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REVIEW PAPER

Removal of pharmaceuticals and personal care products from wastewater using algae-based technologies: a review

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Abstract Pharmaceuticals and personal care products (PPCPs) consist of a variety of compounds extensively used for the treatment of human and animal diseases and for health or cosmetic reasons. PPCPs are considered as emerging environmental contaminants due to their ubiquitous presence in the environment and high environmental risks. In wastewater treatment plants using conventional activated sludge processes, many PPCPs cannot be efficiently removed. Therefore, there is an increasing need for more effective and cost-efficiency ways of removing PPCPs while treating wastewater. Algae-based technologies have recently attracted growing attentions for their potential application in wastewater treatment and hazardous contaminant removal, which are advantages in reducing operation cost while generating valuable products and sequestrating greenhouse gases at the same time. This work reviews the up-to date researches to reveal potential toxic effects of PPCPs on algae and algae-bacteria consortia, identify mechanisms involved in PPCP removal, and assess the fate of PPCPs in algae-based treatment systems. Current researches suggest that algae and algae-bacteria consortia have great potentials in PPCP removal but more works are required before algae-based technologies can be implemented in large scales. Knowledge gaps are identified and further research focuses are proposed in this review.

Keywords PPCPs · Algae · Bacteria · Wastewater treatment · Biodegradation

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1 Introduction

Nowadays, pharmaceuticals and personal care products (PPCPs) are extensively used for the treatment of human and animal diseases and for health or cosmetic reasons. PPCPs can enter wastewater streams after being used, and can be transported into the environment via different pathways (Daughton 2007; Lapworth et al. 2012). As a consequence, PPCPs are frequently detected in many aquatic and terrestrial environments (Luo et al. 2014; Monteiro and Boxall 2010; Verlicchi and Zambello 2015). Increasing

evidences suggest that environmental residues of PPCPs could adversely affect non-target organisms, ecosystem functions, and even human health (Prosser and Sibley 2015; Rosi-Marshall and Royer 2012; Wilkinson et al. 2016). Due to their widespread use and bioactive nature, PPCPs are considered as a group of contaminants of emerging concern, and have drawn growing attention over the past decades from the scientific community and the public (Daughton and Ternes 1999; Evgenidou et al. 2015).

Municipal wastewater is a major source of PPCPs used by human. In developed countries and regions, municipal wastewater is collected through sewer systems and get treated in wastewater treatment plants (WWTPs) before discharging into the environment. Therefore, many previous research efforts have been focused on characterizing the occurrence and fate of PPCPs in WWTPs (Esplugas et al. 2007; Luo et al. 2014; Ternes et al. 2004; Tiwari et al. 2017). The removal efficiency has been found to be compound specific, many PPCPs were poorly removed in WWTPs using conventional activated sludge (CAS) treatment processes, and considerable amount of PPCPs remained in effluent and/or biosolids (McClellan and Halden 2010; Melvin and Leusch 2016; Miège et al. 2009). Therefore, PPCP residues can be introduced into the environment following the discharge of effluent and land application of biosolids.

Animal wastes are important sources of veterinary pharmaceuticals and steroid hormones. Wastewater from intensive animal farming is usually treated in treatment lagoons or constructed wetlands. Incomplete removal of many veterinary pharmaceuticals and steroid hormones has also been reported (Carvalho et al. 2013; Kolz et al. 2005; Liu et al. 2015; Shappell et al. 2007). Additionally, animal manures are commonly land applied as fertilizers in crop fields. As a consequence, veterinary pharmaceuticals and steroid hormones in manures are transported into the environment and their residues have been detected in soil, surface water and groundwater impacted by animal wastes (Bartelt-Hunt et al. 2011; Chen et al. 2010; Wei et al. 2011; Yost et al. 2014).

Many advanced technologies have been investigated to improve the removal of PPCPs from wastewater. Advanced oxidation processes using O_3, H_2O_2, and Fenton (Fe^{2+}/H_2O_2) have been found to be highly efficient in removing PPCPs (Esplugas et al. 2007; Ghatak 2014), which generate hydroxyl radical to breakdown PPCPs oxidatively. Photodegradation and photocatalytic degradation processes have also been found to be able to degrade many PPCPs rapidly, which involve direct photolysis and indirect photolysis (Kanakaraju et al. 2014; Yang et al. 2014). Other advanced technologies such as membrane filtration and activated carbon adsorption have also been investigated and the removal of PPCPs were related to their physical–chemical properties (Rodriguez et al. 2016; Yang et al. 2011). Hybrid processes using a combination of different technologies are able to further enhance the PPCP treatment efficiency (Ahmed et al. 2017). Although these advanced technologies show promising results in PPCP treatment, high cost limits their application on a large scale. Consequently, there is an increasing need for more cost-effective and sustainable approaches for the removal of PPCPs and other contaminants in wastewater.

Biological treatment technologies are generally more environmentally friendly and less expensive compared with those physical and chemical treatment technologies. Membrane bioreactor (MBR) treatment processes showed improved PPCP removal compared with CAS and biological nutrient removal processes (Sipma et al. 2010; Sui et al. 2011). Constructed wetlands based on shallow pond, beds, or trenches containing aquatic macrophytes are considered to be cost-effective in treating wastewater due to a relatively low cost in construction, operation, and maintenance (Wu et al. 2015b). Constructed wetlands include surface horizontal flow, subsurface horizontal flow, and vertical flow systems according to their flow regime, and can be used in combination to take advantage of different systems (Vymazal 2011). Removal of PPCPs in constructed wetlands has been found to be highly dependent on the physical–chemical properties of the PPCPs, and affected by the configuration and operation of the wetland and the environmental conditions (Garcia-Rodríguez et al. 2014; Zhang et al. 2014a).

In recent years, there is a growing research interest in utilizing algae-based technologies in wastewater treatment (Abinandan and Shanthakumar 2015; Wang et al. 2016b; Wu et al. 2012). Nutrients from wastewater are assimilated into algae biomass for their growth, which can be harvested and used as a biofertilizer (Cai et al. 2013; Hwang et al. 2016). Valuable products such as biofuel, proteins,

carbohydrates, pigments, and vitamins can be produced by algae while treating wastewater (Cuellar-Bermudez et al. 2016; da Silva et al. 2014; Milledge 2011; Úbeda et al. 2017). In addition, algae capture CO_2 during photosynthesis, which reduces greenhouse gas emission (Razzak et al. 2013; Subashchandrabose et al. 2011). Therefore, algae-based technologies are more sustainable compared with conventional wastewater treatment technologies and none bioprocesses.

