Review

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Using macroalgae as biofuel: current opportunities and challenges

https://doi.org/10.1515/bot-2019-0065
Received 9 September, 2019; accepted 18 February, 2020

Abstract: The rising global demand for energy and the decreasing stocks of fossil fuels, combined with environmental problems associated with greenhouse gas emissions, are driving research and development for alternative and renewable sources of energy. Algae have been gaining increasing attention as a potential source of bio-renewable energy because they grow rapidly, and farming them does not, generally, compete for agricultural land use. Previous studies of algal biofuels have focused on microalgae because of their fast growth rate and high lipid content. Here we analyze the multiple merits of biofuel production using macroalgae, with particular reference to their chemical composition, biomass and biofuel productivity, and cost-effectiveness. Compared to microalgae, macroalgae have lower growth rates and energy productivity but higher cost-effectiveness. A biomass productivity of over 73.5 t dry mass ha^{-1} $year^{-1}$ with a methane yield of 285 m^3 t^{-1} dry mass would make electricity production from macroalgae profitable, and this might be achieved using fast-growing macroalgae, such as *Ulva*. Taking into account the remediation of eutrophication and CO_2, exploring macroalgae for a renewable bioenergy is of importance and feasible.

Keywords: biofuel; biogas; bioremediation; cultivation; macroalgae; photosynthesis.

Introduction

Today, approximately 85% of the total energy consumed worldwide is provided by fossil fuels (Dudley 2018). Stocks of fossil fuels are declining, and there is growing concern regarding the serious environmental problems associated with their consumption (Ravi et al. 2018, Saratale et al. 2018). Consequently, it is of general importance to search for renewable and cost-effective energy sources with low or zero greenhouse gas emissions (Ravi et al. 2018). Biofuels, mainly extracted from plants, can serve as an attractive source of energy to meet some of the present and future fuel demand (Robertson et al. 2017). Currently, bio-ethanol and biodiesel, which are produced primarily from food crops such as grain, sugar cane and vegetable oils, are the most widely available forms of biofuel (Landis et al. 2017, Adeniyi et al. 2018). However, these first generation biofuels suffer from concerns regarding their possible impact on food supply and security (Herrmann et al. 2018). For instance, the UK consumed an estimated 47 billion liters of transport fuel in 2008, 53% of which was diesel. More than half the land area of the UK would be needed if this diesel were to be produced from oilseed rape (Stephenson et al. 2008). As a result, increasing attention is being focused on non-food based biofuels and feedstocks. In comparison to other feedstocks, such as sugarcane, algae can provide a high-yield source of biofuels without competing with food production (Chisti 2007, Adeniyi et al. 2018). For instance, to meet the energy requirement for transport in the UK in 2008, only 2.17% and 6.63% land area of the UK is needed if diesel is produced from microalgae with 30% lipid content (Chisti 2007) and from *Ulva lactuca* Linnaeus (Bruhn et al. 2011), respectively. In addition, algae also display a potential for CO_2 sequestration (Gao and McKinley 1994, Duarte et al. 2013, 2017).

The first consideration when developing algal biofuels is the choice of species or strains that contain high oil-like precursors. Algae are aquatic photosynthetic organisms which can generally be divided into two groups: microalgae (unicellular plants) and macroalgae (or seaweeds). Microalgae appear to be the only source of biodiesel that has the potential to completely displace fossil diesel

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because of their fast growth rate and rich lipid content (Singh et al. 2011, Jiang et al. 2016, Dickinson et al. 2017). Biomass doubling times for microalgae are generally less than 24 h. In addition, the lipid content of some microalgae is close to 80% dry weight (Spolaore et al. 2006, Chisti 2007, Bwapwa et al. 2017). Thanks to their fast growth rate and high lipid content, biodiesel productivity from microalgae with 30% (w/w) lipid content in algal biomass could be up to 342 or 92 times more when compared to corn or soybean, respectively (Mata et al. 2010). However, based on previous studies, there is still a long way to go to bring biodiesel from microalgae to the market because of the high cost of biomass production and extraction (Sheridan 2013, Ajjawi et al. 2017, Borowitzka and Vonshak 2017, Posewitz 2017, Remmers et al. 2018).

The use of macroalgae for CO_2 bioremediation has been explored previously (Gao and McKinley 1994, Chung et al. 2011, Sondak et al. 2017). Nevertheless, macroalgae have been less studied as a source of biofuel, although there was a pilot project during the 1980s in the USA (North 1987). However, macroalgae are commonly known to have high levels of carbohydrates (Kraan 2010, Gao et al. 2017a, 2018a), which can be fermented into biogas. They are also more cost-efficient with respect to farming and processing compared to microalgae (see the section "Economics of biofuels from algae"). We therefore set out to examine the feasibility of using macroalgae as biofuel by analyzing their chemical composition, biomass productivity and biofuel productivity, and cost-effectiveness. In terms of the macroalgal biofuels, we focus on biomethane as it is deemed to have the highest energy yield in comparison to biodiesel and bioethanol (Harun et al. 2011, El-Mashad 2015, Wu et al. 2019). The literature was obtained from Web of Science for the period from 1980 to 2019 when searching with related keywords. The availability of the data was also considered during this selection of literature. While there are a great number of scientific articles about algal biofuels, we only analyzed and cited those that are most relevant to the topic of this review.

