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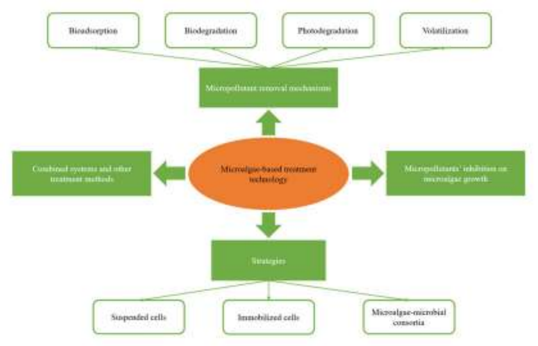
The application of microalgae in removing organic micropollutants in wastewater

Hoang Tam Nguyen[a], Yeomin Yoon[b], Huu Hao Ngo[c], and Am Jang[a]

[a]Graduate School of Water Resources, Sungkyunkwan University, Suwon, Gyeonggi-do, Republic of Korea; [b]Department of Civil and Environmental Engineering, University of South Carolina, Columbia, SC, USA; [c]Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Broadway, NSW, Australia

ABSTRACT



Micropollutants have become a serious environmental problem with several negative outcomes for human health and ecosystems. Many efforts have been made to remove micropollutants using a variety of physical, chemical and biological methods. By far, the most attention has been paid to microalgae-based technologies for wastewater treatment in order to obtain high-quality effluents, recover algal biomass for fertilizers, protein-rich feed, biofuel, and put them to other practical use. This paper reviews the potential of microalgae-based systems for the removal of organic micropollutants from open ponds to closed photobioreactors coupled by suspended microalgal cells, immobilized cells, or microalgae-microbial consortia. The inhibition of micropollutants on microalgae growth as well as micropollutant removal mechanisms performed by microalgae-based systems are also discussed. Other treatment methods for the removal of micropollutants are analyzed to show the advantages and limitations of microalgae-based treatment strategies, from which some possible combined systems can be suggested. Finally, some recommendations for future studies on this topic are proposed.

Abbreviations: AOPs: Advanced oxidation processes; BOD: Biological oxygen demand; COD: Chemical oxygen demand; CWs: Constructed wetlands; EDCs: Endocrine-disrupting compounds; HRAPs: High-rate algal ponds; MBR: Membrane bioreactor; MFCs: Microbial fuel cells; MPs: Micropollutants; PBRs: Photobioreactors; PCPs: Personal care products; PPCPs: Pharmaceuticals and personal care products; WWTPs: Wastewater treatment plants

KEYWORDS Microalgae; micropollutants; photobioreactor

CONTACT Am Jang ✉ amjang@skku.edu 🖃 Graduate School of Water Resources, Sungkyunkwan University, Suwon, Gyeonggi-do, Republic of Korea.

1. Introduction

Micropollutants (MPs) are defined as anthropogenic chemicals found in different water bodies with concentrations remaining at trace levels, up to the microgram per liter range. These emerging organic pollutants mainly consist of pharmaceuticals, personal-care products, steroid hormones, industrial chemicals, pesticides, polyaromatic hydrocarbons and other recently seen compounds (Wanda et al., 2017). Heavy metals, which are considered inorganic MPs, are not targeted in this paper. Herein, MPs refer only to organic MPs. These contaminants are generally associated with adverse effects, such as endocrine disruption, acute and chronic toxicities for different species, and particularly the development of antibiotic resistance in microorganisms (Tijani et al., 2016). Even at very low levels, the continuous discharge of MPs into the environment may elevate the abnormalities in reproduction and development of sensitive species (Lecomte et al., 2017).

At present, few discharge standards and regulations about MPs have been published and implemented. Only some developed countries have made regulations regarding these MPs, mostly surfactants, industrial chemicals and pesticides; pharmaceuticals, personal care-products (PCPs) and steroid hormones are generally not listed (Kim & Zoh, 2016). The detection and analysis of MPs during treatment are generally complicated because of the low concentration and diversity of these compounds (Luo et al., 2014). Although current wastewater treatment processes can reduce the concentrations of many MPs, they are not specifically designed to remove them. Moreover, stable structures make it even more difficult to eliminate these compounds (Rizzo et al., 2019). Because of their distinctive properties, there is no specific treatment for the complete removal of all MP groups. In addition, these treatment processes cannot simultaneously remove both bulk contaminants and MPs very efficiently (Dolu et al., 2017).

During the past few decades, several efforts have been made to remove or degrade MPs using a variety of physical, chemical and biological technologies. Among these, several methods are mainly with reference to microalgae-based wastewater treatment technologies using high-rate algal ponds (HRAPs) and/or photobioreactors (PBRs) as these can recover algal biomass for fertilizers, protein-rich feed, biofuel and provide high-quality effluents from the system (Ravindran et al., 2016). In addition to some common organic and inorganic compounds present in wastewater (e.g. ammonium, nitrate, phosphates), microalgae can biodegrade and assimilate more persistent molecules, such as hydrocarbons, antibiotics, pharmaceuticals and personal care products (PPCPs), endocrine-disrupting compounds (EDCs) and heavy metals (Delrue et al., 2016). Moreover, microalgae can simultaneously sequestrate CO_2 during photosynthesis, which reduces greenhouse-gas emissions (Razzak et al., 2013).

To date, several studies have focused on microalgae's ability to remove some specific MPs. However, only a few reviews on the efficiencies and removal mechanisms of microalgae-based wastewater treatment systems have been published. Especially, there is no comparison between the performances of suspended cells, immobilized cells, and consortia of microalgae species in MP removal. Accordingly, we aim to evaluate and compare the potentials of different microalgae-based systems for MP removal, as noted previously. The inhibitions of MPs on microalgae growth are also discussed. In addition, we provide an overview of treatment methods for MP removal in order to investigate the advantages and limitations of microalgae-based methods compared to others. Finally, some recommendations for future studies on this topic are proposed.

2. MP removal mechanisms in microalgae-based treatment systems

Biodegradation, photodegradation, volatilization and sorption to the biomass are the most relevant contaminant-removal processes occurring in microalgae PBRs (Bilal et al., 2018; Matamoros et al., 2015; Norvill et al., 2016, 2017; Petrie et al., 2015; Tolboom et al., 2019; Wang & Wang, 2016). These four removal mechanisms are illustrated in Figure 1. The most important pathways for the removal of compounds appear to be biodegradation and photodegradation, whereas sorption and volatilization are significant only for hydrophobic compounds ($\log K_{ow} > 5$) and pollutants with a moderately high Henry's law constant ($K_H > 10^{-3}\,\mathrm{atm\ m^3\ mol^{-1}}$), respectively (Duchowicz et al., 2020; Tolboom et al., 2019; Wang, Liu, et al., 2017). The treatment efficiencies of microalgae-based systems and their main mechanisms to remove some MPs are summarized in Table 1.

