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## Algal Research



# Spatially Explicit Life Cycle Assessment: Opportunities and challenges of wastewater-based algal biofuels in the United States



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#### A R T I C L E I N F O

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#### ABSTRACT

This work presented a Spatially-Explicit-High-Resolution Life Cycle Assessment (SEHR-LCA) model for wastewater-based algal biofuel production, by integrating life cycle assessment, GIS analysis, and site-specific Wastewater Treatment Plants (WWTPs) data analysis. Wastewater resources, land availability, and meteorological variation were analyzed for algae cultivation. Three pathways, Microwave Pyrolysis, hydrothermal liquefaction, and lipid extraction were modeled for bio-oil conversion. This model enables the assessment of seasonal and site-specific variations in productivity and environmental impacts of wastewater-based algal bio-oil across the whole U.S. Model results indicate that wastewater-based algal bio-oil can provide an opportunity to increase national biofuel output. The potential production of algal bio-oil can reach to 0.98 billion gallon/yr, nearly 20% of advanced biofuel projection as outlined in the U.S. Energy Independence and Security Act (EISA) of 2007. LCA results shows significant variations among different locations, WWTPs, and operational seasons. Although not competitive to conventional fossil fuel in energy efficiency, wastewater-based algal biofuel could offer significant benefit in controlling GHG emissions. However, spatial analysis shows that only 61% of the total wastewater could be used, based on current land use efficiency for algae cultivation and land availability around each WWTP in a radius where algal biofuel production is energy positive (energy output > energy input). These results indicate that land availability could be a significant challenge for wastewater-based algal biofuels that have not been considered in previous studies. They also suggest that improvement should be made in technological development and system design to increase energy and land use efficiency for full potential of wastewater as a promising resource for algal biofuel production. Although focusing on the U.S. as the case study, the developed methodology could be used for spatially explicit analysis of algal biofuel integrated with wastewater on macro-scale in other regions as well.

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#### 1. Introduction

Algae have unique and desirable characteristics as a source for biofuel, including rapid growth and capability of growing in poor quality water, but there remain a number of challenges before the technology can be deployed at large-scale [1,2]. Key barriers that hinder the utilization of algae biofuels are high cost and limited capacity for scaled-up production of algal biofuel feedstock. Studies indicate that wastewater, currently underused, could be one of the most favorable resources for algae feedstock production, because it (1) provides ample supply of nutrients and water, (2) can support a large capacity for biofuel production (up to 5 billion gallons of algal biofuel per year could be generated with municipal wastewater in the U.S. [3], and (3) can be integrated into existing public infrastructure, rather than creating new industrial systems [4–7].

\* Corresponding author. *E-mail address:* zhangyl@wayne.edu (Y. Zhang). A number of studies have been conducted to investigate the potential of the synergies of algae biofuels and wastewater, from empirical selection of algal strains to pilot-scale algae cultivation systems and energy conversion pathways [8–11]. Despite such progress and promise, however, there has been no large-scale algae-wastewater facilities emerging yet. To better understand the potential performance of integrated algae-wastewater systems, life cycle assessment (LCA) has been applied to assess these integrated systems. LCA is a widely accepted quantitative accounting tool for evaluating the environmental effects of products, process, or services by computing the energy/material inputs and wastes released to the environment, and also assessing the potential environmental impacts of those energy, materials, and wastes [12]. LCA has become an actively researched area and has been increasingly applied in academic and industrial fields for environmental impact assessments.

Early stage of wastewater-algae LCA studies assessed the environmental performance of wastewater-based algal bioenergy system based on process modeling. Clarens et al. (2010) compared environmental impacts of bioenergy from algae and other territorial crops

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including canola, corn, or switchgrass [13]. They found significant environmental benefits of using wastewater in algae cultivation. Similarly, Davis et al. (2011) and Gallagher et al. (2011) reported that the environmental and economic performance of micro-algal biofuel production are unlikely to be competitive with traditional fossil fuel in the near term, without the replacement of energy-intensive commercial fertilizers [14,15]. In all of these studies, the authors pointed to the need for improved access to low cost, low energy-intensity nutrient (nitrogen and phosphorus) sources, such as wastewater resources, to improve the overall environmental and economic bottom lines. Later on, Mu et al. (2014) evaluated the environmental performance of wastewaterbased algal biofuels with a well-to-wheel LCA [16]. Their results indicated that the environmental performance of wastewater-based algal biofuels is generally better than freshwater-based algal biofuels. However, these LCA studies only focused on single site with generalized assumptions, without systematic consideration of geographic diversity, seasonal climate variation, and resource availability. This type of LCA, often referred to as a Static LCA using only peak or lumped data, is unsuited to assess national-scale potential and environmental performance of wastewater-based algal biofuel that depend on many factors including quality/quantity of wastewater, climate variations (solar radiation, temperature, and precipitation among others), and land availability.