Chemical composition

The chemical composition of plant biomass is a primary factor to be considered for generation of biofuels as it determines not only which method should be employed to process the biomass but also the production yield of biofuel. As shown in Figure 1A and Supplementary Table S1, carbohydrates dominate macroalgal chemical

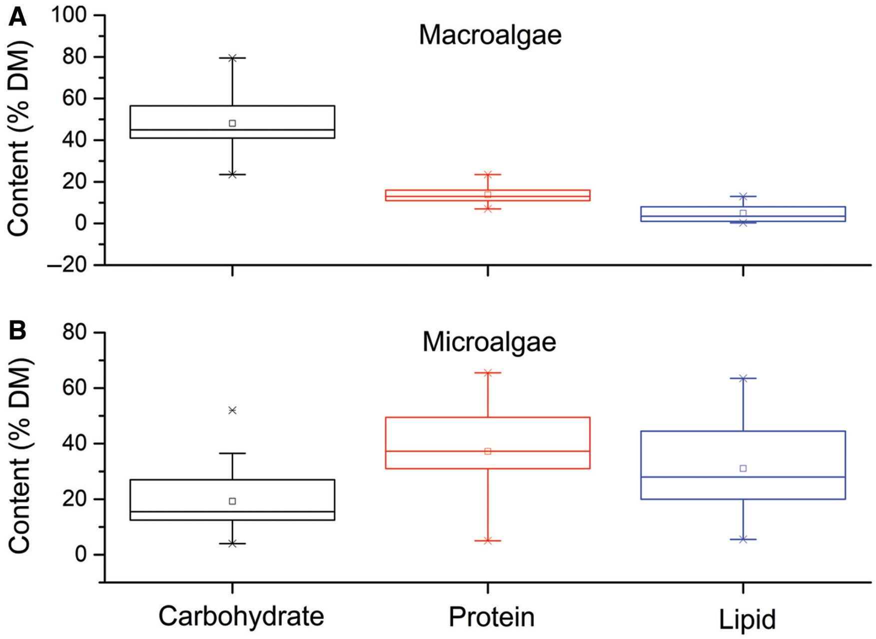

Figure 1: Box charts of main biochemical composition of macroalgae (A) and microalgae (B).
DM, Dry mass. Data were based on Renaud et al. (1994), Horn (2000), Pádua et al. (2004), Chisti (2007), Demirbas (2010), Yoon et al.(2010), Kim et al. (2011), Satyanarayana et al. (2011), Jang et al. (2012), Hong et al. (2014), Khatoon et al. (2014), Tibbetts et al. (2015), Jiang et al. (2016), Zheng et al. (2016), Chan and Matanjun (2017), Cheng et al. (2017), Diprat et al. (2017), Gao et al. (2017a, 2019), Kalita et al. (2017), Shakya et al. (2017), Wang et al. (2017), Abomohra et al. (2018), Gao et al. (2018b), Ishika et al. (2018), Shuba and Kifle (2018), Uribe et al. (2018). Please see Supplementary Table S1 for details.

composition, making up about half of their dry mass in some species, while the lipid content is usually very low (<10% dry mass). Based on the chemical composition, the preferential approach with macroalgae has therefore been to produce biogas by anaerobic digestion or bioethanol by fermentation. In contrast, the major components of microalgae are usually protein and lipid (Figure 1B). The lipid content of microalgae ranges from 1 to 77% of the dry mass and a level of 20–40% is quite common. The high lipid content in microalgae indicates that extracting biodiesel would be the most appropriate for biofuel production.

Despite species and environmental differences, the reasons for a substantially different chemical composition between macroalgae and microalgae are many. Firstly, macroalgae have substantially more compounds with a mechanical function to support the thallus structure (especially carbohydrates such as phycocolloids) leading to a relatively low abundance of lipids (Lee et al. 2017, Rhein-Knudsen et al. 2017). Secondly, microalgae have a larger proportion of chloroplast and thylakoid membranes compared to macroalgae. This is because microalgae are unicellular with little functional differentiation, leading to a larger proportion of photosynthetic membranes over the total cell volume (Han et al. 2003, Gao et al. 2017b). Phospholipid is the main component of cell membranes and a larger proportion of cell membranes in microalgae leads to a higher lipid content (Mooy et al. 2009, Yu et al. 2018). Thirdly, microalgae have a higher percentage of cell membrane by weight and hence a higher lipid content because of their higher surface to volume ratio (Hein et al. 2014). Finally, many microalgal species may have proportionately more lipids to increase their buoyancy and decrease their settling (Khanam et al. 2017, Pančić and Kiørboe 2018).

Biomass production

High biomass productivity is also an important factor when searching for potential bio-renewable resources. Algae possess remarkable advantages over terrestrial plants in terms of their biomass productivity since they grow in aquatic environments, saving land space and offering the potential for continuous cultivation (Jung et al. 2016). In addition, algae have a higher conversion efficiency of light energy to biomass compared to terrestrial plants, up to 5–10% vs. 0.5–3% (Wassink 1959, Laws et al. 1986, 1988, Melis 2009). This could be due to their fast growth with lower photorespiratory carbon loss because most algae operate CO_2 concentrating mechanisms to facilitate carboxylation and suppress photorespiration (Melis 2009, Stephenson et al. 2011).

In addition to the culture conditions, the culture systems determine, to a large extent, the algal biomass yield. Daily biomass productivity of most macroalgae in tank culture is commonly not more than 30 g DM m^{-2} land d^{-1} (Figure 2A and Supplementary Table S2), which is comparable to microalgal productivity in open ponds (OP) and horizontal photobioreactors (HPB) but lower than in inclined photobioreactors (IPB; Figure 2B). Annual biomass productivity of macroalgae in nearshore farms is usually lower than 60 t DM ha^{-1} land $year^{-1}$, and lower than that in tank culture (Figure 2C). Tank culture of *Ulva* species could reach higher biomass productivity (138 t DM ha^{-1} land $year^{-1}$) but is still lower than the maximum from microalgae (Figure 2C, D). Microalgae are usually single-celled organisms, with a diameter of 1–200 μm (Madhu et al. 2017), while macroalgae, in most life stages, are more than 1 cm in length, and some species such as giant kelp can reach up to 90 m in length (Setchell 1908). The smaller microalgae have a higher surface to volume ratio and as a result can benefit from faster access to light and nutrients (Hein et al. 2014). In addition, microalgae do not have nonproductive cells, and all cells of microalgae are identically productive, while some tissues in the basal parts of macroalgae generally grow much slower than apical cells (Gao et al. 2017a). Due to their single-celled construction, microalgae do not utilize energy in producing structural biopolymers as backbones for their multicellular tissue. In addition, photobioreactors are more productive by providing optimal light and temperature conditions compared to ponds or near shore cultivation of macroalgae (Narala et al. 2016).