Other than nutrients, many hazardous contaminants existed in wastewater can be treated with algae-based technologies. Removal of heavy metals from wastewater by algae and algae-bacteria consortia has been demonstrated with the involvement of surface sorption, bioaccumulation, and precipitation (Zeraatkar et al. 2016). Removal of chemical oxygen demand (COD) using algae based technologies can reduce the energy input via O_2 provided by photosynthetic oxygenation (Muñoz and Guieysse 2006; Wang et al. 2016b). Due to the emerging concern on PPCPs and the merit of algae-based technologies, related works are increasingly reported over the past decades. In this work, we provide a critical review on the up-to date works to (1) reveal potential toxic effects of PPCPs on algae and algae-bacteria consortia, (2) explore the PPCP removal mechanisms, (3) assess the fate of PPCPs in algae-based treatment systems, and (4) knowledge gaps are identified and further research focuses are proposed.

2 Potential toxic effects of PPCPs on algae and algae-bacteria consortia

PPCP residues in wastewater may adversely affect the growth and functionality of algae and algae-bacteria consortia, which can adversely affect the application of algae-based technologies. Toxicity of PPCPs to algae and other aquatic organisms has been extensively reviewed previously (Brain et al. 2008; Brausch et al. 2012; Fent et al. 2006; Wilkinson et al. 2016). Results from these reviews indicate that algae are generally more sensitive to antibiotics, antimicrobials, and selective serotonin re-uptake inhibitors (SSRIs) than other classes of PPCPs. Triclosan, clarithromycin, spiramycin, and tetracycline are among the most toxic PPCPs to algae with half maximal effective concentration (EC_{50}) for growth inhibition down to few $\mu g\ L^{-1}$. However, no significant toxicity has been observed for algae exposed to PPCPs belonging to nonsteroidal anti-inflammatory drugs (NSAIDs), analgesic, antiepileptic, fibrate-type lipid regulator, β-blocker, vasodilator, antihyperglycemic, antimetabolite, nitroimidazoles, nicotine, or stimulant at concentration levels below 500 $\mu g\ L^{-1}$. Cyanobacteria such as *Microcystis aeruginosa* are generally more sensitive than green algae such as *Selenastrum capricornutum* (Crane et al. 2006; van der Grinten et al. 2010).

Potential adverse effects of PPCPs on algae-bacteria consortia have also been demonstrated. At 10 $\mu g\ L^{-1}$, triclosan and triclocarban significantly affected the microbial community composition, algal biomass, architecture, and activity of river biofilms (Lawrence et al. 2009). In stream microcosms, significant changes in antibiotic resistance, bacteria abundance and productivity, algae biomass, cyanobacteria, organic biomass, and nematodes have been observed in periphyton exposed to tetracycline as low as 0.5 $\mu g\ L^{-1}$ within 7 days (Quinlan et al. 2011). Exposure to ibuprofen, carbamazepine, furosemide, and caffeine at 10 $\mu g\ L^{-1}$ has been found to exhibit both nutrient-like and toxic effects on riverine biofilm communities (Lawrence et al. 2005). However, how these impacts affect the wastewater treatment ability of algae-bacteria consortia remains unclear and needs to be addressed in future research.

In municipal wastewater influent, concentrations of PPCPs vary considerably from place to place but are generally within the $ng\ L^{-1}$ to $\mu g\ L^{-1}$ range (Luo et al. 2014; Miège et al. 2009). Concentration ranges of PPCPs detected in municipal wastewater and the EC_{50} values for the growth of algae are presented in Fig. 1. For the majority of the PPCPs, their EC50 values are orders of magnitudes higher than their highest concentrations detected in influent. However, triclosan has been detected with the maximum concentration up to 23.9 $\mu g\ L^{-1}$ (Luo et al. 2014), which is more than ten times higher than the EC_{50} value for the growth of algae. In wastewater from animal farms, antibiotics are commonly detected in the $\mu g\ L^{-1}$ range and chlortetracycline has been detected up to 1000 $\mu g\ L^{-1}$ in swine wastewater lagoon (Campagnolo et al. 2002; Wei et al. 2011). Therefore, it is possible that algae and algae-bacteria consortia can be adversely affected by high PPCP residues in wastewater.

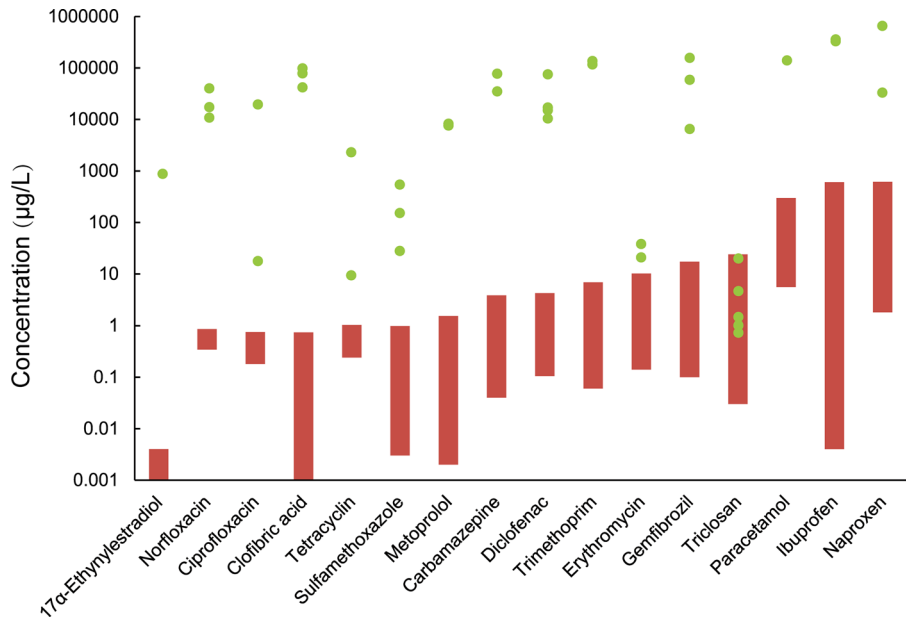

Fig. 1 A comparison of PPCP residues in municipal wastewater (red bars represent maximum and minimum concentrations) and the reported EC50 values for the growth algae (green dots). Data were from Brain et al. (2008), Luo et al. (2014), and Miège et al. (2009). (Color figure online)