More recently, a few limited studies applied Geographical Information Analysis (GIS) to analyze the potential production of algal bioenergy with wastewater [17,18]. However, these studies didn't include LCA in their analyses, and, as such, could not answer the question whether wastewater-algae system is truly environmental friendly at large-scale. Furthermore, the data resolution of these GIS analyses was relatively low: using regional data rather than site-to-site specific data. Finally, there were no co-siting analysis of algae facilities and wastewater infrastructure in these studies. This is a critical research gap, because facility siting is one of the most significant challenges faced by wastewater-based algae systems since wastewater treatment facilities tend to be near metropolitan areas with limited land availability, and it is not practical to transport wastewater over long distances [1].

To address these research gaps, the present work develops a High-Resolution-Spatially-Explicit Life Cycle Assessment (HRSE-LCA) model, by integrating LCA, GIS analysis, and site-to-site specific analysis of Wastewater Treatment Plants (WWTPs) and land availability. This model enables the evaluation of seasonal and site-to-site variations in production and environmental impacts of wastewater-based algal biofuels across the whole U.S. This study is conducted with two specific objectives: (1) assess the realistic potential in production of algal biofuels with municipal wastewater resources across the whole U.S., using site-to-site specific GIS-based analyses of resource availability and algae growth model; and (2) evaluate seasonal and geographic variations in environmental impacts of wastewater-based algal biofuels, by integrating GIS-based algae growth model and life cycle assessment. We focus on municipal wastewater because it is the most studied wastewater resources for algal biofuel production, as well as its available data source [6,17–19]. This work extends the literature by integrating geographic diversity, seasonal climate variation, and resource availability into large-scale life cycle assessment of wastewater-based algal biofuels. Although focusing on the U.S. as the case study, the developed methodology could be used for spatially explicit analysis of algal biofuel integrated with wastewater on macro-scale in any other regions as well

#### 2. Material and methods

The purpose of this study is to evaluate the realistic potential and seasonal/site-to-site variations in production and their implications for environmental performance of wastewater-based algal bio-oil across the whole U.S., based on a HRSE-LCA framework (Fig. 1). The national-scale potential production and environmental performance of wastewater-based biofuels depend on many factors including wastewater resources, climate variations (seasonal and spatial variations), and land availability. To account for these variations, the HRSE-LCA model is composed of four modules, including high-resolution GIS-based spatial resource assessment (Module 1), spatially explicit algae growth model (Module 2), biofuels conversion pathways (Module 3), and LCA (Module 4). Fig. 1 depicts how these four modules are incorporated together and what are the overall processes flows for the modelled system. Specifically, Module 1 (high-resolution GIS-based spatial resource assessment) estimates wastewater resource, nutrient profile, and land availability based on each individual municipal WWTPs across the whole U.S. Module 2 (spatially explicit algae growth model) predicts spatial and seasonal algal biomass production and material/energy input/output by incorporating the results of Module 1 (resource analysis) and spatial/seasonal variations of meteorological data into the algae growth model. Module 3 assesses biofuels production and material/ energy input/output for three biomass-to-bio-oil conversion pathways. Based on the results of Module 1–3, Module 4 performs life cycle impact analysis by calculating environmental burdens associated with process operation and upstream input. GIS information (such as temperature, land coverage, and solar radiation among others) were obtained from PRISM, USGS, and NREL, respectively [20-22]. All modules were built in Microsoft Excel, using Crystal Ball Commercial suite for characterization of input and output uncertainty. The following sections briefly discusses the methodology for each Module. Details are provided in the Supporting Information (SI).

#### 2.1. High resolution GIS-based spatial resource assessment (Module 1)

#### 2.1.1. Municipal wastewater

Spatial wastewater resource data for each individual WWTP, including capacity and population served, was extracted from the Clean Watersheds Needs Survey [19] by using "Exist Total Flow" (wastewater generated by population plus infiltration). Data shows that there are around 17,000 WWTPs for the whole U.S., and the yearly flow rate is roughly about 34,200 million gallon/day ( $1.3 \times 10^8 \text{ m}^3$ /day). By filtering out WWTPs with very small capacity (<0.05 MG/D), 12,452 WWTPs with a total capacity of 33,576 MG/D, accounting for 99.7% of total wastewater flow, were included in this analysis. Primary or secondary wastewater effluent were chosen for algae cultivation, as previous studies suggest that solid material contained in wastewater prior to primary clarifier could damage pumps and reduce their operation life [5,23,24]. The nutrient profile (nitrogen, phosphorous, and COD) of wastewater was determined by literature [25,26].

