



Microplastics with adsorbed contaminants: Mechanisms and Treatment

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ABSTRACT

Plastic pollution has been a significant and widespread global issue, and the recent COVID-19 pandemic has been attributed to its worsening effect as plastics have been contaminated with the deadly infectious virus. Microplastics (MPs) may have played a role as a vector that carries hazardous microbes such as emerging bacterial threats (i.e. antibiotic resistant bacteria) and deadly viruses (e.g., coronavirus); this causes great concern over microplastics contaminated with emerging contaminants. Mitigation and treatment of MPs are challenging because of a range of factors including but not limited to physicochemical properties and composition of MPs and pH and salinity of the solution. Despite the heterogeneous nature of aquatic systems, research has overlooked interactions between contaminants and MPs under environmental conditions, degradation pathways of MPs with adsorbed contaminants, and, especially, the role of adsorbed contaminants in the efficiency of MP treatment through membrane filtration, in comparison with other treatment methods. This review aims to (1) analyze an assortment of factors that could influence the removal of MPs and mechanisms of contaminant adsorption on MPs, (2) identify mechanisms influencing membrane filtration of MPs, (3) examine the fate and transport of MPs with adsorbed contaminants, (4) evaluate membrane filtration of contaminant-adsorbing MPs in comparison to other treatment methods, and (5) draw conclusions and the future outlook based on a literature analysis.

1. Introduction

Increasing plastic production is an unmanageable issue in view of disposing plastic contaminants safely and mitigating their adverse effects on the environment and the risk of deteriorating public health. The majority of plastic debris and microplastics (MPs) are found in almost all aquatic media, ranging from the ocean to freshwater lakes and rivers (Hamid et al., 2018). In the significant amount of plastic debris, MPs (<5 mm in size) are found as the major component of plastic pollution (Wardrop et al., 2016).

Widespread distribution of MPs in the ocean is attributed to the intrinsic properties (e.g., density, surface charge, aggregation, size, and color) of MPs, as well as abiotic (e.g., oxidation) and biotic (e.g., biofouling) factors (Lusher et al., 2015). While primary MPs are defined as tiny particles that were manufactured for commercial use, secondary MPs are classified upon fragmentation of larger plastic items

(Lehtiniemi et al., 2018). Although primary MPs ideally maintain their size, generally from 0.1 to 5,000 μm , they undergo fragmentation and become debris, in which case their particle size ranges from 1 to 100 nm (nanoplastics, NPs). This turns out to be problematic because of difficulty of quantification and identification of nanoplastics (NPs), particularly in edible items (EFSA Panel, 2016).

Sources of MPs range from fragments of plastic products due to environmental degradation to the release of microbeads present in cosmetics that are found in effluents of wastewater treatment plants (Kalcíková et al., 2017). According to a study by Kalcíková et al. (2017) around half of microbeads were retained in activated sludge with smaller particles (up to 60–70 μm) compared to those detected in effluents, when a lab-based sequencing batch biological wastewater treatment plant was operated.

In the manufacture of plastics, various additives are put in to improve their quality, despite potentially negative effects such as re-

Abbreviations: DDT, dichloro-diphenyl-trichloroethane; DM, dynamic membrane; EDCs, endocrine-disrupting compounds; FOSA or PFOSA, perfluorooctane sulfonamide; GAC, granular activated carbon; HDPE, high-density polyethylene; LDPE, low-density PE; MF, microfiltration; MPs, microplastics; MBR, membrane bioreactor; NF, nanofiltration; NOM, natural organic matter; NPs, nanoplastics; OM, organic matter; PAs, polyacrylates; PA, polyamide (nylon); PAHs, polycyclic aromatic hydrocarbons; PBDEs, polybrominated diphenyl ethers; PCBs, polychlorinated biphenyls; PE, polyethylene; PET, polyethylene terephthalate; PFCs, perfluorinated compounds; PFCAs, perfluorinated carboxylates; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonic acid; PFAS, per-/poly-fluoroalkyl substances; PFHxA, perfluorohexanoic acid; POPs, persistent organic pollutants; PPCPs, pharmaceuticals and personal care products; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride; PVDF, polyvinylidene fluoride; RO, reverse osmosis; SR, synthetic rubber; TMP, trans membrane pressure; UF, ultrafiltration.

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calcitance of persistent organic pollutants (POPs) to degradation, and chemical stability Andradý and Neal (2009). MPs may adsorb contaminants present in environmental media because of the lipophilicity of MPs (O'Donovan et al., 2018). Degradation of MPs into smaller plastic particles may enhance adsorption of contaminants on MPs because more of MPs' surface area is exposed, and their chemical reactivity thus increases. Environmental conditions such as weathering, sunlight, pH, long exposure times and hydrophobicity of POPs may significantly influence kinetics of adsorption of contaminants to MPs (Antunes et al., 2013).

Despite the potential impact of MPs with adsorbed contaminants (c-MPs) on the efficiency of MP treatment and other emerging contaminants that co-exist in the aquatic environment, studies on mechanisms of contaminant adsorption on MPs, the fate and transport of MPs with adsorbed contaminants, and efficiency of MP treatment are rarely found. One of emerging contaminants, per- and poly-fluoroalkyl substances (PFAS), has increasingly posed risk on public health because of their widespread production, use, and resistance to degradation in the environment (U.S. EPA, 2019). Among perfluorinated compounds (PFCs), perfluorooctanoic acid (PFOA) and perfluorooctanyl sulfonate (PFOS) are of particular concern because of not only their prevalent detection in the environment and animal, even human bodies, but also their high stabilization and unknown fate of the contaminants (Wang et al., 2008).

Motivated by the critical advanced need for safe disposal and efficient treatment of MPs under various environmental conditions, this review aims to offer a critical analysis of c-MPs in regard to treatment methods (e.g., membrane filtration vs. other treatment methods, including sedimentation and flotation), adsorption kinetics and pathways of c-MPs, and mechanisms influencing membrane filtration of c-MPs. Major findings from a literature analysis are presented, in conjunction with the future research outlook and challenges to be addressed.

2. Mechanisms of contaminant adsorption on microplastics and factors influencing the removal of microplastics

In treatment of MP-laden water and wastewater, various factors may influence the removal effectiveness of MPs, and some of them include mechanisms of contaminant adsorption on MPs, fragmentations and physicochemical properties of MPs, and treatment unit processes. The mechanisms and factors influencing adsorption of contaminants on MPs, as well as the resultant removal efficacy of MPs, are worthy of investigation towards developing new treatment methods appropriate for MPs with adsorbed contaminants. A list of reviewed contaminants focuses on emerging contaminants and includes, but is not limited to, PFAS and PCBs.

2.1. Mechanisms of contaminant adsorption on microplastics

Emerging contaminants of concern such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), per-/poly-fluoroalkyl substances (PFAS) in environmental media could well be adsorbed on MPs. Of recent particular concern is PFAS, because of the widespread detection of PFAS in surface water, drinking water, and wastewater treatment plants, as well as resistance to degradation and chemical stability of PFAS (Boiteux et al., 2017; Zareitalabad et al., 2013; Post et al., 2013; Zhang et al., 2016; Sun et al., 2019; McCormick et al., 2016; Lv et al., 2019). PFAS have two major types (i.e., PFOS and PFOA) with increasing attention because of their harmful effects on public health and the environment (Ng et al., 2019).

Because of their hydrophobic nature, MPs easily adsorb contaminants (e.g., PBDEs, EDCs, and PPCPs), along with other POPs in aqueous media. The adsorption of contaminants may occur on MP surfaces because of the large surface area and hydrophobicity of MPs, further

inducing pollutants attached on MPs upon release of MPs to the environment. The adsorption (desorption) of contaminants on (from) MPs is complicated in heterogeneous environmental conditions, because of a mixture of dynamic factors such as MP characteristics (e.g., composition/type, structure, binding energy, and surface properties), release medium (e.g., pH, temperature, salinity, and ionic strength), and contamination factors (e.g., solubility, redox state, charges, and stability) (Verla et al., 2019; Yu et al., 2019; Wang et al., 2020; Li et al., 2018a).

As an example, upon adsorption of benzo(a)pyrene on polyvinyl chloride (PVC) MPs, a time- and dose-dependent adsorption kinetics was demonstrated to have the highest toxicity, compared with bare MPs or benzo(a)pyrene alone, indicating the significant role of MPs as a vector for organic pollutants in sediments and the potential synergistic effect of contaminant-adsorbing MPs (Gomiero et al., 2018; Caruso, 2019). The contaminants may affect the transformation of MPs into byproducts containing plastic particles in various environmental conditions, but this information is not found in the literature. As well as regarding the mechanisms of contaminants adsorbed on hydrophobic adsorbents, the mechanisms primarily involved in c-MPs could involve hydrophobic interaction, electrostatic repulsion and attraction, pore blockage, and site competition (Ma et al., 2019a; Ma et al., 2019b; Enfrin et al., 2020; Thomas et al., 2016; Hadidi et al., 2014; Huisman et al., 2000; Bellona and Drewes, 2005; Breite et al., 2016a; Breite et al., 2016b; Fotopoulou and Karapanagioti, 2012).

The hydrophobicity (K_{OW}) and weathering/aging of MPs were identified as mechanisms of contaminant adsorption on MPs (Wang et al., 2018). The adsorption/desorption kinetics may differ, depending on types of MPs, e.g., polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC), with PE (rubbery polymer PE) having higher adsorption than those of other types of MPs (Alimi et al., 2018). Fig. 1 (a) illustrates mechanisms of PFAS adsorption on MPs. A PFAS molecule with a negatively charged head and a hydrophobic C-F chain maintains chemical stability even under high temperature (Du et al., 2014; Vecitis et al., 2009). The mechanisms of PFAS adsorption on MPs could involve electrostatic and hydrophobic interactions, which primarily govern adsorption of PFAS on several adsorbent materials (Gagliano et al., 2020).

Hydrogen bonding and covalent bonding may also occur during interactions between PFAS and adsorbents (Gagliano et al., 2020). Depending on the surface charge of adsorbents, electrostatic repulsion (with a negative adsorbent surface charge) or electrostatic interaction (with a positive adsorbent surface charge) occurs. The electrostatic interactions appear to be dominant for short-chain PFAS, whereas adsorption through hydrophobic interactions takes place on longer PFAS, favoring the molecular aggregate of PFAS on the active surface of the adsorbent (Deng et al., 2012a; Deng et al., 2012b; Zaggia et al., 2016). As reviewed in the literature, these mechanisms are likely to depend on not only the types and physicochemical properties of contaminants adsorbed on MPs but also the types of MPs, as factors to consider in removal of c-MPs.