2.1. Bioadsorption of MPs by microalgae

Biosorption is a physical-chemical process that can be simply defined as the removal of substances from solution using biological material (Fomina & Gadd, 2014). Many researchers consider biosorption to be a subcategory of adsorption, where the sorbent is a biological matrix (Michalak et al., 2013). Due to the presence of dominant functional groups (e.g. carboxyl, phosphoryl, amide), the microalgae cell wall is negatively charged. As a result of the electrostatic interaction, contaminants with cationic groups are effectively attached to the microalgal surface, resulting in biosorption. Because adsorption is extracellular, the process depends largely on the

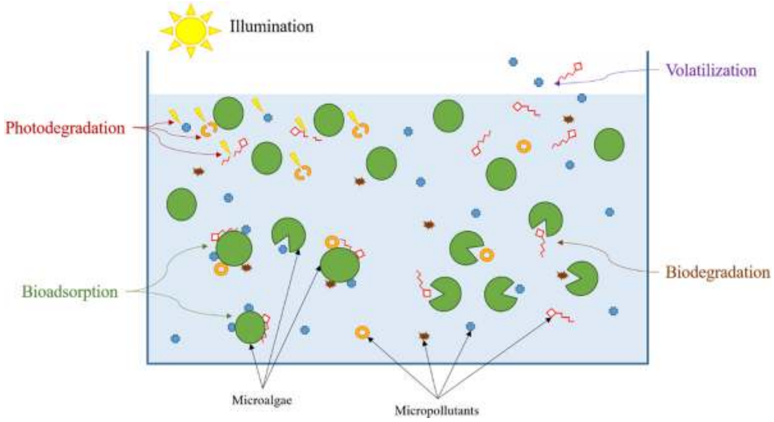


Figure 1. Micropollutant removal mechanisms in microalgae-based system.

hydrophobicity, structure, and functional groups of different MPs and microalgae species (Xiong, Kurade, et al., 2018).

Cho et al. (2019) studied the adsorptive interaction of 30 MPs with microalgae *Chlorella vulgaris* to investigate the environmental fate and transport of pharmaceutical waste. They measured the isotherms between *Chlorella vulgaris* and 30 chemicals in neutral and ionic forms to predict their adsorptive affinities based on the linear free-energy relationship. Dispersive and hydrophobic interactions prompt adsorption of pharmaceuticals toward *Chlorella vulgaris*, whereas the polar, hydrogen-bonding, and anionic interactions of these compounds repel the adsorption (Cho et al., 2019).

The amount of MPs (including diclofenac, ibuprofen, paracetamol, metoprolol, trimethoprim, carbamazepine, estrone, β-estradiol, and ethinylestradiol) adsorbed by microalgae ranges from a lower limit of detection to 16.7% of the removed amount (de Wilt et al., 2016). In addition, adsorption is the only contributing factor of the dead microalgae for MP removal. The dead-cell biomass of *Scenedesmus obliquus* and *Chlorella pyrenoidosa* was described as able to adsorb approximately 10% of two studied MPs, i.e., progesterone and norgestrel (Peng et al., 2014). This is because algae cell walls contain several polymers that are similar to cellulose, pectin, chitin, alginate, glycan, lignin, etc. (Fomina & Gadd, 2014; Xiong, Kurade, et al., 2018). These chemical components furnish important sites for the sorption of organic contaminants. The microalgae cell wall is negatively charged due to the presence of the dominant functional groups such as carboxyl, amine and phosphoryl. Therefore, contaminants with cationic groups are effectively attracted toward the microalgae surface, even the dead cells, through electrostatic interaction, resulting in biosorption.

Compared to other removal mechanisms, sorption onto biomass contributed less to the total removal (de Godos et al., 2012). Norvill et al. (2017)

Table 1. Micropollutant treatment efficiencies and main removal mechanisms performed by different microalgae species.

Main removal mechanism	Microalgae species	Micropollutants	Initial concentrations	Removal (%)	References
Biodegradation	*Phaeodactylum tricornutum*	Oxytetracycline	2.5 mg/L	97	Santaeufemia et al. (2016)
	Selenastrum capricornutum,	17β-estradiol, 17α-ethinylestradiol	5 mg/L	60–100	Hom-Diaz et al. (2015)
	Chlamydomonas reinhardtii			100	
	Scenedesmus obliquus,	Diclofenac	25 mg/L	99	Escapa et al. (2018)
	Chlorella vulgaris,			71	
	Chlorella sorokiniana			67	
	Chlorella sorokiniana	Paracetamol	100–350 µg/L	>99	de Wilt et al. (2016)
		Metoprolol		>99	
	Algal-bacterial consortia	Naproxen	4.2 µg/L	28–52	López-Serna et al. (2019)
		Salicylic acid	63 µg/L	83–97	
		Atrazine	0.01 mg/L	84–95	Derakhshan et al. (2019)
	Inoculum from lake water	Acetaminophen	50 µg/L	>99	Hom-Diaz et al. (2017)
		Ibuprofen	53 µg/L	>98	
		Naproxen	25 µg/L	69	
	Algae pond	Estrone, 17β-estradiol, 17α-ethinylestradiol	1 µg/L	52–56	Shi et al. (2010)
Bioadsorption	Microalgal-bacterial consortia	Tritosan	537 ng/L	85–100	López-Serna et al. (2019)
		Propylparaben	408 ng/L	87–100	
	Selenastrum capricornutu	17β-estradiol	5 mg/L	42	Hom-Diaz et al. (2015)
	Chlamydomonas reinhardtii			54	
Photodegradation	*Chlorella sorokiniana*	Diclofenac	100–350 µg/L	40–60	de Wilt et al. (2016)
		Ibuprofen		100	
		1H-benzotriazole	200 µg/L	79	Gatidou et al. (2019)
		Xylytriazole		>42	
		5-methyl-1H-benzotriazole		>97	
		5-chlorobenzotriazole		52	

indicated that sorption accounted for only 6% of the total tetracycline removal in algae ponds. They also pointed out that the sorption removal of estrogens onto microalgae biomass was apparent only given high initial concentrations (e.g. 5 mg/L). Concentration gradient could be one explanation for this phenomenon since it would facilitate the adsorption rate. Another possible reason is that when given low initial micropollutant concentrations, biodegradation can cause the subsequent release of sorbed contaminants, leading to the decrease in the removed amount by adsorption mechanism. Besides, microalgae's ability to absorb MPs may be affected by pH value and the amount/composition of extracellular polymeric substances in PBRs (Cheng et al., 2019).

2.2. Biodegradation of MPs by microalgae

Biodegradation is defined as the breakdown of organic chemicals catalyzed by enzymes produced by microorganisms (Wang, Liu, et al., 2017). The mechanisms can be either metabolic degradation or co-metabolism. For metabolic degradation, organic chemicals are used as the sole carbon and energy sources. For co-metabolism, on the other hand, the degradation of MPs depends on nonspecific enzymes present in the environment that will catalyze the metabolism of other substrates (Tiwari et al., 2017). Because the concentration of MPs in wastewater is usually within the ng/L to µg/L range, it may be insufficient to maintain the growth of microalgae via metabolic degradation. Therefore, co-metabolism may be the main process responsible for the degradation of MPs (Tran et al., 2013). For example, Quintana et al. (2005) examined the biodegradation pathways of some acidic pharmaceuticals in wastewater treatment process by comparing their biodegradation with and without external carbon source. Bezafibrate, naproxen and ibuprofen were proved to be degraded only co-metabolically. In addition, Tran et al. (2013) reviewed the metabolic and co-metabolic activities of autotrophic and heterotrophic microorganisms in the biodegradation of micropollutants. They also point out that there is still no available information on metabolism by autotrophic microorganisms.

Biodegradation is considered to be the most effective way by which microalgae eliminates organic MPs from the aqueous phase. It can be categorized as intracellular biodegradation and extracellular degradation. Under microalgal activity, complex parent compounds are catalytically degraded and thus simpler molecules are formed. With a complex enzyme system, microalgae have different types of enzymatic reactions with the MPs during biodegradation, for example hydroxylation, carboxylation, oxidation, hydrogenation, demethylation and ring cleavage (Ding et al., 2017). In addition, microalgae can also excrete various extracellular polymeric substances to form a hydrated biofilm matrix. This layer can act as an external digestive system by keeping the extracellular enzymes close to the ells and enabling them to metabolize the organic compounds (Xiong, Kurade, et al., 2018). Within the biofilm, organic compounds (either in dissolved, colloidal, or solid form) imported through the water phase of the matrix can be sequestered, accumulated and utilized owing to the retention of extracellular enzymes in the matrix (Flemming & Wingender, 2010).