#### 2.1.2. Land availability

National Land Cover Database (NLCD 2011) map, published by USGS, was used for land availability analysis and site selection around each individual WWTP of a total 12,452 WWTPs across the U.S. [21]. Suitable land for algae cultivation is non-agricultural, undeveloped or low-density developed, and non-environmentally sensitive, including grassland/herbaceous, shrub/scrub, and barren land [5,27]. Analysis was performed by considering the land availability in 1, 2.5, 5, 7.5, and 10 km radius distance from each WWTPs. This method has been applied in the study of land availability in Kansas up to 2.5 km ([28]. In this analysis, we extended the radius up to 10 km to analyze land availability for the whole WWTPs around the US. To avoid land overlapping around different WWTPs, Thiessen Polygon method from ArcGIS toolbox was used. This study did not include CO<sub>2</sub> constraint in site selection, as previous LCA studies conclude that CO<sub>2</sub> supply plays a negligible role for wastewater-based algae cultivation [16,18]. However, CO<sub>2</sub> supply could affect site selection, if large amount of CO<sub>2</sub> supply was necessary.

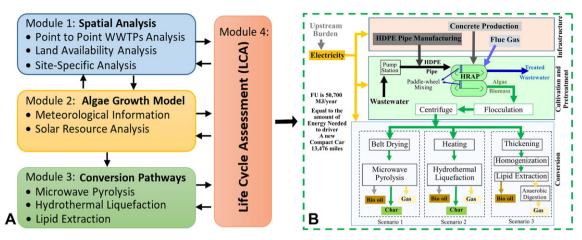


Fig. 1. Module and process flow diagram for HRSE-LCA model. A is the four modules contained in the HRSE-LCA model, and B is the system boundaries and processes for life cycle assessment.

#### 2.2. GIS-based spatial explicit algae growth model (Module 2)

Because of easy operation and low cost, open pond system (OPs) is currently the most promising system for algal biomass production at large scale [29]. Previous studies have reported that the productivity of algae dry biomass ranges from 0.12 to 0.48 g·L<sup>-1</sup>·d<sup>-1</sup>, or 8 to 20 g·m<sup>-2</sup>·d<sup>-1</sup> [24,30]. Likewise, algal oil yield varies from 2.3 to 25 m<sup>3</sup>·ha<sup>-1</sup>·yr<sup>-1</sup> [31]. Among different OPs cultivation strategies, High-Rate Algal Ponds (HRAP) is the most studied system with relatively low environmental impact [32–34]. *Chlorella* sp. is the predominant phytoplankton in HRAPs and WWTP clarifiers [35], and also one of the most studied algae species for biofuel production [36,37]. Therefore, *Chlorella* sp. in HRAP was chosen as the algae cultivation system in this study. Modeling parameter is presented on table S3 of the supporting information (SI).

GIS-based spatial explicit algae growth model was developed based on our previous work [13,38]. Specifically, algal biomass production, water/nutrient demand, material input/output, and energy consumption were computed by site-specific meteorological information (solar radiation, temperature, precipitation, and evaporation) incorporated into a mass and energy balanced algal open pond model [13,38,39], including available wastewater and land resources from the module 1. Algae cultivation was assumed to occur in those months when average monthly temperature are >10 °C [40]. Site-specific biomass yield had a strong effect on land analysis and was calculated based on the formula as a function of solar radiation (Photosynthetically Active Radiation or PAR), temperature, and conversion efficiency [41,42]. Specifically, solar radiation was the average value over surface cells of 10 km in size, and data was extracted from the model developed by Dr. Richard Perez and collaborators at the National Renewable Energy Laboratory and other universities for the U.S. Department of Energy. Temperature variations were obtained based on PRISM Climate data that is a 30 years Normal Mean Temperature database. Model outputs were calculated on monthly basis in operational periods when temperature is above 10 °C. More information about GIS data is available on Section 2.2 and 2.3 of SI.