Biofuel production

Conversion of biomass to energy can be categorized into three main technologies: thermo-chemical, biochemical/biological and physical extraction (with esterification) as shown in Figure 3. Thermo-chemical conversion encompasses four process options: combustion, pyrolysis, liquefaction and gasification. There are two main process techniques in biochemical conversion: anaerobic digestion and ethanol fermentation. Several techniques have been utilized to produce biofuel from macroalgae; ethanol fermentation, anaerobic digestion and liquefaction will be briefly introduced in the following context.

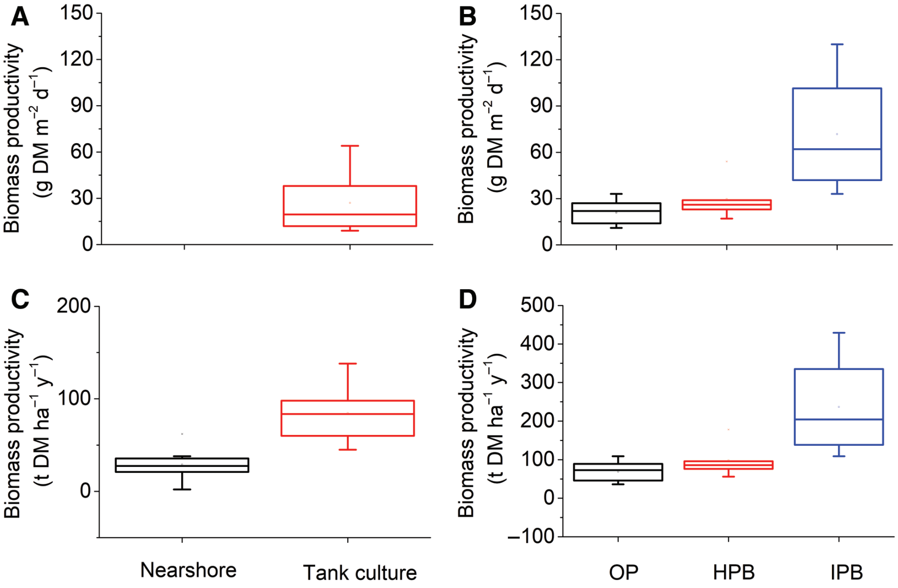

Figure 2: Box charts of biomass productivity of macroalgae (A, C) and microalgae (B, D) using different culture systems. OP, Open pond; HPB, horizontal photobioreactor; IBP, inclined photobioreactor; DM, dry mass. Data were based on Bidwell et al. (1985), Torzillo et al. (1986), Chaumont et al. (1988), Richmond et al. (1990), Lee and Low (1991), Richmond et al. (1993), Buschmann et al. (1994), Lee et al. (1995), Hu et al. (1996), Chynoweth (2002), Jiménez et al. (2003), Moreno et al. (2003), Doucha et al. (2005), Doucha and Lívanský (2006), Bruhn et al. (2011), Chen et al. (2013), Al-Hafedh et al. (2015), Correa et al. (2016), de Mooij et al. (2016), Mata et al. (2016), CFSY (2017), Benavides et al. (2017), Camus et al. (2018), Gao et al. (2018b), Romero-Villegas et al. (2018) and Magnusson et al. (2019). Please see Supplementary Table S2 for details.

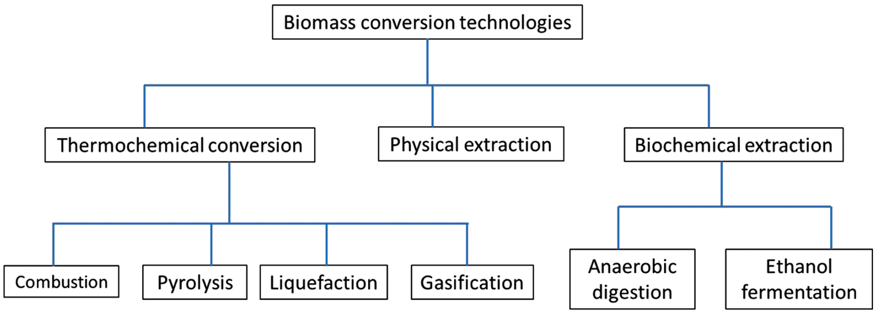

Figure 3: Technologies for conversion of biomass to energy.

Ethanol fermentation

Bioethanol from first generation feedstock, such as corn and sugarcane, has been the preferred choice as an automotive co-fuel and is now widely produced and used in many countries (Khuong et al. 2017). However, this first-generation bioethanol consumes a large amount of food crops and impacts food security. This has led to the development of second-generation bioethanol from lignocellulose biomass. Unfortunately, bioethanol from second-generation feedstock encountered huge resistance due to the difficulties in processing technology and scaling up (Gressel 2008, Ramachandra and Hebbale 2020). One main technological challenge is that biodegradation of cellulose and hemicelluloses by cellulases can be inhibited by lignin (Gressel 2008, Ramachandra and Hebbale 2020). Macroalgae do not usually contain much lignin and therefore polysaccharides in macroalgae can be more easily converted to ethanol (Dave et al. 2019). Accordingly, macroalgae-derived bioethanol has been gaining increasing attention.