The environmental presence of organic matter (OM) may influence the adsorption of both long- and short-chain PFAS on MPs, because of complexation of PFAS with OM or co-sorption (Gagliano et al., 2020; Ateia et al., 2020). Thus, PFAS adsorption on OM in the presence of MPs could occur through the interaction of OM-adsorbed MP surfaces with PFAS through electrostatic or hydrophobic interactions. As illustrated in Fig. 1 (b), when organic matter (OM)—mostly composed of anionic species—adsorbs on materials via repulsive electrostatic interactions, in the meantime PFAS adsorption on OM occurs via hydrophobic interaction between perfluoroalkyl tail and OM-adsorbed adsorbent surfaces (Du et al., 2014; Gagliano et al., 2020). Although hydrophobic OM was found to enhance PFAS retention in granular activated carbon (GAC) adsorption, hydrophilic OM did not appear to significantly affect PFAS adsorption on both GAC and anion exchange (AE) (Kothawala et al., 2017). Nonetheless, several studies indicated that the presence of OM may reduce the sorption of PFAS on active carbon fiber, because OM

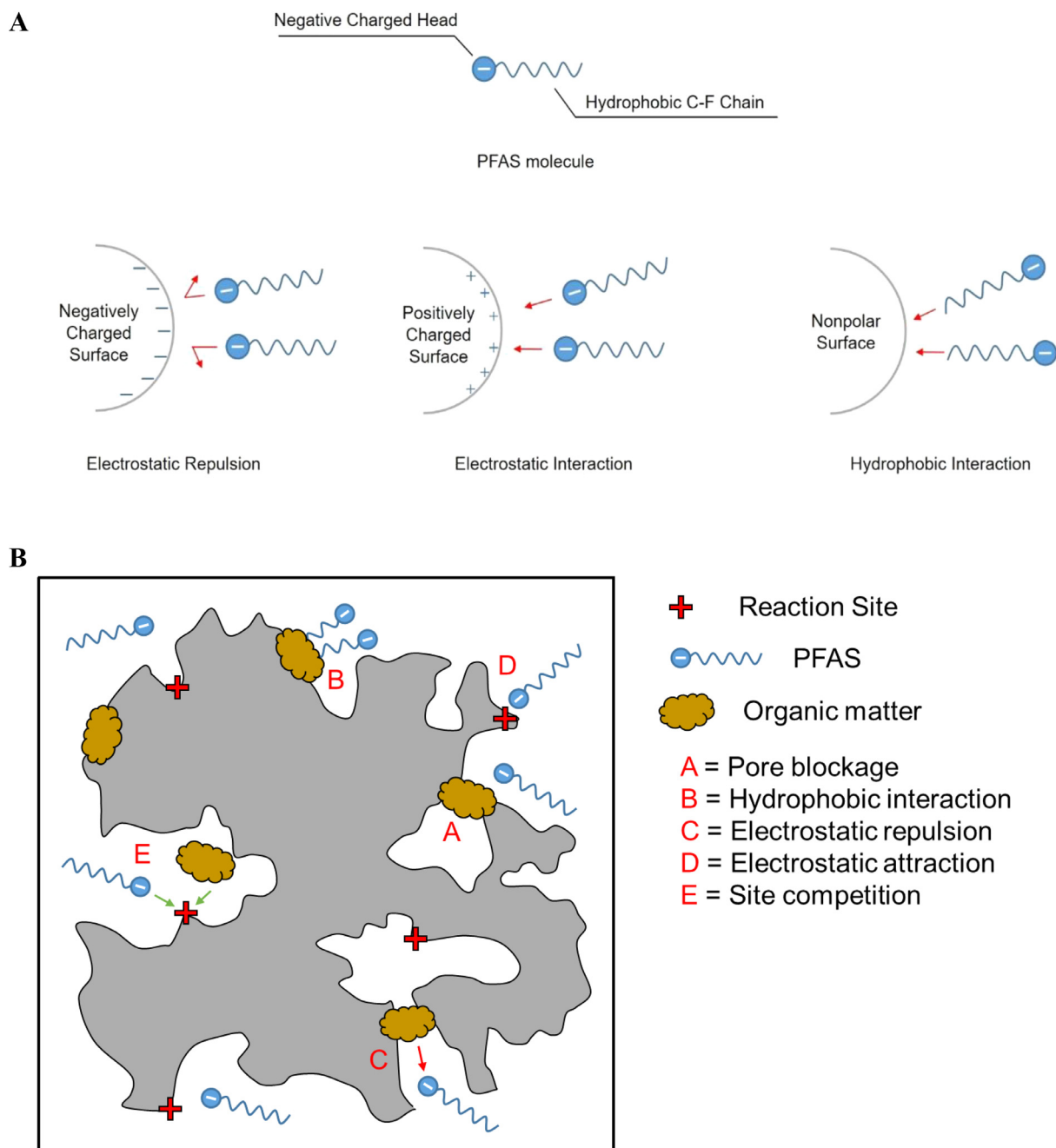


Fig. 1. Mechanisms of contaminant-adsorbing MPs: (A) Proposed mechanisms of PFAS interactions with adsorbents; (B) mechanisms of NOM interaction with a PFAS molecule in an adsorbent (elaborated from Gagliano et al., 2020).

could compete for active sites or cause pore blockage (Wang et al., 2015a; Yang et al., 2013).

The inhibition of PFOA sorption on active carbon fiber by OM was reported in a study where no obvious sorption occurred with increasing OM concentration to 500 mg L⁻¹ (Wang et al., 2015a). These results support competitive sorption between OM and PFOA, and the pore blockage of active carbon fiber by OM. A similar result was presented in another study, which demonstrated a 40% reduction in adsorption equilibrium for the removal of perfluorinated carboxylates (PFCAs) in the presence of OM (Du et al., 2015). Other studies also support the findings, wherein proteins present in OM caused a low retention of PFOA and PFOS through hydrophobic and electrostatic interactions (Pramanik et al., 2017), and the adsorption of PFCAs on Bamboo-derived Activated Carbon (BdAC) was limited by competition for

adsorption sites with co-existing OM (Du et al., 2015). The adsorption mechanisms of contaminants (e.g., PFAS) on MPs appear to depend on the presence of OM. However, the underlying conditions of different mechanisms of contaminant adsorption on MPs are not well understood, and their further investigation is required to assist one in mitigating contaminant adsorption on the MPs and removing c-MPs from water.

2.2. Factors related to the environmental conditions and to the plastic polymer

MPs have diverse characteristics since those can be categorized by their form (e.g., fibers, fragments, and spherical beads), their chemical composition (e.g., PE; low-density PE (LDPE); and polyacrylates (PAs)), particle sizes, and shapes (Silva et al., 2018; Jiang, 2018). The extent

of removal efficiency of MPs could be influenced by the adsorption of contaminants on MPs, solution pH, surface roughness and porosity of MPs, organic matter, and ionic strength and salinity of the solution. Aged MPs, which mostly underwent degradation from photooxidation, seawater corrosion, or other processes, exhibited increased sorption of chemicals, potentially due to decreasing molecular weight of the polymer (Pérez et al., 2010; Wang et al., 2018).

2.2.1. Solution pH and ionic strength

The presence of co-existing inorganic anions or solution pH could influence adsorption efficiency (Du et al., 2014). As reported by several studies (Deng et al., 2012a; You et al., 2010), the adsorption of perfluorocarboxylate anions decreases with increasing solution pH, similarly to most other anionic contaminants. For instance, the sorption of perfluorinated compounds on carbon nanotubes (CNTs) decreased with increasing pH, with negative zeta potential values of all tested adsorbents above pH 3.4, preventing perfluorocarboxylate anions from approaching adsorbents (Deng et al., 2012a). Increasing sorption of contaminants on MPs by decreasing pH (e.g., PFOS sorption on PE and PS MPs) was reported, but it may also depend on the types of contaminants (e.g., no sorption effect of FOSA on MPs due to changes in pH) (Wang et al., 2018).

Sorption mechanisms appear to be dependent on physicochemical properties of MPs and solution chemistry. A case study (Zhan et al., 2016) on PCB sorption on MPs (e.g., model MPs, polypropylene (PP) and 3,3',4,4'-tetrachlorobiphenyl (PCB77)) in synthesized seawater has demonstrated increasing sorption capacity with smaller MP particle sizes and the highest sorption of PCBs on MPs in seawater, compared with MPs in other solution environments (e.g., ultrapure water and n-hexane). MPs (microscale polymer particles) were found to adsorb organic compounds in the increasing order of affinity: polyamide (PA) < polyethylene (PE) < polyvinyl chloride (PVC) < polystyrene (PS) (Hüffer and Hofmann, 2016). However, the order of PA < PE < PVC < PS excludes considering plastic sizes within aqueous dispersion of MPs and can be influenced by other factors such as π - π interactions (Hüffer and Hofmann, 2016). The study also indicated that properties of both the sorbent and the sorbate (e.g., hydrophobicity) influence the sorption kinetics.

The presence of divalent cations in solution is attributed to adsorption even at high pH, as demonstrated in a study by You et al. (2010), where PFOS sorption at pH 8 was higher in sediment, compared to that at pH 7 with the same concentration of CaCl_2 . With increasing CaCl_2 concentration from 0.005 to 0.5 mol L^{-1} at pH 8, the sorption of PFOS increased by a factor of 6 but only a factor of 3 at pH 7 (You et al., 2010). Such an increase in the presence of Ca^{2+} or Mg^{2+} in water is attributed to the adsorbent surfaces having more basic sites to bind cations at high pH—a divalent cation bridging effect (Du et al., 2014; You et al., 2010; Zhou et al., 2013). As demonstrated in a study by Xiao et al. (2011), increasing ionic strength in the presence of Na^+ enhanced PFAS removal through a weakened electrical double layer caused by the aqueous sodium or hydrogen ion concentration (Xiao et al., 2011). The removal of PFAS could be improved by increasing ionic strength and lowering pH of the solution, i.e. mitigating contaminant adsorption on MPs through adjusting pH and ionic strength.

2.2.2. Salinity

Salinity is one of influencing factors on contaminant adsorption on MPs. For instance, the adsorption of PFAS on MPs was demonstrated to be favored by salinity through hydrophobic forces and the salting-out phenomenon (Lohmann, 2012; Sacks and Lohmann, 2011). With high salinity, PFOS was revealed to readily adsorb on PE and PS (Wang et al., 2015b), indicating favorable sorption of PFOS on MPs in seawater (Gagliano et al., 2020; Wang et al., 2015b). Similarly, a high adsorption capacity was observed in fluoropolymers with 10–14 carbon chains in seawater (Llorca et al., 2018). The extent of sorption dependent on types of MPs was examined in a study where PS and polystyrene

carboxylate (PS-COOH) had more affinity for PFAS than HDPE's affinity for PFAS (Llorca et al., 2018). According to the study, decreasing sorption of PFAS on MPs was observed in the presence of salts. For instance, the adsorption onto a surface of HDPE in freshwater was higher than that in seawater (70% adsorption of PFHxA on HDPE in freshwater vs. 20% adsorption of PFHxA on HDPE in seawater) (Llorca et al., 2018). Controlling salinity is one of approaches to decreasing adsorption of PFAS on MPs, although the kinetics depends on types of MPs.