#### 2.3. Biomass harvest and bio-oil conversion model (Module 3)

Mass and energy balance methods were used to develop the biomass harvest and bio-oil conversion model. Processing and modeling parameters were determined based on previous studies [13,16,43–45]. Three conversion pathways were examined for bio-oil production: lipid extraction, microwave pyrolysis, and hydrothermal liquefaction. Lipid extraction (LE) is the most studied conversion pathway, consisting of algal lipid extraction and anaerobic digestion of residual non-lipid biomass for nutrient recycling and by-products generation (bioelectricity and fertilizer) [13,46]. The LE technology is mature, but its energy yield is relative low because lipid is the only energy carrier. Microwave Pyrolysis (MP) uses uniform internal heating of large biomass particles to generate bio-oil, combustible biogas, and biochar. This process does not require agitation or fluidization, and, as such, the bio-oil contains less particles (ashes) [44]. The main disadvantage of MP is the necessity for removing nitrogen and oxygen from crude oil which needs more energy [44]. Hydrothermal liquefaction (HTL) has gained increasing interests as it is more energy attractive. The main advantages of HTL are that it can convert non-lipid compounds to bio-oil and does not requires energy intensive processing such as drying [16]. However, the complexity of the conversion mechanisms, as well as the difficulty of maintaining constant property of biomass feedstock, makes it hard to improve conversion efficiency for higher bio-oil yield [16,47]. Detailed information regarding energy requirement for each conversion pathway are presented in Section 4.4 of SI.

#### 2.4. Life cycle assessment (Module 4)

Results from Modules 1–3 were used for LCA to account for two types of seasonal and site-specific environmental impacts: energy use and greenhouse gas emission. The functional unit (FU) was defined as 50,700 MJ/yr, the average energy embodied in gasoline required for driving a compact car by an American for a year (13,476 miles driving per year) [48,49]. System boundaries were "cradle-to-gate" (Fig. 1B), encompassing all processes associated with algal bio-oil production with wastewater including pond instruction, algae cultivation, bio-oil conversion, by-product generation, and extraction of raw resources for production of required energy/material inputs. The Environmental burdens associated with infrastructure and equipment were calculated by multiplying required material inputs and their corresponding impact factor obtained from the Ecoinvent database [50]. These burdens were divided by the assumed project life time (30 years) for direct comparison with annual impacts arising from operations. All facilities associated with WWTPs were excluded from analysis, because they would already be in place at all WWTPs. However, environmental impacts associated with nitrogen and phosphorous removal by algae were considered as credits, as algae cultivation replaced the corresponding N and P treatment from WWTPs.

#### 3. Results and discussion

#### 3.1. Resource availability

#### 3.1.1. Wastewater resource

As shown in Fig. 2, there are total 12,452 municipal WWTPs with a capacity of 0.05 MG/D (190  $m^3$ /day) and above across the U.S.,

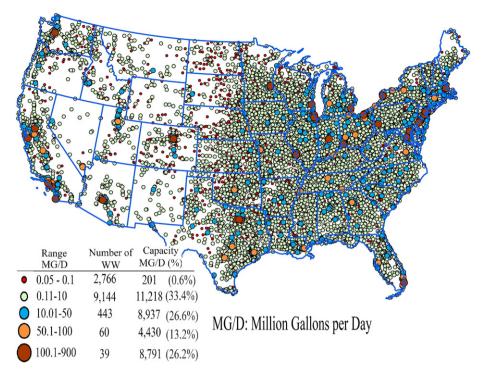


Fig. 2. WWTPs and their corresponding treatment capacity (wastewater flow) across the whole U.S.

accounting for 99.7% of total wastewater flow. Most WWTPs, 73% of the total WWTPs, have the capacity of 0.1–10 MG/D, accounting for 33.4% of the total wastewater flow, followed by WWTPs of 10–50 MG/D (27% of total wastewater flow), >100 MG/D (26% of total wastewater flow), 50–100 MG/D (13% of total wastewater flow), and 0.05–0.1 MG/D (0.6% of total wastewater flow). The majority of WWTPs locate in middle-to-east and west coast of the US, in accordance with population distribution. Large metro areas, such as Detroit, Chicago, Los Angeles, usually have WWTPs with large capacity (>100 MG/D), which indicates the popularity of centralized wastewater infrastructures.

