesium chloride, and 175 mg/L urea.	Membrane with a thin polytetrafluorethylene layer on top of a polypropylene support	Severe fouling after 30 days of operation due to complete retention of organic matter (such as humic-like and protein-like substances) and inorganic salts (calcium/ magnesium phosphate and struvite phosphate and struvite), which resulted in salinity build-up.	36
Synthetic wastewater composed of glucose (308.4 mg/L), ammonium chloride (114.6 mg/L), potassium dihydrogen orthophosphate (44.0 mg/L) and trace nutrients.	Hydrophobic polytetrafluoroethylene membrane	Severe fouling (80% flux decline) after 25 days of operation due to salt build-up, soluble microbial by-products, and extracellular polymeric substances.	195
MBR permeate spiked with four metal salts and 12 micropollutants to simulate acid mine drainage and sewage-impacted groundwater. The MBR feed was prepared from glucose, peptone, urea, monopotassium phosphate, magnesium sulfate, ferrous sulfate, and sodium acetate.	Hydrophobic polytetrafluoroethylene membrane	29–76% flux decline after 5 days of operation due to rejected metals and bulk organics, which resulted in scaling.	248
Flue gas desulfurization wastewater containing extremely high concentrations of heavy ions and unbiodegradable organic pollutants such as humic substances.	Hydrophobic polytetrafluoroethylene membrane	Sharp flux decline (greater than 50%) after 249 h of operation due to the crystallization of calcium, magnesium, silica, and inorganic aluminum salts. Pretreatment reduced fouling.	274

[a]Some flux decline rates were estimated from flux decline curves presented in the cited data using the following equation: $\triangle J(\%) = (1 - J_f/J_0) \times 100$; where $\triangle J$ is change in flux, J_f is flux at the end of filtration run and J_0 is flux at the beginning of filtration run.

charge and blocks membrane-solute repulsive interactions, lowering rejection[116]. To be more relevant to HR-MBRs with respect to feed water type, the removal of TrOCs from fouling studies investigated using wastewater and synthetic wastewater samples are referenced (Table 11).

Strategies to mitigate fouling in HR-MBRs
Several fouling mitigation strategies have been adopted, with some focusing on fouling prevention, whereas others are applied as corrective measures to restore membrane flux after the membrane has fouled. The following fouling alleviation approaches have been reported: pre-treatment of the feed, membrane cleaning, membrane surface modification to make membrane surfaces resistant, and optimization of operating conditions to delay fouling.

Pre-treatment of feed as mitigation for fouling. In conventional MBR, pre-treatment is performed to lower the levels of total suspended solids. However, pre-treatment is limited to sedimentation or chemical dosage of coagulants, flocculants, or adsorbents, implying that similar pre-treatment approaches would be used in HR-MBR. Chemical pre-treatment involves using coagulants or flocculants such as ferric chloride ($FeCl_3$), aluminum sulfate (alum), or adsorbent agents such as activated carbon. At the municipal scale, sedimentation is normally followed by other pre-treatment approaches such as: i) dissolved air flotation (DAF) then dual media filter (DMF); ii) DMF followed by cartridge filter or iii) DAF followed by sand filtration then cartridge filter[117]. Regardless of the processes used, coagulation is done before the influent enters the first unit of the treatment process. Subsequently, anti-scalants are added to prevent membrane fouling by minimizing the deposition of scale forming ions on the membrane[118]. Combining sedimentation with DAF is recommended to obtain desirable water quality especially when the turbidity of the influent increases unexpectedly. Similarly, DMF is preferred over single media filter because it can produce water of high quality. These pre-treatment approaches (which are effective in wastewater treatment at

Table 8 | Membrane fouling studies during wastewater treatment by nanofiltration membrane bioreactors (NF-MBR)

Feed type	Membrane	Impact of membrane flux	Ref.
Real wastewater treated by MBR at a wastewater treatment plant was used as feed for NF.	NF membrane (NF90)	50% flux decline after 6 cycles of fouling by inorganic foulants such as salts and divalent cations. Each cycle lasted for 24 h.	[121]
MBR effluent where the MBR was directly fed from the hospital's sewer system.	Polyamide composite NF membrane (NE 70)	Colloidal fouling occurred at a pH range of 6.05–6.80, and calcium phosphate scaling at a pH range of 7.20–8.27, leading to over 70% flux decline after 8 h of filtration. More fouling was observed at high transmembrane pressure.	[275]
Municipal wastewater from a sewer system.	NF membrane (NF270)	Fouling on the NF270 membrane by inorganic and organic foulants (humic acids) was observed after 1 year of operation.	[276]
Real wastewater treated by MBR and used as feed for NF.	NF membranes (NF270 and NF90)	Sodium polyacrylate and magnesium sulfate led to over 60% flux decline after 40 days of operation due to reversible fouling of the NF membranes.	[277]
MBR-treated wastewater discharged from the treatment plant was used as feed for NF.	NF membrane (NF90)	A slight decrease in permeate flux (less than 10%) after 180 min of filtration due to organic and inorganic fouling when the concentrated amount increased in the feed mixture.	[278]
MBR effluent (treated wastewater) with high salt content was used as feed for NF.	NF membrane	The presence of salts of calcium, magnesium, and silica promoted inorganic scaling.	[279]
Pre-treated leachate from a sanitary landfill with 23 mS/cm conductivity was used as feed for NF.	Polyamide thin film composite NF membrane (NF90)	Up to 70.83% flux decline was recorded after 2 h of filtration due to fouling by inorganic substances containing calcium and magnesium.	[280]
Real wastewater from a pharmaceutical company was pre-treated by MBR and granular activated carbon.	NF membranes (DK and NF90)	About 60% of flux declines after 45 h of filtration due to fouling by soluble microbial by-products, aromatic protein-like substances, and humic acid.	[281]
The effluent of an anaerobic process from a wastewater treatment plant of a pharmaceutical company that produces spiramycin. The feed had chemical oxygen demand of 1460 mg/L.	NF membrane (Desal 5 DK 1812)	Over 30% flux declined after 65 days of operation due to fouling by extracellular polymeric substances and soluble microbial by-products, which increased due to salinity build-up.	[187]
Municipal wastewater which was sieved through a 1 mm opening mesh.	Glutaraldehyde crosslinked layer-by-layer polyelectrolyte NF membrane	Severe fouling of the NF membrane by extracellular polymeric substances, organics (biopolymers) and inorganic salts, including calcium phosphate leading to a sharp increase in transmembrane pressure within 50 days of operation.	[202]
Effluent from MBR treating municipal wastewater with total organic carbon of 4.2 mg/L and electrical conductivity of 355 µS/cm.	NF membrane (NTR-729HG)	Remarkable membrane fouling by combined silica and organic matter resulted in a 50% pressure drop after 60 days of operation.	[282]
Laundry wastewater with chemical oxygen demand of 2800 mg/L and electrical conductivity of 1050 µS/cm. The feed was collected directly from the sewage discharge point.	NF membrane (Desal 5 DL)	Up to 41% flux decline in 140 min of filtration due to scaling by salts, including calcium carbonate.	[283]
Synthetic municipal wastewater prepared from 250 mg/L glucose, 250 mg/L starch, 134.1 mg/L NH_4Cl, 30.7 mg/L KH_2PO_4, 200 mg/L $CaCl_2$, 10.2 mg/L $MgSO_4$, 189 mg/L NaCl, 75 mg/L $NaHCO_3$ and 1 mg/L trace elements.	Novel polyamine NF membrane	Fouling of the NF membrane by humic substances, polysaccharides, and proteins increased transmembrane pressure from 12 kPa to 30 kPa within 120 days of operation.	[284]
The effluent of primary sedimentation of municipal wastewater was treated through MBR and used as feed for NF.	NF membranes (NE70 and NE90)	79% decrease in flux for NE70 and 89% flux decline for NE90 after 96 h of operation due to fouling by polysaccharide-like substances.	[285]
Industrial zone wastewater (total organic carbon of 28.8 mg/L and chemical oxygen demand of 25.4 mg/L) was treated through MBR and used as feed for NF.	NF membrane (NF90)	A minor decrease in permeate flux over 1 year of operation due to the scaling of heavy metal salts.	[286]

municipal scale) can be integrated in HR-MBR to mitigate fouling challenges. Furthermore, lessons learned during conventional wastewater treatment can be applied in HR-MBR for process improvement.

Due to the excessive fouling propensity of high-retention membranes, sedimentation and chemical treatment may not be sufficient, and therefore other pre-treatment options, such as MF or UF membrane, may be necessary. Different studies, including the works of Uzal et al.[119], have shown that MF and UF membranes can remove macromolecules that can potentially foul high-retention membranes, thus reducing membrane fouling[120]. Nonetheless, due to their large pore sizes, MF and UF membranes do not remove all fouling agents from feed solutions[121]. This implies that fouling may still occur even with MF/UF pre-treatment. Further, adding MF/UF as a pre-treatment step increases the capital and operational cost of HR-MBR, but the reduction in membrane cleaning and longer membrane lifespan can compensate for this cost. Nonetheless, several UF membranes have been successfully installed and integrated in full-scale water treatment plants since the mid-2000s in various countries such as Singapore, Spain, China,

Saudi Arabia and United Arab Emirates[122]. Therefore, UF membranes could be used as pre-treatment in HR-MBR to mitigate fouling.

Advanced oxidation processes (e.g., pre-ozonation[123] and UV-oxidation[124]) may also be used as pre-treatment for HR-MBR because advanced oxidation processes can break down organic macro- and micropollutants. However, such processes are not only high-cost but also deactivate the microorganisms responsible for the biodegradation of TrOCs in the reactor. Pramanik et al.[125] found that pre-treatment of anaerobically treated dairy effluent with ultraviolet/persulfate oxidation reduced fouling of FO membranes by decreasing reversible and irreversible fouling, resulting in over 95% flux recovery after three cycles. The use of persulfate as fouling mitigation has also been demonstrated by Asif et al.[11], who illustrated a reduced fouling layer on the membrane surface in the persulfate-assisted direct contact membrane distillation system used to treat secondary effluent spiked with a mixture of TrOCs. Persulfate remarkably degraded total organic carbon (70%) and total nitrogen (40%), leading to membrane fouling during wastewater treatment by HR-MBR. Although advanced

Table 9 | Membrane fouling studies during wastewater treatment by osmotic membrane bioreactors (OMBR)