2.2.3. Surface roughness and porosity

Surface roughness or the presence of a filler in MPs was found to influence the uptake of PFAS (Ateia et al., 2020; Zhang et al., 2018). In removing PFOS in the industrial wastewater using a polymer resin, the sorption rate and sorption capacity of PFOS depended on the polymer matrix and porosity (Deng et al., 2010). Surface roughness may affect sorption ability; however, the adsorption process occurs at the molecular level, and in this level, the porosity of the adsorbent plays a crucial role. HDPE MPs exhibited the worst adsorption capacity for PFAS because of limited intraparticle diffusion due to its granular shape (Llorca et al., 2018). As indicated in Fig. 2 (a) (Wang et al., 2015a; Llorca et al., 2018), hydrophobic interaction between MPs and PFAS generally increased the adsorption affinity of MPs on longer-chain compounds. Although the effect of salinity should not be overlooked, adsorption of PFAS on PS in freshwater was higher than on PE and PVC.

The sorption capacity of various organic materials on MPs is illustrated in Fig. 2 (b) (Fang et al., 2019; Xu et al., 2018; Wu et al., 2019; Liu et al., 2019; Li et al., 2018b; Wang W. and Wang J., 2018). MPs appear as a sorption media for organic compounds, with high sorption capacities of most organic compounds (e.g., tetracycline, pyrene, and trimethoprim). The same chemical has different adsorption affinities depending on the types of MPs, where PE MPs had the most sorption of pyrene and relatively low sorption on PS and PVC MPs. In natural ecosystems, MPs may be found as adsorbents for pollutants, and a recent study revealed that MPs, which were collected from Zhengmingsi Beach and Dongshan Beach in China, adsorbed persistent organic pollutants (POPs, such as PCBs, PAHs and DDTs) from the adjacent environment (Zhang et al., 2015).

Sorption capacity of different types of MPs is reported in several studies (Fang et al., 2019; Xu et al., 2018; Bakir et al., 2014). These studies include a significant sorption capacity of PVC and PE for DDT in freshwater (Bakir et al., 2014) and the equilibrium adsorption capacity of hexaconazole (HEX, 41.56 $\mu\text{g g}^{-1}$), myclobutanil (MYC, 18.94 $\mu\text{g g}^{-1}$) and triadimenol (TRI, 10.48 $\mu\text{g g}^{-1}$), suggesting that the adsorption order is consistent with log K_{ow} (HEX: 4.01, MYC: 3.58 and TRI: 2.91) (Fang et al., 2019). The greater sorption capacity of tetracycline on PS MPs than on PE and PP was reported when there was a coexisting contaminant (e.g., benzene). The interaction may occur through polar and π - π interactions due to the presence of benzene in tetracycline and PS polymers (Xu et al., 2018). The surface property of MPs remained unchanged even in the presence of other organic matter (i.e., fulvic acid), whereas decreasing sorption of tetracycline on PE, PS and PP occurred because of complexations of fulvic acid with carboxylic functional groups and hydrogen bonding in tetracycline through competition of tetracycline with fulvic acid for sorption on MPs (Xu et al., 2018).

The sorption mechanism may be dependent on the types of MPs and contaminants. A hydrophobic interaction was identified for bisphenol analog-adsorbing PVC MPs, whereas ionization of bisphenol F (4,4'-dihydroxydiphenylmethane) in the neutral solution caused sorption inhibition through electrostatic repulsions (Wu et al., 2019). Noncovalent bonds such as hydrogen and halogen bonds increased the adsorption of bisphenols on PVC MPs (Wu et al., 2019). Notably, the adsorption efficiency (%) increased over the PVC MP dosage, before reaching equilibrium regardless of the five bisphenols (BPA, BPAF, BPB, BPF, and BPS) (Wu et al., 2019). At the same dosage of PVP MPs (1.5 g L^{-1}), the adsorption efficiency was in the order of BPAF > BPB > BPA > BPF > BPS (Wu et al., 2019). Polyamide (PA) revealed the

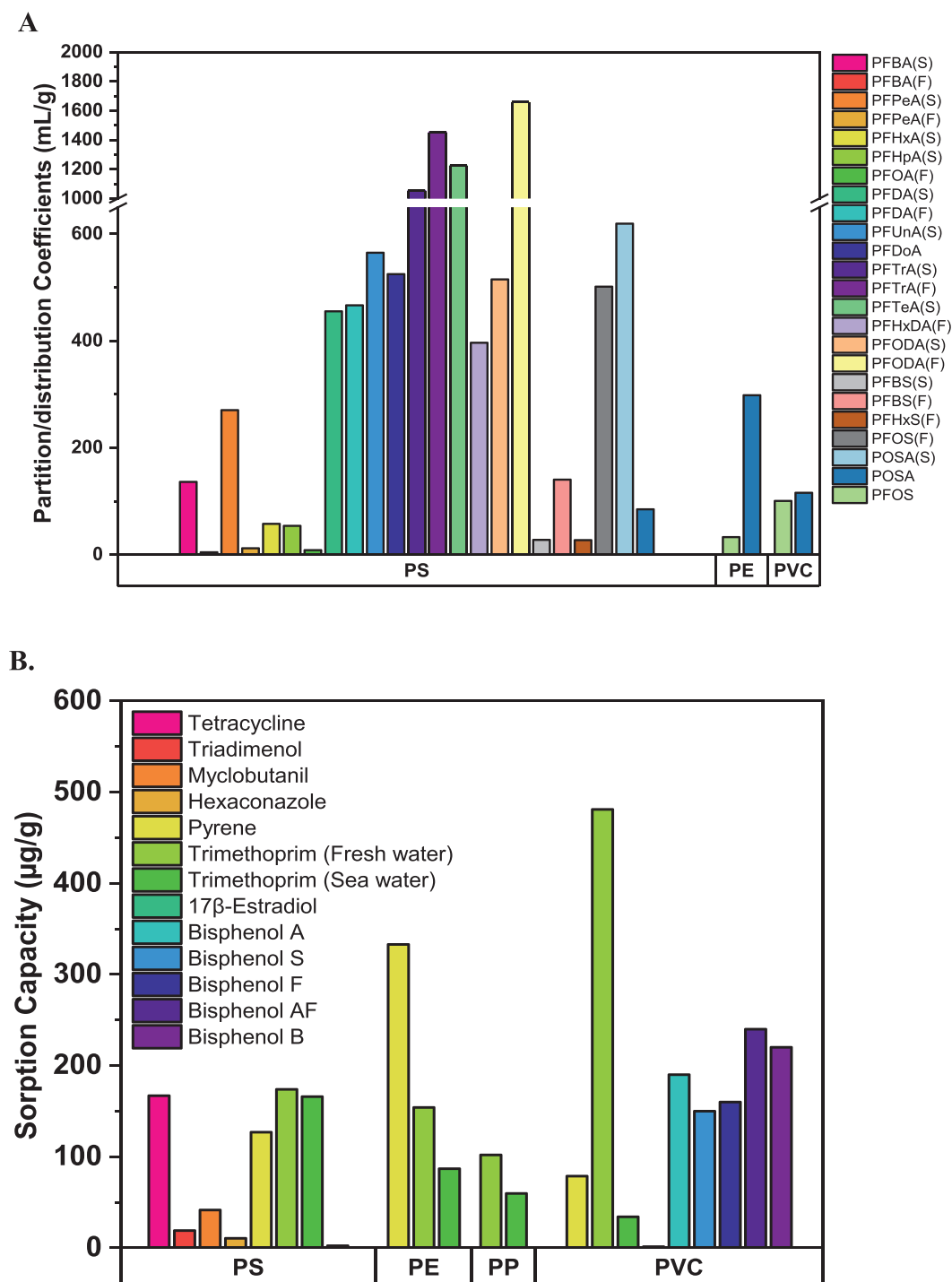


Fig. 2. Sorption of POPs on MPs: (A) Partition and distribution coefficients of PFAS on PS, PE, and PVC MPs. Elaborated from (Wang et al., 2015a; Llorca et al., 2018); (B) Sorption capacity of POPs on PS, PE, and PVC MPs. Elaborated from (Fang et al., 2019; Xu et al., 2018; Wu et al., 2019; Liu et al., 2019; Li et al., 2018b; Wang, W. and Wang, J. 2018). (Note: [S]: seawater and [F]: freshwater).

highest sorption capacity for 17β-Estradiol through hydrogen bonding (Liu et al., 2019) and exhibited high affinity to antibiotics in freshwater through hydrogen bonding (Li et al., 2018b). Sorption of aromatic compounds on MPs was demonstrated to increase in the order of PA < PE < PVC < PS through the π - π interaction Hüffer and Hofmann (2016).

Further, the change of surface properties of MPs influenced by OM may affect the sorption of OM or other co-existing contaminants. For instance, negatively charged HA molecules caused highly negative zeta potentials on PS microspheres (micro-PS), leading to an increase

in the net energy among micro-PS particles through a high electrostatic repulsive force and decreasing micro-PS aggregation (Li et al., 2018c).

The sorption of organic compounds is also affected by the rubbery/glass state theory of polymers (Guo et al., 2012; Guo and Wang, 2019). On PE, complete removal of PCB congeners was seen in water, compared to that on PVC or PS, because of high affinity for PCB by abundant rubbery domains in PE (Pascall et al., 2005). Similarly, the

larger surface area and the rubber-like nature of PE enabled the higher adsorption of chemicals on PE than on PS or PVC (Wu et al., 2019).

2.2.4. Microbial colonization of MPs and the effect of biofilm on plastic aggregates

Significant accumulation of millimeter-sized polymers and pelagic surroundings for microorganisms in aquatic and soil environments are currently emerging as a new surface for biofilm formation and gene exchange (Reisser et al., 2014; Zhang et al., 2019; Arias-Andres et al., 2018). The most important determinant in biofilm for microbial collections on MPs is considered to be biogeography and environmental factors (Oberbeckmann and Labrenz (2020)). Wastewater treatment plant (WWTP) effluent has been one of pathways through which bacteria have stronger affinity for MPs due to the growth of biofilm biomass through high concentration of inorganic nutrition (McCormick et al., 2014). Bacteria have shown strong adsorption on MPs, but MPs also influence biofilm structure, as demonstrated in several studies (Lagarde et al., 2016; Carson et al., 2013; Bravo et al., 2011; Parrish and Fahrenfeld, 2019).

The pattern in bacterial community composition in the diverse microbial community (plastisphere) appears to be influenced by the surrounding water, rather than by detachment of biofilm into surrounding water (Zettler et al., 2013). When different types of extracellular polymeric substances (EPS) were produced, the presence of microalgae on dissimilar polymer resulted in diverse cohesive and sticking properties, enhancing the formation of heteroaggregates (Lagarde et al., 2016). Another study indicated that the presence of microalgae rapidly colonized PP and HDPE polymers, displaying some heteroaggregates in PP but no observable aggregation in HDPE, because of production of different types of EPS by microalgae for the colonization of PP and HDPE fragments (Lagarde et al., 2016).