Other than PPCPs, heavy metals, ammonium, COD, and other organic contaminants can also inhibit algae when their concentrations in wastewater are higher enough (Muñoz and Guieysse 2006; Wang et al. 2016b). Resistant strains or algae-bacterial consortia in the form of biofilm can be used to mitigate the adverse effects of toxic contaminants in wastewater (Pittman et al. 2011), but the tolerance of algae is limited. Therefore, wastewater with very high residues of these contaminants must be pretreated to avoid the inhibitory effects. Pretreatment can employ processes such as dilution, photodegradation, adsorption, and anaerobic digestion (González et al. 2008; Muñoz and Guieysse 2006; Wang et al. 2015). Use of algae-based technologies in tandem with other less sensitive processes such as treatment pond, constructed wetland, and advanced oxidation can also be a good solution (Oller et al. 2011).

3 Processes and mechanisms involved in PPCP removal using algae-based technologies

As illustrated in Fig. 2, algae-based technologies can remove PPCPs from wastewater with the involvement of sorption, biodegradation, photodegradation, and volatilization.

3.1 Sorption

Sorption means the redistribution of a substance from the liquid phase to the solid phase. Surface sorption processes are considered as adsorption while absorption refers to the transfer of a substance into a sorbent. Sorption can involve in treatment processes with the application of any sorbent but the significance of sorption in contaminant removal is determined by the affiliation strength between the contaminants and the sorbents. In algae-based treatment systems, algae and bacteria can act as biosorbents (Gadd 2009). Polysaccharides such as cellulose, chitin, alginate, and glycan exist in the cell wall of algae, while peptidoglycan, teichoic acids and lipoteichoic acids are in the surface of bacteria. Additionally, extracellular polymeric substances (EPS) can be produced by algae and bacteria, which are mostly polysaccharides, proteins, nucleic acids, and lipids (Flemming and Wingender 2010). These chemical components provide important sites for the sorption of heavy metals and organic contaminants (Fomina and Gadd 2014; Wang and Chen 2009).

Sorption of organic contaminants is highly dependent on their physical–chemical properties. Nonpolar PPCPs have been shown to have strong affiliation to

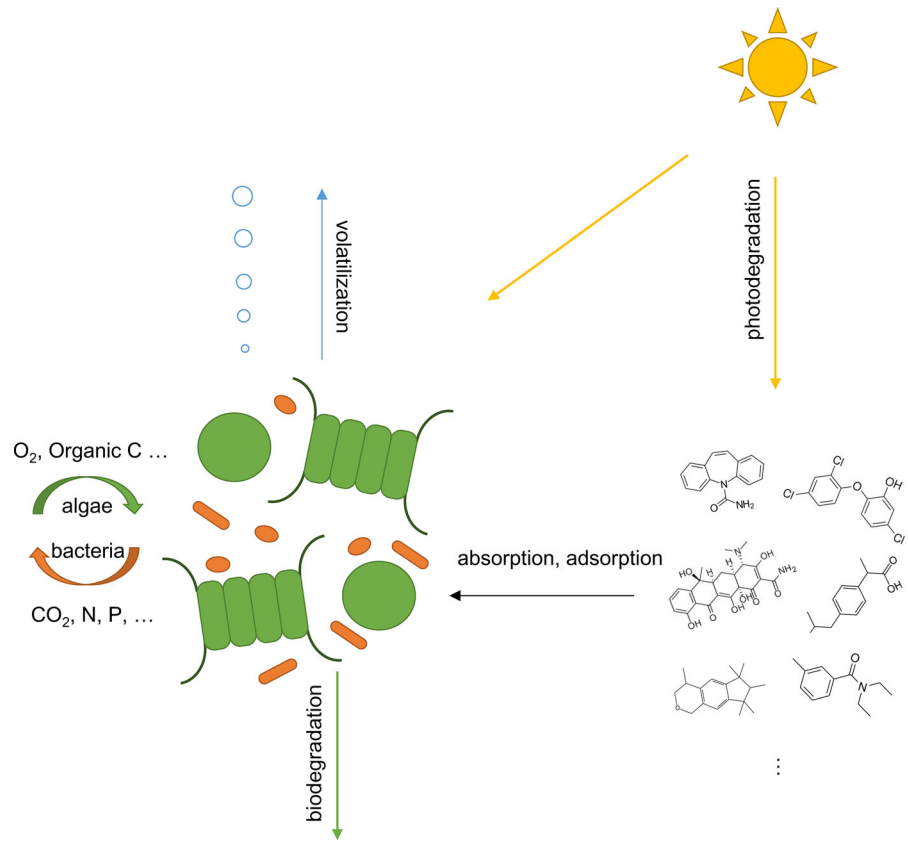

Fig. 2 Processes involved in PPCP removal using algae-based technologies

biosorbents due to hydrophobic interactions, which can cross cell membranes and be absorbed into the organic matrix (Gadd 2009). In a stream receiving wastewater effluent discharge, triclosan and triclocarban have been found able to bioaccumulate in filamentous algae with estimated bioaccumulation factors up to 2100 and 2700, respectively (Coogan et al. 2007). Maes et al. (2014) studied the removal of EE2 by *Desmodesmus subspicatus* and found EE2 can be taken up and biotransformed in algal cells and the ratio of EE2 between algae and water reached a maximum value of 2700 L kg^{-1} (Dry weight) after 24 h.

Sorption of ionizable pharmaceuticals can be more complicated. Basic pharmaceuticals with aliphatic amine groups have been found able to accumulate in algae cells via the ion-trapping effect (Neuwoehner and Escher 2011). Positively charged basic pharmaceuticals can also be adsorbed onto negatively charged surface via electrostatic interactions (Stevens-Garmon et al. 2011). However, acidic pharmaceuticals generally show relatively weak sorption especially when water pH favors deprotonation (Duan et al. 2013). Thus, elevated pH as a result of algae photosynthesis can reduce the sorption of acidic pharmaceuticals. Additionally, some PPCPs such as fluoroquinolones and tetracyclines can exist as zwitterions, which show a strong adsorption to soil via surface complexation (Carrasquillo et al. 2008; Vasudevan et al. 2009). Metal ions precipitated in algae biofilms may act as potential adsorption sites for these zwitterions.