Some previous research has investigated the capability and mechanisms for the removal of MPs using microalgae in laboratory batch experiments. As summarized in Table 1, microalgae are able to remove many MPs belonging to estrogenic hormones, antibiotics, antimicrobials, anti-epileptics and nonsteroidal anti-inflammatory drugs. For most of the investigated contaminants, biodegradation was found to be the main removal mechanism.

Biodegradation by microalgae was the main removal mechanism of diclofenac, a nonsteroidal anti-inflammatory drug with the highest removal efficiency of 99% when using *Scenedesmus obliquus* (Escapa et al., 2018). In other studies, several pharmaceutically active compounds including ibuprofen, caffeine, carbamazepine, and tris(2-chloroethyl) phosphate were biodegraded about 30–80% by microalgae in urban or synthetic wastewater (Ding et al., 2017; Hom-Diaz et al., 2017; Matamoros et al., 2016). However, during biodegradation, if the concentration and/or toxicity of the transformation products exceed those of the parent compounds, a large removal does not warrant an efficient treatment. For example, Phong Vo et al. (2019) investigated the occurrences, toxicity, removal strategies and transformation pathways of acetaminophen micropollutant. More than 20 by-products and intermediates of acetaminophen were detected with different toxicities. Some of them are more detrimental than the parent form such as N-acetyl-p-benzoquinone imine (Liang et al., 2016). Accordingly, they concluded that treated wastewater was not totally free from the toxic effects due to acetaminophen metabolites. Hence, effects of the final effluent on living organisms need to be tested for a comprehensive evaluation of the microalgae-based treatment efficiency. Escapa et al. (2018) confirmed that the removal of diclofenac by three microalgae strains, namely *Chlorella sorokiniana, Chlorella vulgaris*, and *Scenedesmus obliquus,* did not generate compounds toxic to zebrafish embryos.

2.3. Photodegradation

Illumination is required for photosynthesis in a microalgae-based treatment system. Therefore, photodegradation can play an important role in the removal of MPs in wastewater treatment systems using microalgae-based technologies. The principle relies upon solar energy needed for photosynthesis that will also induce a pathway for micropollutant removal through direct or indirect UV photodegradation (Hom-Diaz et al., 2015). Direct photodegradation encompasses the direct absorption of light by the pollutants, which results in a chemical reaction. The micropollutant structure determines whether the radiation absorption occurs, in which the molecular energy increases, leading to bond breaking and degradation (Gruchlik et al., 2018). Indirect photolysis can be explained by the dissolved organic substances in wastewater that can absorb light energy and produce reactive oxygen species (e.g. hydroxyl radicals, singlet oxygen), which subsequently degrade the target contaminants (Norvill et al., 2016). Due to the strong attenuation of light in wastewater, indirect photodegradation contributes much more to the overall removal than does direct photolysis (Zhang et al., 2014). Norvill et al. (2017) found that the rate of tetracycline photodegradation was 7 times greater than the control due to indirect photodegradation.

The photodegradation mechanism depends on the molecule structure of micropollutants and on environmental conditions. Wang et al. (2016) studied the photodegradation of seven micropollutants. The experimental result indicated that sulfamethoxazole and triclosan were susceptible to sunlight exposure, whereas carbamazepine, diuron, simazine, caffeine and 2,4-Dichlorophenoxyacetic acid were not. In addition, Mathon et al. (2019) studied the solar photodegradation of 23 organic micropollutants at different depths. The results showed that photodegradation is most effective in the first 10 cm of the water column. Amongst studied micropollutants, ketoprofen, diclofenac, fenofibric acid, and metronidazole were classified as fast-photodegradable. Five micropollutants defined as medium-photodegradable were acebutolol, theophylline, sulfamethoxazole, sotalol, and isoproturon. Besides, six slow-photodegradable microppolutants included diazepam, atrazine, dimethoate, diuron, simazine, and cyclophosphamide (Mathon et al., 2019). The photodegradation rate of a particular compound is also affected by the change in solar irradiance intensity with both latitude and season. For example, the removal of diclofenac and ketoprofen (fast-photodegradable compounds) was better in summer with the average daily solar irradiation of 282 $\mathrm{W m^{-2}}$ compared to winter with much lower solar irradiation (74 $\mathrm{W m^{-2}}$) (Gruchlik et al., 2018).

The disadvantages of this removal mechanism may include the incomplete degradation of micropollutants as well as the great dependence on the chemical properties of contaminants. The transformation products are important to identify because some intermediate compounds may be more toxic than the parent compounds. For instance, acridine, a photodegradation product of carbamazepine, is mutagenic and carcinogenic. The photodegradation products of naproxen are also more toxic than the parent compound (Rivera-Utrilla et al., 2013). Additionally, not all micropollutants are photodegradable and this phenomenon only occurs predominantly at the upper layer of the water column. In some circumstances, photodegradation could not be considered as the main removal mechanism for micropollutants but a supplemental one.

2.4. Volatilization

Volatilization can contribute to the removal of volatile and semi-volatile MPs in an open microalgae-based treatment system. The elimination of trace pollutants by volatilization during the microalgae-based process depends on Henry's law constant of the analyzed trace pollutants, and becomes significant when this value is greater than 10^{-3} atm m^3 mol^{-1} (Duchowicz et al., 2020). However, most MPs (e.g. pharmaceuticals) are large molecules with low Henry's law constants ($K_H < 10^{-3}$ atm m^3 mol^{-1}) (e.g. diazepam 2.7×10^{-5}, diclofenac 3.42×10^{-5}, naproxen 2.52×10^{-5}, ibuprofen 1.1×10^{-4}, sulfamethoxazole 4.68×10^{-5}, trimethoprim 1.76×10^{-4}, erythromycin 3.96×10^{-5}, roxithromycin 1.8×10^{-5}, etc.) (Suárez et al., 2008). Thus, volatilization is usually considered negligible. This removal mechanism is also affected by the air stripping intensity and temperature in the system. In any case, volatilization might not be a desired outcome for wastewater treatment, since it converts pollutants only from a liquid phase into the atmosphere instead of degrading them into smaller molecules (Wang, Liu, et al., 2017).

The capacity to remove many MPs simultaneously with other pollutants of microalgae-based treatment systems has been demonstrated only somewhat in the research. However, poor removal was also noted for several MPs, such as carbamazepine (30–37%), lorazepam (30–57%), trimethoprim (0%, non-degradable), and verapamil (12–40%), perhaps because of their low biodegradability, lack of functioning UV absorption groups, or physical-chemical properties (Kovalova et al., 2013; Wang, Liu, et al., 2017).

3. MPs' inhibition on microalgae growth

Although microalgae-based wastewater treatment is a potential system being a source for microalgae biomass production, the high concentrations of nutrients and toxic contaminants could, nonetheless, inhibit the microalgae growth. There are contradictory results about the influence of MPs on microalgae growth. On one hand, Li et al. (2016) found that tetracycline concentrations of 0-0.25 mg/L exerted a positive impact on the growth and nutrient removal of *Chlamydomonas reinhardtii*. On the other hand, the inhibitory effect of different tetracycline concentrations on microalgae has been documented. Xiong, Hozic, et al. (2018) stated that the generation of microalgae biomass was not affected at a low tetracycline concentration (≤150 µg/L), but was inhibited significantly when tetracycline increased to 20 mg/L. Pomati et al. (2004) found that the *Synechocystis* growth reduced by 20–22% at a tetracycline concentration of 10 and 100 µg/L, but no negative effect was seen at 1000 µg/L. One possible reason proposed by these authors was that at high tetracycline concentration, the production of exoenzymes was induced, which converted the antibiotics into smaller molecules and reduce their effect on *Synechocystis* growth. The different responses of microalgae toward the presence of MPs could be the consequence of natural variability in MP resistance between different species. The sensitivity and type of response to MP are dependent upon the transport of MP molecules across the cell membrane. Therefore, the opposing responses may suggest differences in the uptake mechanism.

