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Life Cycle Environmental Impacts of Wastewater-Based Algal Biofuels

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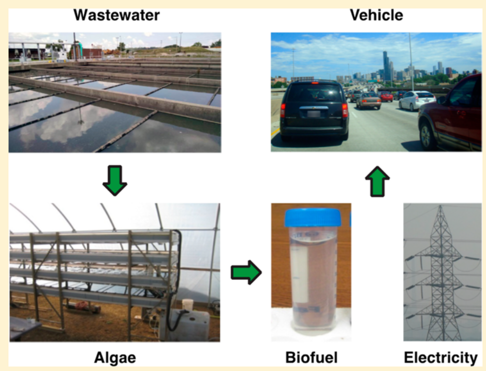
Supporting Information

ABSTRACT: Recent research has proposed integrating wastewater treatment with algae cultivation as a way of producing algal biofuels at a commercial scale more sustainably. This study evaluates the environmental performance of wastewater-based algal biofuels with a well-to-wheel life cycle assessment (LCA). Production pathways examined include different nutrient sources (municipal wastewater influent to the activated sludge process, centrate from the sludge drying process, swine manure, and freshwater with synthetic fertilizers) combined with emerging biomass conversion technologies (microwave pyrolysis, combustion, wet lipid extraction, and hydrothermal liquefaction). Results show that the environmental performance of wastewater-based algal biofuels is generally better than freshwater-based algal biofuels, but depends on the characteristics of the wastewater and the conversion technologies. Of 16 pathways compared, only the centrate cultivation with wet lipid extraction pathway and the centrate cultivation with combustion pathway have lower impacts than petroleum diesel in all environmental categories examined (fossil fuel use, greenhouse gas emissions, eutrophication potential, and consumptive water use). The potential for large-scale implementation of centrate-based algal biofuel, however, is limited by availability of centrate. Thus, it is unlikely that algal biofuels can provide a large-scale and environmentally preferable alternative to petroleum transportation fuels without considerable improvement in current production technologies. Additionally, the cobenefit of wastewater-based algal biofuel production as an alternate means of treating various wastewaters should be further explored.



INTRODUCTION

Algal biofuels have attracted considerable attention as a replacement for petroleum-derived fuels, but there is not yet consensus on their commercial scale production being environmentally beneficial.[1] Numerous studies have suggested using nutrient-laden wastewater in algae cultivation to improve the environmental performance of algal biofuels[2−6] because this reuses waste nitrogen (N), phosphorus (P), and water, which reduces the energy use and emissions from acquiring these inputs, and potentially reduces overall biofuel production costs. This also reduces nutrient loads in treatment facilities, which in turn decreases electricity consumption. Currently, algal cultivation has been examined with various wastewaters, such as municipal wastewater,[7,8] industrial wastewater,[9,10] and animal manures.[11,12]

Wastewater-based algal biofuel production still faces many challenges, however.[2,13] Wastewater composition varies by source, infrastructure, weather conditions, and pretreatment methods, which increases the difficulty and uncertainty in algal biomass cultivation.[2] Furthermore, the nutrient profile of wastewater from some sources may render it unsuitable for algae cultivation. For example, low nutrient levels or the presence of inhibitors can reduce nutrient assimilation and biomass productivity.[14] In addition, characteristics of algal biomass change with different wastewaters,[7] which leads to different biofuel yields, resource use, and environmental impacts in algal biomass conversion. Even when derived from the same wastewater, algal biofuels may vary in their environmental performance depending on which of any number of evolving conversion technologies is used.[15] Therefore, there is a continuing need to examine energy use, resource use, and emissions of wastewater-based algal biofuels to assess their benefit as alternatives to petroleum transportation fuels.

Several studies have used life cycle assessment (LCA) to characterize the environmental performance of wastewater-based algal biofuels. Clarens et al.[16] compared the environmental burden from biomass production between municipal wastewater-grown algae and terrestrial biomass including corn, switchgrass, and canola. They found environmental benefits when conventional fertilizers are replaced by wastewater in algae biomass cultivation. More recently, they compared wastewater algal biodiesel and bioelectricity production via lipid extraction, combustion, and digestion technologies.[17] Their results showed that producing bioelectricity by