Bioethanol has been produced from all kinds of macroalgae, including brown, green and red macroalgae (Ramachandra and Hebbale 2020). Brown macroalgae seem to be the principal feedstock for bioethanol production due to their high polysaccharide content and successful mass-cultivation (Jung et al. 2013, Enquist-Newman et al. 2014). For instance, ethanol production from brown macroalgal sugars by a synthetic yeast platform yields up to 83% of the theoretical maximum (Enquist-Newman et al. 2014), although ethanol conversion up to of 50% of the theoretical maximum from macroalgae is considered ambitious (Roesijadi et al. 2010a). Red macroalgae have also been fermented and yielded 45% of the theoretical maximum (Meinita et al. 2013). Bioethanol yields from the fermentation of macroalgae commonly range from 0.08 to 0.12 $kg \cdot kg^{-1}$ dry mass. However, Wargacki et al. (2012) has reported higher experimental ethanol yields of up to 0.281 $kg \cdot kg^{-1}$ dry mass from brown macroalgae by an engineered microbial platform. Different bioethanol yields can be attributed to species differences and processing methods. The bioethanol production from macroalgal biomass can be divided into two processes: pretreatment and microbial fermentation. The pretreatment step is very crucial and can determine the saccharide generation efficiency for efficient bioethanol production (Dave et al. 2019). The energy-intensive pretreatment leads to the failure of large-scale utilization of bioethanol from macroalgae to a large extent. Therefore, future work should pay more attention to developing a low-cost and scalable method for bioethanol production from macroalgae.

Anaerobic digestion

Anaerobic digestion (AD) technologies have a long history and the first industrial digestion plant was initiated in Bombay in 1859 (Kiyasudeen 2016). The first usage of biogas recovered from a sewage treatment facility was reported for street lamps in Exeter, England in 1895 (Kiyasudeen et al. 2016). Cellulosic materials such as dung and straw have been converted into methane for cooking in China for a long time (Buysman 2009). Anaerobic digestion is also an effective technique to treat sewage bio-solids, livestock manure, and concentrated wastes from the food industry and industrial wastewater (Nasir et al. 2012). Rising fossil fuel prices combined with increasing concerns for greenhouse gas emissions and global warming have prompted interest in further AD research and industrial applications. The total biogas production in the world has almost doubled from 1990 to 2016 (Energy Statistics Database 2019). AD occurs in four stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis, among which, hydrolysis is the rate-limiting step of AD and also determines the final yield of biomethane to a great extent (Montingelli et al. 2016, Thompson et al. 2019).

Compared to biodiesel extraction and bioethanol fermentation, AD has a larger energy output because all macromolecular substances (protein, lipid and carbohydrate) can be utilized for biomethane production (Harun et al. 2011). Meanwhile, the energy input for biogas production from some crops is lower than that for bioethanol production, which results in a higher energy output-to-input ratio for AD (Börjesson and Mattiasson 2008). However, Patterson et al. (2008) demonstrated that the input energy for biomethane from sugar beet or fodder maize was higher than that for biodiesel from oilseed rape and bioethanol from wheat grain due to extra energy requirement for gas upgrading and compression. Another main problem for AD of algae is that biomethane yields from many algae are substantially below the theoretical maximum. Typical methane yields of seaweeds (~200 $l\ CH_4\ kg^{-1}$ volatile solids, VS) are less than 50% of those from common commercially exploited feedstocks (Astals et al. 2015, Chen et al. 2015) although an extremely high yield (480 $l\ CH_4\ kg^{-1}$ VS) was reported from a mixture of *Ulva*, *Cladophora* and *Chaetomorpha* species (Hansson 1983). The relatively low yield is related to high nitrogen, sulfur and salt contents of algae that can lead to potential inhibition for biomethanation (Tedesco et al. 2014). Therefore, there is noticeable room to improve biomethane yield by developing robust AD technology.

Liquefaction

Liquefaction used to involve employing carrier gases such as hydrogen or carbon monoxide to produce liquid fuels from solid material at moderate temperature (573–673 k) and high pressure, and it was initially used for coal liquefaction (Balat 2008). Liquefaction now refers to any thermochemical conversion process that primarily yields liquid products (Balat 2008, Ghadiryanfar et al. 2016). Liquefaction can process materials with any level of moisture content. Thus, this method is particularly suitable for algae since they contain a high level of water. The water may play a positive role in the liquefaction process as the physical and chemical properties of water change when heated near or above the critical point (374°C, 221 bar). The solubility of nonpolar hydrocarbons increases and, therefore, the decomposition of the biomass is improved (Kruse and Dinjus 2007). Current liquefaction processes involve high temperature and high pressure liquefaction, direct catalyst liquefaction, and supercritical liquefaction (Zhuang et al. 2012, Raikova et al. 2019).