Feed type	Membrane	Impact of membrane flux	Ref.
Synthetic wastewater prepared to obtain 100 mg/L glucose, 100 mg/L peptone, 17.5 mg/L KH$_2$PO$_4$, 17.5 mg/L MgSO$_4$, 10 mg/L FeSO$_4$, 225 mg/L CH$_3$COONa, and 35 mg/L urea and spiked with 31 TrOCs at 5 μg/L per organic.	Thin film composite FO membrane	Fouling of the FO membrane by soluble microbial products and extracellular polymeric by-products increased due to salinity build-up, leading to over 60% flux decline after 40 days of operation.	[126]
Synthetic textile wastewater with chemical oxygen demand of 3000 ± 150 mg/L and a color of 1000 ± 100 Platinum Cobalt (Pt. Co.) with chemical oxygen demand: nitrogen: phosphorus ratio of 100:10:1.	Hydrophilic thin film composite membrane with a top poly-amide layer	Over 80% flux decline after 50 days of operation due to biofouling and internal concentration polarization of salts.	[287]
Raw municipal wastewater with chemical oxygen demand, total organic carbon, ammonia nitrogen, total nitrogen, and phosphate of 150–280, 32–98.2, 35–60, 38–70 and 1–4 mg/L, respectively. Salt concentrations of Na$^+$, K$^+$, Ca^{2+} and Mg^{2+} were 50–70, 18–21, 25–30 and 4–5 mg/L, respectively.	Cellulose triacetate FO membrane	Less than 20% flux decline due to inorganic calcium and magnesium salts scaling after 70 days of operation.	[288]
Synthetic wastewater with 439.47 mg/L chemical oxygen demand, 60.23 mg/L total nitrogen and 9.42 mg/L phosphorus.	Cellulose triacetate FO membrane	There was a 30% flux decline after 24 h of operation due to fouling by natural organic matter.	[289]
Real wastewater collected after primary sedimentation in a water recycling plant and sludge centrate from digested sludge dewatering centrifuge.	Cellulose triacetate FO membrane	About 40% flux decline from wastewater after primary sedimentation and over 80% flux decline from sludge centrate after 12 h of operation. Fouling was due to the bulk crystallization of minerals and the deposition of particulate matter on the membrane surface.	[127]
Synthetic wastewater simulating high-strength domestic wastewater. The feed was prepared from sucrose and ammonium bicarbonate to achieve a strength of 1350 mg/L chemical oxygen demand and 160 mg/L ammonium nitrogen.	Cellulose triacetate FO membrane and thin film composite membranes	Over 80% flux decline for cellulose triacetate and thin film composite membranes after 20 h due to inorganic fouling and salinity build-up.	[290]
Simulated inorganic leachate prepared from Na$_2$SO$_4$, CaCl$_2$, MgCl$_2$ and NaCl in water at concentrations of 73.9 mM, 27 mM, 18.5 mM, and 308 mM, respectively.	Aquaporin flat sheet thin film composite FO membrane	Severe scaling of calcium sulfate led to over 80% flux decline after 24 h of filtration.	[291]
Raw municipal wastewater collected from an MBR plant.	Thin film composite FO membrane	Fouling of the membrane by polysaccharides, humic-like substances, and proteins led to about 25% flux decline after 42 days of operation.	[292]
Synthetic wastewater simulating medium-strength domestic wastewater (soluble chemical oxygen demand of 500 mg/L) with a chemical oxygen demand: nitrogen: phosphorus ratio of 100:5:0.7.	Thin film composite FO membrane	Fouling by soluble microbial by-products and minerals/salts resulted in a 45–70% flux decline due to fouling by soluble microbial by-products and metals after 30 days of filtration.	[293]
Synthetic wastewater that had 149 ± 5 mg/L total organic carbon, 26.8 ± 0.8 mg/L total nitrogen (TN), 11.1 ± 0.4 mg/L ammonium (NH$_4$–N), 4.7 ± 0.1 mg/L total phosphorus (TP), and conductivity of 416 ± 48 μS/cm.	Thin film composite FO membrane	Irreversible membrane fouling by hydrophilic foulants comprising proteins and polysaccharides led to over 80% flux decline within 10 days of filtration.	[294]
Simulated sewage consisting of 300 mg/L glucose, 50 mg/L yeast, 15 mg/L KH$_2$PO$_4$, 10 mg/L FeSO4, 60 mg/L (NH$_4$)$_2$SO$_4$, and 30 mg/L urea.	Polyamide thin film composite FO membrane	About 36% flux decline after 7 days of filtration due to salinity build-up, which increased the concentration of extracellular polymeric substances and soluble microbial by-products.	[295]
Wastewater with tetramethylammonium hydroxide concentration of 100 ± 10 mg/L, pH around 11, suspended solids less than 10 mg/L, 500 ± 50 mg/L chemical oxygen demand, and ammonium nitrogen of 2.0–15.0 mg/L. 148 mg/L NH$_4$Cl, 43 mg/L KH$_2$PO$_4$, and 100 mg/L NaHCO$_3$ were added to maintain nutrients and alkalinity.	Thin film composite FO membranes	About 40% flux decline after 10 days of filtration due to fouling by humic-like substances, fulvic acid-like substances, aromatic protein, soluble microbial by-products, and extracellular polymeric substances.	[239]
Synthetic wastewater simulating domestic wastewater constituting of chemical oxygen demand, total organic carbon, total phosphorus, ammonium nitrogen and total nitrogen concentrations of 372.6 ± 7.19, 123.97 ± 2.23, 2.97 ± 0.14, 28.26 ± 0.51 and 42.42 ± 1.19 mg/L, respectively.	Cellulose triacetate membrane	30–50% flux decline after 30 days of operation due to inorganic scaling and biofouling of the membrane.	[296]
Raw sewage with conductivity, pH, and total organic carbon concentration of 1130 ± 53 μS/cm, 7.3 ± 0.2, and 28 ± 5 mg/L, respectively.	Cellulose triacetate FO membrane	36% flux decline after 10 days of filtration due to fouling by proteins, polysaccharides, calcium, aluminum, and iron salts. Periodic microfiltration extraction reduced salinity build-up.	[207]
Secondary wastewater effluent pre-filtered with a microfiltration cartridge filter with a 0.45 μm pore size.	Cellulose triacetate membrane	Over 40% flux decline after 36 days of filtration due to fouling by organic foulants (hydrophilic aromatic proteins, humic- or fulvic-like organics, and soluble microbial by-products).	[297]
Pre-filtered secondary wastewater effluent with biochemical oxygen demand of 5.0 ± 0.6 mg/L, a chemical oxygen demand of 7.5 ± 0.3 mg/L, suspended solids of 0.53 ± 0.12 mg/L, total nitrogen of 8.5 ± 0.9 mg/L, total phosphate of 0.5 ± 0.1 mg/L, and pH of 6.5 ± 0.1.	Cellulose triacetate SWFO membrane	Greater than 55% flux decline after 36 days of filtration due to membrane fouling by hydrophilic and large molecular weight organic matter.	[298]
Refinery wastewater with 36 ± 4 mg/L ammonium, 336 ± 190 mg/L chemical oxygen demand, 75 ± 21 mg/L dissolved organic carbon, 2.3 ± 0.6 mS/ cm electrical conductivity, 30 ± 225 mg/L oils and greases, 17 ± 4 mg/L phenol, total phosphorus 4.2 ± 1.0 mg/L and 54 ± 35 NTU turbidity.	FO membranes manufactured by Hydration Technologies Inc.	Precipitation of inorganic ions and fouling by soluble microbial by-products and extracellular polymeric substances led to about 80% flux decline after 100 days of operation.	[299]

oxidation processes and other advanced pre-treatment methods are used for dense membrane processes, their practical application in MBR may not be feasible[17], thus most of the processes have only been demonstrated at laboratory scale. Figure 9 shows the classification of the reviewed pre-treatment methods based on the cost, efficiency, and associated challenges.

From the image, AOPs have been classified as costly, and efficient but have high challenges which are linked to the formation of disinfection by-products and destruction of the microorganisms in the biological process. Due to the low fouling propensity and energy efficiency of FO processes, fouling of OMBR coupled with RO has been investigated, where the

Table 10 | Membrane fouling studies during wastewater treatment by reverse osmosis membrane bioreactors (RO-MBR)

Feed type	Membrane	Impact of membrane flux	Ref.
Synthetic wastewater with 77 mg/L chemical oxygen demand, 1650 mg/L sulfate, 1230 mg/L sodium, 128 mg/L magnesium and 6083 mg/L total dissolved solids.	Polyamide thin film composite FO membranes	Fouling by humic acid and scaling of $CaSO_4$ and NaCl salt resulted in over 60% flux decline after 20 h of filtration. Internal concentration polarization also reduced membrane flux.	[300]
MBR effluent from a water reclamation plant with pH from 7.12–7.20, 4.2–5.6 mg/L dissolved organic carbon and 92–322 mg/L total dissolved solids.	Thin film composite RO membrane (BW-30)	43% flux decline in RO permeate after 5 days of filtering heated aluminum oxide particles treated effluent, and 62% flux decline on filtering powder activated carbon effluent. The membrane was fouled by biopolymers such as polysaccharides and proteins.	[301]
Industrial zone wastewater with chemical oxygen demand of 1500 mg/L, suspended solids of 650 mg/L and various inorganic salts. The feed was pre-treated in a moving bed bioreactor, dissolved air flotation unit, and chemical treatment unit prior to RO.	RO membrane (BW30XFR-400/34i)	Scaling of inorganic calcium, manganese, zinc, and molybdenum salts on the membrane surface as observed through membrane characterization after 2 years of operation.	[302]
Tertiary wastewater with a dissolved oxygen concentration of 9.1 mg/L and electrical conductivity of 703 µS/cm.	Thin film composite RO membrane (ESPA-1)	Over 45% flux decline after 500 h of filtration due to fouling by effluent organic matter and extracellular polymeric substances.	[303]
Effluent from primary clarifiers of a municipal wastewater plant pre-treated with MBR prior to RO filtration.	Thin film composite RO membrane	Up to 70% flux decline after 300 h of filtration due to fouling by inorganic and organic constituents.	[304]
Effluent from primary clarifiers of a municipal wastewater plant was pre-treated with MBR and had a dissolved oxygen concentration of 4.0 to 6.3 mg/L.	Polyamide thin film composite RO membrane	Fouling of the membrane by live and dead cells, proteins, and carbohydrate-like materials was observed using confocal laser scanning microscopy.	[305]
Municipal wastewater spiked with copper, lead, nickel, and zinc; and treated through MBR.	Polyamide thin film composite RO membrane	Reduction in water permeability after 150 days of filtration due to inorganic fouling, which was increased by the presence of heavy metals.	[306]
Municipal wastewater was pre-filtered through microfiltration and nanofiltration and used as feed for reverse osmosis.	RO membrane	55% flux decline after 14 days of filtration due to fouling by low molecular weight organic foulants, including alcohols, aldehydes, ketones, and amino acids.	[307]
Raw wastewater sewage after settling in a primary settling tank.	Polyamide-urea thin film composite hydrophilic RO membrane	The RO membrane was fouled by organic, colloidal, and inorganic substances.	[308]
Raw wastewater was pre-treated by MBR prior to filtration by RO. The MBR effluent had 27.6–48.6 mg/L chemical oxygen demand and electrical conductivity of 3.580–5.150 µS/cm.	RO membrane (XLE-4040)	Up to 60% flux decline after 200 days of filtration due to organic fouling, scaling, and biofouling.	[309]
Synthetic wastewater simulating municipal wastewater was prepared from the following: CH_3COONa (2.65 g/L), NH_4Cl (0.376 g/L), KH_2PO_4 (0.109 g/L), peptone (0.706 g/L), $FeCl_3.6H_2O$ (0.782 mg/L), $CaCl_2$ (1.56 mg/L), $MgSO_4$ (1.56 mg/L), KCl (1.56 mg/L) and NaCl (1.56 mg/L).	RO membrane (LFC3-LD-D2)	Fouling of RO membrane by microbes, extracellular polymeric substances, proteins and humic and fulvic acid-like substances as visualized through different characterization techniques.	[310]
Synthetic wastewater simulating municipal wastewater and constituted of sucrose (210 mg/L), meat extract (41.7 mg/L), peptone (60 mg/L), NH_4Cl (95.5 mg/L), KH_2PO_4 (22 mg/L), $CaCl_2.2H_2O$ (10 mg/L), $FeSO_4.7H_2O$ (10 mg/L), $MgSO_4.7H_2O$ (10 mg/L) and $NaHCO_3$ (400 mg/L).	RO membrane (ESPA2)	Membrane fouling by biofilms and organic substances resulted in cake layer build-up, leading to increased transmembrane pressure from 6 bar to 8 bar within 12 days of filtration.	[311]
Acid and alkaline, fluoride-containing, and ammonia-containing wastewater was treated through various methods before the MBR process. The MBR effluent was used as feed for RO.	RO membrane (LFC3-LD)	The RO membrane was fouled by inorganics with plate-like morphology and microbial contaminants. The major constituents of foulants were barium sulfate, with lesser amounts of strontium sulfate, and deposits involving calcium, silica, aluminum, magnesium, iron, and zinc.	[312]
Fruit wastewater was treated through MBR. The feed had the following characteristics: biological oxygen demand of 40–90 mg/L, the chemical oxygen demand of 60–110 mg/L, protein of about 1–15 mg/L, humic substances of 40–70 mg/L and carbohydrates of 30–55 mg/L.	RO membrane (BW30XLE)	There was a 58% flux decline within 40 h of filtration due to fouling by soluble microbial products, specifically dissolved organic matter.	[313]

FO served as pre-treatment. The coupled processes are robust, resist fouling and perform better than RO-MBR without the FO process[126]. The inclusion of the FO step induces additional costs. However, the advantages of incorporating FO compensate for the additional capital and operational costs. Firstly, membrane cleaning is minimized, while membrane lifespan increases due to the reduction in fouling. Secondly, the coupled process can ensure simultaneous wastewater treatment and reconcentration of FO draw, thus maintaining a zero liquid discharge—a benefit for water-stressed countries. Treating diluted FO draw-streams with RO will reduce fouling due to concentration polarization and save the RO process energy because low-pressure NF/RO membranes can be used as the final treatment.

Cleaning to restore membrane flux and performance. Depending on the membrane process, foulant type and extent of fouling, membrane flux (and performance) can be restored by cleaning with deionized water. For instance, Liu et al.[93] used deionized water to clean a hydrophobic polytetrafluoroethylene (PTFE) membrane fouled by lithium chloride (LiCl) in direct contact membrane distillation. In another study, Asif et al.[2] restored UF and NF membrane fluxes by backwashing with permeate water. The membranes were reversibly fouled by synthetic wastewater containing TrOCs. To ensure a longer membrane lifespan and achieve high cumulative permeate volume while saving energy, periodic cleaning with water is advised than continuous membrane application until substantial fouling is observed[127].

Chemical cleaning may be required for compact fouling layers. Membranes fouled by inorganic foulants are cleaned with acidic solutions, while alkaline solutions are used to clean membranes fouled by organic foulants. Chemical cleaning still needs to be optimized to reduce operational
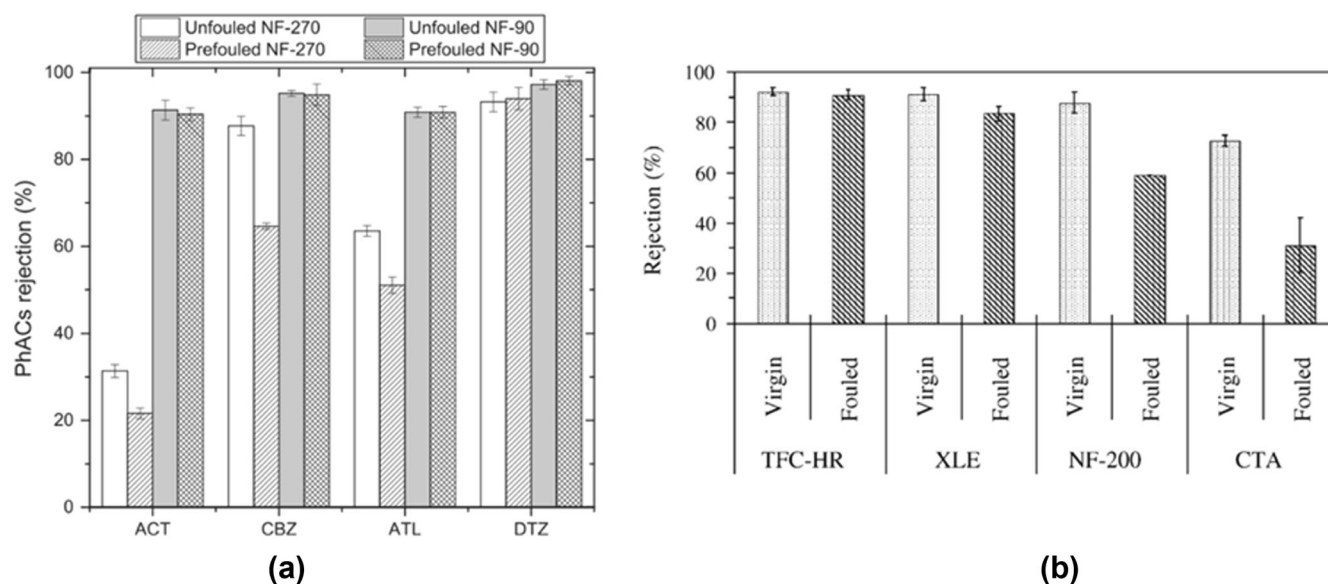
Fig. 8 | Removal of trace organic compounds by NF and RO membranes. Influence of fouling on the rejection of TrOCs by high-retention membranes: **a** TrOCs removal by NF270 and NF90 membranes before and after fouling with effluent organic matter (ACT acetaminophen, CBZ carbamazepine, ATL atenolol and DTZ diatrizoate)[161]; **b** Rejection of primidone by different membranes before and after fouling by effluent organic matter[316]; error bars present standard deviations.

costs. Underdosing chemicals and using ineffective chemicals decreases membrane lifespan and increases membrane replacement costs. Similarly, overdosing on chemicals may result in rapid membrane deterioration because most membrane materials are intolerant of high concentrations of cleaning agents[128]. The cleaning strategies, which include the time for flushing and backwashing, frequency of backwashing, the type and amount of chemicals and chemical cleaning, may depend on the chemistry of the wastewater. Therefore, constant monitoring of the wastewater characteristics becomes key so that the cleaning strategies may be modified accordingly. Observations can also be done by profiling the autopsy of a fouled membrane to determine the key foulants.