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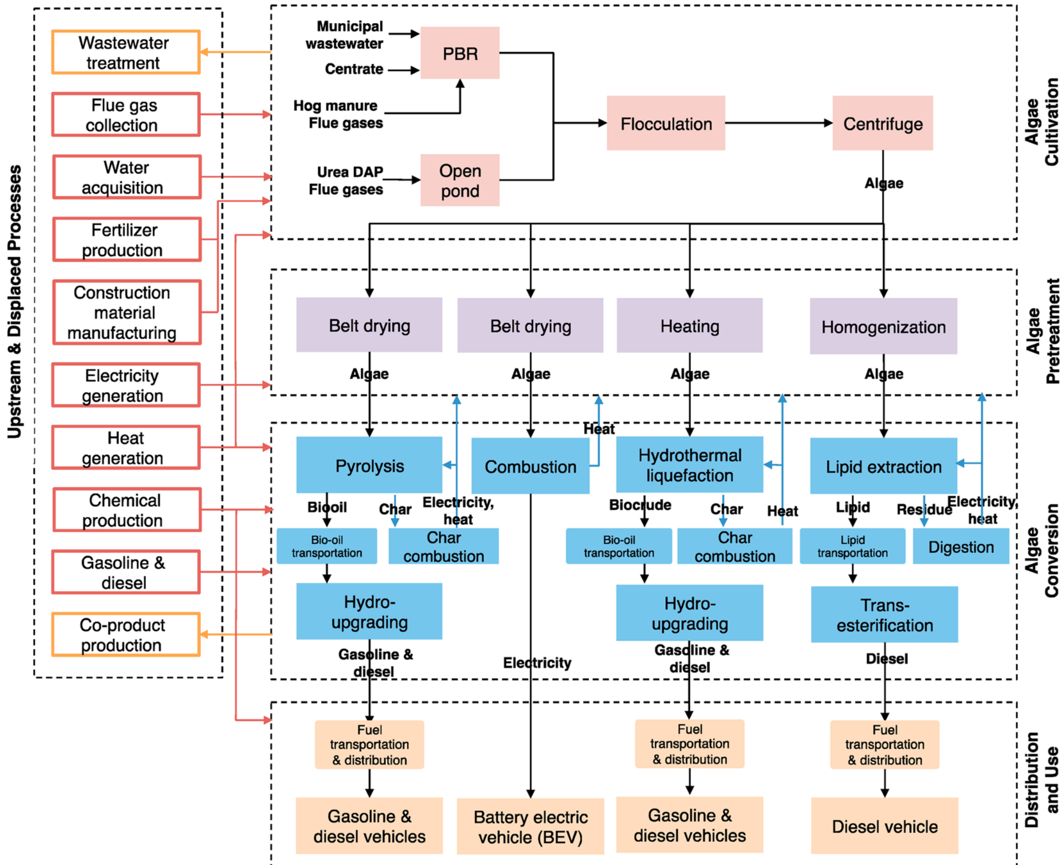

Figure 1. Systems considered and the system boundary for analysis. Included are four nutrients sources (municipal wastewater, centrate, swine manure, and freshwater with fertilizers) and four algae conversion technologies (microwave pyrolysis, direct combustion, hydrothermal liquefaction, and lipid extraction). Upstream processes include chemical and fertilizer production, construction material manufacturing, and electricity and heat generation. Displaced processes include coproducts production and wastewater treatment. The processes for energy recycling (char combustion and digestion) are built in the heat and power system (HPS) that is included in algae conversion systems.

combustion outperformed the other two pathways in energy use and greenhouse gas (GHG) emissions because it requires less upstream electricity and heat, and fewer chemical inputs. Similarly, Sander and Murthy[18] used LCA to examine using effluent of the activated sludge process in wastewater treatment plants. Well-to-pump results showed total energy inputs for algal biodiesel lower than that of petroleum diesel. Total CO_2 emissions were found to be higher with centrifuge drying but lower with filter drying.

Recommendations from these and other LCA studies vary, however, due to differing model scopes, coproduct allocation methods, functional unit definitions, assumptions about wastewater composition, production technologies, and algal growth parameters.[19] Additionally, as interest in algal biofuels has increased, new technologies and pathways have been proposed. For example, hydrothermal liquefaction (HTL)[20] and microwave pyrolysis[21] followed by hydro-upgrading is being explored for the conversion of wastewater-based algae to high quality bio-oil. Also, there is particular interest in improving productivity by feeding algae wastewater with high nutrient loads, such as centrate, which is the wastewater collected in the sludge drying process in a secondary wastewater treatment plant.[22]

This study focuses on the environmental performance of four wastewater streams combined with four promising technologies in the production of transportation energy derived from algae. Sixteen unique production pathways are modeled and compared to petroleum diesel for life cycle fossil fuel consumption, GHG emissions, eutrophication potential, and consumptive water use. This study expands the existing literature on algal biofuels by examining highly concentrated nutrient sources, centrate and swine manure, as well as algal biomass conversion by microwave pyrolysis. Data used in modeling algae cultivation in wastewater are based on pilot-scale experiments and facilities reported in existing literature, differing from many LCA studies that utilize lab-scale data or are based on stoichiometric calculations for nutrient removal. In doing so, this study presents a picture of the environmental impacts and trade-offs of wastewater-based algal biofuel production closer to commercial production.

■ METHODOLOGY

An LCA model was developed to compare production of transportation energy, both liquid and electric, from algae. Four nutrient sources, of which three are nutrient-laden wastewaters, and four conversion technologies are evaluated, as shown in Figure 1. A well-to-wheel analysis was conducted, which accounted for life cycle processes from extraction of raw resources to use of energy in vehicles. The functional unit is one vehicle-kilometer (km) traveled, to allow for fair comparison of different energy carriers. Process modeling is based on a review of current literature, technology reports, design manuals, LCA databases, and interviews with researchers at pilot-scale facilities. To capture uncertainty about production pathways, key design parameters are modeled with the best/likeliest/worst values. The best scenario represents the most favorable operating conditions and is calculated with a combination of the highest algae yields, the least energy demand, the lowest chemical use, and the highest biomass conversion efficiencies in a pathway. Model details and data sources are found in the Supporting Information (SI).