According to de Wilt et al. (2016), the growth of the microalgae *Chlorella sorokiniana* was not inhibited by the presence of MPs at applied concentrations (100–350 µg/L). Escapa et al. (2016) also reported that the presence of diclofenac provided a larger microalgae biomass concentration than did the controls. This phenomenon can be explained that, for the studied strains, the addition of diclofenac meant an organic carbon source supplying for their heterotrophic growth and thus, provided higher biomass concentration. Moreover, microalgae-based treatment systems can simultaneously remove MPs and recover nutrients. Harvested microalgae biomass can be applied as fertilizer in agriculture (de Wilt et al., 2016). The cultivation of microalgae in wastewater has been intensively studied focusing on biomass production and factors affecting the growth of microalgae (Komolafe et al., 2014; Leite et al. 2019; Prandini et al., 2016; Shahid et al., 2020; Wang et al., 2015; Wang, Ho, et al. 2017). Although sewage wastewater is a potential source for microalgae cultivation, high concentrations of contaminants may negatively affect the rate of microalgae growth as well as biomass production. In general, microalgae growth rate depends on initial nutrition levels, cultivation conditions (e.g. light intensity, pH, temperature, cultivation modes), the presence of MPs (types, concentrations), and microorganisms in wastewater (Gatamaneni et al., 2018; Li et al., 2016; Metsoviti et al., 2019; Miazek et al., 2015). Regarding the presence of micro-contaminants in wastewater, their inhibition on microalgae growth varies with MP types, concentrations, and exposed microalgae species as shown in Table 2.

4. Strategies of microalgae-based treatment systems for removing MPs

The removal of MPs has been studied using microalgae-based systems with different configurations, ranging from open ponds to closed PBRs, applying

Table 2. Microalgae growth inhibition in the presence of micropollutants.

			Inhibition		
Micropollutants	Microalgae	Concentrations	Yes	No	References
Tetracycline	*Synechocystis*	10 μg/L	O		Pomati et al. (2004)
		100 μg/L	O		
		1000 μg/L		O	
	Chlamydomonas reinhardtii	0–0.25 mg/L	O		Li et al. (2016)
	Chlorella spp.	≤150 μg/L		O	Xiong, Hozic, et al. (2018)
		20 mg/L	O		
Sulfamethoxazole	*Selenastrum capricornutum*	1.0 mg/L	O		Liu et al. (2011)
Sulfamonomethoxine	*Chlorella vulgaris*	2.5–20 mg/L	O		Huang et al. (2014)
	Scenedesmus obliquus	≤0.5 mg/L		O	Xiong, Hozic, et al. (2018)
Carbamazepine	*Chlamydomonas mexicana*	200 mg/L		O	Xiong et al. (2016)
	Scenedesmus obliquus		O		
Diclofenac	*Chlorella sorokiniana*	147 ± 9 μg/L		O	de Wilt et al. (2016)
Ibuprofen		317 ± 33 μg/L		O	
Paracetamol		337 ± 23 μg/L		O	
Metoprolol		181 ± 62 μg/L		O	
Carbamazepine		117 ± 17 μg/L		O	
Trimethoprim		202 ± 30 μg/L		O	
Diclofenac	*Chlorella sorokiniana* *Chlorella vulgaris* *Scenedesmus obliquus*	25 mg/L		O	Escapa et al. (2016)

suspended cells, immobilized cells, or consortia. This section will discuss each configuration and summarize the results published in previous studies.

4.1. Suspended cells

4.1.1. Open ponds

Open ponds are the most extensively used reactors for large-scale microalgae cultivation because of the reasonable construction cost, low energy usage, easy scale-up, and simple cleaning compared to closed PBRs. High-rate algal ponds (HRAPs), circular ponds, and tanks are the commonly used forms that have received the most attention. HRAPs are shallow raceway reactors in an open space, hence contamination by bacteria is unavoidable. In HRAPs, microalgae and bacteria grow in symbiosis and consequently do not require aeration (Matamoros et al., 2015). Compared to the conventional activated sludge process for wastewater treatment, HRAPs provide an additional advantage in that shallower ponds are used to retain water, thereby allowing better penetration of light through the water column. Optimal depths for HRAPs reported in the literature range from 15 to 100 cm (Grobbelaar, 2012; Park et al., 2011; Sutherland et al., 2014), while the depth for activated sludge process usually ranges from 2.4 to 6 m (Norvill et al., 2016). In accordance with better photocapture by

Table 3. Micropollutants removal by HRAPs compared to conventional activated sludge.

Category	Compound	Initial concentration (μg/L)	Removal efficiency (%)	Conventional activated sludge (%)	References
Analgesics	Acetaminophen	135	98	99	Villar-Navarro et al. (2018)
	Ibuprofen	25	86	96	
	Naproxen	5	20	80	
Lipid regulators	Gemfibrozil	3.2	41	28	
β-blockers	Atenolol	1.5	73	40	
H2-blocker	Ranitidine	2.2	82	45	
Drugs	Furosemide	1.7	76	47	
EDCs	Bisphenol A	00–24	85	63–99	Luo et al. (2014); Matamoros et al. (2015); Stasinakis et al. (2013)
	Octylphenol		93	<97	
Pesticides	Atrazine		85	25	
	Benzothiazole		78	40–60	
	2,4-dichlorophenoxyacetic acid		32	–	
	Diazinon		63	0	
	OH-benzothiazole		84	60	
	Triclosan		95	71–99	
Fragrance agents	Methyl dihydrojasmonate	–	92–99	98	
	Cashmeran	–	61–79	50	
PCPs	Oxybenzone	–	75–99	63-98	
Flavoring agents	Hydrocinnamic acid	–	99	–	
Pharmaceuticals	5-methyl benzotriazole	–	74-95	60	
Antibiotics	Tetracycline	2000	69	–	de Godos et al. (2012)
NSAIDs	Ketoprofen	24	95	–	Garcia-Rodríguez et al. (2014)
Stimulants	Caffeine	9	85–98	50–99	

algae, the shallow depth enhances the photodegradation of photosensitive compounds (Delrue et al., 2016).

Some previous research has investigated the performance of HRAPs systems for MP removal and compared them to the activated sludge process in conventional WWTPs (Matamoros et al., 2015; Villar-Navarro et al., 2018). The removal by HRAPs is about as efficient as that of conventional activated sludge, as can be seen in Table 3.

The most commonly used analgesics and anti-inflammatories (e.g. ibuprofen, acetaminophen, and naproxen) were slightly better removed in the conventional WWTPs. Conversely, HRAPs were more efficient at eliminating some pharmaceuticals that are of major environmental concern such as diclofenac and antibiotics (Villar-Navarro et al., 2018). This phenomenon can be linked to the MP molecular properties, which define their biodegradation abilities by a given strain of microorganism under given operating conditions. Accordingly, some pharmaceuticals that were not appreciably removed in activated sludge (e.g. carbamazepine) were not eliminated by HRAPs either. For example, the presence of a relative complex aromatic structure or chlorine in the molecule are the reasons for the low degradation rates of clofibric acid, dichloprop, and dichllorofluorescein (Grandclément et al., 2017). Some physico-chemical characteristics (e.g. hydrophobicity, functional groups) are important factors governing MP biodegradation. MP removal are, therefore, compound- and process-specific. As a result, hybrid processes are expected to achieve better MP removal, which will be discussed in Section 5.