Sorption processes transfer PPCP residues from wastewater to algae or algal–bacterial consortia instead of breaking their down, which can still be hazardous. Therefore, biomass containing PPCP residues generated from algae-based treatment systems must be disposed appropriately to avoid releasing of the sorbed PPCPs. In addition, contamination of PPCPs should also be considered if biomass from algae-based treatment systems is used for the production of other valuable products.

3.2 Biodegradation

Biodegradation is the breakdown of organic chemicals catalyzed by enzymes produced by microorganisms. Bacteria and archaea are mainly responsible for biodegradation in CAS and MBR treatment processes. The mechanisms can either be co-metabolism in which the degradation of pollutants depends on the presence of non-specific enzymes catalyzing the metabolism of other substrates, or be metabolic degradation in which organic chemicals are used as sole carbon and energy sources (Tiwari et al. 2017). Previous works have showed that many heterotrophs can metabolically degrade PPCPs. For example, 17β-estradiol (E2) can be used as sole carbon and energy source by *Pseudomonas aeruginosa* TJ1 isolated from an aerobic activated sludge (Zeng et al. 2009). Bacteria within the family of Alcaligenaceae have been found to be able to use triclocarban as the sole carbon source (Miller et al. 2010). *Stenotrophomonas maltophilia* KB2 has been found to be able to metabolically degrade Naproxen (Wojcieszyńska et al. 2014). Even though, co-metabolism is believed to be the main process responsible for the degradation of PPCPs in WWTPs due to the fact that PPCPs are usually present in wastewater at low concentration levels within the ng L^{-1} to μg L^{-1} rang, which may not be higher enough to maintain the growth of microbes performing metabolic degradation (Onesios et al. 2009). As reviewed by Tran et al. (2013), autotrophs such as ammonia oxidizers co-metabolize a variety of emerging organic contaminants and heterotrophs degrade emerging organic contaminants via co-metabolism and/or metabolism mechanisms, but co-metabolism is mainly responsible for the biodegradation of emerging organic contaminants.

In algae-based technologies, algae can also actively participate in biodegradation of organic contaminants. Algae contain enzymes that metabolite a variety of xenobiotics and the process can be divided into three phases (Torres et al. 2008). Phase-I involves oxidation, reduction, or hydrolysis, which transforms lipophilic xenobiotics into more hydrophilic compounds to facilitate their excretion. Cytochrome P450, which are microsomal heme-thiolate proteins anchored in membrane, typically catalyze the primary step of detoxification (Zangar et al. 2004). Phase-II is characterized by the addition of hydrophilic moieties to facilitate excretion. Xenobiotics with −COOH, −OH or $-NH_2$ in their structures and metabolites from phase-I can be conjugated with glutathione (GSH) or glucuronic acid catalyzed by glutathione S-transferases (GSTs) or glucosyltransferases (Nakajima et al. 2007; Pflugmacher et al. 1999; Yang et al. 2002). Phase-III involves compartmentation of xenobiotics in vacuoles or cell wall fractions (Dietz and Schnoor 2001; Petroutsos et al. 2008). The ability of algae to detoxicate xenobiotics is similar to that of mammalian liver and thus algae are considered as "green livers" for the detoxification of environmental contaminants (Torres et al. 2008).

In algae-bacteria consortia, microbial interactions can provide mutual benefits. Photosynthesis of algae provides O_2 for aerobic bacteria to degrade organic pollutants while bacterial respiration releases CO_2 in return for algae photosynthesis (Muñoz and Guieysse 2006). Algae also exude organic compounds such as carbohydrates and amino acids, which support the heterotrophic metabolism and provide substrates for the co-metabolism of organic contaminants (Battin et al. 2016; Tran et al. 2013). Bacteria in turn accelerate the regeneration of nutrients and trace elements, and release phytohormones to promote algae growth (Dang and Lovell 2016; Wang et al. 2016a). Interactions between algae and bacteria can also be completive. Competition for nutrients and spaces has been observed in many nutrient-limiting environments (Liu et al. 2016; Rier and Stevenson 2002; Scott et al. 2008), but may not be the case in wastewater. Algae can raise water pH, increase dissolved oxygen content during photosynthesis, and release inhibitory chemicals, which can be detrimental to certain bacteria, while bacteria can in turn affect algae by killing or lysing or by changing the microenvironment (Amin et al. 2012; Subashchandrabose et al. 2011; Wang et al. 2016a). However, how microbial interactions affect the overall biodegradation ability of the consortia remains poorly understood.

3.3 Photodegradation

Many PPCPs have been found to be photodegradable under sunlight irradiation (Hanamoto et al. 2014; Lam et al. 2004; Lin et al. 2006; Matamoros et al. 2008). Photodegradation of PPCPs involves both direct photolysis and indirect photolysis. In direct photolysis, target compound is broken down by absorbing

incident light directly. PPCPs with aromatic rings, conjugated π systems, heteroatoms, and other functional groups can be prone to direct photolysis due to a strong absorption of irradiation within the UV range of the sunlight (Challis et al. 2014). However, indirect photolysis occurs with the generation of free radicals such as hydroxyl radicals (OH·), peroxyl radicals (ROO·), and singlet oxygen ($^1O_2$) produced during sunlight illumination. Free radicals are generated with the presence of photosensitizer such as chromophoric dissolved organic matters (CDOMs), NO_3^-, carbonate, and certain metal ions (Boreen et al. 2003). Indirect photolysis are generally more important for the overall fate of PPCPs in natural waters and wastewater due to the ubiquitous presence of photosensitizers, especially for those without significant absorption of light above 290 nm (Challis et al. 2014). Rate of sunlight induced photodegradation can be related to the latitude and varies seasonally due to angle and duration of the solar irradiance, and more rapid photodegradation can be expected during summer time in low latitude area (Andreozzi et al. 2003). Half-life ($t_{1/2}$) values estimated using the first-order decay model for the photodegradation of PPCPs under sunlight vary considerably from a few hours to hundreds of days (Andreozzi et al. 2003; Wu et al. 2015a; Yamamoto et al. 2009).