Overview of Production Processes. *Nutrient Sources.* Three wastewater streams, municipal wastewater (MW), centrate, and swine manure, are examined, covering both agricultural and municipal wastewater with a range of nutrient concentrations. The first stream, MW, is inflow to the activated sludge process in a secondary wastewater treatment plant.[7] MW is abundant in the United States, with approximately 120 million m^3 generated per day.[23,24] The second stream, centrate, is produced in smaller amounts. The large scale Metropolitan Wastewater Treatment Plant (MWTP) in Saint Paul, Minnesota, for example, which has a capacity of 700 million liters per day of MW, produces only 4 million liters per day of centrate.[22] However, centrate has higher concentrations, usually by an order of magnitude or more, of chemical oxygen demand (COD), suspended solid (SS), N, and P relative to MW. The third stream, hog manure, is another high concentration wastewater[25] generated at a rate of over 0.1 million m^3 in the United States per day.[26,27] A few previous studies[8,18,28] have modeled algal biofuels from the outflow of the activated sludge process. This stream is excluded here as scarcity of carbon leads to low algae productivity.[7,29] The synthetic fertilizers urea and diammonium phosphate (DAP) are the conventional nutrients for algae cultivation in freshwater[30] and are used here for comparison.

Algae Cultivation Systems. Algae cultivation facilities are modeled from pilot-scale facilities.[8,22,25,31] Wastewater-based cultivation facilities modeled in this study use a nontraditional photobioreactor (PBR) design[25] developed at the University of Minnesota. In each unit reactor, four layers of plastic troughs, similar to shallow open ponds, are stacked vertically (Supporting Information, Figure S1), which allows for reduced land footprint and production on sloped terrain.

Algae cultivated in wastewater grow with both organic carbon (C) and inorganic carbon (CO_2), and therefore need less sunlight than algae grown with only CO_2 in freshwater. The multilayer PBR design has been shown not to substantially influence algae yields when wastewater is present.[22] However, when growing algae with just CO_2, the yield of the multilayer PBR is lower than that of the conventional open pond with the same surface area, as the PBR sunlight reaches lower layers less than the surface layer. Therefore, the open pond with freshwater, synthetic fertilizers, and CO_2 injection is modeled as a baseline for comparison with wastewater cultivation in PBRs.

For centrate-PBR and MW-PBR systems, wastewater streams are fed directly into the PBR. In the manure-PBR, hog manure is diluted to avoid inhibiting algae growth from the high turbidity and ammonia concentrations.[25] Scrubbed flue gas collected from coal-fired power plants is injected into the hog manure-PBR to provide CO_2 to compensate for low concentrations of organic C due to the dilution. Inorganic CO_2 is not pumped into the centrate-PBR or MW PBR system as the organic C concentration is sufficient for a high algae yield. All PBR systems cultivate *Chlorella spp*. because of its high productivity and tolerance of high ammonia environments.[32] Studies show that the likeliest biomass yield for centrate (34.6 g m^{-2} d^{-1})[22] is higher than MW (15 g m^{-2} d^{-1})[7] and manure (24.6 g m^{-2} d^{-1}),[25] which is caused by the high nutrient concentration in centrate and a more favorable molecular ratio of C:N:P (100:12:12) for algae growth.[22] The neutral lipid content of wastewater-based algae (2−10%) is lower than algae grown with freshwater and synthetic fertilizers (14%) because the high N concentration and turbidity of wastewater inhibits neutral lipid production.[2,33] In addition, the neutral lipid content of wastewater-based algae in pilot-scale reactors is lower than the same algae cultivated in lab-scale reactors.[8]

The open-pond structure and operation is primarily based on the design of Beneman and Oswald.[34] Algae yield is based on estimates of Murphy and Allen,[31] who report 13.6 to 24.7 g m^{-2} d^{-1} across the United States. A yield of 13.7 g m^{-2} d^{-1} from Minnesota is used as the likeliest value to provide a fair comparison to PBRs operated there. Highest and lowest yields are used in the best and worst scenarios to reflect the possible range of impacts across the U.S. No specific algae species is assigned in the model of the open pond because a variety of species have been described in the literature. Algal characteristics are based on a review of eight species by Clarens et al.[17]

Flocculation and sedimentation processes, based on the design by Lardon et al.,[35] are used to separate algae from water in all cultivation systems. For the manure-fed PBR, supernatant collected after separation is recycled back to the cultivator. This reduces the volume of freshwater needed to dilute the manure stream, but results in nitrogen accumulation in the manure-PBR, which can be detrimental to algae growth.[25] To reduce the nitrogen concentration, more frequent tank washes are applied in the manure-PBR. Recycling supernatant is also applied in the open pond system to recycle nutrients and to reduce freshwater use. After flocculation, algae slurry is centrifuged to further remove water and increase solid content to 20 wt % in all cultivation pathways. Although centrifuging uses more electricity than other dewatering facilities, it has the advantages of being a commercialized technology, not contaminating the surrounding environment, and requiring less space.[36]