There are reports of chemical cleaning changing the physico-chemical properties of polymeric membranes and their separation efficiency. Simon et al.[129] investigated the influence of cleaning nanofiltration (NF270) membrane with citric acid, sodium hydroxide, ethylenediaminetetraacetic acid and sodium dodecyl sulfate. Caustic cleaning resulted in a remarkable increase in the membrane surface hydrophobicity and permeability due to the temporal enlargement of the membrane pores. Cleaning with citric acid and sodium dodecyl sulfate resulted in a slight increase in the rejection of carbamazepine, while the rejection of sulfamethoxazole remained unchanged. However, a decline in sulfamethoxazole rejection was noted at pH less than 8 following chemical cleaning. Nevertheless, this effect has been labeled temporary and could be restored by applying acidic cleaning immediately after caustic cleaning[130]. In another study, Zhou et al.[131] found that membrane cleaning with urea-based chemicals resulted in decrease in the rejection of neutral and negatively charged TrOCs by NF270 membrane. However, TrOCs removal by the NF90 membrane was not affected by chemical cleaning (Fig. 10). The effects of chemical cleaning are less significant for negative TrOCs because their rejection is predominantly governed by electrostatic repulsion between the compound and the negatively charged membrane surface. Therefore, their removal is not influenced by any enlargement of the membrane pores[132].

Membrane surface modification. Recent studies have focused on modifying membrane surface properties to make the membranes repellent to fouling while maintaining high fluxes and TrOCs retention properties[42,133–135]. Depending on the design requirements, the membranes can be more hydrophilic or hydrophobic (for MD purposes). One of the membrane modification techniques is the addition of nanoparticles, including graphene oxide (GO), graphitic boron, zinc oxide

(ZnO), titanium dioxide (TiO_2), copper oxide (CuO), silica dioxide (SiO_2), and many others. For example, Mahlangu et al.[42] modified UF polyethersulfone (PES) membranes with GO-ZnO and noted improvement in rejecting 23 TrOCs. Other reports on membrane modification to improve TrOCs removal are the works of Patala et al.[136], Ojajuni et al.[137] and Kong et al.[138], who demonstrated improvement in the rejection of TrOCs. Although the nanoengineered membranes perform better than the pristine membranes, the nanomaterials may leach from the polymer matrix. For example, the leaching of CuO[139] and silver nanoparticles[140–142] has been reported during filtration and membrane cleaning. Nanoparticle leaching may limit the application of nanoengineered membranes because nanoparticles such as Pb, Cd and Hg have been reported to inhibit the growth and enzyme production of white-rot fungus[143], while silver is well known for its antimicrobial activity. Tan et al.[144] reported that silver nanoparticles caused a remarkable decrease in nitrifying efficiency from 98% to 15% and increased the extracellular polymeric substance content in the activated sludge by 77.8%. Extracellular polymeric substances could promote membrane fouling and lead to more issues with membrane cleaning.

Further, the leaching of nanoparticles has been associated with a decrease in rejection of $MgSO_4$, while hydrophobicity, permeability, surface roughness and porosity increased[139]. This was in agreement with the findings of Simon et al.[129], who studied the influence of chemical cleaning on the physico-chemical properties of an NF270 membrane. Such changes in physico-chemical properties are expected to affect the removal of TrOCs during wastewater treatment.

There are other membrane modification techniques used to improve membrane properties, and they include cross-linking modification[145], tuning interlayer spacing[146], and graft polymerization[147]. Adding nanomaterials improves the membrane's structural and chemical properties resulting in thin-film nanocomposite membranes with enhanced properties compared to their respective pristine thin-film nanocomposite membranes[148]. In graft polymerization, polyethylene glycol is used as a hydrophilic filler to improve the membrane surface for fouling resistance. The main challenge with polyethylene glycol is its stability in complex media where polyethylene glycol degrades via auto-oxidation[149]. Therefore, it may be suggested that studies on graft polymerization utilize polymers with both positively and negatively charged functional groups to obtain membranes with a net neutral surface charge. Other emerging strategies to build multifunctional

Table 11 | Effects of fouling on the removal of TrOCs by high-retention membranes during wastewater treatment by HR-MBRs

Feed type	Trace organic compounds	Effects of fouling on TrOCs removal[a]	Ref.
Municipal wastewater effluent was pre-treated by MBR, and a tertiary treatment. The pre-treated feeds were spiked with 6 TrOCs targeting final concentrations of 100 ng/L per organic.	Clofibric acid, diclofenac, ketoprofen, mefenamic acid, carbamazepine, and primidone	Fouling by tertiary effluent improved the rejection of clofibric acid (75–78%), ketoprofen (55–83%), mefenamic acid (70–80%), diclofenac (60–80%) and primidone (60–70%) by the NF membrane (UTC-60), while the retention of carbamazepine declined slightly (55–53%). Fouling by MBR effluent decreased the rejection of clofibric acid (75–58%), mefenamic acid (70–55%), diclofenac (60–55%) and primidone (60–30%), carbamazepine (55–20%), while the removal of ketoprofen improved slightly (55–58%).	[314]
Secondary effluent wastewater spiked with 4 TrOCs targeting 750 µg/L per compound concentration.	Acetaminophen, atenolol, carbamazepine and diatrizoic acid	Fouling of the NF270 membrane by effluent organic matter resulted in a decrease in rejection of acetaminophen (30–20%), carbamazepine (90–65%) and atenolol (65–50%), while the removal of diatrizoic acid was not affected. On the other hand, fouling of the NF90 membrane did not affect the retention of the TrOCs.	[161]
Synthetic wastewater with a chemical oxygen demand of 640 mg/L and a chemical oxygen demand: nitrogen: phosphorus ratio of 100:10:1. Cyclophosphamide monohydrate and ciprofloxacin hydrochloride were spiked at 100 µg/L and biodegradation was allowed for 40 days.	Cyclophosphamide monohydrate and ciprofloxacin hydrochloride	Fouling of the ceramic NF membrane slightly increased the rejection of cyclophosphamide monohydrate and ciprofloxacin hydrochloride.	[245]
Secondary effluent wastewater spiked with 3 TrOCs at 750 µg/L per organic. The wastewater effluent had chemical oxygen demand of 18 mg/L.	Acetaminophen, carbamazepine and diatrizoate	Fouling of the NF270 membrane improved the removal of acetaminophen (20–50%), carbamazepine (75–92%), and atenolol (50–85%), while the removal of diatrizoate did not change. Improvement in the rejection was attributed to the TrOCs interacting with effluent organic matter on the membrane surface. On the contrary, fouling of the NF90 membrane resulted in the decreased rejection of TrOCs over time.	[164]
Municipal wastewater was pre-treated with MBR and used as feed for the nanofiltration process.	Metoprolol, trimethoprim, carbamazepine, chloramphenicol, diclofenac sodium and indomethacin	The removals of metoprolol, trimethoprim, carbamazepine, and chloramphenicol were enhanced by 4.7%, 5.4%, 5.6% and 0.5%, respectively, upon fouling the NF membrane (DF30) by organic matter and polysaccharides. The removal rates of diclofenac sodium and indomethacin did not change.	[315]
Secondary effluent was collected from a local municipal wastewater treatment plant. The feed was filtered through a microfiltration unit (EW4040F GM).	Primidone	The removal of primidone by the thin film composite (TFC-HR) membrane did not change due to membrane fouling by effluent organic matter. However, fouling decreased primidone rejection by XLE (90–85%), NF200 (90–60%) and CTA (70–30%) membranes.	[316]
Secondary effluent wastewater was collected from a municipal wastewater treatment plant and pre-filtered using a 0.45 µm mixed cellulose filter.	Carbamazepine and naproxen	Fouling the polyethersulfone membrane by effluent organic matter improved the removal of naproxen and carbamazepine. The removal depended on the transmembrane pressure; the highest removals were at 40 and 60 kPa. Naproxen removal increased from 65–75% at 40 kPa and 55–62% at 60 kPa, while carbamazepine rejection increased from 15–28% at 60 kPa.	[317]
Synthetic water spiked with two hormones at the initial concentration of 100 ng/L at 50 mM NaCl. 200 mg/L colloidal silica was used as the model foulant.	Estradiol and progesterone	Estradiol rejection by the RO membrane (LFC-1) decreased from about 98% to about 77% after 100 h of filtration due to membrane fouling by silica colloids. Similarly, the retention of progesterone decreased from about 98% to 78% after 100 h of fouling. The decline in TrOCs removal was attributed to cake-enhance concentration polarization effects.	[318]
Synthetic water spiked with 10 mg/L carbamazepine. Sodium alginate (20 mg/L), latex (30 mg/L), and silica colloids (30 mg/L) were used as model foulants in the presence and absence of calcium.	Carbamazepine	Fouling of the NF270 membrane resulted in a decrease in the rejection of carbamazepine after 72 h of filtration. The decline in rejection was as follows: 70% for fouling with alginate, 41% for fouling with latex and 35% for fouling with silica. The decrease in rejection was ascribed to a decline in permeate flux and cake-enhanced concentration polarization. Fouling in the presence of calcium resulted in a lesser decline in carbamazepine rejection.	[163]

coatings and engineered materials include biomimetic coating with mild and versatile merits, sequential infiltration synthesis as well as atomic layer deposition with precise and controllable features. Yang et al.[150] prepared catalytic-cleaning antifouling membranes using a prebiotic-chemistry-inspired aminomalononitrile (AMN)/Mn^{2+}-mediated biomimetic mineralization technique. The biomimetic mineralized membranes fouled less during oil-in-water emulsion separation (with 99.8% flux recovery rate) and exhibited excellent in situ regeneration efficacy. Atomic layer deposition

Table 11 (continued) | Effects of fouling on the removal of TrOCs by high-retention membranes during wastewater treatment by HR-MBRs

Feed type	Trace organic compounds	Effects of fouling on TrOCs removal[a]	Ref.
Synthetic water spiked with 12 TrOCs, each dosed at 2 µg/L. Fouling experiments were conducted with 100 mg/L humic acid and 1 g/L colloidal silica in separate experiments at different initial fluxes.	Amitriptyline, trimethoprim, sulfamethoxazole, diclofenac, bezafibrate, caffeine, atrazine, primidone, carbamazepine, pentachlorophenol, linuron and triclosan	Fouling of the thin film composite FO membrane by humic acid at the initial flux of 9 L/m²h improved the rejection of amitriptyline, trimethoprim, sulfamethoxazole, diclofenac, bezafibrate, caffeine, atrazine, primidone, carbamazepine, pentachlorophenol, linuron and triclosan. In comparison, the removal of the TrOCs decreased when fouling was conducted at 20 L/m²h. Similar observation was made for membrane fouling with colloidal silica, where improvement in rejection was noted for fouling at the initial flux of 9 L/m²h, while the decrease in rejection was noted when fouling was conducted at 20 L/m²h. The most noticeable changes in rejection were made for caffeine, atrazine, primidone, and carbamazepine, all neutral compounds.	[319]
Synthetic municipal wastewater spiked with TrOCs at 25 µg/L per compound. Primary wastewater was used for biofilm formation on the FO membrane for 24 h.	Amitriptyline hydrochloride, atenolol, atrazine, caffeine, carbamazepine, primidone, and sulfamethoxazole	Biofouling of the polyamide thin film composite FO membrane improved the rejection of caffeine (90–98%), atrazine (90–100%), primidone (98–100%), carbamazepine (98–100%), sulfamethoxazole (95–100%) and atenolol (55–85%) while the retention of amitriptyline decreased from 98% to 88%. Improvement in removal was attributed to the biodegradation of the TrOCs and an increase in mass transfer resistance by the biofilm, while the decrease in amitriptyline removal was ascribed to solute build-up on the membrane surface, leading to its poor removal.	[320]
Municipal wastewater obtained from a wastewater treatment plant and synthetic wastewater prepared by adding either 200 mg/L sodium alginate or 200 mg/L humic acid to a background electrolyte solution of 1 mM CaCl₂. Two TrOCs were spiked in the feed at 2 mg/L per organic.	Carbamazepine and sulfamethoxazole	After 720 min of filtration using a thin film composite FO membrane, sulfamethoxazole removal increased for fouling with humic acid (98.5–99%) and sodium alginate (98.5–99.5%), while its removal decreased slightly for membrane fouling by municipal wastewater (98.5–98%). On the other hand, the retention of carbamazepine decreased for all fouling types as follows: 99–98.5% for humic acid, 99–98% for sodium alginate and 99–97% for municipal wastewater.	[321]
Synthetic wastewater was prepared by spiking 5 TrOCs at a concentration of 800 µg/L each. Fouling was performed with 20 mg/L humic acid, 60 mg/L sodium alginate or 1.0 g/L silica at background electrolyte 20 mM NaCl and 1 mM NaHCO₃.	Sulfamethoxazole, ibuprofen, triclosan, sulfadiazine and carbamazepine	The operational pH controlled the removal of TrOCs by the fouled membranes. At pH 8, the removal of simazine, triclosan and carbamazepine by the NF270 membrane declined for fouling by humic acid, silica, combined humic acid + sodium alginate and combined humic acid + silica. For the NF90 membrane, fouling remarkably improved the retention of all TrOCs at pH 8 except for sulfadiazine which decreased from 80% to about 75%. Fouling of the XLE membrane by all foulants improved the removal of TrOCs except for sulfadiazine, triclosan and carbamazepine, whose retention decreased upon fouling with silica (sulfadiazine) and combined humic acid + silica (triclosan and carbamazepine).	[322]