Several studies revealed that diatom abundance on rough surfaces and that at sites with high plastic density were apparently increased, whereas bacterial abundance was inconsistent, but it instead increased only on foamed PS (Carson et al., 2013; Bravo et al., 2011). There are studies that suggest that morphology and surface texture of MPs, as well as water source, influence the microbial community structure (Parrish and Fahrenfeld (2019)).

2.2.5. Microbial assemblage

MPs with their buoyancy, hydrophobic surface, and long transport distance could serve as a new substrate for selection and dispersion of unique microbial assemblage (Mammo et al., 2020). One of notable features of the MP-associated microbial assemblages is relatively high abundance of the bacterial family *burkholderiales incertae sedis* (Pal et al., 2012), as detected in all 14 wastewater treatment systems (Wang et al., 2012). *Pseudomonas* sequences, prevalent on MP-associated bacterial assemblages, are found in an urban river (McCormick et al., 2014; Ibekwe et al., 2013). This microorganism is associated with the degradation of plastic polymers such as HDPE (Balasubramanian et al., 2010), LDPE (Tribedi et al., 2015), PE (Arkatkar et al., 2010), PP (Shimao, 2001), and polyvinyl alcohol (Harrison et al., 2014). The consecutive formation of plastisphere-specific bacterial assemblages found within coastal marine sediments is due to bacteria that rapidly colonize LDPE MPs (Viršek et al., 2017), which can selectively enrich hydrocarbon-degrading bacteria (Oberbeckmann and Labrenz, 2020).

Several factors, such as solution pH, ionic strength of the solution, natural organic matter, and surface roughness and porosity of MPs, affect adsorption kinetics of contaminants on MPs, which consequently influences removal of c-MPs or MPs. Particularly, properties of MPs such as size, shape, and polymer type are considered to be significant properties of c-MPs (Fred-Ahmadu et al., 2020) and should be counted as operation unit parameters for efficient c-MP removal.

3. Evaluation of membrane filtration for the removal of microplastics in various environmental conditions

Evaluation of treatment technologies for MPs is essential for development and improvement of existing treatment units to eliminate recalcitrant contaminants such as MPs in the environment. Understanding the ultimate fate and transport of MPs, especially c-MPs, are not well understood. Thus, the fate and transport of MPs under various environmental conditions and treatment technologies for MPs are examined, and their pros and cons are described.

3.1. Fate and transport of microplastics with adsorbed contaminants

MPs may correspond to a significant habitat and transport medium for microbes. As a potential pollutant vector, MPs alone or c-MPs could be transported directly to even the food chain (Zarfl and Matthies, 2010; Mammo et al., 2020). During their transport to the environment, MPs may undergo a mechanical breakdown, photochemical degradation, hydrolysis, biodegradation by organisms, biofouling, and coagulation, leading to a multiplicity in density, size (nano-, micro-, and millimeters), shape (fiber, spherical, etc.), and surface characteristics found in water and sediments (Zhang (2017)).

Their mechanical degradation through shredding into smaller particles due to friction forces could result in an increased surface area of polymer particles, thereby causing rapid degradation through increasing chemical reactivity (Klein et al., 2018). Quantification of MP removal in each state of the treatment process may assist in developing treatment processes for MPs in a multifaceted water system. Few studies are found that elucidate sources, the fate, and transport of MPs in wastewater treatment plants where various types of contaminants co-exist.

MPs are accumulated through their environmental release from various sources to land (through littering, ineffective waste management, and sewage sludge), freshwater (from the terrestrial environment), ocean (through rivers, fishing waste, and mismanaged maritime), and the atmosphere (suspended and transported within the air, ultimately leading to deposition of MPs to land or aquatic environments) (Horton and Dixon (2018)). Several mitigation and controlling measures for impacts of MPs are suggested. For instance, avoidance of massive MP accumulation through sustainable agricultural practices and recycling could minimize land accumulation of MPs. However, in freshwater, there are MPs that are difficult to mitigate because of complex characteristics of freshwater, and in the ocean, most MPs are formed from macroplastic waste within the marine environment. The extent of ultimate sinkage of MPs from the atmosphere to land or aquatic environments is unknown and warrants further research.

Several models (e.g., emission-based mass flow models, global river models, multimedia models, and spatiotemporally explicit models) were introduced as a tool for evaluating risk assessment of plastic pollutants (Wagner and Lambert (2018)). For instance, using emission-based mass flow models, wastewater treatment plant was estimated to retain between 40 and 96% of MPs (van Wezel et al., 2015). According to global river models, most rivers are reported to be more polluted over time globally, considering change in several features such as urbanization and hydrology (Fekete et al., 2010; Bouwman et al., 2009; Van Drecht et al., 2009). With multimedia models, increasing removal and more settling of MPs from water and soil are estimated with an increase in particle diameter (Wagner and Lambert (2018)).

Of two models (i.e. NanoDUFLOW and INCA-Plastic model) presented from spatiotemporally explicit models, the INCA-Plastic model estimated lower retention of smaller MP particles in the Dommel River simulation, thereby exporting those particles to sea, whereas the NanoDUFLOW hydrological model predicted increasing retention of MP particles smaller than 5 μm , resulting in increasing sedimentation through capturing small MP particles shown in heteroaggregates (Wagner and Lambert, 2018; Besseling et al., 2017; Nizzetto et al., 2016).

However, such models are very limited in their simulation of transport and the fate of MPs in aquatic systems. Despite the importance of effluent filters in the removal of residual MPs, significant amount of MPs are still accumulated from WWTPs. The MP transport mechanisms in water were proposed as a complex combination of processes involving surface drifting, vertical mixing, beaching, settling, etc. (Li et al., 2020). Three principal transport mechanisms of MPs are proposed as (a) gravity-driven transport, most commonly found in sediment-laden flows; (b) settling or conveyance of materials floating on a surface or suspended in a water column through biological processes; and (c) transport by thermohaline currents during settling or by deviation of deposited MPs Kane and Clare (2019).

Notably, despite significant concern about MPs as a potential vector for carrying pollutants, there is little known about the fate and transport of c-MPs. Among the physical properties of MPs (size, density, morphology, etc.), the density of MPs appears to significantly influence the distribution of MPs (e.g., larger-sized MPs much are less affected by turbulent mixing), and Stokes drift primarily causes coastal MP transport (Li et al., 2020). In addition to settling of MPs in sediment by the density effect, MPs are subject to biofouling, which increases their MP densities, leading to slow sinking of MP particles (Li et al., 2020).

Retention of MPs was dependent on particle sizes, being lowest (18%–25%) for medium-sized particles ($\sim 5 \mu\text{m}$) with the lower limit of 100 nm, compared to smaller submicron particles or larger micron- and millimeter-sized plastics (Besseling et al., 2017). As illustrated in Fig. 4, the plastic concentration decreases over increasing flow distance, but wide distribution of plastic concentration is apparent in the order of $100 \text{ nm} > 1 \mu\text{m} > 5 \mu\text{m}$, implying that the removal of larger MP particles occurs earlier within a shorter flow distance. The shape/morphology, aggregation, and attachment efficiency of NPs/MPs may also influence the fate of NPs/MPs, along with particle concentration and flow distance.

The degree of near-shore MP transport is likely to be controlled by hydrodynamic processes including estuarine circulation, Stokes drift and bed-load transport Zhang (2017). For the transport of MPs in coastal seas, the following equation (Eq. 1) is applied to model the dispersal of floating objects, and Eq. 2 is used to model turbulent vertical mixing Zhang (2017).

$$\vec{U}_P = \vec{U}_A + \xi \sqrt{\frac{2\vec{K}_H}{\Delta t}} + W_d \vec{V}_W \quad (1)$$

[The floating velocity of a MP particle (\vec{U}_P), surface current (\vec{U}_A), computation time step (Δt), random coefficient (ξ : $-1 \sim 1$), and horizontal eddy mixing parameter (\vec{K}_H), wind drift coefficient (W_d), near-surface wind velocity field (\vec{V}_W)]

$$w = \frac{(\rho_w - \rho_p)gd^2}{18\eta} \quad (2)$$

[Terminal velocity (w), seawater viscosity (η), and gravitational acceleration (g)]

In benthic transport of MPs, deposition of MPs onto the sea bed is likely to occur because of high density of MPs through coagulation with natural suspended solids or fouling with microbes, although resuspension and transport of MP particles may take place during turbulent flows (Zhang, 2017). Biofouling caused by the interaction of MP particles with various marine organisms is attributed to the ultimate sinking of MPs to the benthos, making the transport pathways a determinant factor for assessing the ecological risk of MPs (Zhang, 2017). Since the solubility of hydrophobic contaminants is influenced by salinity, temperature and the presence of OM, such environmental conditions are likely to affect the fate and transport of POPs on MPs.

The sorption behavior and transport of POPs by MPs in estuarine conditions was examined in a study by Bakir et al. (2014). The salinity did not affect sorption and desorption of phenanthrene (Phe) on MPs (e.g., PVC and PE MPs), whereas a slightly decreasing sorption capacity

of DDT on MPs was demonstrated; this indicates that physical properties of the polymer, including plastic density and other environmental conditions such as particle residence time in aquatic media (e.g., estuaries) may govern the transport of contaminated plastics (Bakir et al., 2014). In addition, the transport model of POPs-MPs from freshwater and brackish water toward marine conditions, which followed the order of Phe-PE >> DDT-PVC = DDT-PE >> Phe-PVC, indicates more sorption capacity of MPs in estuaries due to higher concentrations of contaminants than in marine waters.

In a sediment, the presence of OM could limit the transport of highly hydrophobic organic chemicals (e.g., the partition coefficients of octanol/water, $\log K_{ow} > 6.5$); however, OM-adsorbed MPs enhanced mobility to carry pollutants (Zarfl et al., 2009; Gouin et al., 2011). The interaction between MPs and bacteria could result in transport of either MPs or bacteria in any case. A study by He et al. (2018) revealed increasing bacteria transport and decreasing deposition of bacteria in quartz sand by plastic particles at high ionic strength conditions (e.g., 50 mM NaCl and 5 mM in CaCl_2). This is well explained with the adsorption of MPs onto the cell surface and the repellent effect caused by suspended plastic particles, which increase cell transport. Fouling microorganisms could still increase the density and decrease buoyancy of the particle, with fast reaching in its critical sinking density by the smaller particles (Lagarde et al., 2016).