Algal Biomass Conversion Technologies. Four promising algal biomass conversion pathways are examined. Design parameters are based on literature,[17,20,21,37,38] technical reports from Argonne National Laboratory (ANL),[30] the National Renewable Energy Laboratory (NREL),[39] and the GREET, 2012 model.[40] The first pathway includes microwave pyrolysis followed by hydro-upgrading processes. Microwave pyrolysis offers several advantages over conventional pyrolysis: uniform internal heating of large biomass particles, ease of control, and no need for agitation or fluidization resulting in fewer particles in the bio-oil.[21] Biochar, syngas, and wastewater are coproduced with bio-oil in the pyrolysis process. Biochar is combusted in the heat and power system (HPS) powering the algae conversion system. Wastewater is assumed to discharge to local wastewater treatment facilities. Bio-oil is stored at the pyrolysis facility and then sent to a centralized biofuel refinery to be upgraded by hydrotreating and hydrocracking to produce gasoline and diesel.[39]

The second pathway is direct combustion of dried algae for electricity to power battery electricity vehicles (BEVs). In process modeling, net electricity is assumed to power BEVs after meeting internal plant demands. Belt drying[41] is the pretreatment option for the pyrolysis and combustion conversion processes as they require a solid content greater than 85%.[21] Heat generated during direct algae combustion or from the HPS is reused in the belt drying process. If this heat is insufficient, the drying process uses heat from natural gas.

The third pathway, hydrothermal liquefaction (HTL), has attracted interest as it does not need energy intensive water evaporation processing (drying), and can convert the nonlipid portions of algae into bio-oil.[42,43] The HTL pathway includes a pretreatment process that increases water temperature from 25 to 150 °C, and an HTL process operating at 300 °C and 2000 psig.[37] HTL is followed by an extraction process to separate bio-oil from water. Bio-oil is sent to the centralized biofuel refinery and upgraded by hydrotreating and hydrocracking processes.[39] Solid residue is combusted in the HPS to generate heat for in-plant use.

The fourth pathway, lipid extraction, uses technologies of lipid extraction followed by transesterification, which is a commercial-scale technology for biodiesel production.[17] Pressure homogenization pretreatment is used to break cell walls and facilitate lipid extraction.[17] The pathway is also called "wet lipid extraction" as no intensive drying process is applied. This frequently modeled technology is included to provide a comparison to the other three thermochemical conversion technologies.

Algal Bioenergy Distribution and Use. Conventional petroleum fuels are assumed to be used in transportation of bio-oil, wastes, and final products. In the biofuel use stage, three different passenger cars with different engines are modeled: gasoline engine vehicles, diesel engine vehicles, and BEVs. Efficiencies and manufacturing impacts of these three vehicles are based on GREET, 2012 model.[40] The GHG emissions of algal gasoline and algal diesel during vehicle operation are assumed to be equivalent to petroleum gasoline and diesel, respectively, on an energy basis. Biofuel distribution is based on report from ANL.[30]

Life Cycle Inventory (LCI) for Production Pathways. Sixteen pathways are examined, shown in Figure 2. The LCI for each includes direct emissions of processes and vehicles, upstream impacts of process inputs and materials for constructing the cultivation systems, and displaced processes of coproducts and wastewater treatment. Impacts from labor, capital, and machinery production are excluded. Environmental burdens of blowdown (that is, wastewater discharged during cleaning of cultivation reactors) are excluded from all cultivation systems owing to a lack of available data. Impacts are allocated between coproducts using the displacement method. Algae cultivation displaces treatment of sewage from residence in the secondary wastewater treatment plant. Displacement processes are counted as credits that reduce impacts of pathways. All electricity generated in HPS is utilized within the plant such that none is exported to the grid. Excess heat generated in HPS is not counted as a coproduct as no market currently exists. In GHG emissions calculations, CO_2 captured by algae in cultivation is counted as a credit when it is removed from the atmosphere and the flue gas, and then as an emission when returned to the atmosphere in use. Credits are also given for N and P removal by algae in wastewater when calculating eutrophication potentials. Impact factors are from the Ecoinvent 2.2 database[44] and the GREET model.[40]