[a]Some TrOCs removal rates were estimated from rejection graphs presented in the referenced articles.

and/or sequential infiltration synthesis allows for the regulation of material properties to obtain thin films of desirable thickness, stoichiometry as well as physical and chemical properties using atomic engineering[151]. In a more recent work, Yang et al.[152] demonstrated unprecedented formation of acid-tolerant ultrathin membranes with finely tuned sub-nanopores for energetic-efficient molecular sieving. The membranes were prepared from polyurethane (PU)-based materials and the highly crosslinked structure and unique electron-absorbing ability enabled the novel membranes to withstand strong acidic conditions. The membranes have great potential for energy efficient environmental remediation and resource recovery. Although a lot of work has been done to improve membrane properties, there is still little information regarding the performance of the modified membranes using real wastewater. For commercial membranes, there is sufficient information on the ideal/optimal operating conditions that has helped to build consumer confidence, while such information is still lacking for the newer generation membranes. Further, some modification techniques are costly, not green (i.e., use toxic chemicals) and hard to reproduce.

System optimization and operating conditions. Optimizing the design configuration and operating conditions can also reduce membrane fouling. For instance, Hai et al.[153] placed a bundle of hollow fibers within a non-woven coarse pore to prevent the direct deposition of macromolecules onto the membrane surface. Further, the authors optimized operational parameters such as temperature, pH, applied Hydraulic Retention Time (HRT) and flux to reduce fouling and maintain high performance over a prolonged time. When tuning operational temperature to alleviate fouling, it must be considered that the performance of MBR regarding removing TrOCs is influenced by temperature. For example, Hai et al.[154] studied the removal of TrOCs by MBR under temperature variation and found that operating under 45 °C significantly

Fig. 9 | Membrane pre-treatment options. Classification of pre-treatment techniques according to efficiency, cost, and associated challenges.
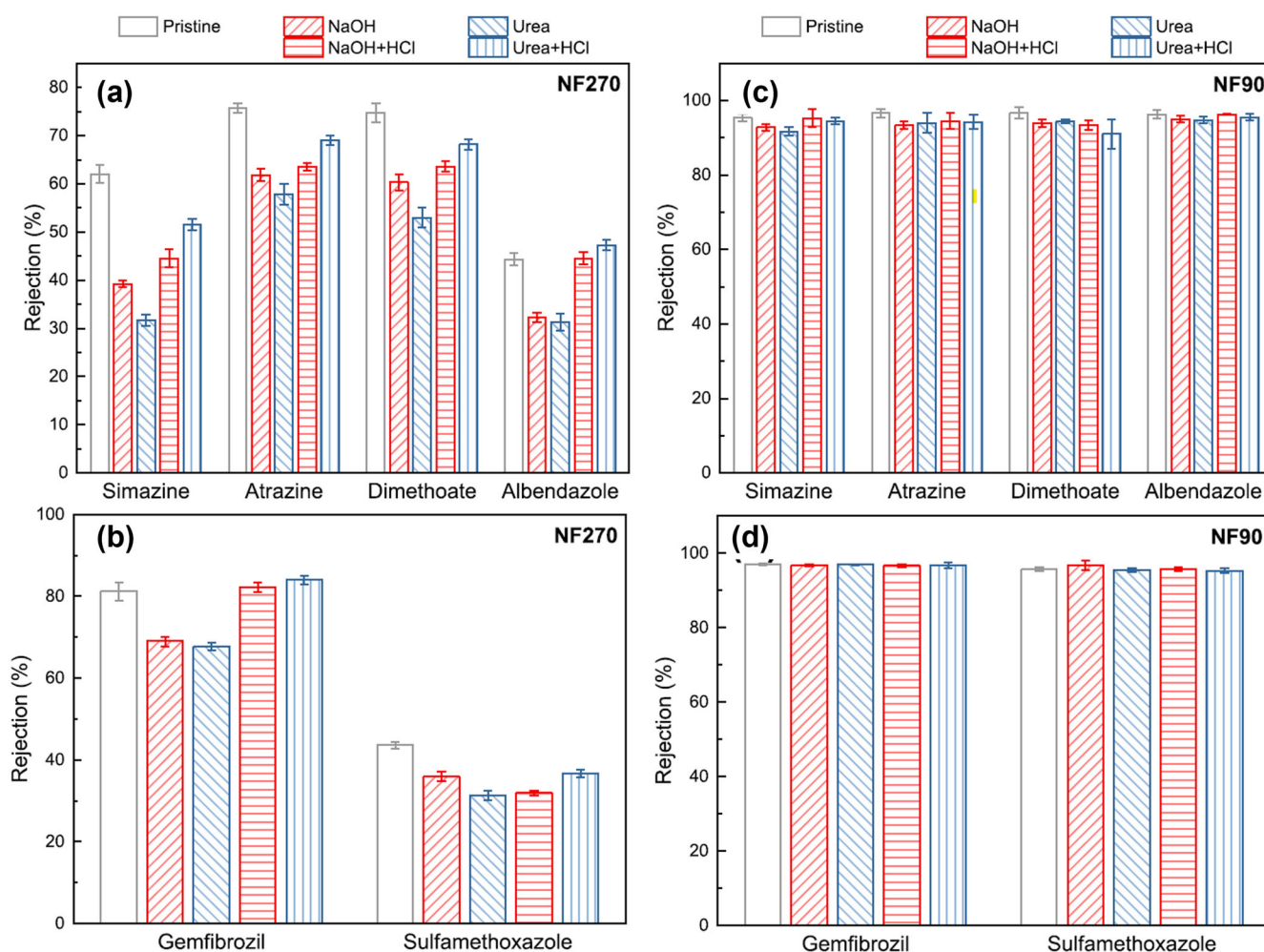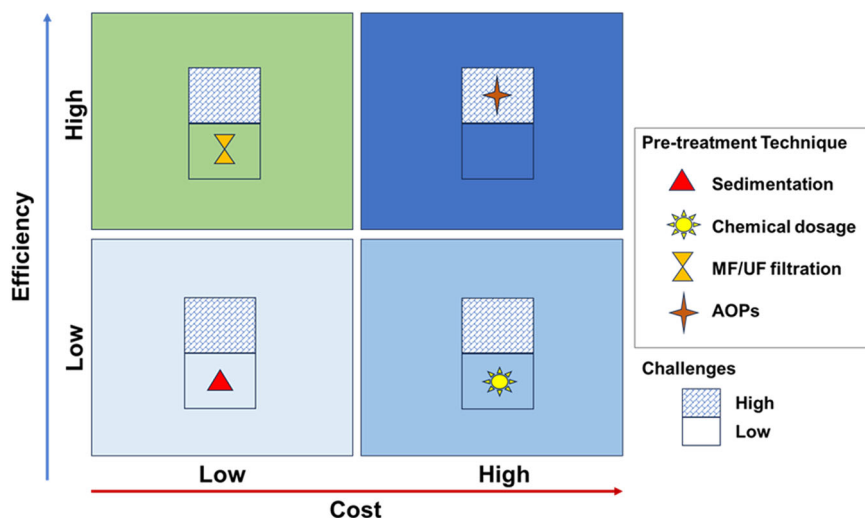
Fig. 10 | Rejection of trace organic compounds by virgin and fouled NF membranes. Effects of urea-based chemical cleaning on the rejection of TrOCs by NF membranes: removal of neutral (**a**) and negatively charged TrOCs (**b**) by NF270 membrane; and removal of neutral (**c**) and negatively charged TrOCs (**d**) by NF90 membrane[131]; error bars present standard deviations.

influenced the removal of certain less hydrophobic micro-pollutants possessing strong electron-withdrawing functional groups.

On the one hand, the removal of most hydrophobic compounds was stable when the temperature was maintained between 10–35 °C. On the other hand, their removal deteriorated when the temperature increased to 45 °C. Plattner et al.[56] studied the role of temperature (40, 55 and 70 °C) on the removal of pesticides by a commercial hydrophobic PTFE flat sheet membrane in membrane distillation and observed a decrease in rejection at high feed temperature. This was more noticeable for dichlorvos, while the removal of atrazine and parathion-methyl declined slightly. Furthermore,

elevated temperatures are believed to result in the thermal expansion of the active membrane layer and an increase in both membrane pore size and the diffusion coefficient of the TrOCs[155]. Some studies have reported probable distortion of the microstructure of MD membranes at high temperatures. Saffarini et al.[156] studied the effect of temperature variation (25–70 °C) on microstructure and stability of commercial PTFE membranes in relation to the liquid entry pressure (LEP) under MD conditions. Differential scanning calorimetry (DSC) measurements displayed potential structural modifications resulting in the relaxation of internal stresses in the interconnected continuous fibrils of the expanded PTFE membranes. This microstructural evolution was due to the distortion of fibril, gap and node as the temperature was elevated.

Further, it was found that the membrane pore diameter increased with temperature. Similar observations of the pore size of NF membranes becoming larger as the feed temperature was raised were reported by Xu et al.[157]. Their work investigated the influence of temperature on the retention of pharmaceuticals and personal care products (PPCPs) by NF membranes. In another work, the removal of TrOCs decreased with an increase in temperature[158], and the authors reported that the pore size of an NF membrane depended on the feed solution temperature. The effective pore radius increased from 0.39 to 0.44 nm when the feed temperature was raised from 20 to 40 °C. Long-term effects of the temperature variations on pore or void structure, membrane flux and TrOCs removal have not been well investigated and require systematic investigations.

Therefore, there must be a balance where fouling is kept at a minimum while high removals are prioritized because fouling can be mitigated by simple backwash (depending on the fouling type), while poor removals will require additional treatment steps that may be costly. Studies have already shown the role of operating conditions on the removal of TrOCs, and the governing mechanisms explaining the removal trends have been reviewed by Mahlangu et al.[51] and Khanzada et al.[52], amongst other researchers. For example, the following has been reported: i) elevated calcium concentration reduced the removal of halogenated acetic acid by NF270 membranes[159]. The presence of divalent cations in the feed (calcium concentrations of 5 and 10 mM) improved the rejection of positively charged solutes and decreased the rejection of negatively charged TrOCs by NF membranes due to shielding of the surface charge, while the removal of neutral TrOCs was not affected[63]; ii) NF270 membrane had poor removal of perfluoro carboxylic acid at low pH, while the removal of the same compound was improved for NF90, XLE, BW30, and SW30XLE membranes[160]. The removal of sulfamethoxazole, carbamazepine and ibuprofen by NF270 decreased when the pH was changed from pH 10.5 to pH 3.5[161]; iii) increasing the pressure from 10–20 bar resulted in a 15% decline in the removal of estrone[162], while other studies noted improved rejection with increasing pressure but up to a limit[163]; iv) the rejection of TrOCs increase with flux but at high flux rejection may decline due to the effects of concentration polarization[164]; v) the rejection of neutral and positively charged solutes (benzotriazole, hydrochlorothiazide, carbamazepine, metoprolol, trimethoprim, and sulpiride) by NF membranes (VNF1 and VNF2) decreased with an increase in temperature from 5 °C to 25 °C, while the removal of negatively charged TrOCs was not affected due to the presence of stronger electrostatic repulsions between the negatively charged compounds and NF membrane surface[157]; and vi) the presence of macromolecules (foulants) may result in membrane fouling and have various effects on TrOCs retention (Table 11).

The choice of HR-MBR process and enzyme in the bioreactor can also be used as a fouling mitigation strategy. Regarding the choice of the HR-MBR process, the idea is to adopt a system that would have an inherently low membrane fouling tendency, in this case, OMBR. Concerning the option of enzymes, Tufail et al.[165] used horseradish peroxide enzymes to reduce fouling in an enzymatic membrane distillation bioreactor coupled with a membrane distillation process. The authors observed lesser fouling of the MD membrane (28%) after 240 h for the enzyme-assisted MBR compared to the conventional MBR without adding horseradish peroxidase (50%) and emphasized the importance of the choice of enzyme in enzymatic MBR. To the best of our knowledge (on writing this review), a comparison of different

enzymes' efficiency in reducing membrane fouling in HR-MBRs has not been reported.