In membrane filtration of MPs or c-MPs, fouling microorganisms may govern the fate and transport of c-MPs or MPs; however, very limited information is available on the fouling process. Furthermore, there has been scanty research on membrane biofouling mitigation by MPs with adsorbed contaminants (c-MPs). The biofilm–plastic interactions could influence removal of MPs, warranting future research to improve understanding of key mechanisms and other environmental factors of such removal and to develop a new treatment method and efficient management of MPs in different environments.

MP-mediated transport of PCBs and the effect of MP on PCB removal were investigated by exposing *Daphnia magna* with PCB (congeners, 18, 40, 128 and 209) to a mixture of MPs and algae, with daphnids exposed to only algae used as a control (Long et al., 2015). The removal of PCB209 was shown to be efficient when MPs were involved (e.g., $0.1 \mu\text{g}$ PCB/g *Daphnia* with PCB+MP, compared to $0.4 \mu\text{g}$ PCB/g *Daphnia* with PCB only), whereas no difference for other tested congeners was observed (Long et al., 2015); this indicates that MPs are a sink for hydrophobic organic contaminants.

Under various environmental conditions, pollutant-adsorbing MPs could influence the MP fate and transport mechanisms involving MPs' gravity-driven move, settling or floating by interaction with microbes, or Stokes drift, since environmental conditions (salinity, temperature, OM, etc.) affect the fate and transport of contaminants. Nonetheless, the currently available models do not appear to reflect such variables including environmental conditions, especially pollutants that have high adsorption affinity on MPs.

Given the high probability of MP accumulated in WWTPs, the fate of MPs may depend on unit operation processes and parameters influencing each treatment unit. The removal of MPs (plastic particle sizes < 5 mm) and small anthropogenic litter (SAL) in WWTPs depended on WWTP configuration (Michielssen et al., 2016). For instance, the highest removal of MPs was achieved through tertiary granular sand filtration and membrane filtration (Michielssen et al., 2016; Poerio et al., 2019; Talvitie et al., 2017; Liu et al., 2021).

Another case study indicated that 99.5% of influent MPs were removed by a membrane bioreactor (MBR) system, whereas a slightly lower removal rate (97%) of MPs was found in the oxidation ditch system, based on influent MPs that consisted of PET (47%), PS (20%), PE (18%) and PP (15%) (Lv et al., 2019). The slightly improved MP removal by the MBR system could be due to a synergistic effect of biodegradation and membrane filtration. As revealed in the study, most MPs were accumulated in a sludge phase (e.g., 74% MPs contained in

the sludge after a secondary settling tank or 80.5% MPs contained in the sludge from the MBR tank) (Lv et al., 2019).

The significant accumulation of MPs in sewage sludge is one of major sources of soil MPs, which pose a high risk to food security and public health, along with combined contamination of MPs with other toxic pollutants (Wang et al., 2019a). The overview regarding interactions of MPs with contaminants in the environment is presented in Fig. 5 (schematic of the interactions of MPs with contaminants in the environment). In the figure, plastic debris through physical, chemical and biological interactions is dispersed into soil, water and the atmosphere and further broken into smaller particles and then reacts with other contaminants present in the environment. The presence of OM, microbes, algae, earthworms, and other organisms may even assist interactions of contaminants with MPs, leading to environmental accumulation of metabolic products, degradation by-products, and residual MPs.

3.2. Membrane filtration of microplastics with adsorbed contaminants in comparison to other treatment methods

The treatment processes for MPs in freshwater and wastewater may not offer the desired efficiency of MP treatment because the treatment efficacy may depend on the applied treatment methods, even if other factors may influence it. Although MBRs exhibited higher removal efficiency of MPs (e.g., 99.4%) compared to other methods (e.g., a conventional active sludge (CAS)-based process: 98.3%) (Lares et al., 2018), several factors (e.g., biofilm thickness and pore blockage) remain challenging in the membrane filtration. In the present study, membrane filtration for the treatment of MPs or c-MPs is reviewed and compared with other treatment methods that focus on MP removal, thus identifying their pros and cons.

3.2.1. Treatment methods other than membrane filtration

As with the fundamental process of sedimentation, which removes solid particles from water through gravitational settling (Cheremisinoff, 2002), the removal rate of MPs in wastewater was around 92 %, based on the report from a wastewater treatment plant (WWTP) located in Vancouver, Canada (Ngo et al., 2019). Yet, contaminants, particularly MPs, are not completely removed, and the remaining contaminants simply get trapped or sink in waste sludge, causing concern about the accumulation of MPs in wastewater treatment processes from the turbulent environment.

Furthermore, MPs present in sewage sludge in a landfill could be released back to WWTP as leachate, or they may contaminate the natural water environment through storm water runoff. An alternative sustainable solution for removal and management of MPs is required urgently. One of factors influencing the sedimentation process is density, which affects the removal rate of MPs (e.g., buoyancy vs. sinking). MPs left in wastewater and sludge for whatever reasons (e.g., longer retention time during treatment unit operation) may cause adsorption of various toxic contaminants, creating a high risk to the environment (Hermabessiere et al., 2017).

Air flotation applies air bubbles to enhance buoyancy of contaminants for floating on the water surface, and with flotation, the removal rate of MPs was reported to be 95% at a WWTP (located in Hameenlinna, Southern Finland) (Ngo et al., 2019). Contrary to sedimentation, flotation can control low-density MPs (e.g., PE, PP, synthetic rubber (SR)) and moderate-density plastic pieces (e.g., PS and PA) that are otherwise hard to capture through the sedimentation process (Ngo et al., 2019). However, flotation is still not ideal for the removal of MPs, since high treatment efficacy is difficult to achieve because of poor removal potential if the densities of MPs are not low enough for the flotation method.

The activated sludge process, which is one of the commonly applied methods in municipal WWTPs, could further remove MPs after sedimentation, aerated grit chamber or dissolved air flotation (Ngo et al., 2019). The primary issue with the activated sludge process is potentially

increasing accumulation of MPs by sludge flocs or bacterial extracellular polymers in an aerobic tank, and microorganisms might even digest the plastic debris present in sludge flocs (Scherer et al., 2018).

During the activated sludge process, several factors, such as the retention time and nutrient level in wastewater, could influence the removal rate of MPs (Carr et al., 2016; Rummel et al., 2017). For instance, increasing retention time and nutrient level may increase the risk of cross-contamination, thereby decreasing the efficacy of MP removal. Further, the removal efficacy of MPs could be significantly diminished by increasing reagent dosage, inhibiting nitrogen conversion rate, and membrane fouling in wastewater treatment processes (Wu et al., 2021).

Rapid sand filtration may offer speedy and efficient MP removal at a low operational and maintenance cost (Ngo et al., 2019). Retrofitting of existing secondary WWTPs by granular sand filtration (97.2%) or membrane filtration (99.4%) proved to yield the highest possible removal of small anthropogenic litter (SAL) (Michielsen et al., 2016). Yet, the MP removal efficacy was highest at MBR as 99.9% removal of MPs was achieved, compared to a rapid sand filter (97% MP removal) in the study (Talvitie et al., 2017) where several advanced final-stage treatment technologies were applied in removal of MPs in wastewater. Such a lower MP removal by the rapid sand filter is potentially due to higher porosity of the filter material (e.g., a mixture of anthracite coal and sand) after the operation period (Ngo et al., 2019).

As a material-based treatment method, a nano-coating technology using ZnO nanorods (Tofa et al., 2019) has exhibited 30% degradation of MP fragments, low-density polyethylene (LDPE) film, in water. The degradation was accelerated by visible light excited heterogeneous ZnO photocatalysts. Since degrading MPs with photocatalysis results in reduced by-products, this could be a cost-effective method for MP removal in water. A polymer coating-based elongated mesh screen as a modification of a membrane proved to be an effective MP removal tool with good durability and easy fabrication from vast materials, along with other advantages including the absence of electrical power and mechanical devices (Li et al., 2018d; Ward, 2015).

Pyrolysis involving thermal, catalytic, and microwave-assisted pyrolysis is another treatment method that could be applied for degradation of MPs. A recent study examined treatment of HDPE plastic waste with microwave-assisted pyrolysis (Juliastuti et al., 2017). Under the microwave heating, the best treatment was achieved at 400 °C with a 1:1 feed-to-catalyst weight ratio for 45 min of operation. The highest methane formation (7.4%) occurred at 400°C of operation with the liquid product, mostly comprising n-paraffin (~53%) and olefin (32%). Despite several benefits (reduced by-products and an excellent alternative fuel source from pyrolysis), further development of MP treatment is necessary to overcome hurdles such as complexity and high operational investment costs.

As a hybrid treatment method (e.g., combined coagulation and membrane filtration), Fe-based coagulation in conjunction with UF processes was investigated for the removal of PE (main component of MPs) in drinking water (Ma et al., 2019b). In the study, low efficiency (around 15%) using Fe-based salt was observed, whereas addition of polyacrylamide (PAM) at high dosage significantly increased the removal efficacy up to 90%, through dense floc formation and high adsorption capacity (Ma et al., 2019b). For instance, at 2 mmol L⁻¹ of FeCl₃•6H₂O, the removal efficiency of MPs was 15% (MPs with $d < 0.5$ mm) and 2.5% ($2 < d < 5$ mm), whereas at the same dosage of FeCl₃•6H₂O, adding cationic or anionic PAM (6 mg/L) significantly increased the removal efficacy up to 65% (for cationic PAM) or 90% (for anionic PAM) for $d < 0.5$ mm of PE MPs and 5% (for cationic PAM) or 18% (for anionic PAM) for $2 < d < 5$ mm of PE MPs (Ma et al., 2019b).

Notably, smaller MP particles achieved higher removal efficacy than that of larger MP particles, and the addition of anionic PAM improved performance and reduced fouling compared with cationic PAM because of anionic PAM's opposite charge to that of Fe-based flocs. At high dosage of FeCl₃•6H₂O (2 mmol/L), the removal efficacy of PE MPs increased with increasing solution pH, although the PE removal efficacy

was slightly influenced by other factors such as ionic strength, OM, and turbidity (Ma et al., 2019b).

As cost-effective treatment widely applied in industrial, commercial and municipal waste treatment, electrocoagulation could be an alternative treatment technique to chemical coagulation. Applying electrocoagulation revealed more than 90% PE MP removal over a wide range of pH (3–10), with the highest removal rate (99.24%) being achieved at pH 7.5 under optimum NaCl concentration (2 g L^{-1}) at a current density of 11 A m^{-2} (Perren et al., 2018).

3.2.2. Membrane processes

MBR, which has high removal capacity for recalcitrant contaminants (Ngo et al., 2019), could be ideal treatment of MPs because it involves both biodegradation and membrane filtration (with the smallest pore size of around 0.08 nm), which prevent most MPs to pass through, as exemplified in a case study where the removal rate of MPs was 99.9% during the MBR process at WWTP in Mikkeli, Finland (Ngo et al., 2019).