In algae-based treatment systems, illumination is required for the photosynthesis of algae. Sunlight is preferred light source to reduce extra energy cost. Therefore, photodegradation can play an important role in PPCPs removal during the wastewater treatment using algae-based technologies. As depicted in Eqs. (1–2), algae are able to facilitate photodegradation by increasing the free radical yield (Collén et al. 1995; Liu et al. 2004; Peng et al. 2006). The photodegradation of Norfloxacin was enhanced with algae concentration and was influenced by temperature and pH of the solution (Zhang et al. 2012a). Chlorophyll from dead algal cells has been found to be able to catalyze the photodegradation of benzo[a]pyrene by acting as a photosensitizer (Luo et al. 2015). Extracellular secretions from *M. aeruginosa* have also been found to be able to accelerate the photodegradation of triclosan (Huang et al. 2016). As photodegradation depends on light penetration, the extent of photodegradation can be affected by the configuration of algae-based treatment systems. Some close systems in which algae biofilm attaches to the inside walls can block the light. Therefore, photodegradation may be less significant in those systems.

$$O_2 + \text{cell organelles of algae} + \text{hv} \rightarrow {}^1O_2/O_2^- \quad (1)$$

$$O_2 + \text{cell secretion of algae} + \text{hv} \rightarrow \cdot\text{OH} \quad (2)$$

3.4 Volatilization

Volatilization can also contribute to the removal of those PPCPs with relative high Henry's law constant (H) values, and the contribution of volatilization to the overall loss depends on the air flow rate in the aeration tank in CAS systems (Suárez et al. 2008). In a stirred-tank aerobic reactor, volatilization contributed to 3–16% of the total removal for musk Celestolide (ADBI) with a H value of 7.3×10^{-1} mg mL^{-3} air/mg mL^3 wastewater but volatilization was found to be negligible for Galaxolide (HHCB) and Tonalide (AHTN) and other PPCPs with H values above 5.1×10^{-3} mg mL^{-3} air/mg mL^3 wastewater (Suarez et al. 2010). In another study, 4-octylphenol, HHCB, and tributyl phosphate with H values higher than 3×10^{-1} Pa m^3 mol^{-1} were removed by volatilization as a result of air stripping in an aerated microalgae batch reactor (Matamoros et al. 2016). Therefore, volatilization can participate in the removal of volatile and semi-volatile PPCPs in open algae-based treatment system and the contribution is relative to the H values of the compounds, and affected by the air stripping intensity and temperature in the system. However, volatilization can only transport pollutants from the liquid phase into the atmosphere instead of breaking them down, it might not be a desired outcome for wastewater treatment.

4 PPCP removal by algae and algae-bacteria consortia

Many previous works were performed to investigate the capability and mechanisms of PPCP removal by algae using laboratory bench experiment. As summarized in Table 1, algae have been found able to remove many PPCPs belonging to estrogenic hormones, antibiotics, antimicrobials, antiepileptics, and NSAIDs. Biodegradation was found to be responsible for the removal for most of the investigated PPCPs.

Table 1 Removal of pharmaceuticals and personal care products by algae in laboratory experiment and associated mechanisms

Compound	Species	Removal	Major mechanism	References
Hormones				
17α-estradiol	*Scenedesmus dimorphus*	85%, 8 days	Biotransformation	Zhang et al. (2014b)
17β-estradiol	*Chlamydomonas reinhardtii*	100%, 7 days	Biodegradation	Hom-Diaz et al. (2015)
	Selenastrum capricornutum	88–100%, 7 days	Biodegradation	Hom-Diaz et al. (2015)
	Scenedesmus dimorphus	95%, 8 days	Biotransformation	Zhang et al. (2014b)
Estrone	*Scenedesmus dimorphus*	85%, 8 days	Biotransformation	Zhang et al. (2014b)
Estriol	*Scenedesmus dimorphus*	95%, 8 days	Biotransformation	Zhang et al. (2014b)
17α-ethinylestradiol	*Chlamydomonas reinhardtii*	71–100%, 7 days	Biodegradation	Hom-Diaz et al. (2015)
	Selenastrum capricornutum	60–95%, 7 days	Biodegradation	Hom-Diaz et al. (2015)
	Desmodesmus subspicatus	68%, 3 days, $k = 0.009\ h^{-1}$	Biodegradation	Maes et al. (2014)
Progesterone	*Scenedesmus obliquus*	>95%, 5 days, $k = 0.044\ h^{-1}$	Biodegradation	Peng et al. (2014)
	Chlorella pyrenoidosa	>95%, 5 days, $k = 0.018\ h^{-1}$	Biodegradation	Peng et al. (2014)
Norgestrel	*Scenedesmus obliquus*	>95%, 5 days, $k = 0.017\ h^{-1}$	Biodegradation	Peng et al. (2014)
	Chlorella pyrenoidosa	60%, 5 days, $k = 0.009\ h^{-1}$	Biodegradation	Peng et al. (2014)
Antibiotics				
Ciprofloxacin	*Chlamydomonas mexicana*	13–56%, 11 days, $k = 0.0121–0.079\ day^{-1}$	Co-metabolite, enhanced with electron donors	Xiong et al. (2017b)
Levofloxacin	*Chlorella vulgaris*	9.5–91.5%, 11 days, $k = 0.011–0.257\ day^{-1}$	Biodegradation, enhanced with NaCl	Xiong et al. (2017a)
Trimethoprim	*Nannochloris* sp.	0%, 14 days	Non-degradable	Bai and Acharya (2016)
Sulfamethoxazole	*Nannochloris* sp.	32%, 14 days	Algae-mediated photolysis	Bai and Acharya (2016)
7-amino cephalosporanic acid	*Chlorella* sp.	100%, 5 days	Hydrolysis, photolysis, and adsorption	Guo et al. (2016)
	Chlamydomonas sp.	100%, 5 days	Hydrolysis, photolysis, and adsorption	Guo et al. (2016)
	Mychonastes sp.	100%, 5 days	Hydrolysis, photolysis, and adsorption	Guo et al. (2016)
Antimicrobials				
Triclosan	*Nannochloris* sp.	100%, 7 days	Uptake and biodegradation	Bai and Acharya (2016)
	Microcystis aeruginosa	46%, 8 days, $k = 0.0034\ h^{-1}$	Biotransformation, photolysis, and adsorption	Huang et al. (2016)
Antiepileptics				