The onset of fouling can be delayed by operating below the critical flux, a benchmark flux at which membranes can be operated without remarkable fouling, whereas filtration above the critical flux results in aggravated fouling. The critical flux concept has been used to control membrane fouling in MF[166], UF[167], NF[168,169], RO[170] and FO[171]. Therefore, the same idea can be applied to HR-MBRs to prevent membrane fouling. However, operating at critical flux may not produce enough permeate to meet consumer demand. Further, the rejection of TrOCs at lower fluxes could be compromised due to the dependency of the rejection of TrOCs to flux[172–174]. Table 12 presents studies that have used various strategies to mitigate the fouling of high-retention membranes. These approaches include pre-treatment, introduction of advanced oxidation processes, cleaning of the fouled membranes, modification of the membrane surface and optimizing operational parameters. Some of the pre-treatment methods are filtration using MF/UF membranes[175] and activated carbon adsorption[176]. Advanced oxidation processes such as Fenton processes[177], photooxidation[178], electrochemical coagulation[179] and sonication-assisted filtration[180,181] have shown great potential in reducing membrane fouling. In photooxidation, UV irradiation is used to degrade compounds that can potentially foul the membrane. Electrochemical coagulation involves the application of an electric field coupled to a low voltage to reduce membrane fouling through repulsive interactions between the membrane and charged foulants such as natural organic matter, colloids, and bacteria. Sonication or ultrasound-assisted filtration involves the irradiation of ultrasound (cavitation) to detach foulants from the membrane surface. Membrane cleaning can be performed through chemical cleaning or physical cleaning (e.g., hydraulic flushing and osmotic backwashing). Membrane fouling can also be delayed by fine-tuning the operational conditions. Optimization of operational conditions ensures that the conditions are not favorable for fouling to take place. For example, intermittent aeration removes loose foulants on the membrane surface thus preventing cake build-up[182]. Membrane surface modification is another method adopted to reduce the affinity of foulants for the membrane surface (i.e., reduction in hydrophobic interactions). Furthermore, the modification aims at making membrane surfaces smoother thus preventing attachment and accumulation of foulants on the membrane surface. Adoption of the fouling mitigation strategies (currently implemented in municipal wastewater treatment) can improve the technical feasibility and the economic viability of HR-MBR which is envisaged to produce high quality effluent compared to conventional biological treatment coupled with MF/UF and advanced oxidation processes.

The different cleaning strategies have various effects on NF/RO, FO, and MD processes. For example, repeated chemical cleaning to remove deposited sediments on the surface of MD membranes can lead to surface wetting and polymer degradation[183]. Polymer degradation is also possible for pressure-driven membranes, however, the effect on membrane performance may not be as severe as the effects of pore wetting which could lead to immediate failure in MD. Residual deposits may remain after cleaning. In NF/RO and FO, the effects would be lower flux recovery, while in MD the residual deposits may lead to premature wetting creating hydrophilic bridges and/or becoming nucleation sites thus promoting scaling. Cyclic cleaning has different effects to MD compared to pressure-driven membranes, where in NF/RO high permeate quality and module performance are restored after each cleaning cycle whereas in MD there is degradation of permeate quality with each cycle due to pore wetting.

Generally, FO has more fouling propensity than NF/RO and MD due to cake-enhanced concentration polarization. However, physical cleaning is more efficient in FO and less efficient in MD due to lesser reversible fouling in MD. Jang et al. found that the flux recovery after cyclic cleaning was higher for FO and RO and lowest for MD[184]. This observation was ascribed to stronger hydrophobic MD membrane-foulant interactions, whereas FO and RO membrane surfaces are hydrophilic.

Several pre-treatment steps are needed to prevent fouling in NF/RO processes. Contrary, a single pretreatment step may be enough to maintain

Table 12 | Some of the strategies adopted to mitigate membrane fouling during wastewater treatment using HR-MBRs

Category	Description of control measure	Assessment of effectiveness	Ref.
Pre-treatment	A micro granular adsorptive filtration consisting of a 7-bore ceramic membrane as the primary membrane was constructed to pre-treat MBR effluent prior to RO filtration. Heated aluminum oxide particles or powdered activated carbon were used as the pre-deposited dynamic membrane.	Reduction in fouling of the thin film composite RO membrane (BW-30) from 63–43% after 120 h of filtering MBR effluent from a water reclamation plant.	[301]
	A combined coagulation-disk filtration system was used as pre-treatment to prevent membrane fouling. The combined coagulation-disk unit treated secondary effluent prior to UF and RO filtration.	The combined coagulation-disk filtration was ineffective in removing various bacteria and algae due to the larger mesh size. However, the microbes were removed by the UF membrane preventing fouling of the polyamide thin film composite RO membrane.	[323]
	Three UF membranes with molecular weight cut-off (MWCO) values of 4, 10, and 250 kDa were used as pre-treatment for the effluent of a wastewater treatment plant. UF permeate was used as feed for NF and RO membranes.	Aggravated biofouling in UF membranes with smaller MWCO (4 kDa and 10 kDa) while metal precipitates fouled the UF 250 kDa membrane. Pre-treatment with UF reduced fouling of NF and RO membranes (NF90, NF270, XLE, AD90 and BW30) applied downstream.	[175]
	Pre-treatment of synthetic wastewater (MBR effluent) with FO membranes prior to filtration by RO membranes. The simulated wastewater was prepared to obtain 100 mg/L glucose, 100 mg/L peptone, 17.5 mg/L KH_2PO_4, 17.5 mg/L $MgSO_4$, 10 mg/L $FeSO_4$, 225 mg/L CH_3COONa, and 35 mg/L urea and 31 TrOCs.	FO process prevented foulants from permeating into the draw solution, and this reduced fouling of the downstream RO membrane process in a combined OMBR-RO system. Direct filtration of MBR effluent without the FO membrane resulted in severe fouling of the RO membrane (LFC3).	[126]
	Before final treatment with RO, raw municipal wastewater was pre-treated with novel low-pressure antifouling polydopamine/poly-ethylenimine NF membranes. Prior to NF, the raw water was filtered through a 0.45 µm microfiltration membrane.	The polydopamine/polyethyleneimine-modified low-pressure NF membrane removed organic and inorganic foulants without fouling. This prevented fouling of the subsequent RO process fed with the permeate from the novel NF membrane resulting in better performance due to the improved NF effluent quality.	[307]
	Granular activated carbon (GAC) was used as an adsorption technique to pre-treatment real wastewater. Adsorption was conducted by adjusting the pH of the soluble microbial by-products to 7, and the temperature was maintained at 25 °C. Granular activated carbon was added at different dosages and stirred, and the supernatant was filtered through a 0.45 µm membrane after settling.	Granular activated carbon alleviated fouling of an aromatic poly-amide RO membrane by adsorbing protein and humic substances in soluble microbial by-products.	[176]
	Before the RO process, biochar was used as an adsorbent treating MBR effluent (municipal wastewater with 400 mg/L chemical oxygen demand).	RO membrane fed with MBR permeates had a 15% flux decline after 180 min of filtration, while the flux declined by 5% when effluent from biochar adsorber was filtered. Fouling was prevented through ammonium recovery and removal of divalent cations.	[324]
Advanced oxidation processes (AOPs)	An electrochemical process was integrated into the MBR, treating synthetic wastewater with a chemical oxygen demand of 556 mg/L and 284 µS/c electrical conductivity. The external voltage gradient was varied between 1 V/cm and 3 V/cm.	The addition of electrochemical processes into the MBR improved the treatment performance achieving high nutrient removal. It reduced the fouling of the polyvinylidene fluoride membrane by 54% due to increase in floc hydrophobicity and decrease in concentration of membrane fouling precursors.	[325]
	An electroosmotic membrane bioreactor was employed at the feed side of a forward osmosis process for municipal wastewater treatment. Municipal wastewater and aerobic sludge were obtained from a wastewater treatment plant.	Fouling potential was lower in the presence of an electric field. Further, the effects of internal concentration polarization were reduced upon introducing an electric field, thus reducing the fouling of the FO membrane.	[326]
	A Fenton process was used as a pre-treatment of MBR effluent (landfill leachate) prior to NF filtration. The pH of the MBR effluent was an adjustment with sulfuric acid (H_2SO_4), followed by the addition of iron salt and, finally, 30% hydrogen peroxide solution.	The Fenton process removed 87% of chemical oxygen demand and 93% of organic matter. However, fouling of the NF90 membrane was observed due to the contribution of new ions to the effluent by the Fenton process. Fe^{2+} and Fe^{3+} caused incrustation in the NF membrane and disturbed the performance of the process due to the flux decline.	[177]
	Photooxidation through UV irradiation was used to pre-treat confectionery wastewater with a chemical oxygen demand of 10800 mg/L, total organic carbon of 4475 mg/L and electrical conductivity of 848 µS/cm. The pre-treated wastewater was used as feed for nanofiltration.	Pre-treatment with UV photooxidation reduced the extent of fouling of the subsequent nanofiltration processes utilizing NF90, NF270 and TM610 membranes. Further, the performance of the NF membranes in removing total organic carbon and color was enhanced.	[178]
	An applied electric field was coupled to a low voltage in the membrane system, where an electric field was applied across the membrane with electrodes installed on either side of the membrane. The feed was synthetic wastewater prepared from 2 g NH_4Cl, 1 g KH_2PO_4, 0.125 g $CaCl_2.H_2O$, 0.3 g $FeSO_4.7H_2O$ and 20 g sucrose.	Application of an electric field improved the permeate flux of poly-propylene membrane and polyester membranes at applied electric fields of 0.036 V/cm and 0.073 V/cm and reduced the production of extracellular polymeric substance and membrane fouling. Further, this extended the operating cycles.	[327]
	Electrical coagulation was integrated with MBR as pre-treatment for industrial zone wastewater prior to RO filtration. The industrial wastewater had chemical oxygen demand of 2000 mg/L.	Addition of the electrocoagulation step improved the removal of chemical oxygen demand by 4% and alleviated the fouling of RO membranes. Further, settling was improved, thus enhancing the dewaterability and filterability of the sludge.	[179]
Cleaning	Membranes fouled after industrial wastewater filtration were cleaned with hydrochloric acid (HCl) and sodium hydroxide (NaOH) by varying the pH of the cleaning solution.	Cleaning of the BW30XFR RO membrane with acid weakened the adhesion forces between the fouling layer and the membrane, and this removed foulants on the membrane surface.	[302]
	A cellulose acetate FO membrane fouled by raw municipal wastewater was cleaned by hydraulic flushing 1% sodium hypochlorite, 0.8% sodium ethylenediaminetetraacetic acid, and 0.1% sodium dodecyl sulfate.	Hydraulic flushing and chemical cleaning were more effective in recovering the water permeability of the fouled cellulose triacetate FO membrane.	[288]

Table 12 (continued) | Some of the strategies adopted to mitigate membrane fouling during wastewater treatment using HR-MBRs

Category	Description of control measure	Assessment of effectiveness	Ref.
	Physical cleaning (hydraulic flushing and osmotic backwashing) and chemical cleaning (ethylenediaminetetraacetic acid, nitric acid, and sodium hydroxide) of a polyamide thin film composite membrane fouled by 100 mg/L alginate, 1 g/L silica with total ionic strength of 50 mM and 1 mM calcium. 35 mM $CaCl_2$, 20 mM Na_2SO_4 and 19 mM NaCl were used to prepare solutions for scaling experiments.	Osmotic backwashing of the polyamide thin film composite FO membrane was more effective than hydraulic flushing for all fouling types except for fouling by silica colloids. Chemical cleaning restored 68%, 90% and 85% for cleaning with ethylenediaminetetraacetic acid, nitric acid, and sodium hydroxide, respectively.	[328]
Intermittent filtration with UF/MF	Intermittent osmotic relaxation was performed by replacing the draw solution in the draw tank with deionized water, and all other operational conditions were kept the same.	A 4-h filtration with 20 min of backwash reduced the resistance of a cellulose triacetate FO membrane by 30% and achieved 100% flux recovery.	[329]
	Periodic filtration using a polyvinylidene fluoride microfiltration membrane was adopted to mitigate salinity build-up during MBR treatment of real wastewater. Physical cleaning with water was also performed.	Microfiltration extraction prevented salinity accumulation, thus slowing the production of soluble microbial by-products. This, together with physical washing, reduced the fouling of the FO membrane.	[239]
Optimizing operating conditions	Membrane flushing, air scouring, and ultrasonication were systematically investigated to remediate cellulose triacetate FO membrane fouling. Accelerated membrane fouling was performed to simulate long-term and intensive pre-concentration scenarios needed for phosphorus recovery from digested sludge centrate.	High cross-flow velocity flushing and ultrasonication effectively prevented fouling of the cellulose triacetate FO membrane under accelerated fouling and maintained stable fluxes. Frequent membrane cleaning helped achieve a higher cumulative permeate volume and lower energy usage.	[127]
	Hexamethylene diamine tetra (methylene phosphonic acid) was added in the draw solution of a forward osmosis system as a control measure to inhibit scaling during landfill leachate treatment with chemical oxygen demand 1803 mg/L and electrical conductivity of 56 mS/cm.	The organic phosphonic acid scale inhibitor chelated with Ca^{2+} and prevented the nucleation and growth of gypsum scale crystals. This alleviated scaling on the aquaporin-thin film composite FO membrane. Further, the scaling inhibitor prevented wetting the super-hydrophobic polytetrafluoroethylene MD membrane used to reconcentrate FO draw solution.	[291]
	A fixed bed biofilm was used to replace the activated sludge as the bio-phase in an osmotic membrane bioreactor treating raw municipal wastewater.	The addition of the fixed bed biofilm provided a vent for the suspended growth to be continuously removed from the bioreactor, and this reduced fouling of the thin film composite FO membrane. A 24.7–54.5% reduction in FO fouling was achieved due to the reduced bacteria deposition and colonization.	[292]
	Biocide solution (5 mg/L) and anti-scalant (5 mg/L) were added to the raw textile wastewater to prevent biofouling and scaling of a RO membrane.	The addition of anti-scalant and biocide did not prevent fouling of the XLE RO membrane; however, this helped improve the membrane cleaning efficiency.	[309]
	Ozonation was applied on MBR effluent (wastewater) to prevent fouling of the downstream RO process.	Even at low doses of 1.5 mg/L, Ozonation minimized fouling of the downstream ESPA-2 RO membrane treating MBR effluent.	[330]
	The performance of different FO membranes in treating high-strength wastewater of sugarcane molasses distillery was compared for proper membrane selection based on membrane properties during wastewater treatment.	Physical cleaning restored 70–82% flux for thin film composite (HTI-TFC) and cellulose triacetate (HTI-CTA) membranes and 65–52% for aquaporin-thin film composite membranes. All membranes fouled, but the ease of cleaning was better for HTI-TFC, followed by HTI-CTA and aquaporin-TFC membranes. These findings suggested that the HTI-TFC membrane was most suited for concentrating distillery wastewater, thus indicating the importance of proper membrane selection.	[331]
	Membrane orientation was altered to investigate TrOCs removal and fouling propensity during OMBR treatment of synthetic wastewater with an 800 mg/L chemical oxygen demand.	There was negligible flux decline when the active layer of the cellulose triacetate FO membrane faced the feed solution, while severe flux decline was observed when the active layer faced the draw solution. The findings showed the importance of optimizing membrane orientation in FO processes.	[259]
	Response surface methodology was used to optimize the operating conditions of a forward osmosis process to achieve high water flux and removal efficiency while reducing salt reverse flux during the treatment of high salinity landfill leachate.	Response surface methodology provided optimum conditions for high pollutant removal and reduced reverse salt flux during landfill leachate treatment by a thin film composite FO membrane.	[332]
	Organic draw solutes (sodium acetate and ethylenediaminetetraacetic acid disodium salt) were compared to NaCl in OMBR integrated with RO. The integrated system treated synthetic wastewater spiked with 31 TrOCs to simulate medium-strength municipal sewage.	Using sodium acetate and ethylenediaminetetraacetic acid disodium salt draw solutes significantly reduced salinity build-up in the bioreactor in comparison with NaCl during OMBR operation. Lesser organic fouling of the cellulose acetate FO membrane was observed when the organic solutes were used compared to NaCl.	[12]
	Membrane fouling was investigated by monitoring transmembrane pressure at different initial fluxes of 3 L/m²h and 40 L/m²h during the treatment of secondary wastewater effluent with electrical conductivity of 2.1 mS/cm.	There was negligible membrane fouling in 48 days of operation when direct NF treatment of secondary wastewater effluent was performed at the initial flux of 3 L/m²h. The cake layer on the surface of the composite polyamide NF membrane was easily removed by simple physical cleaning. The findings showed the importance of operating at critical flux to reduce fouling.	[333]
	Aeration was controlled based on the dissolved oxygen set-point, and the air scours flow rate was maintained at 10 m³/h to control membrane fouling during NF-MBR treatment of primary wastewater effluent.	Membrane flux was maintained, and there was minimum fouling of the NF90 membrane.	[255]
	Different strategies were adopted to control membrane fouling during MBR treatment of secondary effluent wastewater. The techniques	Intermittent aeration, frequent effluent discharge and dosing of sodium hypochlorite prevented fouling of the RO membranes (LFC1,	[182]