The efficacy of MP treatment by the advanced MBR system (consisting of anaerobic and aerobic tanks and a membrane filtration tank with a submerged flat-sheet membrane unit) was compared with that by the conventional activated sludge (CAS) (Lares et al., 2018). According to the study, 2.5 times more MP particles were retained in the effluent from the CAS process (overall 98.3% removal rate) compared to those retained using advanced MBR technology (total 99.4% removal rate). Different types and shapes of MP polymers appear to depend on treatment unit processes. For instance, more than 80% of PES (polyester) fibers were present in activated sludge and MBR sludge, whereas 60% of PES fibers and 30% various polymers were contained in MBR permeate (Lares et al., 2018).

Another case study (Lv et al., 2019) compared MBR with an oxidation ditch and found superior efficacy of MBR (e.g., membrane tank (83.5%) > oxidation ditch (15%)), due to the pore size of $<0.1 \mu\text{m}$ from an MF membrane and an anaerobic/anoxic/aerobic (A/A/O)-MBR system. Nonetheless, the efficacy of MBR may still depend on limiting factors including control of biofilm and membrane congestion (Lares et al., 2018; Nicoletta et al., 2000). Although there was a case study demonstrating the most effectiveness of removing MPs in wastewater at a WWTP in Mikkeli, Finland (Hermabessiere et al., 2017), significant amounts of MPs were found to be discharged into the natural water source from WWTP (e.g., 13,500,000 pieces day^{-1} from a total flow rate of $\sim 270,000 \text{ m}^3 \text{ day}^{-1}$) (Talvitie et al., 2017).

As an attractive membrane-based treatment method for municipal wastewater, oily water, etc., a dynamic membrane (DM) has been applied, based on the principle that the formation of a cake layer (DM) plays a role as a secondary membrane filter; yet, membrane performance becomes less effective because of dense fouling and thicker layers compared those in MF/UF processes (Poerio et al., 2019; Li et al., 2018d). The efficacy of MP removal by DM was investigated along with the effect of influent flux and influent particle concentration on DM performance (Li et al., 2018d).

Superior filtration of MPs was demonstrated in only 20 min, as indicated in the significant decrease in turbidity from 195 NTU (influent) to 1 NTU (effluent) under low TMP and low filtration resistance in wastewater (Li et al., 2018d). Such high removal efficiency (>99.9% after 20 min operation) was achieved when DM (cake layer) was formed on a $90 \mu\text{m}$ mesh under gravity (Li et al., 2018d). TMP increase and resistance were dependent on influent flux over time. The TMP and resistance were in the order of $21 \text{ L h}^{-1} > 18 \text{ L h}^{-1} > 15 \text{ L h}^{-1} > 12 \text{ L h}^{-1} > 9 \text{ L h}^{-1}$ (Li et al., 2018d). For instance, at 50 min of operation, 350 and 100 TMP (mm), as well as 6 and $3.8 (\times 10^9 \text{ m}^{-1})$ resistance from 21 L h^{-1} and 9 L h^{-1} , respectively, at a constant MP particle concentration of 8 g L^{-1} were reported (Li et al., 2018d).

Notably, TMP increase was more significant at a higher MP concentration over time (e.g., around 200 vs. 100 TMP (mm) from 1 and 0.1 g L^{-1} MP concentration, respectively, in 2 h at a constant influent flux of 10 L h^{-1}) (Li et al., 2018d). More resistance was observed from

higher MP concentration (e.g., 7 vs. $3 (\times 10^9 \text{ m}^{-1})$ resistance from 1 and 0.1 g L^{-1} of MP concentration, respectively, in 2 h at a constant flux of 10 L h^{-1}) (Li et al., 2018d). Several studies indicated that DM formation is facilitated when influent flux and concentrations of MPs are high (Li et al., 2018d; Ersahin et al., 2017; Horton and Dixon, 2018). The treatment efficacy of MPs with DM could be maximized by increasing influent flux at high concentrations of MPs.

Reverse osmosis (RO), which uses a partially permeable membrane to remove ions, large objects, and pollutants from drinking water, can be implemented to remove MPs in drinking water. In a case study performed with RO at a WWTP in Sydney, Australia, around 90.4% MP removal (Ziajahromi et al., 2017) was reported after tertiary treatment. As revealed in the study (Ziajahromi et al., 2017), the number of MP particles (MP particles L^{-1}) was in the order of primary (~ 1.5) > secondary (~ 0.5) > tertiary (~ 0.4) > RO (~ 0.2). Notably, the majority of MPs was fibers (especially in primary treatment) with relatively small portion of granular and some irregular shapes of MPs (in secondary and tertiary). After RO treatment, only a small fraction of MP fibers remained. The data suggest that a significant number of MPs are still released to the aquatic environment even after four treatment stages, including primary, secondary, tertiary and RO processes.

In a case study Mrowiec (2018), although municipal WWTPs are one of the primary sources of MPs in aquatic environment, most MP particles were removed through the primary treatment zones (e.g., solid skimming and sludge settling processes), followed by additional removal through tertiary treatment processes including membrane filtration and dissolved air flotation (DAF). However, due to a continuous intake of large volumes, even tertiary-level effluent may still contain a significant amount of MPs, along with smaller sizes (e.g., $<20 \mu\text{m}$) of MPs and MP fragments (e.g., NPs). Further, because of the hydrophobic nature of MPs, the POP (PCB, PBDE, PFAS, etc.)-adsorbing MPs raise great concern for effective treatment, although degradation processes (e.g., biodegradation through interaction with microbes, photooxidation through interaction with sunlight, and hydrolysis through interaction with water) still occur.

In a case study conducted at WWTPs in China (Yang et al., 2019), characteristics and removal rates of MPs throughout each treatment process were reported. According to the study, the majority (>70%) of detected MPs were PET, PS, and PP, and the microfiber shape was predominantly found in both influents and effluents. The overall removal rate achieved was 58.3% after the primary sedimentation tank, 77.5% after the secondary sedimentation tank, and 91.7% after combined denitrification/ultrafiltration/ O_3 /ultraviolet processes. The advanced treatment processes enhance the removal rate of MPs. Nonetheless, even with combined treatment processes, the removal of MPs remains imperfect.

Another case study (Ziajahromi et al., 2017) on the various treatment involving primary, secondary, tertiary and RO processes at WWTPs revealed that the MP composition was in the order of PE (42%) > PET (36%) > PS (15%) and PP (8%) from the primary effluent with MP sizes between 100 and $190 \mu\text{m}$. As per the MP shape, the significant decrease of MP fibers was notable from the RO effluent (e.g., 6 times reduction compared to the MP fibers from the primary effluent (MP particles L^{-1}) (Ziajahromi et al., 2017), indicating that even with RO, the MP removal is incomplete.

A case study exhibited that biological sewage treatment facilities (STFs), which consisted of anaerobic-anoxic-aerobic (A2O), a sequence batch reactor (SBR), and media processes, were effective as they removed more than 98% of MPs (e.g., 98.5% from A2O, 99% from SBR, and 98% from the media processes), yet significant amounts of MPs were still reported as being released into the marine environment Lee and Kim (2018). Among the three processes applied for MP treatment efficiency, the media process has the lowest efficiency (e.g., around 95% removal rate for SS and MP particle sizes greater than $300 \mu\text{m}$) Lee and Kim (2018). Higher treatment efficiency associated

with the biological STF process appears primarily due to the physical adsorption of MPs by the biofilm or floc.

Four municipal WWTPs applying different advanced treatment technologies (e.g., MBR treating primary effluent, disc filter (DF), rapid sand filtration (RSF), and DAF treating secondary effluent) were explored for the removal efficacy of MPs (Talvitie et al., 2017). Results indicate MP removal ranges from DF (40%–98.5%) to DAF (95%), RSF (97%), and MBR (99.9%), indicating that all four advanced treatment technologies were effective in treating MP-laden water, particularly removing all MP fractions by RSF, DAF, and MBR technologies, and treating all shapes of MPs, including the dominant fibers, employing the four treatment methods (Talvitie et al., 2017). Even with the highest efficacy of MBR demonstrated by Talvitie et al. (2017), there is lack of information regarding the RO treatment of MPs in comparison to MBR under the field conditions at different physicochemical properties of MPs.

For treating MP particle sizes more than 10 μm , a Disc Filter (DF) was found to have 89.7% (by number of particles [MPs L^{-1}]) and 75.6% (by mass of particles [$\mu\text{g L}^{-1}$]) removal efficiency (Simon et al., 2019). Different surface properties of MPs could influence their treatment efficacy, in particular using flotation, and recycling plastic waste through flotation is a sustainable approach to minimizing MP pollution. For example, per a lamellar shape of hydrophobic MP or a regular shape of hydrophilic MP, floatability is increased (Wang et al., 2019b), and the following equation was used to predict the floatability of MPs, based on the shape, density, and bubbles (Shen et al., 2001):

$$\rho_{bp} = \frac{1.59d_{pe}\rho_p\Phi}{1.59d_{pe}\Phi + 4Kd_b} \quad (3)$$

[ρ_{bp} : density of bubble–particle aggregation; ρ_p : particle density; d_{pe} : diameter of an equivalent sphere of an MP particle; Φ : shape parameter (surface area of the equivalent sphere: real particle); K : bubble coverage % of the MP particle surface related to chemical composition; d_b : bubble diameter].

Based on the review of MP treatment technologies, their pros and cons are identified (Table 2). First, although MBR has high removal efficacy of MPs due to its synergistic effect with biodegradation and membrane filtration, it is more costly and energy intensive than sedimentation. Second, sedimentation and flotation are relatively simple to operate through density difference; however, their treatment efficacies are relatively low, and residual pollutants still remain with MPs in these methods. RO can also effectively remove MPs; however, the lower efficacy of MP removal by RO than that by the MBR process (Ngo et al., 2019; Ziajahromi et al., 2017) may be controversial and require further evaluation. In addition, even after RO, residual plastic debris remains. Activated sludge with sedimentation is one of the popular methods widely applied for municipal wastewater treatment and is easy to operate, but the MP treatment efficacy is low and depends on retention time and nutrient level.

RSF has low operation and maintenance costs, and although it could offer rapid MP removal, its removal efficacy is lower than that of the MBR process. Photocatalysis is another feasible and non-toxic treatment method, yet few studies have examined the efficacy of MP removal with photocatalysis; thus, this process warrants further research. Overall, while membrane filtration appears to offer efficient treatment for MPs, such removal efficiency depends on various factors, including membrane's durability (e.g., mechanical strength and reusability), influent flux, and size and concentration of MPs.

3.3. Mechanisms influencing membrane filtration of microplastics

Significant amounts of MPs in the ocean could be from wastewater treatment plants where unit processes suitable for removing MPs are not operated properly. Membrane filtration is widely used in water treatment. Nonetheless, only membrane bioreactors (MBRs) were demonstrated to be efficient in removing MPs, and performance of the other membrane processes were found to be insufficient (Poerio et al.,

2019). By contrast, a study (Enfrin et al., 2019) revealed that ultrafiltration (UF) is a suitable method for removing not only MPs but also NPs in water, because of the pore size of membranes, which is in the same size range with NPs/MPs (Enfrin et al., 2019). Several mechanisms involved in membrane filtration for MP removal include size exclusion, hydrophobic interaction, biofilm formation, and electrostatic interaction, which are reviewed and described further in this section.