During photosynthesis, microalgae release oxygen as a by-product which can be utilized by aerobic microorganisms to further degrade the remaining organic loads. This lessens the energy cost for aeration compared to conventional wastewater treatment system. Craggs et al. (2014) stated that daytime algal photosynthesis can supersaturate the concentration of dissolved oxygen to over 20 g m^{-3} in the HRAP. Therefore, HRAP paddlewheels consume only one-tenth of the energy required for mechanical aeration in conventional WWTPs. While approximately 1 kWh of electricity is needed to remove 1 kg of BOD in the conventional activated sludge process, microalgae do not require energy input to remove the equivalent amount from brewery wastewater when exposure to sunlight. Moreover, 1 kWh of electric power could be produced through methane production by algal biomass (Amenorfenyo et al., 2019). With comparable removal efficiencies and the added advantage that they produce microalgae biomass without the need for aeration, HRAPs represent an alternative technology for the treatment of wastewater containing MPs. However, one of the main drawbacks for implementing HRAPs in wastewater treatment is that they require a large surface area. This is necessary to promote satisfactory removal efficiency and biomass productivity.

4.1.2. Closed PBRs

Closed-type PBRs generally require high installation and maintenance costs but they are more efficient in terms of light and gas distribution when they are appropriately designed and operated (Chang et al., 2017). They overcome the defects of HRAPs, which have poor mass-transfer efficiency and highly susceptible to contamination. In their easily controlled environment, closed PBRs are better at removing MPs than are open systems (Lavrinovičs & Juhna, 2018; Tolboom et al., 2019; Vo et al., 2019). Details of MP removal by suspended microalgae cells in closed PBRs are described in Table 1S (Supplementary material). Several closed PBRs types have been developed for wastewater treatment and microalgae cultivation. For example, these include flat panel PBR, tubular PBR, rotating algae biofilm PBR, stirred-tank PBR and membrane PBR. Wang, Liu, et al. (2017) summarized the characteristics, different advantages and disadvantages of these systems (Table 2S, Supplementary material).

Closed PBRs can supply a controlled environment for investigating how well isolated microalgae can remove pollutants, in order to identify the species that eliminate them well (Table 1S, Supplementary material). For example, Escapa et al. (2016) carried out their comparative assessment of diclofenac removal from water using a variety of microalgae strains.

Among three selected strains, namely *Chlorella sorokiniana, Chlorella vulgaris* and *Scenedesmus obliquus*, *Chlorella sorokiniana* showed the quickest growth, but *Scenedesmus obliquus* was the most efficient in removing diclofenac from water. The presence of diclofenac provided a higher biomass concentration than did the controls, indicating that the three studied microalgae strains were not inhibited by the presence of diclofenac (Escapa et al., 2016). This phenomenon meant that the studied microalgae species had utilized the MP diclofenac as an additional organic carbon source for their growth, leading to the increased biomass.

Subashchandrabose et al. (2013) summarized a list of organic MPs that have been successfully removed by suspended microalgae cells in an extensive review on how microalgae degrade organic pollutants. Many types of emerging contaminants, including EDCs and PPCPs, can be eliminated effectively by microalgae-based treatment systems (at the surveyed concentrations ranging from 9 to 24 g/L). However, only low pesticide removal has been obtained from this kind of system. Compared to microalgae-based polishing ponds, closed PBR systems were more efficient at removing MPs even at high concentrations (Ahmed et al., 2017). In order to improve the removal efficiencies of pesticides, closed PBRs can be integrated with a biological activated sludge process.

4.2. Immobilized cells

Algal biomass harvesting and separation from treated wastewater is one of the major drawbacks in microalgae-based wastewater treatment. With negatively charged cells and tiny size (e.g. 2–50 µm), microalgae are easily suspended in the medium since their negative charges prevent assemblage. Additionally, their concentration in the cultures is relatively low, ranging from 0.2 to 3.0 g/L (Acién Fernández et al., 2018). Therefore, it is highly energy intensive and costly to collect such small cells from the culture. Conventional harvesting techniques based on mechanical, electrical and chemical processes such as filtration, centrifugation, coagulation and flocculation are currently used for concentrating microalgae from 0.02 to 0.25% (w/w) in mixed liquor effluent to 1–5% (w/w) in solid-liquid separation stage (Gutiérrez Martínez et al., 2016). Microalgae sludge, thereafter, needs further processing. In the context of wastewater treatment, only low-cost techniques capable of managing large volumes of water and biomass can be applied. Herein, immobilization technology can be a good choice for harvesting or separating microalgae biomass from reactors.

The immobilization method could be a potential alternative to conventional remediation due to three key features: (1) highly retained cell density and cell catalytic activity; (2) improved survival in extreme enviroments;

and (3) easy separation and reusability (Abdelmajeed et al., 2012; Lam & Lee, 2012; Nie et al., 2020). Immobilization has been considered for more flexibility in the reactor design than in the traditional suspension systems. How to choose suitable materials for microalgae immobilization and the characterization of the immobilized system are the major contents that most previous studies have concentrated on (Ting et al., 2017). Six different immobilization types have been defined: adsorption, capture behind semi-permeable membrane, affinity immobilization, confinement in liquid-liquid emulsion, covalent coupling, and entrapment in polymers. Immobilization in polymers is the most common and effective method used for microalgae, in which these microorganisms are fixed alive within the gel matrix. The material must be hydrophilic regardless of the polymers used, in order to allow wastewater to diffuse into the bead.

Several synthetic polymers (acrylamide, polyurethane, polyvinyl, resins) and natural polymer (alginate, carrageenan, agar, agarose, chitosan) have been experimentally utilized. Although natural polymers are less stable in wastewater than are synthetic polymers, alginate and carrageenan are the most commonly used materials in immobilization techniques. The major issues with immobilized microalgae systems are: (1) light usage; (2) gel stability; and (3) substrate penetration across the gel matrix. These constrain decisions on the efficiency of the systems needed to remove pollutants, cost of the polymer and cost of the immobilization process. The immobilized microalgae are not only highly resistant to toxic compounds within wastewater, which reduces their effects on algae growth, but also concentrate high biomass that can be used as a valuable by-product.

Low-concentration wastewater is the main target of the immobilized microalgae-based treatment systems. This kind of system has been generally employed to treat municipal wastewater, sewage wastewater, domestic wastewater, and secondary and tertiary effluents. This also satisfies the condition that the initial concentrations of total nitrogen and total phosphorus in the wastewater must always stay below 70 mg/L and 13 mg/L, respectively (Ting et al., 2017).

Given that abundant studies on the use of immobilized microalgae in wastewater treatment systems have been carried out, the majority focused only on nitrogen and phosphorus treatment. Only a few papers on the removal of MPs using immobilized microalgae have been published. Because immobilization can influent the metabolic activity of the cells, it will affect MP removal efficiency. Solé and Matamoros (2016) assessed the effect of free and immobilized microalgae on the removal of six ECDs at a concentration of 10 μg/L using secondary treated wastewater. Both the free and immobilized cells were able to remove up to 80% of the studied EDCs within 10 days of incubation. Compared to suspended microalgae cells, immobilized microalgae in alginate beads were more efficient in removing bisphenol AF, bisphenol F and 2,4-dichlorophenol (Solé & Matamoros, 2016). This study showed that the use of immobilized microalgae increased the removal efficiency for some MPs from wastewater. As reported by Abdullah (2017), immobilized *Chlorella vulgaris* in alginate beads could remove up to 98% of pendimethalin (10 µg/L) and 99% of carbofuran (20 µg/L) after being cultured for 7 days. Recently, Ferrando and Matamoros (2020) demonstrated the suitability of using microalgae immobilized in luffa sponge and polyurethane foam to attenuate some MPs, namely sulfacetamide, sulfamethazine, sulfamethoxazole, bromacil, atrazine, diuron, bentazone, and mecoprop. The result showed that immobilized microalgae in luffa and foam enhanced the attenuation of studied MPs from 36% to 51%, on average, after 10 days of operation. Gao et al. (2011) found that immobilization of microalgae in alginate beads improved nonylphenol removal (at a spiked concentration of 1 mg/L) for a short time (12–24 h) in a culture medium, but free microalgae cells worked more efficient over a longer time (96–168 h).

