Title: Implications of widespread algal biofuels production on macronutrient fertilizer supplies: nutrient demand and evaluation of potential alternate nutrient sources

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Abstract

Biofuels from microalgae are currently the subject of many research projects to determine their feasibility as a replacement for fossil fuels. In order to be a successful candidate, there must be enough fertilizers available to support large scale production. Commercial fertilizers are available for biofuel production from the world fertilizer surplus, but due to nitrogen and phosphorus future production limitations, biofuels would ideally not use any of these resources to be a long term sustainable fuel. Nitrogen, phosphorus and potassium requirements were determined for two algal species, *Chlorella* and *Nannochloropsis*, to produce 19 billion liters per year (BLPY). At this scale, both algal species would use 32-49%, 32-49% and less than 1% of the world surplus values of nitrogen, phosphorus and potassium, respectively. Nutrient recycling options and alternative sources of nutrients were evaluated to determine their potential contribution of lowering the synthetic fertilizer requirement. Results show that all of the recycling scenarios reduce the nutrient requirements, but catalytic hydrothermal gasification has the largest reduction of 95% of the nitrogen and 90% of the phosphorus. Contributions from all alternative sources can also provide only 5% or less of the required nitrogen when produced in the gulf region. For phosphorus in the same region, poultry concentrated animal feeding operations can provide up to 28% of the requirement of *Chlorella*. To find the least amount of nitrogen that may be used, catalytic hydrothermal gasification was combined with all of the alternative nutrients available in the gulf region. The maximum amount of biofuels that could be produced in this location without using any synthetic fertilizers is 50 ± 20 BLPY from *Chlorella* and 45 ± 19 BLPY from *Nannochloropsis*. This study shows that the nutrient requirement for biofuel production from microalgae will not be a limitation if recycling methods within the process chain and alternative sources of nutrients are utilized.

Keywords

Sustainability; fertilizers; microalgae to biodiesel; alternative fuels

1.0 Introduction

Biofuels produced from microalgae are currently being investigated as an alternative to fossil fuels. They are a ‘third generation’ feedstock that does not require arable land, therefore removing competition with food production land area. Algae can also sustain high growth rates per unit area [1], but require significant quantities of nutrients for growth, including nitrogen, phosphorus, potassium, carbon dioxide and a wide range of micronutrients. If biofuel produced from algae is to be sustainable, there must be enough fertilizers available for large scale production and must not use a significant amount of the fertilizers currently being used for food production.

Nutrient markets and availabilities are determined by the raw materials used to create them and the current production processes. For instance, nitrogen fertilizers depend on fossil natural gas reserves for production [2], which increases their environmental burden. As the world considers biofuel production, natural gas production is expected to peak in 2035 and this could have serious implications on any biofuel that relies on man-made fertilizers [3]. The United States imports approximately eight times the amount of nitrogen fertilizers that it exports [4]. In 2012, the largest imports were of 7.5 million tons of urea and 6.9 million tons of anhydrous ammonia, of which the most came from Canada and Trinidad and Tobago, respectively [4]. Ammonium sulfate is the largest source of nitrogen exported, on a mass basis, with 35% sent to Brazil [4].

Phosphate is an already strained resource that is produced through mining [5], with only eight countries controlling the significant majority (95%) of phosphate reserves [6]. In addition to the United States and China, the largest reserve is in the Sahara, with Morocco controlling the largest quantity [7]. Reserves of phosphorus are expected to be depleted within 50 to 100 years [7] and one controlling country, China, is already imposing a large tariff on phosphorus to reduce exports [8]. The United States exports twice as much as it imports [4]. The largest import is of phosphate rock, of which 54% comes from Morocco [4]. Diammonium phosphate (DAP) and monoammonium phosphate (MAP) comprise the largest export amounts. Approximately 29% of the DAP is sent to India, and 25% and 34% of the MAP is sent to Brazil and Canada, respectively [4].

Potassium fertilizers are also produced through the mining of potash. The United States imports more than 36 times more potassium containing fertilizers than it exports, with most of it coming from Canada [4]. Microalgae need approximately 1.8 – 2 kg of carbon dioxide for each kg of microalgae grown [9,10]. The carbon source can be provided as a pure gas bubbled into the system or a carbon-containing flue gas, allowing the microalgae to use a power plant waste stream normally sent to the atmosphere [11]. The algae require numerous micronutrients, which can include forms of copper, manganese, zinc, molybdenum, cobalt and iron [12,13]. The micronutrient requirements are species dependent on both the specific nutrient and amount needed [12].

Synthetic fertilizers are produced through chemical processing or through direct mining, and each year the Food and Agriculture Organization (FAO) of the United Nations reports the world surplus values [14]. If biofuels produced from microalgae use large amounts of national fertilizer surpluses, then there may be an impact on other markets, such as food production.

Greenwell, et al., [9] determined that fertilizer supplies would limit the extent to which algae-based biofuels could replace all of the fossil fuels in the European Union. Another study also evaluated biofuels from microalgae and found that large scales would cause restraints on both nitrogen and phosphorus sources needed for growth [15]. These studies show a need for ways to reduce the overall nutrient amount and/or find alternative nutrient sources during microalgae cultivation.