Due to their high alkali content, macroalgae are suitable for liquefaction because high alkali has been showed to have a catalytic function on bio-oil production during liquefaction (Anastasakis and Ross 2015). Many macroalgal species in all groups have been examined for bio-crude production via liquefaction and, up to now, the most intensively studied macroalgal species have been from the genera *Laminaria* and *Ulva* (Raikova et al. 2019). A maximum bio-oil yield of 19.3% dry weight with a higher heating value (HHV) of 36.5 MJ kg^{-1} was obtained from *Laminaria saccharina* Linnaeus at 350°C without the presence of the catalyst (Anastasakis and Ross 2011). Zhou et al. (2010) used direct catalyst liquefaction to recover bio-oil from *Ulva prolifera* OF Müller and a maximum yield of bio-crude of 23% dry weight was achieved at 300°C with 5% Na_2CO_3 (w/w) by hydrothermal liquefaction and HHV of bio-oils were approximately 29 MJ kg^{-1}. The yield of bio-oil from *U. prolifera* OF Müller was significantly improved up to 84.81% with heating value of 15.05 MJ kg^{-1} via microwave-assisted direct liquefaction and it could be further enhanced to 93.17% with HHV of 17.36 MJ kg^{-1} by optimizing conditions (Zhuang et al. 2012, Liu et al. 2013). Although hydrothermal liquefaction has higher energy output and lower energy input for processing algae compared to gasification and pyrolysis (Vardon et al. 2012, Chen et al. 2014), it is likely that energy requirements for upgrading bio-crudes to usable biofuels would be high because bio-crudes have a higher concentration of heteroatoms oxygen, nitrogen, and sulfur (Mathimani et al. 2019). Few studies have been conducted on upgrading macroalgal bio-crudes to date, and therefore more work should be conducted in the future to improve algal liquefaction.

A comparison of several of the most used technologies for processing biofuels from algae is shown in Table 1. Here we compare methane from macroalgae with biodiesel from microalgae in terms of energy production and cost, since these two methods have been considered to have the potential to compete with fossil diesel (Milledge et al. 2014, Dickinson et al. 2017, Wu et al. 2019). The biofuel yields from macroalgae and microalgae were 157–480 m^3 CH_4 $tonne^{-1}$ DM and 85–360 kg biodiesel $tonne^{-1}$ DM, respectively; when converted to energy yield, they were 6.3–19.2 and 3.2–13.6 MJ $tonne^{-1}$ DM, indicating that macroalgae have a higher energy yield when normalized to dry mass (Figure 4A, B and Supplementary Table S3). Energy productivity (excluding the energy put into biofuel production) from macroalgae is in the range 96–677 GJ ha^{-1} $year^{-1}$, which is again comparable to that from microalgae cultured in open ponds but lower than that from those cultured in photobioreactors (Figure 4C, D). This is due mainly to the high biomass productivity in photobioreactors.

Economics of biofuels from algae

Cost-effectiveness has to be considered when algae are used for the production of biofuels. The main costs can be generally split into two parts: cultivation and processing. The cultivation cost of macroalgae can be lower than \$100 t^{-1} dry mass in Asian countries where the labor cost is low (Chynoweth 2002, Roesijadi et al. 2010b). The cultivation cost of microalgae cultured in open ponds (\$220–5940 t^{-1} dry mass) is usually lower than for photobioreactors (\$430–7152 t^{-1} dry mass; Figure 5A). The higher cultivation cost for microalgae is determined by the culture methods. Large scale microalgal cultivation is conducted in open ponds or photobioreactors, in which the supply of water, CO_2 and nutrients is required and mixed using paddle wheels, which also require energy input, which is costly (Hoffman et al. 2017). In addition, the photobioreactor tube system itself represents a significant capital investment accounting for around 80% of the total capital cost (Davis et al. 2011), leading to higher cultivation costs

Table 1: Comparison of methods used for converting algal biomass to biofuels.

Methods	Principle	Advantage	Disadvantage
Anaerobic digestion	$\text{Organic matters} \xrightarrow[\text{bateria}]{\text{Anaerobic}}$ $CH_4 + CO_2 + H_2 + NH_3 + H_2S$	1. Utilize the whole algal cell 2. High energy output 3. Do not need drying	1. Lack robustness at industrial scale 2. Products need to purify and compress
Ethanol fermentation	$(CH_2O)_n \xrightarrow{\text{Yeast}} C_2H_5OH + CO_2$	1. Do not need drying 2. Product is easy to collect	1. Pretreatment is energy-intensive 2. Only utilize polysaccharides 3. Low energy output
Transesterification/ Biodiesel	$\longrightarrow$	1. Product is easy to extract 2. Product is easy to collect	1. Only apply to algae with high lipid content 2. Biomass needs drying
Liquefaction	$\text{Organic matters} \xrightarrow[\text{\& pressure}]{\text{High T}}$ $\text{Bio-oil} + \text{biochar} + \text{gas}$	1. Utilize the whole algal cell 2. Relatively high energy output 3. Do not need drying	1. Products need to be upgraded 2. High energy input