3.3.1. Size exclusion

In membrane filtration, size exclusion is a fundamental mechanism of removing contaminants. Membrane selectivity depends on a membrane pore size. Contaminants whose sizes are larger than the membrane pore size are excluded through size exclusion Mulder (2012). In general, the pore size of membranes is smaller than the particle size of MPs. Thus, size exclusion may occur on the membrane surface in the removal of MPs. In the meantime, treated MPs could interact with membranes. For the membrane filtration process, fouling caused by contaminants in feed has been well regarded to influence treatment performance (Kimura et al., 2004; Marshall et al., 1993; Mi and Elimelech, 2010).

Significant amounts of MPs could damage membranes, leading to fouling and ultimately low treatment performance (Ma et al., 2019a; Ma et al., 2019b). Yet, there is a paucity of research on mechanisms/pathways influencing membrane performance in MP removal. A recent study by Enfrin et al. (2020) investigated the membrane fouling effect of MPs extracted from cosmetics on UF. In kinetics and mechanistic aspects of UF, up to 38% decrease in the permeate water flux was shown, compared to pure water filtration with cross-flow filtration of MPs. In the study (Enfrin et al., 2020), the fouling mechanisms of interaction between membranes and MPs were investigated by fitting the reduction kinetics into Hermia's fouling model. In general, Hermia's fouling model plays a significant role to investigate the fouling mechanisms of membranes at different time intervals (Abbasi and Mowla, 2014).

3.3.2. Hydrophobic interaction

MPs with hydrophobic surfaces may have a hydrophobic interaction with hydrophobic polymer membranes in water filtration processes. Such a hydrophobic interaction occurs through a noncovalent force, forming a cluster of nonpolar species in water (Thomas et al., 2016). Flux recovery ratios for hydrophobic membranes were found to be less than those for hydrophilic membranes at pH values of 5, 7, and 9 Hadidi and Zydney (2014). Based on the result Hadidi and Zydney (2014), interactions between hydrophobic materials (foulant) and hydrophobic membranes are likely to influence membrane fouling. Ultimately, such interactions, along with that with other foulants (e.g., proteins) (Huisman et al., 2000; Maximous et al., 2009; Xu et al., 2006), cause fouling by MPs in membrane filtration (hydrophobic membrane filters).

3.3.3. Biofilm formation

Biofilm formation is one of factors influencing membrane treatment efficacy. Few studies are found regarding the influence of MPs on biofouling in membrane filtration, although there is ongoing research in regard to interactions between MPs and microorganisms (Lagarde et al., 2016; Oriekhova and Stoll, 2018; Harrison et al., 2018; Long et al., 2015; Michels et al., 2018; Parrish and Fahrenfeld, 2019). The interaction between MPs and aquatic species (e.g., marine phytoplankton freshwater microalgae) formed heteroaggregates (Lagarde et al., 2016; Oriekhova and Stoll, 2018; Harrison et al., 2018; Long et al., 2015; Michels et al., 2018; Parrish and Fahrenfeld, 2019). The formed heteroaggregates may assist in the removal of MPs or c-MPs in membrane filtration and require evaluation regarding mechanisms and removal kinetics of MPs or c-MPs.

In a wastewater treatment plant (WWTP), bacteria exhibit stronger affinity on MPs from the effluent of WWTP than those from other pathways, because the effluent contains a high concentration of inorganic nutrition, supporting the growth of biofilm biomass (McCormick et al.,

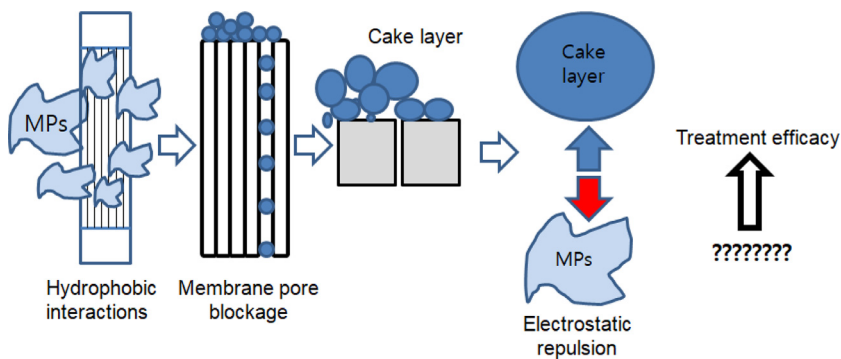


Fig. 3. Schematic of the interaction between MPs and membranes [elaborated from (Enfrin et al., 2019; Enfrin et al., 2020)].

2014). Biofilm formation on MPs influences MP density, thereby causing sinking of MPs in water due to the resultant heavier density of MPs than water; otherwise, MPs float in water (Sun et al., 2019; Juliastuti et al., 2017). The impact of biofilms on the behavior of MPs in water should be explored, but it is challenging because of diverse biological, physical and chemical parameters affecting MP colonization by microbial biota (Enfrin et al., 2020; Harrison et al., 2018).

3.3.4. Electrostatic interaction

Ionized features on the membrane and the foulant cause electrostatic interactions (Breite et al., 2016b). Attractive forces occur between oppositely charged surfaces, whereas repulsive forces become dominant between surfaces with equivalent charges. The intermolecular repulsion between the adsorbed and approaching NP/MP particles indicated a reduction of NP/MP adsorption on membranes, corresponding to a lower flux decline over 48 h (Enfrin et al., 2020). Feed water at high pH causes increasing negative surface charge of the membrane, thereby increasing the rejection rate of pollutants through electrostatic repulsion (Bellona and Drewes, 2005).

When electrostatic repulsion between membranes and MPs occurs, one can expect a high rejection rate of MPs. For example, anionic PS beads were not adsorbed by the membrane at high pH, because of electrostatic repulsion from equally negatively charged membrane and PS beads (Breite et al., 2016b). However, when contaminants adsorbed on PS beads are positively charged at high pH, c-MPs may remain on the membrane surface, thus increasing the rejection rate of c-MPs at an initial stage of membrane filtration, with possible biofouling under long-term treatment. Nonetheless, there is lack of studies regarding MPs with adsorbed contaminants in membrane filtration at different pH levels and types of MPs.

Fouling occurs with decreasing pH values and a decreasing absolute value of zeta-potential to the crucial value (Breite et al., 2016a). In other words, fouling is minimal under electrostatic repulsion conditions (e.g., evenly charged surfaces at low salinity and the corresponding high absolute values of zeta-potential). An electrostatic interaction between polyethersulfone (PES) membranes (e.g., tetraethylene pentamine (TEPA) for cationic membrane and polystyrene sulfonate (PSS) for anionic membrane) and cationic and anionic PS beads was explored (Breite et al., 2016b).

According to the study (Breite et al., 2016b), fouling takes place in oppositely charged surfaces of the membrane and MP particles through electrostatic attraction, whereas electrostatic repulsion under the combination of evenly charged surfaces resulted in no fouling. For zwitterionic membranes, pH could be a critical factor influencing fouling. Such an example is found when at lower pH, repulsion is dominant between a positively charged membrane and cationic PS beads, whereas at higher pH, anionic PS beads are attracted to the positively charged membranes. Based on these studies, not only pH in freshwater and wastewater but also different types of polymers and their zeta-potentials could influence the removal of MPs.

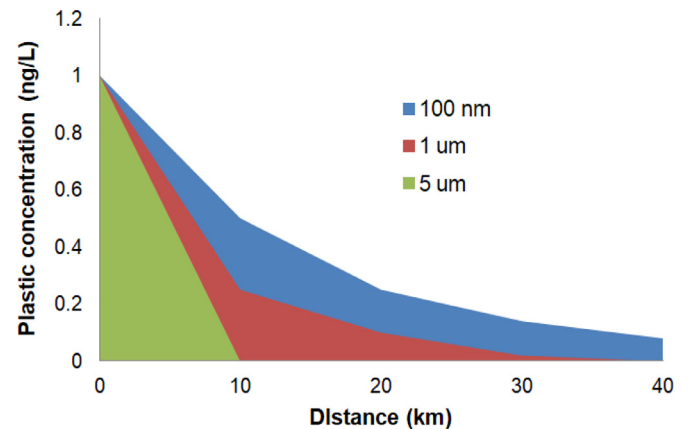


Fig. 4. Spatial distribution of different plastic particle sizes and concentration over a 40 km river flow distance [elaborated from (Besseling et al., 2017)].

As illustrated in Fig. 3, hydrophobic interaction between membranes and MPs is initiated, and because of adsorption of MPs on membrane surfaces, membrane pore blockage may occur. In the meantime, a cake layer is formed among MPs that were adsorbed on membrane surfaces, which later causes a repulsive force between MPs and the cake layer through electrostatic repulsion. Ultimately, this has proved increasing removal efficacy of MPs (Enfrin et al., 2019; Enfrin et al., 2020). Further exploration of the mechanisms of fouling through interactions between a membrane and MPs is encouraged before a suitable membrane filtration method for MP removal is developed.

The removal of MPs in membrane filtration is influenced by several mechanisms such as size exclusion, hydrophobic interaction, biofilm formation, and electrostatic interaction. The influence of the mechanisms on treatment performance is summarized in Table 1. The smaller membrane pore sizes compared to MP sizes result in removal of most MPs (size exclusion), although the treatment efficacy can be low due to fouling and resultant membrane pore blockage. When MPs and the membrane are not charged or under minimal electrostatic repulsion, hydrophobic interaction appears to control the overall MP treatment performance. Biofilm formation on MPs is likely to influence MP removal efficacy because of modification of MP surfaces, which results in a perfect environment for microbes. Electrostatic repulsion from evenly charged membranes and MPs can prevent fouling in MP filtration, thereby maximizing MP treatment efficacy through alteration of surface charges of membranes and MPs and changing pH in feedwater.