From the investigated results, the use of immobilized microalgae technology for wastewater treatment can either improve or worsen MP removal, depending on several factors, these being contaminant properties, immobilization methods, or operating parameters. Therefore, further research is needed to address the technical issues, for example the fabrication of different polymers to create a stronger, more efficient matrix for microalgae cells.

4.3. Consortia

Real wastewaters are contaminated with several microorganisms that can damage the growth of microalgae. Also, microalgae have to tolerate and adapt to the constant changes of environmental conditions, including climate and wastewater characteristics. Meanwhile, the association between different species (e.g. microalgae and bacteria) makes a consortium less sensitive to fluctuations in environmental conditions and more resistant to contamination. Microalgal-microbial flocs settled more easily, creating a natural bio-flocculation that increased efficiency in harvesting biomass (Delrue et al., 2016). Mixed populations in consortia can perform functions that are difficult or even impossible for individual strains or species. Therefore, several studies have concentrated on application of consortia in treating wastewater. This section will analyze the association of microalgae and bacteria, which is the most commonly studied consortium in eliminating MPs.

Biofilms, most of which are bacteria-microalgae colonies, have more advantages for microalgae collection than do suspended PBRs. In this kind of system, CO_2 and O_2 can achieve a balance and do not need additional aeration when the activities of the bacteria and microalgae are stable. Even though microalgae, bacteria and their consortia can remove a variety of MPs, little analysis has been done on the removal of MPs from wastewater using microalgae-based systems on a pilot scale.

Gao et al. (2015) suggested that the pollutant-removal efficiencies in the biofilm PBR were higher than those in the suspended mode. Boelee et al. (2014) confirmed that no extra CO_2 and O_2 were needed for a bacteria-microalgae biofilm PBR, because the supplementation of carbonate and light kept the CO_2 demand of microalgae and the O_2 demand of bacteria in equilibrium. Although the bacteria-microalgae system performed better than did a microalgae system alone for COD removal, it is difficult to keep the system steady because of the complicated intraspecies relationships between the different microbiologies within this system. López-Serna et al. (2019) studied a newly emerged PBR configuration and examined the removal rate of five PPCPs that are typically found in urban wastewater, including ibuprofen, naproxen, salicylic acid, tritosan and propylparaben. Anoxic-aerobic and anaerobic-anoxic-aerobic microalgal-bacterial PBRs were proposed. Although these novel PBR configurations have not been specifically designed for eliminating MPs but for nitrogen removal, MP removal efficiencies were improved more than by single-stage HRAPs. Results from these studies suggest that microalgae-bacteria consortia have a great potential for removing MPs from wastewater.

Examined consortia-based treatment systems are listed in Table 4, including algal ponds and PBRs of different configurations and scales. Instead of pure cultivation, mixtures of microalgae and other microorganisms were used in these treatment systems.

5. Overview of treatment methods and combined systems for MP removal

A variety of technologies, including physical, biological and chemical processes for the removal of MPs from wastewater, have been extensively investigated, as illustrated in Figure 2. Since biological degradation processes have been elaborated, other treatment methods for MP removal will be covered in this section to compare the advantages and disadvantages of different technologies (Table 5).

5.1. Physical processes

Trace organic pollutants in water can be adsorbed, which is the most prevalent physical process for removing MPs in the environment. In order to increase the adsorption capacity for MPs, different adsorbents have been researched and developed, including activated carbon, clay, graphene and

Table 4. Removal of micropollutants in microalgae consortia treatment systems.

Treatment system	Setup	Treatment efficiency	References
Multi-tubular PBRs with *Scenedesmus* sp. and microbial consortium	An enclosed 1200 L multitubular PBR made of low density polyethylene, total working volume of 0.24 m^3, HRT 8–12 days	17β-estradiol 100%	Hom-Diaz et al. (2017); Parladé et al. (2018)
Full scale horizontal tubular PBRs with *Chlorella* sp., *Pediastrum* sp., *Scenedesmus* sp., and the cyanobacteria *Gloeothece* sp.	Two tanks made from polypropylene (3.5 m × 1 m × 0.4 m), working volume of 0.612 m^3 each. HRT 16 days	Tonalide 73%, galaxolide 68%, anti-inflammatory compounds 61%	García-Galán et al. (2018)
HRAP with *Chlorella vulgaris* and other microorganisms	Two cylindrically shaped stainless steel tanks ($0.4 \times 0.2 \times 0.3\ m^3$), UV-AB and PAR radiations at the water surface were 0.8–0.9 and 10 W m^{-2}, HRT 7 days	Tetracycline 69 ± 1%	de Godos et al. (2012); Wang, Liu, et al. (2017)
Stirred-tank PBR with a consortium of *Chlorella* sp. and four Gram negative bacteria	Five liter glass tank stirred at 200 rpm, illumination 5000 lx, temperature 30 ± 2 °C, HRT 3 and 4 days	Acetaminophen 80–100%, Aspirin 100%, Ketoprofen 20–98%, Salicylic acid 80–100%	Ismail et al. (2017); Wang, Liu, et al. (2017)
Multi-tubular PBR with a consortium of microalgae and bacteria	Volume of tubes 0.24 m^3 (230 mm diameter, 1 mm thick, 7 m long), outdoor, velocity 0.13 m s^{-1}, HRT 8 and 12 days	Anti-inflammatory drugs >98%, diuretics >84%, antibiotics >48%, psychiatric drug 30–57%	Hom-Diaz et al. (2017); Wang, Liu et al. (2017)
Continuous stirred tank PBR with a consortium of *Chlorella vulgaris* and bacteria *Pseudomonas*	A 1100 mL conical glass PBR stirred at 200 rpm, illumination 5000 lx, temperature 25 ± 2 °C, HRT 4 days	Salicylate 100%, phenol 97%, thiocyanate 95%	Essam et al. (2014)
Consortium of *Scenedesmus obliquus* and four bacteria species	Flasks incubated at 25 ± 1 °C, constant shaking at 150 rpm, light-dark time regime of 14:10, illumination 165 µmol photons/m^2s	Alkylcycloalkanes and alkylbenzenes 100%; naphthalene, fluorene and phenanthrene 100%	Tang et al. (2010), Subashchandrabose et al. (2011)
Consortium of *Synechocytis* sp. and bacteria *Pseudomonas*-related strain	Flasks incubated at 28 °C, constant shaking at 100 rpm under 12 light/12 darkness for 12 days	Phenanthrene 0.8 µg day^{-1}	Abed (2010)

graphene oxide, carbon nanotubes, sand filtration, coagulation and flocculation. Teh et al. (2016) indicated that coagulation-flocculation processes were inefficient at removing most MPs except for some musks and pharmaceuticals such as diclofenac and nonylphenol. Kovalova et al. (2013) and Grover et al. (2011) studied the removal of some MPs during adsorption using powdered activated carbon and granular activated carbon. The results ranged from very low (17%) to nearly complete removal (~100%) depending on the contaminants, dosage and scale of the experiment.