Table 12 (continued) | Some of the strategies adopted to mitigate membrane fouling during wastewater treatment using HR-MBRs

Category	Description of control measure	Assessment of effectiveness	Ref.
	included intermittent aeration, periodic effluent withdrawal, and continuous sodium hypochlorite dosing.	ESPA1). This enhanced the performance of the membranes in removing estrogens to the concentration of 1.3–8.3 μg/L.	
	Air scouring was used to restore membrane flux after fouling cellulose triacetate FO membranes during the treatment of synthetic wastewater with chemical oxygen demand, total nitrogen, and phosphorus concentrations of 439.47 mg/L, 60.23 mg/L, and 9.42 mg/L, respectively.	Air scouring removed loose foulants and recovered 89.5% of the original permeate flux of a cellulose triacetate FO membrane. Air scouring was more effective than chemical cleaning of the fouled membranes.	289
	MgCl₂ was used as a draw solute with low diffusivity to reduce salinity build-up and prevent membrane fouling during the treatment of refinery wastewater (chemical oxygen demand of 336 mg/L) and electrical conductivity of 2.3 mS/cm) with OMBR.	$MgCl_2$ had minimal impact on microbial activity, and this favored biodegradation of poorly degradable organic compounds. Introducing a UF membrane and using NaCl resulted in stable salt concentration and flux of a cellulose triacetate FO membrane over 330 days; however, the microbial activity was impacted due to the reverse salt flux of NaCl.	334
Membrane surface modification	Commercial FO membranes were coated with electroactive carbon nanotubes and black carbon to prevent fouling during the treatment of synthetic wastewater prepared using the bacterial solution and real wastewater samples.	The modified thin film composite FO membranes had a good anti-fouling performance with about a 63% reduction in flux loss compared to the pristine membrane.	335
	Hydrophilic sulphonated polyethersulfone was anchored in the polyethersulfone matrix to improve the performance of thin film composite FO membranes.	Membrane surface modification enhanced osmotic water flux by 4 folds and reduced reverse salt leakage by about half. This was attributed to the enhanced wettability, which reduces membrane fouling.	336
	A thin film composite RO membrane was modified by surface grafting of polyvinyl alcohol through cross-linking with glutaraldehyde to enhance fouling resistance. The modified membrane was used to treat synthetic water with different salts, including 93 mg/L of sodium bicarbonate, magnesium sulfate, calcium sulfate and ammonium sulfate at 60 mg/L.	The flux of the virgin membrane declined from 62 L/m²h to 34 L/m²h (45.2%), while the modified membrane flux decreased from 49 L/m²h to 36 L/m²h (26.5%). Further, the unmodified membrane had a flux recovery of 54%, while the modified membrane had a 74% flux recovery. This was due to poor adhesion of biological foulants on the membrane surface.	337

stable operations in MD. This implies that although MD has higher fouling propensity, it could still be more easier, cost-effective and environmentally friendly to manage than NF/RO and FO[185].

Salinity build-up in HR-MBRs
Salinity build-up: causes and effects on the removal of TrOCs
HR-MBRs are severely affected by salinity build-up in the bioreactor due to the membranes' high salt rejection capabilities and reverse salt flux in OMBR. The impact of salinity on the performance of HR-MBR has been reviewed by Lay et al.[17]. The effects of salt accumulation were not linked to the removal of TrOCs but to physiological, microbial and membrane aspects, which are mutually engaged in the removal of TrOCs.

Elevated salinity in the bioreactor affects the physical and biochemical properties of microbes, and this is detrimental to the microbial community as it deteriorates or inhibits biological activity[186–188] by reducing the diversity of microbial colony[189] and cell plasmolysis[48]. In OMBR, salinity build-up decreases the effective driving force due to increased osmotic pressure in the feed solution, resulting in low permeate flux. Further, the high salt concentration in the bioreactor can damage membranes and reduce performance and lifespan[190]. Salinity inhibits or slows down microbial kinetics leading to lower growth yield and higher endogenous decay of halophobic bacteria in the bioreactor[17,189]. However, it is believed that the bacterial population may acclimatize to the elevated salinity environment by rapidly reproducing halotolerant or halophilic bacteria[126,191].

There are contradictory reports regarding the influence of increasing salinity on microbial diversity. Luo et al.[191] found similar α-diversity metrics in the bioreactor of the control and saline-MBRs implying that salinity did not affect microbial diversity. This was attributed to the succession of halophobic microorganisms by halotolerant or halophilic bacteria as the bioreactor salinity increased. However, hierarchical clustering showed noticeable differences in bacterial community structure between the control and the saline-MBR systems. The elevated salinity was believed to promote the development of different bacterial communities.

Further, there was a reduction in the abundance of various bacterial phyla (including *Planctomycetes*, *Verrucomicrobia*, *Bacteroidetes*, *Armatimonadetes* and *Gemmatimonadetes*) in the saline-MBRs. The effect of salinity accumulation on microbial activity depends on the type of microbes and anions in the feed. Chapple et al.[192] used 15 cations and anions from common inorganic salts to show that the enzymatic activity of laccase was not inhibited by monovalent cations such as Na^+, NH_4^+ and K^+. In contrast, multivalent cations (e.g., Mg^{2+}) had variable effects from negligible to complete inhibition of both enzyme activity and the degradation of TrOCs.

Reports of high salinity increase the concentration of soluble microbial by-products and extracellular polymeric substances[193,194]. Extracellular polymeric substances result in inefficient oxygen transfer and elevated viscosity[126]. Thus salinity build-up may aggravate membrane fouling, leading to poor fluxes[195] and shorter membrane lifespan, resulting in membrane replacement when the membrane life is spent[187]. Salinity build-up may not only change the microbial diversity and aggravate fouling, but it may also decrease the overall removal of TrOCs. Wijekoon et al.[10] noted a decrease in the biodegradation of TrOCs with electron-withdrawing functional groups (carbamazepine, triclosan and atrazine) due to salinity build-up in a membrane distillation thermophilic reactor. This was attributed to the unfavorable physical and chemical parameters for oxygen transfer, density, turbidity, viscosity, salt precipitation, solute interaction, and colloid chemistry at elevated salt concentrations. Such changes affect the biological activity of nitrification bacteria[17] and lead to lower degradation of carbamazepine and other nitrogenous compounds that nitrifying bacteria remove[48,196].

There are more constraints of salinity build-up on removing biological nutrients (i.e., nitrogen and phosphorus removal by nitrifiers, denitrifiers and phosphorous accumulating organisms). These microorganisms are affected by increasing salinity, with denitrifiers being the most salt tolerant, while phosphorus-accumulating organisms are the least tolerant[197]. Uygur and Kargi[197] observed a continuous decline in phosphorus removal efficiency with salt accumulation, and this was attributed to the increase in the microbial cells' osmotic pressure, which diminishes their phosphate accumulating capability. Similarly, nitrifiers are also impacted by salinity build-up. However, it is not clear whether nitrite oxidizers are more susceptible to

Fig. 11 | Biological removal of trace organic compounds. Effects of salinity build-up on the removal of TrOCs by MBRs[48]; error bars present standard deviations.
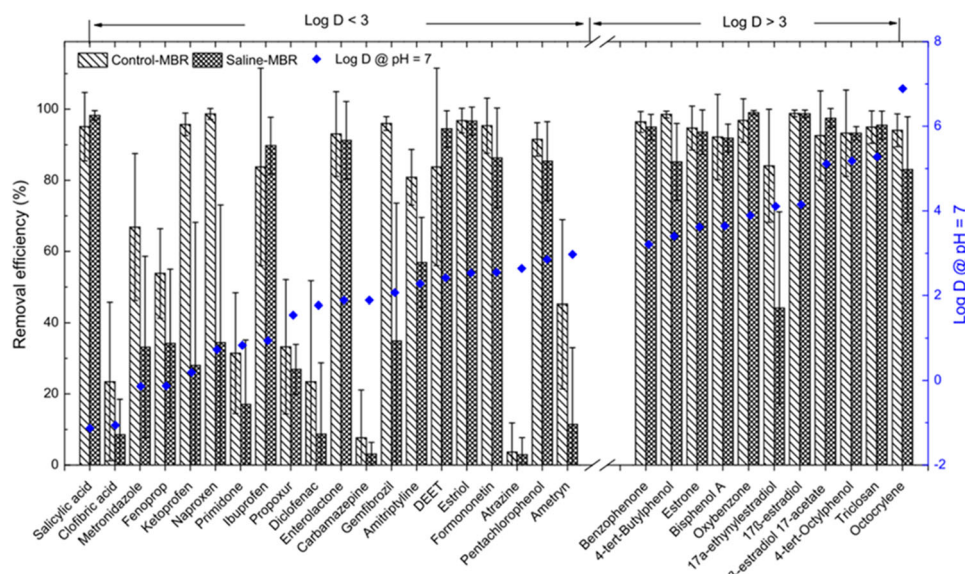
salt effects than ammonia oxidisers[198,199]. A decrease in ammonia removal due to salinity build-up has been reported and ascribed to a loss in biological activity[200,201].

The effects of salinity build-up on the rejection of TrOCs during wastewater treatment have not been well investigated. However, the performance of biological treatment for wastewater under salty conditions (30–200 mg/L NaCl) has been reviewed by Luo et al.[15], who linked salt build-up to the removal of total Kjeldahl nitrogen and chemical oxygen demand. Halophilic microbes removed 63–95% of chemical oxygen demand and 20–96% of total Kjeldahl nitrogen from high-salinity wastewater. In another study, Tay et al.[202] did not find any impact of salinity build-up on the performance of NF-MBR as the system had similar biodegradation efficiency as well as excellent organic (>97%) and ammonia removal (>98%) at low and high salinity conditions. However, salt accumulation aggravated membrane fouling. Although these studies did not investigate TrOCs removal, the performance of the halotolerant microbes under high salt concentrations encountered in these works still provides useful and relevant information for our objective of relating high salt effects on removal of TrOCs by HR-MBRs. Salinity build-up may reduce the removal of TrOCs by promoting fouling, thus decreasing flux due to cake-enhanced concentration polarization effects[81]. To some extent, the rejection of solutes, including TrOCs by RO and NF membranes, is controlled by flux as predicted by the solution-diffusion model[46,58]. Based on the the van't Hoff equation ($\pi = \sum(v_i c_i RT/M_{wi}$; where π is osmotic pressure, v_i is factor for mole increase due to dissociation of the dissolved salts, c_i is concentration in g/L, R is resistance, T is temperature and M_{wi} is molecular weight), at 50 g/L NaCl, the osmotic pressure is greater than 40 bar[17], and this will affect flux in RO (flux decline) unless the applied pressure is increased, leading to more operational costs. Further, this salt concentration and osmotic pressure would be problematic in OMBR, which operates on an osmotic pressure driving force. Linking the retention of TrOCs to membrane flux, Mahlangu et al.[163] found that carbamazepine and NaCl rejection by fouled NF270 membranes decreased with flux due to fouling and cake-enhanced concentration polarization. However, fouling may also promote the removal of TrOCs by high-retention membranes (Table 11), but if the biological activity is compromised, the problems with TrOCs in our ecosystem will be transferred from water treatment to sludge management (unless halotolerant microbes are used). In OMBR, the removal of low molecular weight TrOCs may be affected by a decline in microbial activity because their elimination depends mostly on biological degradation[186]. Luo et al.[48] investigated the impact of salinity build-up on MBR's performance regarding removing TrOCs, nutrients and biomass characteristics. They

noted that salt accumulation decreased the removal of hydrophilic TrOCs, organic carbon and nutrients by MBR, while the removal of hydrophobic TrOCs was unaffected (Fig. 11).