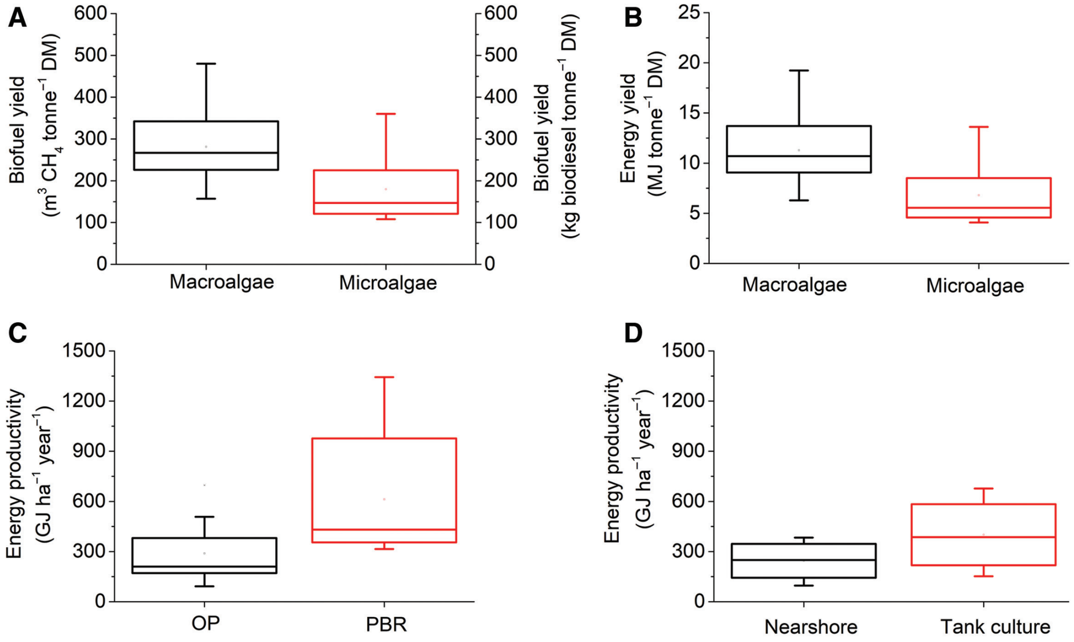

Figure 4: Box charts of yield of biofuel (A) and energy (B) and of gross energy productivity of macroalgae (C) and microalgae (D) in different culture systems.
OP, Open pond; PBR, photobioreactor; DM, dry mass. Data were based on Hansson (1983), Bruhn et al. (2011), Nascimento et al. (2014), Abomohra et al. (2016), Matsumoto et al. (2017), Tabassum et al. (2017), and Gao et al. (2018b). Please see Supplementary Table S3 for details.

compared to open ponds. In contrast, nearshore macroalgal cultivation takes advantage of natural seawater, CO_2, nutrients and mixing, with lower capital costs. Culture systems for macroalgal farming are relatively simple and cheap. In addition, the costs of harvesting and dewatering are more for microalgae compared to macroalgae (Aitken et al. 2014, Fasaei et al. 2018). Processing costs (Figure 5B) are also lower for macroalgae (\$8–18 t^{-1} dry mass) compared to microalgae (\$74–183 t^{-1} dry mass) as shown in Supplementary Table S4. This may be due to the chemicals used in oil extraction while aerobic digestion for biogas production from macroalgae does not consume additional chemicals.

To estimate what biomass productivity and methane yield in macroalgae, or lipid content in microalgae, is required to compete with fossil fuels, we have modeled the costs using the data of Dave et al. (2013) to define the relationship between the minimum electricity selling price (MESP), biomass productivity and methane yield for macroalgae, and the data of Davis et al. (2011) and Batan et al. (2016) to define the relationships between the minimum fuel selling price (MFSP), biomass productivity and lipid content for microalgae. This model excludes co-products, as there is currently no successful commercial operation that combines biofuels and co-products. The price of co-products is also highly variable and was therefore excluded from this model. The equations are based on Sen et al. (2012):

$$\mathrm{MESP\,(or\ MFSP)} = \frac{\mathrm{OC+ROI+IT}}{\mathrm{P\times Y\times A\times C}} \tag{1}$$

$$\mathrm{ROI} = \frac{\mathrm{DR\times(1+DR)^{ELS}}}{\mathrm{(1+DR)^{ELS}-1}}\times\mathrm{TPI} \tag{2}$$

$$\mathrm{IT} = \mathrm{TR\times(BR-OC)} \tag{3}$$

where OC is operating costs, ROI is return on investment, IT is income tax, P is biomass productivity (t ha^{-1} year^{-1}), Y is lipid content (microalgae) or biomethane yield (macroalgae) (m^3 t^{-1}), A is cultivation area, C is conversion coefficient (80% for algal lipid to biodiesel; 40% for CH_4 to electricity), DR is discount rate (10%), ELS is equipment life span (20 years), TPI is total project investment, TR is tax rate (35%), and BR is biofuel revenue.

To make MESP equivalent to the current electricity price of \$0.1043 kWh^{-1}, the biomass productivity for macroalgae should be 73.5 t ha^{-1} year^{-1}, with a methane yield of 285 m^3 t^{-1} dry mass (Figure 6). Based on the previous studies (Figures 2 and 4), it seems possible for some species of *Ulva* to achieve this when cultivated in tanks. It is worth noting that this model is based on the cost of nearshore

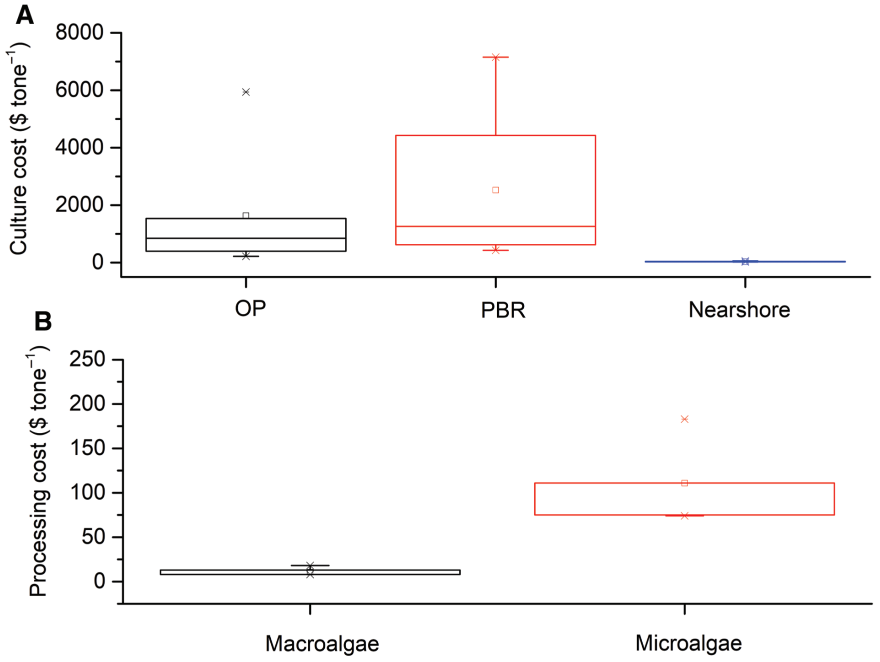

Figure 5: Box charts of culture (A) and processing costs (B) for biofuel production from macroalgae and microalgae in different culture systems.