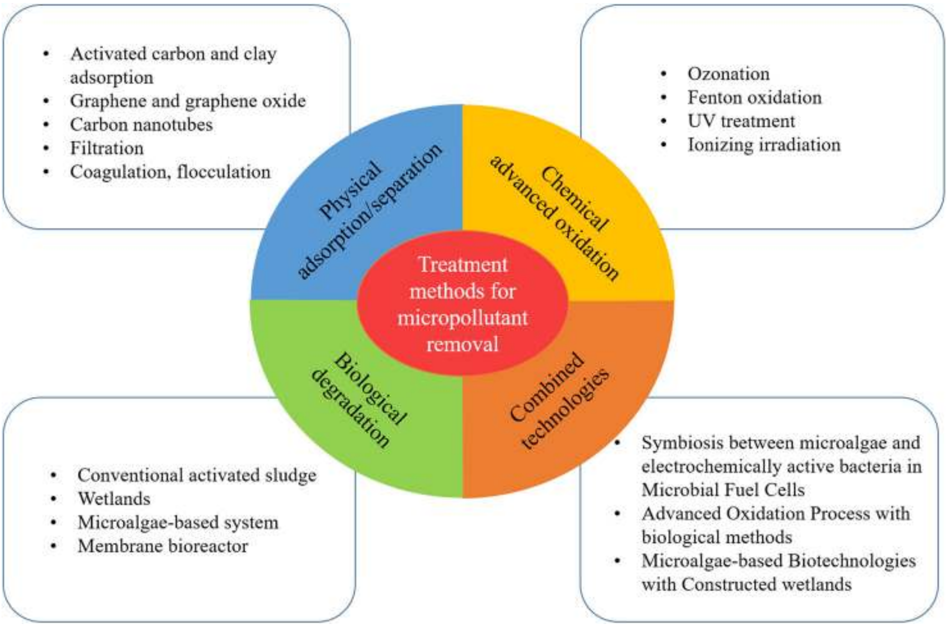


Figure 2. Removal processes for micropollutants in wastewater.

Although microfiltration and ultrafiltration are very effective for eliminating turbidity, they are insufficient for removing MPs, because of the contaminants' molecular size. Nevertheless, MPs can be removed through interaction with natural organic matter and adsorption onto the membrane surface (Dolu et al., 2017). Some hybrid systems such as ultrafiltration-activated biochar, ultrafiltration-powdered activated carbon, and osmotic membrane bioreactor-microfiltration, have been studied and demonstrated a good MP removal rate (Kim et al., 2019; Pathak et al., 2018). However, reduced water flux and fouling are still significant obstacles when applying these systems.

5.2. Chemical advanced oxidation

It has been stated that conventional physicochemical and biological treatments are not sufficient for removing all MPs because of their persistent structure. In such circumstances, ozonation and advanced oxidation processes (AOPs) can be considered as a solution, since they have a high degradation rate. Moreover, this technology is not selective for pollutants removal (Dolu et al., 2017).

Ozonation is a promising method for removing MPs in full-scale WWTPs (Bourgin et al., 2018). Ozone degrades contaminants mainly by producing hydroxyl radicals (·OH), which are strong and less selective for emerging compounds. The presence of H_2O_2, Fenton reagent and

Table 5. Advantages and challenges of different technologies in micropollutants removal.

Treatment process	Advantages	Challenges	Reference
Physical			
Biological activated carbon	- A wide range of MP removal from wastewater - Removal of residual disinfection/oxidation products - Not generating toxic active products	- Relatively high cost in operation and maintenance - Regeneration and disposal issues in high sludge - Sludge processing can increase total cost by 50-60%	Benner et al. (2013); Luo et al. (2014); Rivera-Utrilla et al. (2013)
Coagulation	- Reduced turbidity arising from suspended inorganic and organic particles - Increased sedimentation rate through suspended solid particles formation	- Ineffective MP removal - Large amount of sludge - Introduction of coagulant slats in the aqueous phase	Luo et al. (2014); Töre et al. (2012)
Micro- or ultra-filtration	- Can remove pathogens - Applicable for heavy metal removal	- Not fully effective in removing some MPs as pore sizes larger than the pollutants (from 100 to 1000 times) - High cost of operation	Deegan et al. (2011)
Nanofiltration	- Useful for treating saline water and WWTP influents - Can remove dye stuff and pesticides	- High energy demand, membrane fouling and disposal issue - Limited application in pharmaceutical removal	Benner et al. (2013); Luo et al. (2014)
Reverse osmosis	- Useful for desalination and treating WWTP influents - Can remove PCPs, EDCs and pharmaceuticals	- High energy demand, membrane fouling and disposal issue - Corrosive nature of finished water and lower pharmaceutical removal	Benner et al. (2013); Deegan et al. (2011); Luo et al. (2014)
Chemical			
Ozonation	- Strong affinity to MPs in the presence of H_2O_2 - Selective oxidant favoring disinfection and sterilization properties	- High energy consumption, formation of oxidative by-products - Interference of radical scavengers	Benner et al. (2013); Luo et al. (2014)
AOPs	- Major ancillary effects on removal of MPs such as EDCs, pharmaceuticals, PCPs and pesticides - Short degradation rate	- Energy consumption issues, operational and maintenance cost - Toxic disinfection by-products formation - Radical scavengers interference	Benner et al. (2013); Luo et al. (2014)
Fenton and photo-Fenton	- Degradation and mineralization of MPs - Sunlight can be used by avoiding UV light	- Decrease of $OH^{\cdot}$ forming chloro-, sulfato-Fe(III) complexes or due to scavenge of $OH^{\cdot}$ forming $Cl_2^{\cdot}$ and $SO_4^{\cdot-}$ present in ion forms	Deegan et al. (2011); Malato et al. (2012)
Photocatalysis (TiO_2)	- Degrading persistent organic compounds - High reaction rates upon using catalyst - Low price and chemical stability of TiO_2 catalyst and easier recovery	- Difficult to treat large volume of wastewater - Cost associated with artificial UV lamps and electricity - Complicated process to separate and reuse photocatalytic particles from slurry suspension	Prieto-Rodriguez et al. (2012)
Biological			
Activated sludge	- More environmental friendly than chlorination - Lower capital and operational costs than AOPs	- Low efficiencies for pharmaceuticals and beta blockers - Large amount of sludge containing MPs - Unsuitable when COD levels are higher than 4000 mg/L	Deegan et al. (2011); Luo et al. (2014)

Constructed wetland	- Low energy consumption and low operational and maintenance costs - High performance on removal of estrogens, PCPs, pesticides and pathogens	- Clogging, solids entrapment and sediments formation - Biofilm growth, chemical precipitation and seasonal dependence - Require large land area and long retention time	Töre et al. (2012)
Microalgae reactor	- High quality effluent and no acute toxicity risk associated with MPs - Resource recovery of algal biomass	- Removal efficiencies affected by cold season - EDCs cannot be degraded properly	Matamoros et al. (2015)
MBR	- Effective for the removal of biorecalcitrant and MPs - Small foot print	- High energy consumption and fouling - Hard to control heat and mass transfer - High aeration cost and membrane roughness - Low efficiencies in pharmaceuticals removal	Deegan et al. (2011); Luo et al. (2014)
Combined			
Microalgae-based technologies and AOPs	- Complete removal of resistant compounds - Reduce the cost and ecological risks	- The biodegradability of intermediates produced from AOPs - Over-oxidation - The complicated relationship between the AOPs effluent and PBR performance	Marsolek et al. (2014); Xiong, Kurade, et al. (2018)
Microalgae-based technologies and CWs	- Low cost of construction, operation, and maintenance - Reduce the risk of the eco-toxic buildup - Enhanced pollutant removal	- Complicated relationship in redox conditions - Large space required - Unconfirmed MP removal efficiency	Ding et al. (2016); Huang et al. (2013); Xiong, Kurade et al. (2018)
Microalgae-based technologies and MFCs	- Low energy consumption - Produce clean electrical energy and valuable products	- High construction cost - Unconfirmed MP removal efficiency - Scale-up difficulty	Li et al. (2013); Zhang et al. (2011); Xiong, Kurade et al. (2018)

ultraviolet also promote the production of hydroxyl radicals. Hernández-Leal et al. (2011) investigated the elimination rates of MPs in biologically treated gray water by ozonation; all selected MPs were treated to substantial levels. Sui et al. (2010) carried out the experiment under the similar conditions, with the only difference being in a higher ozone dose of 5 mg/L, results showed higher removal efficiencies for most MPs. In detail, the removal rates of carbamazepine, diclofenac, indomethacin, sulpiride and trimethoprim exceeded 95%. However, the removal rate of bezafibrate was evaluated at only 14%, because of bezafibrate's stable molecular structure (Dolu et al., 2017). In general, oxidation processes cannot provide a complete mineralization of these compounds, so by-products and metabolites may arise from these reactions.