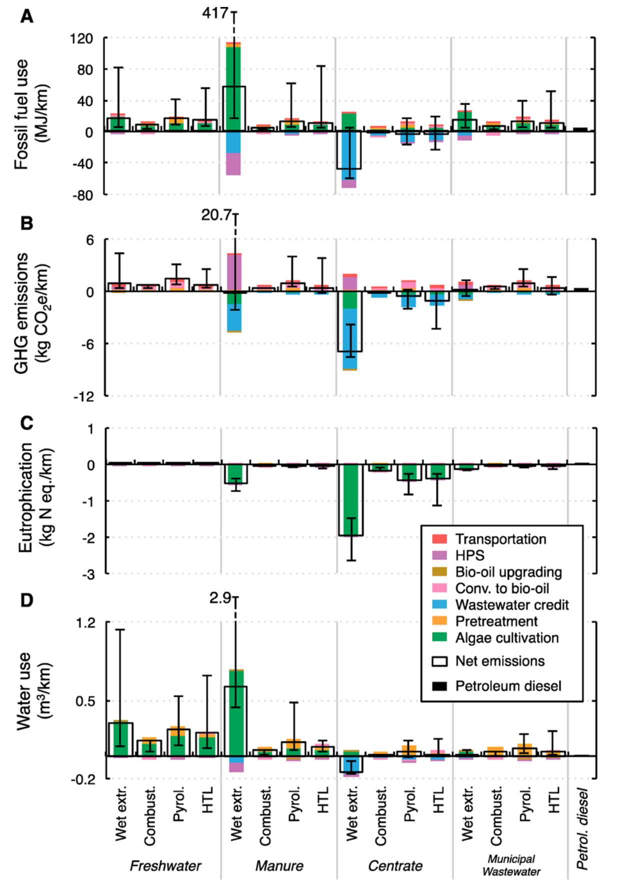

Figure 2. LCA results of 16 algal biofuel pathways and petroleum diesel. "Conversion to bio-oil", "bio-oil upgrading", and HPS are all in the algal biomass conversion system. "Conversion to bio-oil" refers to the pyrolysis process in the pyrolysis pathway, combustion process in the combustion pathway, the hydrothermal liquefaction process in the HTL pathway, and the lipid extraction process in the lipid extraction pathway. "Bio-oil upgrading" refers to hydrotreating and hydrocracking processes followed the pyrolysis and HTL process and the transesterification process followed the lipid extraction. Error bars show the best and worst case scenarios of pathways analyzed.

■ RESULTS AND DISCUSSION

Comparison of Various Production Pathways. The LCA results in Figure 2 show large differences in impacts among pathways. For example, fossil fuel use of the manure + wet lipid extraction pathway is over 10 times that of the manure + combustion pathway and over three times that of MW + wet lipid extraction. In addition, there is a large difference between the best and the worst scenarios in all pathways. For instance, in the manure + wet lipid extraction pathway, fossil fuel use of the worst scenario is over 20 times greater than the best scenario. These results demonstrate great uncertainties in the environmental performance of algal biofuels.

Algal biofuel from centrate can provide environmental benefits relative to conventional transportation fuels. Of the pathways examined, the centrate + wet lipid extraction pathway has the best performance in all impacts examined. This pathway has −47.7 (absolute negative) MJ km^{-1} fossil fuel use, −6.9 kg of CO_2 equiv km^{-1} GHG emissions, −0.1 m^3 km^{-1} water use, and −2.0 kg of N equiv km^{-1} eutrophication potential based on the likeliest case. Both centrate + wet lipid extraction and centrate + combustion pathways have better performance than petroleum diesel. Wastewater treatment credits play a major role in reducing impacts as growing algae removes N and P from wastewater, which reduces eutrophication. Another two centrate-based pathways perform better than diesel in all categories except water use. Higher water use is mainly from

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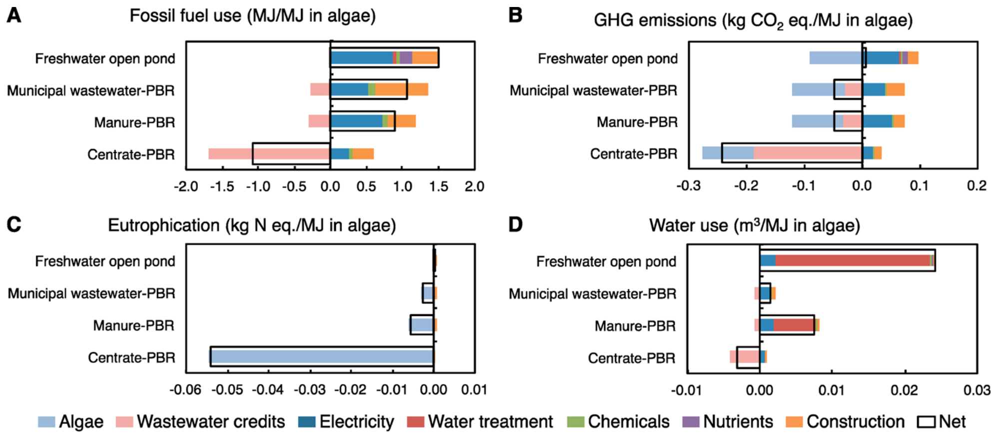

Figure 3. Comparison of the impacts of four algal cultivation systems with different nutrient sources. The normalizing unit is 1 MJ of energy produced in dry algae. Positive values indicate an increase in environmental impact. "Chemical" refers to polyacrylamide production in the flocculation process. "Nutrients" refers to urea and DAP production for water open pond. "Algae" refers to CO_2 captured and nutrients removed by algae. "Wastewater credits" are the wastewater treatment displacement credits. "Water treatment" refers to water treatment process producing clean water for process use.

upstream water use for heat generation in pretreatment processes.

Algal biofuels from MW and manure are likely beneficial only under the favorable conditions considered. Under the best performance scenarios, most freshwater-based algal biofuels have higher impacts than petroleum diesel in all categories; therefore, algal biofuels from freshwater are unlikely to offer environmental benefits given the technologies examined here. In the MW + combustion and the manure + combustion pathways, the best performance scenarios have lower impact than petroleum diesel, and the likeliest impacts have higher impact in categories examined.