Table 1 continued

Compound	Species	Removal	Major mechanism	References
Carbamazepine	*Chlamydomonas mexicana*	37%, 10 days, $k = 0.0424\ day^{-1}$	Biodegradation	Xiong et al. (2016)
	Scenedesmus obliquus	30%, 10 days, $k = 0.0329\ day^{-1}$	Biodegradation	Xiong et al. (2016)
NSAIDs				
Diclofenac	*Chlorella sorokiniana*	29.99%, 9 days	Biodegradation	Escapa et al. (2016)
	Chlorella vulgaris	21.58%, 9 days	Biodegradation	Escapa et al. (2016)
	Scenedesmus			
Obliquus	79.09%, 9 days	Biodegradation	Escapa et al. (2016)	

Estrogenic hormones can be removed rapidly by algae with biodegradation as a major mechanism. As mediated by algae, 17α-estradiol and 17β-estradiol can be biotransformed into estrone, which can be biotransformed into estriol by hydroxylation and further degraded into other unknown products (Zhang et al. 2014b). Biodegradation of progesterone and norgestrel by algae has also been reported, and biotransformation products identified during degradation indicate that hydroxylation, reduction and oxidation were involved in the degradation process (Peng et al. 2014).

Removal of antibiotics by algae was found to be compound specific. Completely removal of 7-amino cephalosporanic acid has been reported in 5 days, which was mainly removed by hydrolysis, photolysis, and adsorption onto microalgae (Guo et al. 2016), while trimethoprim was found to be non-degradable by *Nannochloris* sp. (Bai and Acharya 2016). Ciprofloxacin can be co-metabolized by *Chlamydomonas mexicana* and the removal rate increased from 13 to 56% in 11 days with the addition of sodium acetate as an electron donor (Xiong et al. 2017b). Biodegradation of levofloxacin by *Chlorella vulgaris* enhanced with the addition of NaCl, and the enhanced removal of levofloxacin by salinity was related to the enhanced bioaccumulation and subsequent intracellular biodegradation (Xiong et al. 2017a).

Triclosan was completely removed by *Nannochloris* sp. in 7 days and its removal was attributed to algae mediated uptake and photolysis (Bai and Acharya 2016). However, removal of triclosan by *M. aeruginosa* was mainly attributed to biotransformation with methylation as a major biotransformation pathway and 51% of the removed triclosan was transformed into methyl-triclosan (Huang et al. 2016). Removal of carbamazepine by algae is relatively slow with only 30–37% of the compound removed in 10 days, and two metabolites (10,11-dihydro-10,11-expoxycarbamazepine and n-hydroxycarbamazepine) were identified likely due to the presence of cytochrome P450 in algae (Xiong et al. 2016). Removal of diclofenac varied considerably by different algae species (Escapa et al. 2016). *Scenedesmus obliquus* removed 79.09% of the diclofenac in 9 days while *C. vulgaris* removed only 21.58% with biodegradation as a major mechanism.

Many bacterial strains showing PPCP degradation ability have been isolated from soil, water, activated sludge, and membrane bioreactor (Table 2). These bacteria can degrade certain PPCPs either metabolically or via co-metabolic pathways. Strains belong to *Pseudomonas* sp. were reported most, which showed the ability of degrading a variety of PPCPs. Three bacterial strains belonging to *Stenotrophomonas* and *Pseudomonas* were found to be able to utilize acetaminophen as the sole carbon, nitrogen, and energy source, and a consortium with three strains resulted in a significant enhancement of the degradation rate (Zhang et al. 2013). *Ochrobactrum* sp. MC22, which is a plant-growth promoting bacteria, was found to be able to degrade triclocarban under both aerobic and anaerobic conditions via initial hydrolysis and subsequent metabolism (Sipahutar and Vangnai 2017).

Previously, removal of PPCPs using biofilms with algae-bacteria consortia has been studied as well. Winkler et al. (2001) found that ibuprofen can be

Table 2 Bacteria showing PPCP degradation ability isolated from environment

Strain	Compound	Origin	Mechanism	References
Escherichia sp. HS21	Sulfapyridine, sulfathiazole	Coastal zone	N/A[a]	Zhang et al. (2012b)
Acinetobacter sp. HS51	Sulfapyridine, sulfathiazole	Coastal zone	N/A	Zhang et al. (2012b)
Achromobacter sp. S-3	Sulfamethazine	Aerobic sequence batch reactor	N/A	Huang et al. (2012)
Pseudomonas psychrophila HA-4	Sulfamethoxazole	Activated sludge	Metabolic degradation	Jiang et al. (2014)
Stenotrophomonas maltophilia DT1	Tetracycline	Water	Detoxification	Leng et al. (2016)
Pseudomonas sp. I-24	Iopromide	Activated sludge	Co-metabolism	Xu et al. (2014)
Bacillus thuringiensis B1	Ibuprofen	Soil of a chemical factory	Co-metabolism	Marchlewicz et al. (2016)
Bacillus thuringiensis B1	Naproxen	Soil of a chemical factory	Co-metabolism	Marchlewicz et al. (2016)
Stenotrophomonas maltophilia KB2	Naproxen	NA	Metabolic degradation	Wojcieszyńska et al. (2014)
Variovorax Ibu-1	Ibuprofen	Activated sludge	Metabolic degradation	Murdoch and Hay (2015)
Delftia tsuruhatensis	Acetaminophen	Membrane bioreactor	Metabolic degradation	De Gusseme et al. (2011)
Pseudomonas aeruginosa	Acetaminophen	Membrane bioreactor	Metabolic degradation	De Gusseme et al. (2011)
Stenotrophomonas sp. f1	Acetaminophen	Airlift sequencing batch reactor	Metabolic degradation	Zhang et al. (2013)
Pseudomonas sp. f2 and fg-2	Acetaminophen	Airlift sequencing batch reactor	Metabolic degradation	Zhang et al. (2013)
Pseudomonas aeruginosa HJ1012	Acetaminophen	NA	Metabolic degradation	Hu et al. (2013)
Streptomyces MIUG 4.89	Clofibric acid	Soil	N/A	Popa Ungureanu et al. (2016)
Pseudomonas aeruginosa TJ1	17β-estradiol	Activated sludge	Metabolic degradation	Zeng et al. (2009)
Pseudomonas sp. CBZ-4	Carbamazepine	Activated sludge	Metabolic degradation	Li et al. (2013)
Pseudomonas beteli	Parabens	Non-sterile paraben solution	Metabolic degradation	Amin et al. (2010)
Burkholderia latens	Parabens	Non-sterile paraben solution	Metabolic degradation	Amin et al. (2010)
Pseudomonas putida TriRY	Triclosan	Compost	Metabolic degradation	Meade et al. (2001)
Alcaligenes xylosoxidans subsp. *denitrificans* TR1	Triclosan	Compost	Metabolic degradation	Meade et al. (2001)