#### 3.1.2. Land availability

A high-resolution analysis of land resource around each WWTP was conducted to assess the land availability in 1, 2.5, 5, 7.5, and 10 km radius, respectively. The required land of algae open-pond for each WWTP was determined by pond depth, evaporation, infrastructure land usage (pump station etc.), and pond hydraulic retention time. The land analysis was first performed for 1 km radius around the WWTP. If no enough land available, then 2.5 km radius was analyzed, followed by 5, 7.5, and 10 km radius, respectively. Our Analysis results shows that algae facility located in further than 10 km of the WWTP is not likely to be energy favorable due to the increasing amount of energy required for wastewater pumping. Therefore, land resource in 10 km radius would be first considered for algae cultivation. For those WWTPs where land requirement can't be met in the range of 10 km, energy efficiency was used as the criteria for site selection. Specifically, wastewater would be pumped further for algae cultivation until energy return on investment (EROI), determined by LCA module, reached to 1.0. Results of land analyses show that only 8507 WWTPs, accounting for 16% of total wastewater flow, have the capacity to locate algae facility in 1 km radius (Table 1). These WWTPs usually serve small community/population with low wastewater capacity [51]. The number of WWTPs with available land in 2.5, 5, 7.5, and 10 km radius is 2401, 808, 197, and 58, respectively. In sum, 11,971 of the 12,452 WWTPs could co-site algae facilities in 10 km radius, accounting for 69% of total wastewater flow. Consequences of land availability on LCA are described in Section 3.3.

These results imply the importance of land resources for co-siting algae facilities when using municipal wastewater for algal biofuels.

This constraint has not been fully considered in previous LCA or GIS studies [31]. For example, Orfield et al. (2014) performed a GIS analysis to estimate algal bio-oil production potential through flue gas and wastewater co-utilization without land analysis. Chiu et al. (2013) analyzed water availability, wastewater resources, and suitable lands in the development of algal bio-oil [17,18]. However, they assumed all the wastewater effluent can be used for algae cultivation without considering the co-siting of algae and wastewater facilities.

Interestingly, for most WWTPs with small wastewater capacity, the land demanding for algae cultivation could be met within 1 km radius. The larger capacity the WWTP has, the less land demanding could be met. This raises the question of how to scale the facilities: centralization or decentralization? There have been much debates regarding this issue, both for bioenergy facilities and wastewater infrastructures. Some studies found that large-scale centralized facilities are more cost efficient, especially from economic perspective; others argued that decentralized facilities could have more environmental benefits [14,33,52]. The results of this study suggest that de-centralization could have greater potential for wastewater-based algae bioenergy systems, which aligns with the increasing interest of decentralized water infrastructures for wastewater reclamation [53,54]. However, further research is warranted to investigate to what extent the scale could be optimized for both environmental and economic benefits.

#### 3.2. Production potential of wastewater-based algal bio-oil

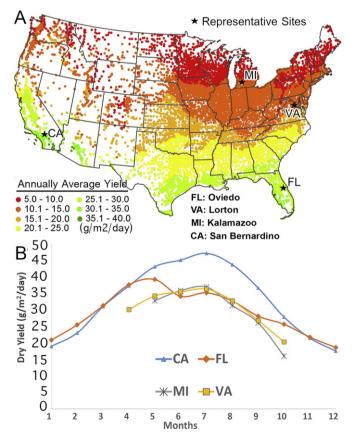
The annually average yield of algae biomass ranges from 8 g/m<sup>2</sup>/day for cold climate to 35 g/m<sup>2</sup>/day for warm climate (Fig. 3A). The results for four representative WWTPs in different climate are shown in Fig. 3B. These four WWTPs are located in San Bernardino (CA), Oviedo (FL), Kalamazoo (MI), and Lorton (VA). Two reasons contributed to the variations of biomass production from cold climate to warm climate. Frist, the colder the climate is, the less seasons are suitable for algae cultivation. For example, in Michigan, only half of the year (May to October) would be suitable for algae cultivation. In the contrast, algae cultivation could be operated in all seasons in warm climate, such as California and Florida. Second, even in suitable cultivation seasons,

Table 1		
Land availability	for WWTPs in	different radius.

Radius, km	Number of WWTPs with enough land	Capacity $(10^{+6} \text{ G/d})$	Percentage of total wastewater flow
0–1 km	8507	5250	16%
1–2.5 km	2401	6150	18%
2.5–5 km	808	5810	17%
5–7.5 km	197	2830	8%
7.5–10 km	58	850	3%
>10 km	481	12,692	38%

there would be higher biomass production in warm climate than that in cold climate due to higher solar radiation (Fig. 3B).