While not presently able to perform better than petroleum diesel environmentally, pathways using wastewater for nutrients generally perform better than pathways using synthetic fertilizers. When identical conversion processes are used, centrate-based and MW-based algal biofuels perform better than biofuels from synthetic fertilizers. Manure-based algal biofuels generally perform better than those that are fertilizer-based if thermochemical conversion systems are used.

This study shows higher impacts than some previous studies of algal biofuels. For example, with the same wet lipid extraction pathway of freshwater-based algal biofuels, fossil fuel use of the likeliest case is about 10 times greater, and GHG emissions are about six times greater, than those reported from the GREET model.[30] This is mainly due to the lower algae yield (13.7 g m^{-2} d^{-1}) and lower lipid content (14 wt %) used in this study; GREET uses a 25.0 g m^{-2} d^{-1} yield with 25 wt % lipids. The low yield is partially attributable to the study location of Minnesota. However, even when reasonable yields in southern states are used, such as 24.7 g m^{-2} d^{-1} in Florida,[31] fossil fuel use and GHG emissions modeled here are still higher than those modeled in GREET. This is because this study calculates potential fossil fuel displacement based upon the content of neutral lipid, triacylglyceride (TAG), that can be actually converted to diesel in transesterification.[45] GREET uses total lipid content in its calculation, which overestimates production because the total lipids value includes lipids that cannot be transesterified to biodiesel.

Results of the freshwater + HTL pathway in this study are more than four times higher for fossil fuel use and GHG emissions than the study by Liu et al.[38] One reason is different upstream impacts related to electricity production. This study uses average United States mix electricity (10.3 MJ fossil fuel kW h^{-1} and 0.8 kg of CO_2 equiv kW h^{-1}) based on Ecoinvent 2.2, which is close to values in the GREET model (8.5 MJ fossil fuel kW h^{-1} and 0.68 kg of CO_2 equiv kW h^{-1}),[40] whereas the study by Liu et al. uses data from the Ecoinvent database embedded in SimaPro (2.50 MJ fossil fuel kW h^{-1} and 0.2 kg of CO_2 equiv kW h^{-1}), which is lower than average United States stack emissions calculated with eGRID (0.56 kg of CO_2 equiv kW h^{-1}).[46] In addition, as this study uses a different characterization of hydro-upgrading, GHG emissions (0.26 CO_2 equiv $liter^{-1}$) of this stage are higher than in Liu's study (0.16 CO_2 equiv $liter^{-1}$).

Impacts of MW-based biofuels in this study are 4−8 times higher than those from previous studies by Clarens et al.[17] This is mainly caused by this study's lower yield (15 g m^{-2} d^{-1}) and higher electricity use for pumping and mixing (72 774 MJ ha yr^{-1}) than the study by Clarens et al. (27.9 g m^{-2} d^{-1}; 41 989 MJ ha yr^{-1}). Fossil fuel use of MW-based biofuels here is also higher than that of Sander and Murthy,[18] where the lipid content applied is 30 wt % and construction impacts are not counted.

When examining impacts by production stages, algae cultivation is the most energy intensive in all pathways compared, consuming over 50% of total life cycle fossil fuels. Second is pretreatment, which consumes 10−50% of fossil fuels. Together, these two stages typically contribute over 90% of the fossil fuel burden. Biomass pretreatment and conversion systems are major contributors to GHG emissions, accounting for 70−90% of the emissions in algal biofuel production. For water use, almost all pathways have higher water use than petroleum diesel except the centrate + wet lipid extraction and the centrate + combustion pathways. This is due to direct water use in algae cultivation and upstream water use for heat generation in pretreatment. For eutrophication, all wastewater-based pathways have negative eutrophication potentials because of nutrient removal from wastewater in algae cultivation. In general, our analysis shows that the environmental burdens of algal biofuel systems depend largely on algae cultivation and pretreatment technologies, in agreement with previous studies.[17,18,30,47]

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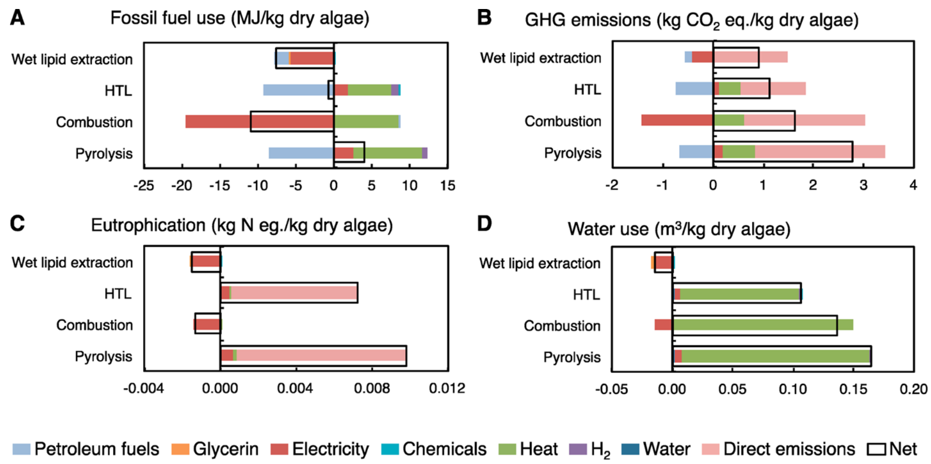