Many research teams have critically reviewed the potential of homogeneous AOPs to degrade several MPs (Barbosa et al., 2016; Ribeiro et al., 2015; Rizzo et al., 2019). However, to the best of our knowledge, no sustainable application and operation of these processes in real scale has been reported so far due to their by-products' toxicity.

5.3. Combined technologies

Realizing the advantage of integrated processes, some combined technologies have been proposed for the removal of MPs in wastewater. Firstly, microalgae-based technologies can integrate with AOPs for a better removal of MPs. In detail, AOPs can transform the resistant compounds into biodegradable intermediates. Then, the following microalgae processes can completely mineralize these by-products. This combined system can both reduce the toxicity of incompletely oxidized products from single-step AOPs and improve the total MP removal efficiency.

Secondly, microalgae-based technologies can combine with constructed wetlands (CWs) for a more sustainable solution. In this kind of system, microalgae are able to enhance dissolved oxygen in water through photosynthesis, and the algal debris can provide as the organic substrate for plants in the CWs. Thus, adding a microalgae pretreatment unit within a CW water treatment system has been considered as an attractive option.

For the third integration, the combination between microalgae and microbial fuel cells (MFCs) may increase the system's pollutant removal efficiency as well as saving energy consumed. According to Yang et al. (2018), contaminant removal in the algal biofilm-assisted MFC was better than that in the MFC or algal biofilm alone. Also, bioenergy of 0.094 kWh m^{-3} of wastewater was obtained in continuous operation of this system. One of the most attractive advantages of MFCs is their ability to extract electric energy directly from organic contaminants in wastewater. Different from other energy products derived during anaerobic digestion processes (e.g. CH_4 and H_2), electricity is a cleaner and more widely utilizable energy form (Li et al., 2013). It has been reported that an amount of 0.080 kWh energy per kg chemical oxygen demand (COD) could be recovered from municipal wastewater. Besides, an MFC is estimated to consume only 0.076 kWh kg^{-1}-COD in average based on the current practice for domestic wastewater treatment (Ge et al., 2014; Zhang et al., 2013). Thus, it is theoretically achievable to perform a positive energy balance in domestic wastewater treatment by MFCs on a liter scale (McCarty et al., 2011). Integrating them with microalgae-based technologies may reduce the total energy usage as well as facilitating the production of valuable biomass products.

However, the integration of microalgae-based technologies with other processes has rarely been reported and is still being researched. There is little data on the use of integrated systems for the removal of MPs in wastewater. Therefore, more analyses are required to fully understand the operations of these integrated systems for treating MPs wastewater. The configuration design, hydraulic mode, pH, oxygen, and redox potential are the key factors that should be carefully considered during the construction of the integrated systems.

6. Recommendations for future research

Several MPs can be effectively removed by means of different microalgae-based systems, but there are still deficiencies in the complete removal of MPs from wastewater. Among the several treatment methods, conclusive evaluation of the most suitable and cost-effective solution for wastewater treatment is not yet possible. Some further research areas are suggested as follows.

The potential of microalgae for degrading MPs is high; however, each microalgae species can degrade different types of contaminants. More studies of new microalgae species will be necessary to find alternatives to degrade resistant MPs. Besides, additional work is needed to evaluate MP removal in microalgae-based treatment systems with different configurations as well as how operating conditions affect their treatment efficiencies. Moreover, removal mechanisms should be better demonstrated for a superior system design. In the future, more comprehensive studies on practical applications of microalgae-based treatment systems in reality will be required to assess their overall performance, benefit and feasibility.

MPs include a variety of compounds with different physical-chemical properties. Therefore, it is unrealistic to treat all MPs sufficiently with microalgae-based technologies. Thus, it is vital to establish a priority list of

MPs exhibiting noticeable and high environmental risks in wastewater. MPs on the priority list will be the main targets for developing and optimizing microalgae-based technologies.

There are difficulties in species control in the microalgae-based treatment systems for real wastewater because of contamination. Hence, the use of microalgae-bacteria or microalgae-fungi consortia can yield advantages over using specific algae species for real systems. It is also beneficial in the way that multiple catabolic pathways from different microorganisms are involved, leading to superior MP removal efficiency. Therefore, instead of trying to maintain certain species in microalgae-based treatment systems, the roles of different microorganisms within consortia in MP removal should be better understood and the optimization of microbial community structure should be carried out. In addition, the ratio for microalgae-bacteria combinations as well as optimal operating conditions need to be investigated further.

Research has shown that microalgae and fungi can execute impressive MP removal rates in separated systems. However, no study has yet investigated the potential of their association in microalgae-fungi consortia. Some fungi can produce extracellular enzymes with low substrate specificity and are very suitable for degrading some MPs even at low water solubility (Pozdnyakova, 2012). Ahmed et al. (2017) showed that fungal-based bioreactors (also termed mycoreactors) can remove up to 100% of several MPs, such as pharmaceuticals beta blockers (atendol, propranolol, and sotalol), gastroesophageal and anticancer drugs (crimetidine, famotidine ranitidine, acridone, and citalopram), anti-inflammatory drugs (acetaminophen, including stimulants such as butalbital), and antibiotics (azithromycin, erythromycin, sulfathazole, sulfapyridine, and sulfamethazine). Within the microalgae-fungi relationship, fungi meet their organic carbon requirement from algal photosynthesis. In return, fungal filaments provide moisture, nutrients, protection and anchor to microalgae cells (Lutzu and Turgut 2018). With the both-sided benefits, the association of microalgae and fungi could be an interesting topic to research on MP removal.

No research has yet examined the direct influences of environmental and operating parameters on MP removal in microalgae-based treatment systems. Existing studies evaluate only the factors affecting microalgae growth as well as nutrient removal performance. Therefore, only indirect effects of the removal of MPs can be drawn out. More research is needed to elaborate the influences of environment and operation mode on the elimination of trace organic contaminants by microalgae.

The integration of microalgae-based technologies with other treatment methods is receiving more attention in recent times. However, the MP removal efficiencies of those combined systems have rarely been reported and are still being researched. There are limited data on the use of integrated systems for the removal of MPs in wastewater. Therefore, more research is required to fully understand the behavior of these integrated systems for wastewater containing MPs.

This review highlights the possibilities of applying microalgae-based systems in various wastewater treatment plants in terms of MP removal. Microalgae-based treatment has proven to be more effective to some extent as well as more efficient and eco-friendly than are conventional wastewater treatment technologies. In addition, microalgae not only have the potential to treat MPs but also can be used to produce biomass energy and biofuel. The removal of trace organic pollutants by microalgae were found to be compound-specific. Compared to other treatment alternatives, microalgae-based systems have several advantages yet also have some limitations that need to be overcome. More comprehensive work is required to fill in the knowledge gap on this type of treatment system.

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