Bio-oil production was examined for three conversion pathways: LE, MP, and HTL (Table 2). The results indicate that HTL yields the highest productivity with a total energy output of  $1.75 \times 10^{11}$  MJ/yr (0.98 billion gallon/yr bio-oil, 1.8 million ton/yr biochar, 1.45 million ton/yr biogas), followed by MP with a total energy output of  $1.61 \times 10^{11}$  MJ/yr (0.77 billion gallon/yr bio-oil, 1.8 million ton/yr biochar, 2.44 million ton/yr biogas), and LE with a total energy output of  $1.15 \times 10^{11}$  MJ/yr (0.57 billion gallon/yr bio-oil, 0.74 million ton/yr biogas). This is because that HTL process can convert non-lipid compounds to bio-oil, and the maximum bio-crude yield could achieve 50-60% of the total biomass [16,47]. The maximum productivity via HTL conversion is 20% of advanced biofuel projection as outlined in the U.S. Energy Independence and Security Act (EISA) of 2007 [55]. Previous studies have reported varied estimations of algal-oil productivity with municipal wastewater, from 0.45 to 2.38 billion gallon/yr [17,18]. Our results tends to be in compliance with the lower estimation of Orfield et al. (2014) [18]. This could be attributed constraints applied in both analyses. Orfield et al. (2014) used competitive price, \$80/barrel, as the



**Fig. 3.** Variations in algal biomass productivity across the whole country. A: annually average yield. B: monthly average yield in four representative WWTP sites. Cultivation seasons are those months when average temperature is above 10 °C. Four stars represent four representative WWTPs.

selection criteria, while we applied land availability in this study [18]. This indicates that, although wastewater could be promising resources for algal oil production, various constraints could limit their utilization. It should be noticed that, for the LE process, the digestate from the anaerobic digestion is used as bio-fertilizer. Based on algae's stoichiometry, 45 kg of nitrogen and 4 kg of phosphorus per ton dry algal biomass is considered as fertilizer production [56]. The energy avoidance is assumed to be 29.9 MJ/kg N and 3.3 MJ/kg P [57], which is considered as energy offset and is counted toward the energy return on investment (EROI) as discussed in Section 3.3.1.

Spatial analysis suggest that California, Florida, and Texas represent the most productive locations, accounting for nearly 50% of the total bio-oil production (0.47 billion gallons/yr in these three states) (SI). These results are consistent with previous reports that, under current technologies for algae cultivation, southern regions have higher potential for algal biofuel production. Davis et al. (2014) and Venteris et al. (2013) suggested Gulf Coast and Florida peninsula as the two most suitable regions when considering productivity and freshwater availability [52,58]. However, with technological development, especially algae cultivation in cold climate, some northern regions could also emerge as promising locations considering their abundant water resources. For instance, Wigmosta et al. (2011) identified Great Lakes as one of the three most promising locations (Gulf Coast, southeastern seaboard, and Great Lakes) for algal biofuel production [59]. Additionally, other resources such as seawater and saline water could be used for algae cultivation as well. Venteries et al. 2013 estimated that 25 billion gallon per year (BGY) of algal biodiesel could be produced by using freshwater, saline groundwater and seawater in the United States, but the productivity of algal biodiesel from seawater and saline groundwater would be limited to approximately 2.0 BGY to be cost efficient [58].

#### 3.3. Environmental impacts

In this work, the developed HRSE-LCA model allowed variation of environmental impacts to be studied in more detail because environmental impacts can be calculated for each individual WWTP and every month, avoiding large area and long-time averaging. Energy use and greenhouse gas (GHG) emission were chosen as two environmental impact factors. Energy use is discussed in detail for seasonal and site-specific variation. For GHG emission, only total emissions is presented here, since GHG emission ties with energy efficiency and shows the same variation pattern.

#### 3.3.1. Energy efficiency

The LCA results (Fig. 4) show large variations in energy efficiency among different conversion pathways, cultivation season, and wastewater treatment plants.

 Table 2

 Energy production from wastewater-based algae in US.

Conversion	Biocrude oil, BG/yr	Biochar, 10 <sup>6</sup> ton/yr	Biogas, 10 <sup>6</sup> ton/yr
pathway	(energy: 10 <sup>9</sup> MJ/yr)	(energy: 10 <sup>9</sup> MJ/yr)	(energy: 10 <sup>9</sup> MJ/yr)
S1 - MP S2- HTL S3 - LE	0.77 (104.7) 0.98 (133.5) 0.57 (77.8)	1.80 (18.0) 1.80 (18.9)	2.44 (37.9) 1.45 (22.5) 0.74 (37.0)

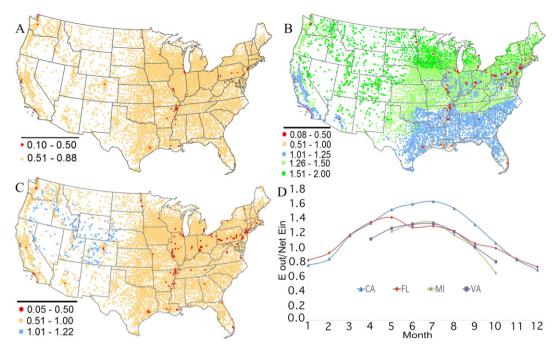


Fig. 4. Variations of energy efficiency among different conversion pathways, cultivation season, and wastewater treatment plants. Energy efficiency (EROI): the ratio of energy output to energy input with greater value being more energy favorable. A, B, C: energy efficiency (yearly average) of individual WWTP across the whole U.S. for MP (A), HTL (B), and LE (C), respectively. D: Monthly variations of energy efficiency in four representative WWTPs for the best performance scenario (HTL).