In OMBR, salinity build-up is also due to the reverse salt flux of solutes from the draw solution across the membrane interface to the feed side (bioreactor)[203]. Reverse salt flux results in the loss of the draw solute. Draw solutions made from divalent cations such as MgCl₂ and MgSO₄ result in lesser reverse salt flux compared to draw solutions prepared from monovalent ions such as NaCl and KCl. This is due to divalent ions' lower diffusion coefficient than monovalent ions. In addition, to reverse salt flux, OMBRs have problems associated with the contamination and dilution of draw solution, and this reduces the osmotic driving force leading to lower fluxes. Dilute draw solutions are reconcentrated using desalination techniques such as RO and MD, but this adds to the capital and operational costs. However, the reconcentration of draw solutions eliminates the challenges associated with brine disposal in NF/RO and MD systems. Thus, coupling OMBR with NF/RO or MD would be more beneficial, except that desalination processes also reject other solutes besides salts, and these concentrated solutes may contaminate the draw solution and change its chemistry.

Besides the effects of salt build-up on the biological process, the membrane processes can also be affected. In pressure-driven processes (NF/RO), salt build-up leads to permeate flux decline through concentration polarization. Similarly, in FO the accumulation of salts in the feed (due to reverse salt flux) may lower the concentration gradient leading to the reduction in permeate flux. However, unlike NF/RO processes, salinity build-up can be prevented by changing the draw solute. In MD, salt build-up could lead to rapid flux decline due to crystal deposition and scale formation on the membrane surface and this reduces membrane permeability. Generally, the main issue in MD is temperature polarization which affects crystallization. Therefore, controlling temperature polarization may reduce the effects of salt build-up on the performance of the membrane. In MD, there are two main challenges associated with the treatment of high salinity feed streams. The first problem is attachment of fouling agents onto the hydrophobic membrane surface leading to blockage of the membrane pores, and consequently cause significantly reduced water vapor flux. Secondly, when MD is used to treat saline water containing amphiphilic molecules, the hydrophobic tails of the amphiphilic molecules attach onto the hydrophobic membrane pore surface, leaving the hydrophilic head exposed and eventually rendering the membrane pores hydrophilic[108]. This eventually promotes wetting of the membrane pores leading to immediate failure in the separation performance of the membrane. Salinity build-up together with

membrane fouling impact energy efficiency in NF/RO processes, while they result in immediate failure in membrane separation in MD.

4.2. Alleviating salinity build-up and reverse salt flux in HR-MBRs

4.2.1. Maintaining optimum concentration factor.
The concentration factor can be loosely defined as the concentration increase of inorganic salts in the bioreactor and can be used to control and optimize salinity build-up in HR-MBRs[17]. Since the concentration factor is related to the sludge retention time and hydraulic retention time, maintaining a lower concentration factor can reduce salinity build-up, which also implies lower water recovery[15]. As Lay et al.[17] recommended, the optimum concentration factor should be 10–30, corresponding to 5–15 g/L total dissolved solids (assumed for wastewater with total dissolved solids of 500 mg/L). These values were estimated for 10–30 days of sludge retention time and hydraulic retention time lower than 1 day.

The frequent withdrawal of the sludge can also control salinity build-up. However, this may not be ideal for HR-MBRs, which need to operate at longer sludge retention time. Further, periodic sludge withdrawal may disturb the biological media and lead to other problems associated with sludge management. Sludge treatment is not a focus of this work. However, details on sludge treatment and handling can be found in the work of Zhang et al.[204].

Intermittent filtration with MF or UF membranes.
Intermittent filtration with MF/UF membranes may be performed to control salinity build-up in HR-MBRs. An additional MF/UF is incorporated in this process, and liquid media is ejected periodically through the MF/UF membranes[205]. This process not only removes salts (due to poor salt removal capabilities of MF/UF membranes) but also reconcentrates the microbes rejected by the MF/UF membranes, which is expected to maintain high biodegradation activity. Holloway et al.[206] used a unique strategy to mitigate salt accumulation where an ultrafiltration membrane was applied parallel to a forward osmosis membrane in the same OMBR. This approach ensured low salinity was maintained in the activated sludge (implying uninterrupted microbial activity).

The recovery of nutrients was not compromised, and membrane fouling was prevented (by reducing the concentration of cations that exacerbate fouling). These benefits may outweigh the additional capital, operational and maintenance costs associated with incorporating MF/UF process in HR-MBRs to control salt concentrations in the bioreactor. A similar approach of periodic MF extraction for nutrient recovery and salinity maintenance in OMBR coupled with RO was adopted by Luo et al.[207]. The authors made similar conclusions to the works of Holloway and co-workers. Since acclimation of microorganisms is sensitive to fluctuations in salt concentration, intermittent MF ensures operation at constant salt concentration, thus allowing the system to be operated at longer sludge retention time. The gradual accumulation of extracellular polymeric substances must be monitored under such circumstances to ensure negligible fouling. However, a remarkable increase in extracellular polymeric substances is not envisaged under low salinity conditions.

Optimization of draw solute in OMBRs.
Reverse salt flux in OMBRs can be reduced by using divalent ions in draw solutions, bearing in mind that water fluxes from such solutions would be lower than those of draw solutions prepared from monovalent ions at similar osmotic pressure[188,208]. Similarly, salts containing sulfate ion (SO_4^{2-}) may produce hydrogen sulfide and sulfuric acid, resulting in scaling that fouls the membrane[209]. Exacerbated fouling with the sulfur build-up was reported by Song et al.[210], and this was ascribed to the release of carbohydrates and proteins (constituents of soluble microbial by-products and extracellular polymeric substances, respectively) at elevated sulfur concentrations. As a result, membrane fouling occurred through cake formation and pore blocking[211]. Further, the build-up of sulfur has been associated with a decrease in the removal of hydrophilic TrOCs[210]. The extent of reverse salt flux can be reduced by adding small concentrations of salts having divalent or organic ions (e.g., magnesium chloride and sodium acetate) to draw solutions prepared from salts having monovalent ions (e.g., NaCl)[208,212].

Using ammonium carbonate (NH_4HCO_3) is another option to reduce salinity build-up in OMBRs because nitrification and denitrification processes in the bioreactor remove both ammonia (NH_3) and carbon dioxide (CO_2). Although this may be a solution, a decrease in the pH of the activated sludge may occur due to the fast diffusion of CO_2 across the membrane interface to the bioreactor, affecting nitrifying microorganisms at lower pH[213]. Organic draw solutions such as glucose, glycine and sodium acetate have been proposed to reduce the intoxication of microorganisms and fouling due to inorganic scaling. Organic draw solutions have lower reverse solute flux compared to inorganic salts. Further, their escape from the draw solution to the bioreactor would not affect microbial activity since they are biodegradable[214]. However, their main challenge is lower fluxes due to lower osmotic pressure.

Alleviating losses in biological activity.
Losses in biological activity due to salinity build-up can be mitigated or compensated for by adopting one of the two strategies: acclimation and inoculation of halophilic or halotolerant microorganisms, where the former is suggested for salt concentrations up to 30 g/L. At the same time, the latter is recommended for salt concentrations of 30–150 g/L[17]. Due to the extremely longer time required for acclimation, the inoculation of halotolerant microorganisms may be a better option[215]. However, the challenge here is to get the right type of microbe according to the salt range[17]. Luo et al.[189] found that halotolerant bacteria (such as *Methylibium*, belonging to the family *Comamonada ceae*) proliferated and became more abundant with the increase in salinity from 0.4 to 13.3 mS/cm in the bioreactor permitting continuous biodegradation of pollutants. It must be noted that some halotolerant microbes have a threshold salt concentration limit[17]. For instance, some genus from the family *Cytophagaceae* increased up to 45.5% when the feed electrical conductivity was elevated to 11 mS/cm but decreased to 32.6% as the conductivity of the mixed liquor was raised further[189]. The concept of halotolerant microbes in HR-MBRs has not been well investigated. Therefore, more research is needed to understand all the influencing factors better.

Incomplete biodegradation and removal of TrOCs
Incomplete degradation of TrOCs in the bioreactor
Conventional MBRs use bacteria which have limitations to degrade TrOCs completely. This leads to the build-up of rejected TrOCs in the bioreactor and transfers the problem to sludge management. In HR-MBRs, partial biodegradation may increase TrOCs concentration in the feed, potentially leading to their poor rejection by high-retention membranes[114]. Insufficient biodegradation of TrOCs can be mitigated by making biological agents stronger. This has motivated the introduction of the concept of enzymatic membrane bioreactors where enzymes such as laccase have shown better performance in degrading TrOCs than conventional bacteria[216–219]. Although complete degradation of some compounds is achieved, recalcitrant TrOCs still resist complete degradation by enzymes hinting at the need for further improvement. This has prompted redox-mediators' inclusion in enzymatic MBR to enhance biodegradation further[220–222].

Another option to improve the biodegradation of TrOCs is to choose microorganisms with high activity in degrading the target TrOCs. In a recent study, horseradish peroxide enzyme was used in an enzymatic MDBR and degradation greater than 99% was achieved for some antibiotics[165]. Horseradish activity was improved further when hydrogen peroxide (H_2O_2) was added. The performance of different microorganisms on the same feed may need to be compared for the accurate selection of appropriate microorganisms. Newer research studies have investigated the use of an additional concentrate treatment step utilizing advanced oxidation processes. The works of Tufail et al.[109] investigated the removal of TrOCs by an enzymatic MBR incorporating a membrane distillation process coupled with UV photolysis. The photolysis step was used to degrade the concentrate

from the MD process, and some TrOCs, such as sulfamethoxazole, bisphenol A and diclofenac, were completely degraded, thus solving the challenges of incomplete degradation in the bioreactor.

The enzymatic activity of ligninolytic enzymes and their catalytic efficiency is inhibited by some organic and inorganic compounds derived from wastewater. For example, oxalic acid and ethylenediaminetetraacetic acid at 1 mM inhibit white-rot fungus and their extracellular enzymes[223]. A detailed review of the mechanisms and factors controlling the interactions of interfering compounds with microbes and their enzymes can be found in the literature[143]. It is important for future research to fully characterize all the parameters that affect enzymatic activity so that complete biodegradation of TrOCs can be achieved in enzymatic MBRs without the need for additional expensive processes.

Incomplete rejection of TrOCs by membranes

Although HR-MBRs perform better than conventional activated sludge and MBRs, incomplete removal of TrOCs has been reported[47,210,216,224]. TrOCs in the permeate of the high-retention membranes (though at a lower concentration) and biodegradation by-products may pose a health risk to humans. To ensure the complete removal of toxic TrOCs, the latest research trend has focused on coupling HR-MBRs with other techniques, such as advanced oxidation processes. The advantage of such techniques is that they enhance the biodegradability index of pollutants by decomposing complex compounds, which in turn increases.

García–Gómez et al.[225] coupled MBR with electro-oxidation (an advanced oxidation process). They achieved almost complete removal of carbamazepine (a recalcitrant compound) through the oxidation of carbamazepine with the hydroxyl radicals ($^*OH$) formed near the high voltage O_2-overvoltage anode. Electrocoagulation processes have also been combined with MBR processes to enhance the removal of TrOCs while reducing membrane fouling by increasing floc size and particle polarization due to electrocoagulation and the applied electric field. Improvement in removing acetaminophen, clarithromycin, and carbamazepine was achieved in an MBR-electrocoagulation coupled process[226]. Another example of utilizing an advanced oxidation process is incorporating a bio-electrochemical system to target TrOCs that are not easily degraded by microorganisms. In this process, the TrOCs are transformed into intermediate products that are then biodegraded by the microorganisms. The concept of coupled membrane bio-electrochemical reactor has already been reported to improve the removal of sulfamethoxazole by 21%[227]. Ultraviolet (UV) photolysis is another advanced oxidation process that can be incorporated into HR-MBRs. A study on removing TrOCs by an enzymatic MBR preceding UV photolysis reported complete degradation of the contaminants while producing fewer by-products[9]. These findings demonstrated that advanced oxidation processes could be used as a polishing step for TrOCs that permeate through the membrane during wastewater treatment by HR-MBRs. The addition of persulfate in the bioreactor seems to improve the degradation of TrOCs. A study focusing on the persulfate oxidation-assisted membrane distillation process found that persulfate-assisted direct contact membrane distillation removed more than 99% of all the investigated TrOCs. In comparison, the control without adding persulfate had lower removals (86–99%)[11].

Although advanced oxidation processes successfully degrade TrOCs, their effectiveness may be negatively impacted by the presence of halogens in the feed. For instance, bicarbonate ion inhibits the photodegradation of sulfamethoxazole, while diclofenac degradation is reduced in the presence of chloride and nitrate ions[228]. Further, most advanced oxidation processes are costly and may produce degradation by-products that may be more toxic than the parent compound[109].