Figure 4. Comparison of impacts of four algal biomass conversion systems. The normalizing unit is 1 kg of dry algae treated. Algae are from the centrate PBR system with 20 wt % solid content. “Chemicals” refers to all chemicals used in lipid extraction and transesterification. “Petroleum fuels” include energy used in bio-oil and waste transportation. “Water” is the water-treatment process producing clean water for process use. The products (algal biofuels or bioelectricity) produced in the systems are treated as displacing petroleum diesel production or electricity production; thus, they are integrated into the “Petroleum fuels” or “Electricity” categories.

Algae Cultivation Systems. Relationships among inputs, energy productivity, and environmental impacts are examined for four cultivation systems, with the likeliest scenario shown in Figure 3. Burdens from fossil fuel use and GHG emissions are mainly from electricity used for pumping, mixing, and centrifuging (40−70% of total impacts), and from construction materials (20−50%). Water use is mainly (>80%) caused by direct consumption of freshwater for dilution and tank cleaning. Fertilizer production is not a major contributor to environmental burden (<11%) in the freshwater open pond. Environmental benefits arise from displacement of wastewater treatment, absorption of CO_2, and removal N and P from wastewater. Wastewater treatment displacement is a major environmental benefit ranging from 40−100% of total benefits in wastewater-based systems.

The performance of algae cultivation depends on wastewater characteristics. With a favorable nutrient profile, centrate-PBR has the highest energy productivity (0.7 MJ m^{-2} d^{-1}) compared to MW-PBR (0.3 MJ m^{-2} d^{-1}), manure-PBR (0.5 MJ m^{-2} d^{-1}), and freshwater open pond (0.3 MJ m^{-2} d^{-1}). Meanwhile, centrate-PBR has the lowest electricity input and highest wastewater treatment displacement credits per kilogram of dry algae produced. As a result, centrate-PBR has the lowest impacts in all categories. Swine manure has high nutrient concentrations, favorable for high yields, but operates with flue gas injection and supernatant recycling because of high turbidity and ammonia concentrations, which increases electricity use. Also, manure dilution and frequent tank cleaning and washing increase freshwater use. Therefore, the manure system has higher impacts than centrate cultivation systems. The MW system has low nutrient concentrations, which leads to low energy productivity; thus, it has the highest impacts of all wastewater-based cultivation systems.

Based on this analysis, algae cultivation performance could be improved by increasing algae productivity and reducing electricity use. Productivity depends on biomass yield and lipid/heat content. Ideally, increased yields and lipid content/heating value (LHV) would lead to production of more biofuel and thus reduce life cycle environmental burdens. However, for algae, lipid/heat content and yield tend to be inversely related; that is, algae with higher lipid/heat content normally have lower yields. Improving both lipid content and yields tends to require more inputs (for example, more electricity for artificial light and more intensive mixing, or more nutrients or carbon sources), which increases environment burdens. Therefore, algae productivity and electricity use are often a trade-off in algae cultivation, which complicates improvement in environmental performance.

Algal Biomass Conversion Technologies. Figure 4 identifies environmental burdens and benefits of four conversion pathways for converting one kilogram of centrate-based algae. Results show that environmental burdens of fossil fuel use are mainly (>80%) from heat generation in pretreatment. GHG emissions are mainly from direct emissions from conversion processes (70−100%). Water use is for heat generation in pretreatment, and eutrophication is from wastewater generation in the pyrolysis and HTL processes. Impacts of H_2 in pyrolysis and HTL, and of chemicals in lipid extraction are not major contributors. Environmental benefits arise from production of algal biofuels and coproducts such as electricity and heat. Generally, the environmental performance of algal biomass conversion could be improved by increasing conversion efficiency to obtain higher fuel yields, reducing electricity and heat use in pretreatment, and recycling energy in wastes.

Of the conversion pathways considered, wet lipid extraction produces less transportation energy (1.5 MJ diesel kg^{-1} centrate-based algae, equivalent to 0.6 km travel distance) than the other three thermochemical conversion technologies because the neutral lipid content in wastewater-based algae is low and thermochemical conversion can convert nonlipid carbon in algae into bio-oil. However, wet lipid extraction has better environmental performance than thermochemical conversion because it uses the lowest amount of heat in pretreatment and more energy is recycled from residue as electricity and heat in HPS. In addition, in wet lipid extraction, a portion of the carbon in algae remains in the digestate in the digestion process and is disposed of in a landfill, which results in lower GHG emissions. Of all three thermochemical conversion pathways, combustion produces the highest amount of transportation energy (3.7 MJ electricity kg^{-1} centrate-based algae, equivalent to 5.3 km travel distance) and performs best on fossil fuel use and eutrophication. The HTL process produces less fuel (3.1 MJ diesel + gasoline kg^{-1} centrate-based algae, equivalent to 3.1 km travel distance) than combustion but performs best on GHG emissions and water use. Pyrolysis produces almost the same amount of fuel (2.8 MJ diesel + gasoline kg^{-1} as centrate-based algae, equivalent to 2.8 km travel distance) as HTL, but has the highest impact in all categories since pyrolysis requires grid electricity for microwave generation and needs more heat in the pretreatment process. Both pyrolysis and HTL processes discharge wastewater, which leads to high impacts in eutrophication.