3.3.1.1. Bio-oil conversion pathway. All conversion pathways are independent from location, and their modeling is based upon the total amount of algal biomass production. Among the three conversion pathways (Fig. 4A, B, C), HTL is the best performance scenario, where most WWTPs can generate positive energy output (EROI > 1). This is because HTL has the best energy output (0.98 billion gallon/yr biooil + 1.9 million tons biochar + 1.4 million tons biogas) and relatively low energy input compared to MP and LE, since HTL does not require intensive energy procedure such as drying and can convert 50-60% of the total biomass to bio-crude oil [16,47]. MP produces second large energy output (0.77 billion gallon/yr bio-oil + 1.8 million tons biochar + 2.4 million tons biogas), but has the worst energy performance (no WWTP producing positive energy output). This is mainly due to high heat and electricity requirement for pretreatment and microwave generation. LE produces least energy (0.57 billion gallons/ vr + 0.74 million tons biogas) among the three conversion technologies, because lipid composition in algae is lower than carbon content that can be converted into bio-oil via thermochemical conversion. Nevertheless, compared to MP, LE has better energy performance (some WWTPs have net energy output), because it requires less heat and electricity. When compared to conventional fossil fuel (EROI: 13) (GREET 2015), wastewater-based algal bio-oils are not energy competitive  $(\text{EROI} \leq 2)$  [60], but they do perform much better than pathways with synthetic fertilizer and fresh water [4,54].

3.3.1.2. Site-specific and seasonal variations. When examining the sitespecific variations (take HTL scenario as the example), it is surprising that energy performance is opposite to the productivity. For example, warm climates have higher yearly productivity but exhibit poorer energy performance compared to cold climates. Further analysis reveals that this is mainly due to seasonal variation. Fig. 4D shows that there is a large variety in energy efficiency among different seasons. Because of lower productivities, the EROI in winter season (December, January, February) decreases >50% compared to summer season. Therefore, warm climates with all-season operation have lower yearly average energy efficiency than cold climates where oil production only occurs in optimal months (April to October). If winter operation is shut down, energy efficiency in warm climates will outperform that in cold climates (data not shown). The regression between algal biomass yield and energy performance (SI) suggests that it will not be energy favorable if the productivity is below  $20 \text{ g/m}^2$ -d (based on operational days). Our results suggest that winter shutdown may be necessary even in warm climates if winter productivity remains low. These results indicate that it is warranted to develop cultivation technology in cold weather for productivity improvement.

3.3.1.3. Energy allocation. To understand the driving force for energy efficiency, four WWTPs in different climate (from very cold to very warm) were selected to analyze the energy allocation for different processes including wastewater pumping, algae cultivation, biomass harvesting and pretreatment, bio-oil conversion, and energy credits from by-products (biochar and biogas) and wastewater treatment (Fig. 5). These four WWTPs have the same distance for wastewater pumping (5 km) and the same wastewater flow (around 100,000 m<sup>3</sup>/day). For all cases in different locations, WWTPs, and bio-oil conversion scenarios, the top two driving forces for energy burden are biomass harvesting/pretreatment and bio-oil conversion (contributing to 60–80% of total energy use), mainly from the electricity and heat used for process operation. The MP conversion pathway is the most burdensome process, accounting for about 50% of total energy use. In HTL and LE, biomass harvesting/ pretreatment is the top contributor (30–50% of total energy use). In contrast to freshwater-based system, energy use for cultivation has much less impact on total energy use. This is mainly due to the replacement of synthetic fertilizer that is very energy intensive. Previous studies indicate that energy burden associated with fertilizer could contribute up to 30% of total energy use [13,61].