4. Future research outlook

Most studies have focused on MPs in the aquatic environment, yet the fate and transport of c-MPs, the influence of contaminant-laden MPs on MP treatment efficacy, and the treatment-associated mechanisms un-

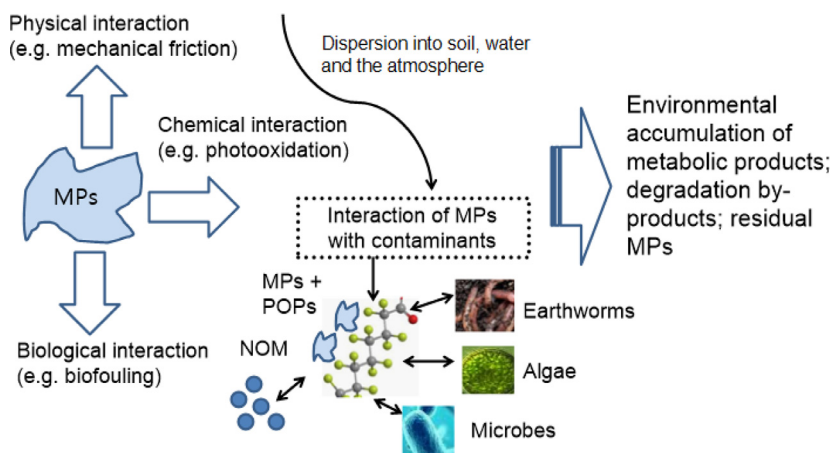


Fig. 5. Schematic of the interaction of MPs with contaminants in the environment.

Table 1
Mechanisms of the removal of MPs in membrane filtration.

| Mechanisms | Treatment performance | Comments | References |
|---------------------------|--|---|--|
| Size exclusion | <p>(a) Removal efficiency of PE MPs over dosage of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ or $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ at pH 7 showed the highest removal of PE MPs with small particle sizes ($d < 0.5 \text{ mm}$) (Ma et al., 2019b).</p> <p>(b) UF membrane filtration of MP size range of 13–690 nm resulted in 38% reduction of permeate water flux, and 25% of NPs/MPs in feed were adsorbed on membrane surfaces within 48 h (Enfrin et al., 2020).</p> | <p>Generally, membrane pore sizes are smaller than MP sizes; thus, most MPs are excluded through membrane filtration (Thomas et al., 2016).</p> <p>Nevertheless, fouling may occur and block membrane pores, resulting in lower treatment efficacy.</p> | [Ma et al., 2019b; Enfrin et al., 2020; Thomas et al., 2016] |
| Hydrophobic interaction | <p>(a) The electrostatic and hydrophobic interactions between MPs (or foulants) and membrane affect the degree of fouling (Hadidi et al., 2014). On a highly hydrophilic zwitterionic membrane, very little fouling is observed (Hadidi et al., 2014), indicating that the hydrophobic interactions between a hydrophobic membrane and hydrophobic foulants (e.g., MPs) may influence membrane fouling.</p> <p>(b) At the initial stages of filtration, hydrophobic interactions between hydrophobic foulants and the membrane appear to determine the overall performance, whereas at later stages of filtration, foulant–foulant interactions (e.g., protein–protein interactions) control the overall performance (Huisman et al., 2000).</p> | Hydrophobic interaction may become dominant and control the overall treatment performance of MPs, where charges of MPs and the membrane become zero or electrostatic repulsion is minimal. | [Hadidi et al., 2014; Huisman et al., 2000] |
| Biofilm formation | <p>(a) Microbial colonization and biofilm formation could be accelerated by MPs that have hydrophobic surfaces (Zettler et al., 2013).</p> <p>(b) MPs from WWTP effluent may enhance more biofilm biomass than MPs from other pathways, because of high levels of nutrients in the effluent (McCormick et al., 2014).</p> <p>(c) Dense bacterial biofilms tended to colonize MP particles—a unique feature of the MP biofilms having significantly less diverse bacterial assemblages in the river than those from other sources (e.g., suspended organic matter and the downstream water column) (McCormick et al., 2014).</p> | <ul style="list-style-type: none"> The removal of MPs could be influenced by modifying the physical properties of MPs through biofilm–MP interactions (Rummel et al., 2017). Because of the hydrophobic nature and large surface area of MPs, they appear to be a perfect habitat for microbes. | [McCormick et al., 2014; Zettler et al., 2013; Rummel et al., 2017] |
| Electrostatic interaction | <p>(a) Combinations of evenly charged membranes and PS beads prevented fouling by electrostatic repulsion (e.g., no fouling in the filtration of anionic PS beads through the anionic PVDF-PSS membrane) (Breite et al., 2016b).</p> <p>(b) When pH increases, the zeta potential of the membrane becomes highly negative, and solutes are increasingly deprotonated (the removal of a proton), thereby achieving a high rejection rate of contaminants due to electrostatic repulsion (e.g., at pH 9, the zeta potential value of the NF-200 membrane was -22 mV, and the highest rejection ($\sim 90\%$) of 1,5-naphthalenedisulfonic acid was observed) (Bellona and Drewes, 2005).</p> | Maximizing treatment performance of MPs could be achieved through alteration of surface charge (Fotopoulou and Karapanagioti, 2012) of the membrane and MPs, pH change in feed. | [Bellona and Drewes, 2005; Breite et al., 2016b; Fotopoulou and Karapanagioti, 2012] |

Table 2
Removal of MPs using membrane filtration vs. other primary treatment methods.

| Membrane filtration vs. other methods | Treatment Performance Pros. | Cons. | References |
|--|---|--|---|
| MBR [Removal rate: 99.9%] | (a) High removal efficiency of MPs as one of commonly applied membrane technologies (b) Good exclusion of MPs due to the smallest pore size (c) A combined method of biodegradation and membrane filtration | (a) Difficulty to control various parameters influencing the treatment performance (e.g., which depends on various types, sizes, and concentrations of MPs) (b) More costly and energy intensive than sedimentation | [Lv et al., 2019; Ngo et al., 2019; Lares et al., 2018; Talvitie et al., 2017] |
| RO [Removal rate: 90.4%] | (a) Removal through semi-permeable membrane (b) Ease of treatment | (a) Lower removal efficiency of MPs than that by the MBR process (b) Even after RO, plastic debris still remains in WWTPs | [Ngo et al., 2019; Ziajahromi et al., 2017] |
| Sedimentation [Removal rate: 91.7%] | (a) Straightforward method of treatment through gravitational settling (b) Removal of high density of MPs | (a) Residual pollutants with MPs (b) Removal efficacy depends on the density of MPs (c) Issues with MPs present in landfill leachate and storm water runoff returning to WWTPs | [Cheremisinoff, 2002; Ngo et al., 2019; Hermabessiere et al., 2017] |
| Flotation [Removal rate: 95%] | (a) Simple treatment method (floating by air bubble) (b) Removal of low density of MPs | (a) Potential residual pollutants with MPs (b) Removal efficacy depends on the density of MPs | [Ngo et al., 2019] |
| Activated Sludge + Sedimentation [Removal rate: 66.7%] | (a) Popular technology in municipal WWTPs (b) Uncomplicated treatment (by microorganisms) | (a) Treatment efficacy depends on retention time and nutrient level (b) Leach out or adsorption of toxic chemicals (in long retention time) (c) Low removal efficiency of MPs | [Ngo et al., 2019; Hermabessiere et al., 2017; Scherer et al., 2018; Rummel et al., 2017] |
| Rapid sand filtration [Removal rate: 97%] | (a) Rapid and efficient removal method (b) Low operational and maintenance cost | (a) Removal efficiency is lower than that by the MBR processes. (b) Filter pore size can be larger when reuse of this process is caused by sand friction. | [Ngo et al., 2019; Talvitie et al., 2017; Michielssen et al., 2016] |
| Photocatalysis [Removal rate: 30%] | (a) Feasible and non-toxic process (b) Ease of treatment (coating photocatalytic material) (c) Clean technology | Few studies have examined the efficacy of MPs with photocatalysis, and this requires further research | [Ngo et al., 2019; Tofa et al., 2019] |

der various environmental conditions are not fully understood because of lack of relevant research. Given that significant amounts of MPs and additives associated with MP products are detected and accumulated in soil and sewage sludge (Wang et al., 2019a), more research should be conducted on MPs in soil, sewage sludge, and biosolids and on treatment of residual pollutants adsorbed on MPs. Furthermore, there is a paucity of research on MPs in membrane retentate (concentrate), even if membrane filtration of MPs was proved to be effective. High salinity present in membrane retentate could influence the fate of MPs or c-MPs.

Recently, electrospun nanofibers, which can be applied in MF, UF, NF, and RO, indicated improvement in flux and fouling resistance and improved performance of UF membranes (Dobosz et al., 2017). Nanofibers produced by electrospinning offer various benefits, including a cost-effective method for environmental remediation, a low pressure drop, greater surface adsorption of contaminants due to the large exposed surface area of nanofiber membranes, easy surface modification, and long lifespan of the filtration media (Thavasi et al., 2008). Nonetheless, few studies are found on MP removal efficacy using electrospun nanofibers applied in membrane filtration, and the application of electrospun nanofibers deserves future research. Among treatment methods for MP removal, combined coagulation/flocculation and membrane filtration could be vital and even more effective for MP removal. Thus, the implication of the presence of OM adsorbed on MPs in water treatment processes (e.g., coagulation/flocculation) deserves further research.

Development of analysis technologies for detecting and characterizing trace levels of MPs in the environment, particularly in sewage

sludge, biowaste composts, warrants further research, along with the risk assessment of biosolids because of a significant concern regarding plastic-containing fertilizers applied in agriculture. The properties of the MP load are also imperative to the assessment of negative environmental impacts of MPs and should be considered in risk assessment of MPs. There has been little attention regarding MPs (particularly MPs of small sizes) in potable water (Novotna et al., 2019), and these should be addressed in future research. Regulations and implementation to control MP pollution should also consider various factors including geographic locations and hydrodynamic conditions due to the diversity of MP accumulation (Hamid et al., 2018).

5. Conclusions

MPs and emerging contaminants of concern (e.g., PFAS) are recalcitrant to degradation in natural environmental systems. Through literature analysis of MPs or c-MPs, several findings are summarized as follows.

- (1) Characteristics of MPs (e.g., size, shape, and polymorphism) and environmental conditions (e.g., solution pH, ionic strength, OM, microbes, and temperature) affect the fate of MPs and c-MPs and the interactions between contaminants and MPs, through several mechanisms including hydrophobic and electrostatic interactions.
- (2) In membrane filtration, four primary mechanisms including size exclusion, biofilm formation, and hydrophobic and electrostatic interactions influence the MP treatment efficacy, which could control the MP removal through modification of MP surface prop-

- erties, changes in pH and membrane pore size, or even addition of bactericidal agents.
- (3) Different MP sizes and concentrations influence MP transport, with smaller MPs widely distributed and difficult to remove in the environment. MPs that have undergone physical, chemical, and biological interactions are released into the environmental media and have different sorption capacities of contaminants, ultimately leading to environmental accumulation of metabolic by-products, degradation by-products, or residual MPs.
 - (4) A hybrid treatment technology such as MBR or a combined coagulation and membrane filtration appears to be the most effective treatment for MPs or c-MPs. In field case studies at WWTPs, MBR exhibited the highest MP removal rate among other tested treatment methods (e.g., RO, sedimentation, flotation, and RSF). However, given significant amounts of MP accumulation in wastewater, enhancing pretreatment should be precedent to MBR, alleviating shortcomings of MBR.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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