When examining life cycle impacts in terms of final transportation energy, the preference of conversion technologies changes with different nutrients sources, as shown in Figure 2. Wet lipid extraction is still the best conversion technology for centrate-based algae. With MW-based algae, combustion has the best performance in fossil fuel use, whereas wet lipid extraction is the best in GHG emissions, eutrophication, and water use. With manure-based algae, combustion has the best performance in fossil fuel use and water use, and HTL has the best in GHG emissions. The manure + wet lipid extraction pathway has extremely high impacts as the low lipid content of the algal biomass (2 wt %) results in impacts of cultivation being allocated to a comparatively small amount of final product. Finally, the freshwater-based algae followed by combustion pathway has the best performance in fossil fuel use and water use, and freshwater-based algae followed by HTL has the best in GHG emissions. This is similar to what was found with manure-based algae pathways. This study suggests the importance of considering conversion technologies carefully to maximize environmental benefits from the production of algal biofuels.

As all conversion pathways examined are not yet at commercial scale, there are still uncertainties in modeling these technologies. For example, few studies examine wastewater-based algae with the HTL process at the pilot scale. Our study modeled HTL pathways from a lab-scale study[20] combined with studies of freshwater-based algae.[37,38,42,43] In addition, although hydrotreating and hydrocracking are commercialized technologies for petroleum, they have not been well studied for algal bio-oil. Each existing study[37,38] uses different assumptions in establishing a hydro-upgrading LCI. To establish a more accurate LCI of algal biofuels in future work, conversion technologies should be examined at pilot or industrial scales, and be specific to wastewater-based algae.

Application of Wastewater-Based Biofuels. This study shows that algal biofuels produced from centrate can have lower environmental impacts than petroleum diesel. However, the amount of centrate available in wastewater treatment plants cannot support large-scale production. For example, the centrate generated in the MWTP could produce only 400 L per day of algal biodiesel with wet lipid extraction or export 13 000 kW h electricity with algae direct combustion under the best growing and operating conditions. If this level of energy productivity from centrate is applied to wasterwater treatment plants across the U.S., maximum petroleum diesel displacement is only 1.05 million L per day, or 0.17% of the United States petroleum diesel consumption.[48] MW is abundant in the United States but MW-based algal biofuels are environmentally better than petroleum diesel only under the most favorable production scenario in the combustion pathway. Even if all MW-based facilities are assumed to operate under conditions presented in the best scenario, and issues related to siting production in urban areas[49] were resolved, the petroleum diesel displaced by using BEVs is only 64.6 million L per day, or 10.5% of United States' petroleum diesel use. Similarly, animal manure is shown to produce biofuels with environmental benefits over petroleum diesel in the best scenario of the combustion pathway. Maximum petroleum diesel displacement is around 2.12 million L per day, or 0.34% of the United States' petroleum diesel use. Therefore, wastewater-based algal biofuels are unlikely to be a viable large-scale replacement for fossil transportation fuels regardless of their environmental impact.

This study also suggests that development of wastewater-based algal biofuels could prove beneficial in reducing environmental impacts of wastewater treatment. In conventional municipal wastewater treatment, removing 1 kg of COD requires 21.2 MJ of fossil energy[44] with most of that being consumed for aeration during the activated sludge process.[50] In contrast, removing 1 kg of COD in a centrate-PBR requires only 9.2 MJ of fossil energy. In a conventional municipal wastewater treatment plant, centrate is sent back to the activated sludge process, which increases COD and nutrients loading, and significantly increases the energy needed for removal.[22] If centrate is fed to algae before being sent back to the activated sludge process, energy consumption in the activated sludge process can be reduced. In addition, algae can be used to generate bioenergy for in-plant use, which further reduces energy use in wastewater treatment facilities, and, with optimistic assumptions, could enable facilities to become net energy producers.[51] Using algae to remove nutrients in manure with algae may have similar benefits, especially as the spreading of manure as fertilizer on agricultural land has come under increased scrutiny because of potential water contamination by excessive nutrients, microbial pathogens, and pharmaceuticals.[52]

While some wastewater-based algal biofuels show considerable environmental benefits over freshwater-based algal biofuels, large-scale production will likely be restricted by the limited amount of wastewater available to support high production volumes and by production costs. However, wastewater-based algal biofuels may prove beneficial for cleaning various wastewaters, consequently reducing the environmental impacts of wastewater treatment, and potentially creating a new source of revenue to municipal wastewater plants and livestock producers. Future research should focus on optimizing cultivation systems to maximize nutrient removal, algae yield, and biomass density, while minimizing energy use for mixing and pumping. In addition, further effort should be put into wastewater-based algae conversion to improve energy productivity and into establishing more thorough environmental inventories.

■ ASSOCIATED CONTENT

Supporting Information

Model details. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

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