Wastewater pumping is a considerable contributor for energy use in wastewater-based algae systems (20–30% of total energy use), from both electricity used for pumping and upstream burden associated with pipe construction. This indicates that land availability around WWTPs has significant impact on the performance of wastewateralgae systems. Further analysis suggests that energy efficiency will drop below 1 if land is not available in 10 km (SI). Ironically, WWTPs with abundant wastewater resources usually located in well-developed metro areas, where land is limited. As discussed in Section 3.2, about 40% of wastewater resources could not be utilized due to the short of

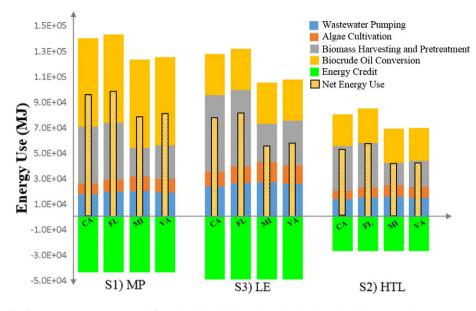


Fig. 5. Allocation of energy use for four representative WWTPs in California (CA), Florida (FL), Michigan (MI), and Virginia (VA). Y axis is the energy use per functional unit (50,700 MJ/yr). MP, LE, HTL

land resource. Land availability plays a significant role for wastewateralgae systems, because it not only determines the feasibility of co-siting algae facilities but also affects the overall cost. This is evidenced in Fig. 4A–C, as most of the large WWTPs (red dots) in metro areas are not energy favorable. According to the EPA survey [62], U.S. needs \$271 Billion investment to maintain and/or improve the nation's wastewater infrastructures. Algal cultivation is an opportunity for wastewater treatment and bioenergy generation. This study suggests that, for those WWTPs need redesign or reconstruction, decentralization could be one solution for wastewater utilization/reclamation such as algal biofuel production.

#### 3.3.2. Greenhouse gas emission

The main processes contributing to greenhouse gas emission include pipe production, concrete production, and  $CO_2$  emission from electricity used for operations. Upstream impact of GHG emissions from electricity and construction materials are calculated based on US mix electricity (0.8 kg of CO2 equivalent/kWh<sup>-1</sup>) and Ecoinvent Database [16]. Similar to energy efficiency, the total GHG emissions vary significantly among different scenarios (MP, HTL, and LE) and locations (Fig. 6), from - 2677 to 29,486 kg/FU, with the best performance scenario as HTL,

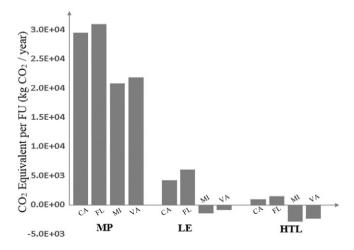


Fig. 6. Total greenhouse gas emissions per functional unit in four representative WWTPs in California (CA), Florida (FL), Michigan (MI), and Virginia (VA). Y axis is the greenhouse gas emission per functional unit (50,700 MJ/yr). MP, LE, HTL.

followed by LE and MP. High electricity demand for MP is the main reason for large GHG emission. The sit-specific differences are in accordance with the variations of energy efficiency, better performance in colder climate (MI, VA) than in warmer climate (CA, FL). This is attributed to the same reason causing the variations of energy efficiency, all-season operation in warm climate with lower average productivity while optimal-season operation in cold climate with higher productivity.

While not competitive to conventional fossil fuel in energy efficiency, wastewater-based algal could offer significant benefits in GHG control. GHG emissions for the best performance scenario (HTL) are 4–7 times lower than that of conventional fossil fuels (GREET 2015), with negative GHG emissions in some cases (LE and HTL in MI and VA). Flue gas uptake by algae biomass and wastewater treatment credit play a major role in reducing GHG emission.

#### 4. Conclusion

By developing a SEHR-LCA, this work presents the first study of point-to-point analysis of wastewater-based algal bio-oil for each individual WWTP across the whole US. The result indicates that there is a great potential for wastewater-based algal biofuel production. The total production of algal crude oil could be 0.98 billion gallon/yr, 20% of advanced biofuel projection as outlined in the U.S. Energy Independence and Security Act (EISA) of 2007. LCA results show that environmental impacts vary significantly among different locations, WWTPs, operational seasons, and bio-oil conversion pathways. Although not competitive to conventional fossil fuel in energy efficiency, wastewater-based algal biofuel could offer significant benefit in GHG control. However, spatial analysis indicates that land availability could be a significant challenge for wastewater-algae systems as it affects both the feasibility of co-siting algae facilities and energy cost. These results suggest that improvement should be made in technological development and system design to increase biomass productivity, energy efficiency, and land use efficiency for full potential of wastewater as a promising resource for algal biofuel production.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx. doi.org/10.1016/j.algal.2016.08.008.

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