Table 2 continued

Strain	Compound	Origin	Mechanism	References
Ochrobactrum sp. MC22	Triclocarban	Soil	Metabolic degradation	Sipahutar and Vangnai (2017)
Pseudomonas sp. CE21	Cefalexin, Caffeine, Salicylic acid, Chloramphenicol, Sulfamethoxazole, Naproxen	Activated sludge	N/A	Lin et al. (2015)
Pseudomonas sp. CE22	Cefalexin, Caffeine, Salicylic acid, Chloramphenicol	Activated sludge	N/A	Lin et al. (2015)

[a] Not mentioned

degraded biologically in a river biofilm reactors but clofibric acid was recalcitrant. Writer et al. (2011) demonstrated that stream biofilm play an important role in the removal of steroidal hormones with the involvement of both biodegradation and sorption processes. Periphyton cultured in laboratory was found to be able to remove parabens and their chlorinated by-products rapidly, and higher incubation temperature enhanced the degradation (Song et al. 2017). Combining *Chlorella* sp. and a ketoprofen degrading bacterial consortium showed a high biodegradation efficiency and tolerance for ketoprofen (Ismail et al. 2016). Results from these works suggest that algae and algae-bacteria consortia have a great potential to be used in PPCP removal from wastewater.

5 Removal of PPCPs in algae-based treatment systems

Many algae-based systems have been developed for the treatment of wastewater and algae cultivation. Features of typical algae-based treatment systems are summarized in Table 3. Although many previous works have showed that algae, bacteria, and their consortia are capable of removing a variety of PPCPs, very few works have been performed to study the removal of PPCPs from wastewater using algae-based treatment systems. As listed in Table 4, investigated algae-based treatment systems include algal pond and photobioreactor (PBR) of different configurations at laboratory or pilot scales. Mixtures of algae or algae-bacteria consortia were used in these treatment systems.

Removal of three estrogens (estrone, 17β-estradiol, 17α-ethinylestradiol) from wastewater was studied in algae and duckweed pond systems (Shi et al. 2010). In algae pond, a mixture of six different algae genera (*Anabaena cylindrica*, *Chlorococcus*, *Spirulina platensis*, *Chlorella*, *Scenedesmus quadricauda*, and *Anaebena* var.) was used. Presence of both algae and duckweed enhanced the removal of three estrogens, and continuous flow tests showed that the algae and duckweed ponds effectively removed all three estrogens with treatment efficiency over 80%.

Two studies investigated the removal of PPCPs using high rate algal pond (HRAP) both with *C. vulgaris* and other heterotrophic microorganisms. de Godos et al. (2012) found that the removal of tetracycline stabilized at around $69 \pm 1\%$ in the HRAP after 62 days, and photodegradation and biosorption are main mechanisms responsible for the removal. Moreover, deflocculation of the HRAP biomass in the presence of tetracycline was observed, which might hamper biomass recovery during the full-scale treatment. Matamoros et al. (2015) showed that the removal of 26 emerging organic contaminants varied considerably from no removal to over 90% removal when treated using a HRAP. Higher removal efficiency was observed during warm season, while longer hydraulic retention (HRT) only showed noticeable enhancement in cold season. Biodegradation and photodegradation were believed to be the most important pathway for the removal of the compounds, while volatilization and sorption were only significant for hydrophobic compounds with a moderately high Henry's law constant.

Removal of PPCPs has also been studied using PBRs with different configurations. In a stirred-tank photobioreactor with a microalgal–bacterial consortium, the removal of ketoprofen, acetaminophen, and aspirin mixture was poor with a HRT of 3 days but

Table 3 Features of typical algae-based treatment systems

System	Characteristics	Advantages	Limitations
High rate algal ponds	Shallow, open raceway ponds with mixing provided by paddlewheels illuminated with sunlight	Low-cost to install and maintain, simple to operate, easy to scale up	Low light utilization efficiency, high risk of contamination, difficult in species control, large land requirement, poor environmental condition control, high evaporation
Algal Turf Scrubber	Attached algae biofilm growing on screens in a shallow trough or raceway illuminated with sunlight	Low-cost to install and maintain, simple to operate, easy to harvest, low energy consumption, easy to scale up	Difficult in species control, large land requirement, mutual shading, poor environmental condition control, high evaporation, poor mass transfer
Rotating algae biofilm photobioreactor	Semi-immersed disks with attached algae biofilm	Simple to operate, easy to harvest, good gas transfer, easy to scale up	High-cost to install, difficult in species control, poor environmental condition control, poor mass transfer
Stirred-tank photobioreactor	Mechanically agitated tanks with suspended algae	Better environmental condition control, low risk of contamination, high productivity, space saving	Low light utilization efficiency, high-cost to install and maintain, difficult to scale up, complicated to operate
Flat panel photobioreactor	Closed cuboidal shape chambers with suspended or immobilized algae	High light utilization efficiency, better environmental condition control, low risk of contamination	High-cost to install, difficult to scale up, complicated to operate
Tubular photobioreactor	Closed tubing systems placed vertically or horizontally with suspended or immobilized algae	High light utilization efficiency, better environmental condition control, low risk of contamination, reduced photoinhibition	High-cost to install, complicated to operate, decrease of illumination surface area upon scale-up, difficult to clean
Membrane photobioreactor	Membrane contactor for biomass retention and/or gas sparger	High productivity, better environmental condition control, low risk of contamination, easy to harvest, good gas transfer, easy to scale up	High-cost to install, complicated to operate, membrane fouling

enhanced greatly with a HRT of 4 days (Ismail et al. 2017). Continuous illumination showed the highest removal efficiency. An enclosed multitubular PBR was used for toilet wastewater treatment and PPCP removal (Hom-Diaz et al. 2017). Removal of the investigated PPCPs varied from 98% for anti-inflammatory drugs to 30–57% for the psychiatric drug lorazepam. Higher removal efficiencies were obtained during period-II with a HRT of 12 days than period-I with a HRT of 8 days, although the temperature and light irradiation were lower in period-II. An open photobioreactor with a mixture of wild freshwater green algal species was operated under natural light and aerated with flue gases, and its ability to remove 79 PPCPs was studied (Gentili and Fick 2017). Among the investigated 79 PPCPs, 27 were not detected, and very high (>90%), moderate (50–90%), low (10–50%), and very low or non-quantifiable (<10%) removal were observed for 9, 14, 11, and 18 PPCPs, respectively. Removal efficiency was positively correlated with light intensity inside the culture.