Capital and Operational cost

Another challenge with HR-MBR is high cost associated with capital, operational and maintenance of the biological and membrane processes. The cost of each process is controlled by several parameters such as plant capacity, power requirements, plant configuration, chemistry of the feed, labor and treatment requirements amongst others[229]. There is limited information on the peer-reviewed information on capital expenditure (CAPEX) or cost of biological processes as well as high-retention membrane processes, while there is focus on energy demand. In wastewater treatment, energy demands contribute to 27–34% of the operational expenditure (OPEX) for treatment and aeration energy[230] in a large MBR (19–49 megaliters per day, capacity). Working at full capacity helps lower OPEX associated with aeration by minimizing the specific aeration demand while maximizing flux. Further, the energy for mixing and biological aeration can be reduced by operating at low solid concentration. Based on the design capacity, the capital cost of MBR with flow capacity of 100, 500 and 2 500 m^3/d is estimated at 0.6, 0.48 and 0.38 \$/L/d, respectively; while the operational costs are approximately 0.58, 0.45 and 0.4 \$/m^3 for 100, 500 and 2500 m^3/d plants, respectively[230]. Interestingly, small plants have higher energy demands. For example, a 100 m^3/d plant has specific energy demand of about 2 kWh/m^3 while a 2500 m^3/d plant requires half of that. DeCarolis et al.[231] performed cost analysis of MBR plants supplied by Koch Membrane Systems, Huber Technologies Inc., Parkson Corporation and Kruger Inc with design capacities of 4 000 to 20 000 m^3/d. The estimated CAPEX ranges from \$7 990–\$9 850 for a 4 000 m^3/d plant and \$32 270–\$38 790 for a 20 000 m^3/d plant. The yearly total OPEX was estimated at \$3 350–\$4 649 and \$14 974–\$20 344 for plants with design capacities of 4 000 and 20 000 m^3/d, respectively.

High-retention membrane processes are also associated with high CAPEX and OPEX. In the literature, the cost of high-retention membranes is presented for desalination technologies, while this work focuses on wastewater treatment where the salt content is lower. Therefore, the operational costs are expected to be lower than those of brackish and/or seawater desalination plants using high-retention membranes. The CAPEX for RO plants with treatment capacity of 1 000 m^3/d to 290 000 000 m^3/d ranges between 0.1 and 293 million US\$, while the OPEX for the same plants is between 0.2 and 1.2 US\$/m^3. Such plants have total energy requirements between 0.5 and 4.0 kWh/m^3. NF plants with design capacity of 2 400 to 100 000 m^3/d require 1–17 million US\$ to install and OPEX of 0.16 – 0.30 US \$/m^3. Their energy requirements range from 0.01–2.4 kWh/m^3. MD plants are assumed to have lower capital cost than NF/RO due to the use of inexpensive plastics as construction material[232]. Khan et al.[233] performed a techno-economic assessment of MD integrated to an anaerobic digestion biogas plant. The plant with design capacity of 190 m^3/d in full-scale operation had CAPEX of about 375 000 US\$ with thermal energy demand of 22 MWh/d. Further, the maintenance and operating cost was 375 700 US \$/year for the MD. External thermal energy costs contributed about 90% of the total maintenance and operating expenditure. Installation of FO is also costly. However, CAPEX makes up a large part while operating and maintenance costs contribute a small fraction to the total cost[234]. According to Le and Nunes[235], high CAPEX is associated with membranes that have low power density due to the requirements for huge membrane area and high membrane installation costs. A membrane with a power density of 1 W/m^2 has a capital cost of 20 000 US\$/kW but the capital cost of a membrane with power density of 5 W/m^2 would be 4 000 US\$/kW[236].

Feasibility assessment and effectiveness of control strategies

Table 13 summarizes the challenges and mitigation strategies in HR-MBRs. The feasibility and effectiveness of the control strategies are also assessed. The proposed solutions appear effective in mitigating problems associated with HR-MBRs; however, their main common drawback is the high cost which would make them unsustainable. Further, some of the processes still need further investigation for improvement before commercialization.

Research gaps and future directions

This work highlighted the major challenges associated with HR-MBRs and their respective potential corrective measures. In the past few years, substantial work has been conducted to enhance the performance of HR-MBRs.

Table 13 | Summary of challenges and their associated effects in HR-MBR as well as corrective approaches

Challenge	Effect on HR-MBR	Corrective measures and effectiveness	Drawbacks
Fouling of high-retention membranes	○ Fouling leads to flux decline, making it hard to meet consumer demands. ○ Fouling results in membrane degradation, which leads to a shorter membrane lifespan. ○ Fouling may potentially decrease the removal of TrOCs.	○ Pre-treatment of feed with UF/MF or low-energy FO membranes reduces the fouling of high-retention membranes. ○ Membrane cleaning with chemicals or clean water restores membrane performance when the fouling is reversible. ○ Membrane surface modification reduces membrane fouling by making membrane-foulant interactions repulsive. ○ Optimization of system and operating conditions delay fouling onset, e.g., operating below critical flux.	○ Adding pre-treatment steps increases capital and operational costs. ○ Chemical cleaning degrades membranes and shortens their lifespan. ○ The potential leaching of nanoparticles when nanoengineered membranes are used may lead to secondary pollution. ○ Operating at critical flux may not be sufficient to meet consumer demands.
Salinity build-up in the bioreactor due to high salt rejection by membranes and reverse salt flux in OMBR	○ Salt build-up in the bioreactor deteriorates microbial activity leading to poor degradation of TrOCs in the bioreactor. ○ Salt accumulation decreases the driving force in OMBR, resulting in low flux. ○ Salt build-up increases membrane fouling by promoting the formation of extracellular polymeric substances and soluble microbial by-products. ○ An increase in salinity may potentially decrease the removal of TrOCs.	○ Acclimation and inoculation of halotolerant or halophilic microorganisms ensure that biological activity is maintained. ○ Maintaining the optimum concentration factor reduces salinity build-up. ○ Frequent sludge withdrawal helps maintain low salt concentrations. ○ Intermittent filtration with MF/UF membranes discharges salty streams and prevents salt accumulation. ○ Using organic salts instead of inorganic salt reduces reverse salt flux in OMBR. Alternatively, salts with divalent ions may be used.	○ Additional capital and operational costs when MF/UF are added as pre-treatment steps. ○ Frequent sludge withdrawal affects the concentration factor due to low sludge retention time. ○ Low osmotic pressure when organic salts or salts with divalent ions are used leads to low permeate flux.
Incomplete biodegradation of rejected recalcitrant TrOCs in the bioreactor	○ Incompletely degraded TrOCs in the bioreactor transfers the problems associated with toxic TrOCs from water treatment to sludge treatment and management. ○ Poor degradation of TrOCs may negatively affect their removal by membranes due to concentration polarization effects.	○ Using stronger biological agents (e.g., enzymes) improves biodegradation. ○ Appropriate selection of microorganisms according to the characteristics of the target TrOCs can help achieve high degradation. ○ Coupling HR-MBR with advanced oxidation processes improves the quality of the final effluent. ○ Adding appropriate mediators enhances the activity of biological agents.	○ Commercial biological agents are costly, and the purification process is lengthy and complex. ○ Appropriate selection of microbes is a clever idea, except that the water quality changes over time. The feed will need to be monitored frequently. ○ Advanced oxidation processes are costly; their application would result in high operational costs. ○ Potential formation of by-products when integrating HR-MBR with advanced oxidation processes may limit their application when the by-products are more toxic than the parent compound.
Capital and operational cost	○ The high cost of HR-MBRs (capital and operational expenditure) make them unaffordable by low-income countries.	○ Development of more robust membranes that will have longer life-span. ○ Developing energy-efficient membranes to lower energy demands and cost of water production. ○ Adopt less expensive technologies that can be constructed from inexpensive plastic material (e.g., MD).	○ Tailoring membrane properties to achieve low energy demands often compromises their TrOCs removal efficiency. ○ High-pressure membrane processes cannot be designed using inexpensive plastic materials.

However, challenges and research gaps still need further investigation for improvement.

There is a need to improve cleaning approaches to ensure that the membrane integrity is not compromised after repeated cycles of cleaning. Where possible, environmentally-friendly cleaning techniques must be adopted to ensure longer membrane service life-span. Ideally, fouling prevention methods must be utilized to reduce fouling and avoid chemical cleaning. Amongst these methods, advanced oxidation processes (AOPs) such as sonication-assisted filtration and electrochemical coagulation hold great potential for future application. Therefore, more research is needed for further optimization before commercialization of these methods.

More research ought to be conducted on the use of more economical yet effective polymers or their mixtures with more established membrane materials to lower capital costs. A blending of hydrophobic polymers could enhance hydrophobicity in MD membranes. In this case, a hydrophobic polymer is mixed with a small amount of a more hydrophobic polymer. For example, Su et al.[237] fabricated PTFE hollow fiber membranes for MD application. PTFE was chosen because of its high chemical resistance, thermal stability, and hydrophobicity. The fabricated membrane was highly hydrophobic (water contact angle of 164°) and had high flux. The membrane flux was stable over long-term experiments and elevated salinity, indicating its potential for MD application and treatment of high-salinity wastewater. In a recent study, Burnwal et al.[238] prepared a polyvinylidene fluoride–polytetrafluoroethylene (PVDF–PTFE) hybrid flat sheet membrane to separate oil-in-water emulsions through MD. PVDF was used as the base polymer while PTFE was added as a copolymer in varying mass between 1 and 2 wt%. Mixing the polymers made the membranes more hydrophobic, with contact angles increasing from 88° for the pure PVDF membrane to 117° for the blended membrane, making it more suitable for MD operation. These findings hint at the possibility of combining polymers to enhance membrane properties. However, more research is still needed to evaluate the cost and applicability of membranes with polymer blends in wastewater treatment. There are preliminary studies in line with these concepts, but these are still far from commercialization.

In OMBR processes, the regeneration of the draw solution is energy intensive and reduces the advantage of energy saving by FO. The use of

alternative energy sources, such as harvesting and applying geothermal and low-grade heat energy (e.g., industrial waste heat and solar), needs further investigation to reduce the total energy costs of OMBR and MDBR processes[189]. Another issue with OMBR is severe reverse salt flux when monovalent salt ions are used. Using monovalent salt as a draw solution produces higher flux than divalent salt, but higher reverse salt flux for monovalent salt decreases the driving force across the membrane leading to lower permeate flux. Holloway et al.[208] lowered reverse salt flux and maintained high flux by adding small concentrations of salts having divalent or organic ions (e.g., magnesium chloride and sodium acetate) to NaCl in the draw solution. This was attributed to the low diffusivity of divalent ions, which inhibited sodium and chloride ions from diffusing through the membrane pores at a high rate. The water flux was also enhanced slightly when a low concentration of $MgCl_2$ was introduced into a draw solution of NaCl. In another study, Nguyen and co-workers[212] kept the reverse salt flux to a minimum of 0.13 g/m²h by combining a surfactant (Triton X100) and sodium phosphate in a forward osmosis desalination system. Further, the mixture of Triton X100 and sodium phosphate was easily recovered through a UF–NF system that rejected 98% of the solutes. The use of draw solutions combining various solutes in OMBR needs further investigation, and the effects on the performance of the bioreactor and the high-retention membranes must be evaluated.

Salt can gradually build up in all HR-MBR due to salt retention in raw wastewater and deteriorate biological activity[186,187]. Intermittent withdrawal of bioreactor media through MF/UF membrane can be done to prevent the build-up of salt[205,239]. Further, halotolerant or halophilic microorganisms can be used to ensure that biodegradation is not compromised[215]. Frequent sludge withdrawal has been proposed as another option to prevent salinity build-up by maintaining a low concentration factor. However, this approach may not be suitable as it implies a lower water recovery[15].

Incomplete degradation of rejected recalcitrant TrOCs can be mitigated by using microbes of special degrading capacity—this has led to the concept of using enzymes from suitable microbes in the bioreactor (enzymatic membrane bioreactors)[9,217,218,222,240]. Redox mediators are added in the bioreactor to strengthen the biological activity of enzymes further; however, some reports show that the mediator-enhanced enzymatic process may degrade the parent compound but produce toxic effluent streams. For example, Nguyen et al.[221] studied laccase-syringaldehyde meditated degradation of TrOCs in an enzymatic bioreactor: the treated effluent showed remarkable toxicity at syringaldehyde concentrations greater than 10 μM. Therefore, more research is needed to establish safe working concentrations for the redox mediator-enhanced enzymatic process. On the other hand, membrane bioreactor processes can be coupled to other treatment techniques, such as AOPs; however, such concepts are still in their infancy. Further, incomplete biodegradation of TrOCs retained within HR-MBR shifts the problem from wastewater treatment to sludge treatment and management. The option of using advanced oxidation processes (AOPs) to treat the concentrated TrOCs-sludge may be considered; however, some inherent limitations of AOPs, such as high cost and the potential production of disinfection by-products, need to be tackled.

The application of separation membranes is influenced by the material surface wettability and microstructure amongst other parameters. The membrane vapor flux is controlled by microstructural properties including average pore size and pore size distribution, pore structure, and porosity. In addition to desirable microstructure, hydrophobicity is another feature determining the application of separation membranes. For potential MD applications, hydrophobic materials with high water contact angles and small maximum pore size are required to avoid wettability of the membrane pores. However, this often leads to low MD permeability. Contrarily, high-pressure-driven (NF/RO) and osmotic pressure-driven (FO) processes require hydrophilic materials with small pore sizes to achieve high flux and separation performance. The challenges in HR-MBR (pore wetting in MD, membrane fouling and incomplete retention of TroOCs amongst other challenges) are associated with membrane properties (material

wettability, microstructure, thickness etc). Therefore, there is a need for research to further fine-tune the membranes to achieve the desirable properties to advance the separation efficiency (targeting poorly removed TrOCs like carbamazepine) and fouling prevention. This could also be achieved through developing novel membrane materials with the desirable physicochemical properties for the respective application.

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The authors declare no competing interests.

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