OP, Open pond; PBR, photobioreactor. Data were based on Roesijadi et al. (2010b), Norsker et al. (2011), Richardson et al. (2012), Nagarajan et al. (2013), Davis et al. (2014), Dave et al. (2013), Hoffman et al. (2017), Soleymani and Rosentrater (2017). Please see Supplementary Table S4 for details.

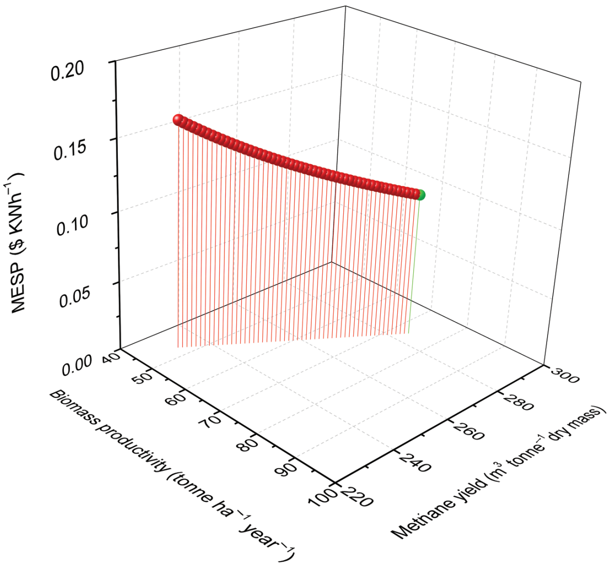

Figure 6: Effects of biomass productivity and methane yield of macroalgae on minimum electricity selling price (MESP).

Green circle indicates current electricity price. MESP calculation was based on data from Dave et al. (2013) using equation from Sen et al. (2012) with assumptions that discount rate is 10%, equipment life span is 20 years and tax rate is 35%.

systems. Until now, the highest biomass productivity in nearshore systems is 62 t ha^{-1} year^{-1} for *Macrocystis pyrifera* (Linnaeus) C. Agardh (Supplementary Table S2). Therefore, biomass productivity in nearshore systems needs to be improved to make MESP equal to the current electricity price.

The MFSP for microalgae cultured in photobioreactors (\$8.36 l^{-1}) is much higher than in open ponds (\$3.88 l^{-1}). To compete with diesel from fossil fuel (currently \$0.82 l^{-1}), the biomass productivity and lipid content for microalgae cultured in open ponds must be more than 61.8 g DM m^{-2} day^{-1} and 71% (Figure 7A). From the previous studies (Figures 1 and 2), this seems very difficult for microalgae cultured in open ponds. For microalgae cultured in photobioreactors to compete with diesel from fossil fuel, even higher biomass productivity (232 g DM m^{-2} d^{-1}) and lipid content (80.2%) are required (Figure 7B). Crucially, this is well above the theoretical maximum microalgal productivity of 196 g m^{-2} day^{-1} (Weyer et al. 2010). The target lipid content of 80.2% also seems currently impossible.

To optimize culture conditions for microalgae so that both high biomass yield and high lipid yield are achieved is always a challenge because the optimal culture conditions for cell growth and lipid production are not the same. Accordingly, a two-stage culture method is proposed to resolve this problem, in which growth and lipid production are split into separate phases. This culture system has been proven successful in *Nannochloropsis oculata* (Droop) D. J. Hibberd (Aléman-Nava et al. 2017), *Chlorella* sp. (Nayak

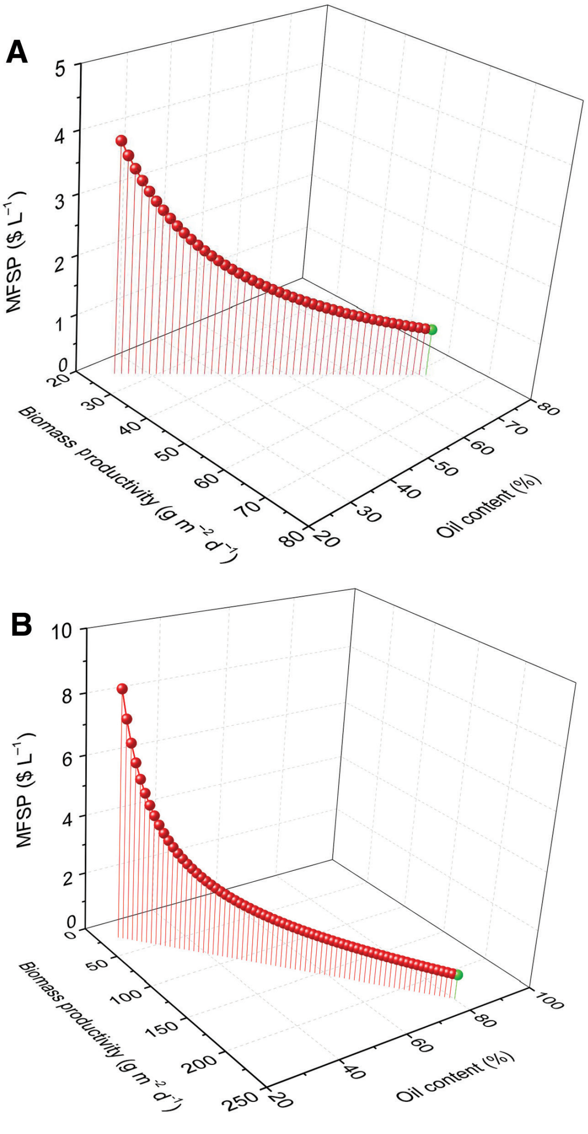

Figure 7: Effects of biomass productivity and lipid content of microalgae cultivated in open pond (A) and photobioreactor (B) on minimum fuel selling price (MFSP).
Green circle indicates current diesel price from fossil fuel. MFSP calculation based on data from Davis et al. (2011) using equation from Sen et al. (2012) with assumptions that discount rate is 10%, equipment life span is 20 years and tax rate is 35%.