Limited works have demonstrated that many PPCPs can be removed from wastewater simultaneously with other pollutants using algae-based treatment systems. Biodegradation, photodegradation, and sorption are main processes involved in PPCP removal during treatment but also depending on the properties of the compounds. Poor removal was also observed for several PPCPs such as carbamazepine, lorazepam, trimethoprim, and verapamil, which can be attributed to their low biodegradability, lack of UV absorption functional groups, or physical–chemical properties unfavorable for sorption and volatilization.

The performance of algae-based treatment systems can be affected by environmental and operation conditions. Enhanced treatment efficiency was observed in warm season likely due to higher microbial activities at higher temperature (Matamoros et al. 2015). Temperature conditions are well known to affect the enzyme activities of microbes and different temperature favors the development of different algae and bacteria species, which will result in functional

Table 4 Removal of PPCPs in algae-based treatment systems

Treatment system	Setup	Treatment efficiency	References
Algal pond inoculated with a mixture six algae	Continuous flow, three aquaria ($50 \times 29 \times 25$ cm^3), temperature 20 °C, 12-h light and dark regime illuminated with high pressure mercury lamp at 100 μE m^{-2} s^{-1}, HRT 15 days	Estrone 83.9%, 17β-estradiol 91.2%, 17α-ethinylestradiol 86.8%	Shi et al. (2010)
High rate algal pond with *Chlorella vulgaris* and other microorganisms	Two cylindrically shaped stainless steel tanks ($0.4 \times 0.2 \times 0.3$ m^3), PAR[a] and UV-AB radiation at the water surface were 10 and 0.8–0.9 W m^{-2}, HRT 7 days	Tetracycline 69 ± 1% from day 62	de Godos et al. (2012)
High rate algal pond inoculated with non-axenic *Chlorella vulgaris* culture	Recirculation velocity, 11 cm s^{-1}, surface area of 1.54 m^2, depth 0.3 m, volume 0.5 m^3, outdoor, HRT 4 and 8 days	26 pollutants including PPCPs 0 to >90%, higher in warm season, higher HRT enhanced removal in cold season	Matamoros et al. (2015)
Stirred-tank photobioreactor 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%, 24 h illumination at short HRT led to the best results	Ismail et al. (2017)
Multitubular photobioreactor 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)
Open photobioreactor with a mix of freshwater green algae	Surface area 2.72 m^2, 3 m long, 1.45 m wide, 0.4 m deep, volume 650 L, aerated with flue gases at 3 L min^{-1} in the daytime, outdoor, 1 week retention for each batch	Very high (>90%), moderate (50–90%), low (10–50%), and very low or non-quantifiable (<10%) for 9, 14, 11, and 18 pharmaceuticals, respectively	Gentili and Fick (2017)

[a] Photosynthetically active radiation

differences of algae-based systems (Boulêtreau et al. 2012; Webster et al. 2011). Illumination is also important for algae-based systems due to the requirement of photosynthesis for algae growth and the possibility of photodegradation. Previous research showed that continuous illumination was preferred for the removal of analgesic pharmaceuticals but dark condition favored the removal of chlorinated parabens (Ismail et al. 2017; Song et al. 2017). HRT has been studied most as an important operational parameter, and better PPCP removal was achieved with longer HRT (Ismail et al. 2017). However, the relevant information is still very limited. The influences of illumination strategies (light source, intensity, and illumination regime), treatment system configurations (open or closed; flat, tubular, or spiral), and microbial selection (single or mixed; suspended or biofilm) on PPCP removal are largely unknown. Therefore, more researches are needed to solve these problems in future.

6 Conclusions and future perspectives

In the past decades, algae-based technologies have received growing attention for their potential application in wastewater treatment. Meanwhile, environmental pollution of emerging contaminants such as PPCPs attract increasing concern due to their extensive use and potential environmental risks. As reviewed in this work, algae and algae-bacteria consortia have been demonstrated with great capability in PPCP removal. Therefore, algae-based technologies can be used as an alternative measure for removing PPCPs from wastewater. Algae-based technologies show several advantages over conventional wastewater treatment technologies including reduction in energy consumption, biological sequestration of industrial flue gas, and algae biomass production for valuable products. Even though, there are still many research gaps need to be filled before algae-based technologies can be implemented in large scales:

1. Considering that PPCPs include a variety of compounds of different physical–chemical properties. It is unrealistic to treat all PPCPs efficiently with algae-based technologies. It is important to develop a priority list of PPCPs exhibiting high occurrence and high environmental risks in wastewater. Algae-based technologies should be developed and optimized primarily targeting PPCPs among the priority list.
2. Many studies reported the degradation of PPCPs by pure algal and bacterial culture. However, only few studies have identified the complete degradation pathways and assess the fate and toxic effects of the degradation products. Therefore, it is recommended that more attention should be focused the degradation pathways and potential risks of the degradation products to ensure not only the removal of parent compounds but also a reduction in toxicity.
3. Using algal–bacterial consortia can be advantages over using specific algae species for PPCP removal due to the involvement of multiple catabolic pathways from different microorganisms and the difficulty in species control in the algae-based treatment system for real wastewater due to contamination. Therefore, roles of different microorganisms in algal-bacterial consortia in PPCP removal should be better understood and the microbial community structure should be optimized for PPCP removal instead of trying to maintain certain species in algae-based treatment systems.
4. Configuration and operation conditions of algae-based treatment systems affect the microbial composite and biomass accretion and thus affect the performance of algae-based treatment systems in pollutants removal. Additional works are needed to investigate the removal of PPCPs in algae-based treatment systems with different configurations and to study the how their treatment efficiency affected by operation conditions. Mechanisms behind should be better revealed for a better design of the treatment systems for PPCP removal.
5. Most of the works studied the removed of PPCPs in algae-based treatment systems along but overlook the impact of PPCPs on conventional pollutant removal and subsequently valuable product generation. In future, more comprehensive works are required to assess the overall performance and benefit of algae-based treatment systems to allow their practical application in reality.

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