et al. 2019) and *Skeletonema costatum* (Greville) Cleve (Gao et al. 2019). Currently, the culture cost is the main obstacle for biodiesel from microalgae to achieve a positive energy balance. Because of the difficulty of near shore or off shore cultivation, it would be difficult to reduce the farming costs for microalgae. Although genetic engineering approaches have been used to improve lipid content in microalgae without significantly affecting biomass yield, commercially producing biodiesel from microalgae remains difficult without subsidies (Sheridan 2013, Ajjawi et al. 2017, Posewitz 2017, Remmers et al. 2018). In addition, de Boer et al. (2012) proposed several pathways to reduce the cost of biofuel from microalgae, while more research is still required to increase the efficiency of these pathways and to apply them at commercial scale. It seems that commercially viable biofuel production from microalgae can only be possible if the other (more valuable) constituents of the algal biomass are exploited as co-products (Borowitzka 2013, Foteinis et al. 2018).

Conclusion

Compared to microalgae, the biomass productivity of macroalgae is lower. However, the lower cultivation and processing costs make the production of biomethane from macroalgae very close to profitability. The main reason for the lower annual biomass productivity of most macroalgae is their periodic reproduction (Wei et al. 2013, Gao et al. 2017c). This is particularly obvious in summer when most macroalgae have to survive via the form of microscopic propagules. Therefore, to obtain a stable and high biomass yield, it would be beneficial to obtain some species without reproduction to improve the biomass productivity (Gao et al. 2017c) and some strains which are tolerant of high temperatures. Compared to *Porphyra* and *Laminaria*, *Ulva* has higher tolerance to high environmental pressures, making it feasible for year-round cultivation (Carl et al. 2016, Gao et al. 2016a,b). In addition, the biomass of *Ulva* can be directly collected from green tides that commonly occur in eutrophic waters worldwide (Ye et al. 2011, Paumier et al. 2018), which can further reduce the cost of the biomass. For instance, around 20 million tons (fresh weight) of *Ulva* biomass were produced by the green tide occurring in the Yellow Sea of China in 2008 (Gao et al. 2010, Ye et al. 2011). In addition to green tides, another type of macroalgal bloom termed golden tides is also on the rise (Smetacek and Zingone 2013, Milledge and Harvey 2016, Xu et al. 2017). The coverage area of the golden tide in 2017 even exceeded that of most of the green tides in the Yellow Sea during past 10 years (Qi et al. 2017). Although there are some studies on extracting biofuels from *Sargassum* (Li et al. 2012, Borines et al. 2013, Soto et al. 2015), biofuel production from *Sargassum* is still in its infancy. Using bloom-forming macroalgae as biofuel should be given priority because of their high growth rate and direct availability from the field. In addition, to develop engineered microbial platforms is an effective approach to improve biofuel yield from macroalgae (Wargacki et al. 2012, Enquist-Newman et al. 2014, Camus et al. 2016).

The lower growth rate of macroalgae compared to microalgae means that larger areas are required to produce the same biomass. For instance, to meet the global natural gas demand (3848.9 bcm) in 2018 (Dudley 2019), an area of 315 million ha is needed to culture *Ulva lactuca* (Bruhn et al. 2011), which is equivalent to the whole ocean area of China. In addition, there is a possible future shortfall in phosphate supplies globally which would adversely affect the ability to fertilize mass cultures (Raven 2017, Gao et al. 2018b). Meanwhile, macroalgae are ideal materials for wastewater bioremediation because of their strong capacity to absorb nutrients and heavy metals (Gao et al. 2018c, Nardelli et al. 2019). To use bioremediating macroalgae for biofuel is thus a way to reduce the cultivation area and add ecological services. Therefore, integrated farming of macroalgae and other commercial marine animals needs further development as it can enhance the productivity of both macroalgae and animals (Pedra et al. 2017, Gao et al. 2018c, Laramore et al. 2018) as well as reducing eutrophication.

Acknowledgements: This study was supported by the National key R&D program of China (2016YFA0601400), the Lianyungang Innovative and Entrepreneurial Doctor Program (201702), the Jiangsu Planned Projects for Postdoctoral Research Funds (1701003A), the Postgraduate Research & Practice Innovation Program of Jiangsu Province (SJCX19_0951), and the Priority Academic Program Development of Jiangsu Higher Education Institutions of China.

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Supplementary Material: The online version of this article offers supplementary material (https://doi.org/10.1515/bot-2019-0065).

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Graphical abstract

Guang Gao, James Grant Burgess, Min Wu, Shujun Wang and Kunshan Gao
Using macroalgae as biofuel: current opportunities and challenges

https://doi.org/10.1515/bot-2019-0065
Botanica Marina 2020; x(x): xxx–xxx

Review: Compared to microalgae, the biomass productivity of macroalgae is lower. However, the lower cultivation and processing costs move the production of biomethane from macroalgae closer to profitability.

Keywords: biofuel; biogas; bioremediation; cultivation; macroalgae; photosynthesis.