With the acknowledgment that nitrogenous and phosphorous sources may become constrained in the near future due to sustainability limitations, this work attempts to analyze alternate sources of nutrients while also covering the broad implications of current practices. In this study, the total estimated nitrogen, phosphorus and potassium requirements to produce biofuel from microalgae in the United States are estimated to determine the possible effect of algal biofuels on each fertilizer market. Production is examined at regional scales most likely to be used for cultivation and for the entire United States. The predicted nutrient requirements are compared to current estimates of U.S. fertilizer use and current world fertilizer surplus values for primary commodity fertilizers. This paper will determine whether or not microalgae will require synthetic fertilizers, or require additional nutrient sources like wastewater. Algae biomass processing methods that may serve to recycle nutrients, like anaerobic digestion, hydrothermal liquefaction, and catalytic hydrothermal gasification will also be evaluated to estimate their ability to reduce synthetic nutrient requirements through recovery of nutrients from residual biomass after or in place of lipid extraction and energy generation. Results of this work will help determine which areas of future research and investments into new technologies will benefit from further research in the future.

2.0 Establishing major nutrient requirements for large scale algal biofuels production

2.1 Algal fuel production target

In 2007, the United States established production targets for alternative fuels that meet various standards of environmental performance in the Energy Independence and Security Act (EISA) [16]. In 2022, the EISA mandates that advanced biofuel production, defined as having a life-cycle greenhouse gas emissions factor less than 50% of petroleum fuel greenhouse gas emissions, should reach 79 billion liters annually. In this analysis, we examine the potential impact of using synthetic fertilizers for widespread and significant algal cultivation activities to contribute to this goal. The baseline scenario in this work assumes that algal oil-based fuels will constitute 5 billion gallons per year or 19 billion liters per year (BLPY) towards this goal, similar to other high-level sustainability assessments of this emerging industry [17].

2.2 Algal and Nutrient Requirements to Meet 19 BLPY Target

Gross nutrient demand for this level of algal biofuel production will depend on assumed N, P and K requirements for algae species under cultivation. There are various representations for marine phytoplankton, one being the Redfield ratio of C:N:P at 106:16:1 [15]. Since different species have different elemental compositions, the effect of algal species choice on sustainability was evaluated by comparing freshwater and marine species of algae commonly studied for biofuels production, *Chlorella* sp. and *Nannochloropsis* sp. Compositions of *Chlorella* sp. and *Nannochloropsis* sp. were taken directly from Handler, et al. [18]. To determine the amount of nutrients required for the 19 BLPY target, the total mass of algae was needed. This was found by multiplying an average biofuel density of 0.88 g/mL [19] and the lipid content for various

composition scenarios for each algal species [18]. In these calculations it is assumed there would be no algal mass loss during dewatering or lipid loss during extraction and biofuel conversion. Nutrient amounts required for *Chlorella* were 0.05 ± 0.03 kg-N/kg-algae, 0.0009 ± 0.0008 kg-P₂O₅/kg-algae and 0.00018 ± 0.00015 kg-K₂O/kg-algae. *Nannochloropsis* had nutrient requirements of 0.06 ± 0.01 kg-N/kg-algae, 0.019 ± 0.004 kg-P₂O₅/kg-algae and 0.0011 ± 0.0008 kg-K₂O/kg-algae. Further details on these calculations, along with the entire nutrient requirement to produce 19 BLPY, can be found in the Supplementary Data, Section S.1.

2.3 Current Fertilizer Use and Availability for Large Scale Algae Production

In order to provide context for the N and P requirements for large scale algae biofuels production, we rely on two comparisons. First, the annual United States nutrient consumption for crops during the year 2010 was 11.1 billion kg N, 3.72 billion kg P₂O₅ and 4.04 billion kg K₂O [4]. Compared to the nutrient values calculated above, cultivation of both algal species on a 19 BLPY scale would consume 26-28% of current U.S. N use, 15-23% of current P₂O₅ use, and 0.2-1.5% of current K₂O use. The other metric for comparison is the current world fertilizer surplus of these nutrients, as major fertilizers are global commodities. Recent estimations of fertilizer surpluses are found in the report by the Food and Agricultural Organization of the United Nations report [14]. According to this assessment, for the year 2013, there are estimated world surpluses of 6.91 billion kg N, 1.75 billion kg P₂O₅, and 10.7 billion kg K₂O. If these surplus fertilizers were used to satisfy the 19 BLPY target, microalgae would require 41-56% of surplus N, 32-49% of surplus P₂O₅ and less than 1% of surplus K₂O.

The levels of macronutrient demand for algal biofuels are massive in scale, placing algae production on par with major domestic commodity crops like corn and soybeans [20]. Significant

increases in demand for N and P fertilizers have the potential to raise fertilizer prices, reducing economic incentives for algae production. The nutrient requirements detailed above were tabulated for just 23% of the 2022 advanced biofuel requirement. If this amount was doubled, then there may not be enough fertilizers in the world surplus to meet the needs of the microalgae. Also, these calculations were for the stoichiometric requirement, and the amounts of fertilizer would most likely be higher due to nutrient competition from grazers [21] or lack of nutrient uptake if media recycling is not used. Based on this, the world surplus of nutrients cannot be expected to meet the needs of microalgae based biofuels in the United States, and alternative sources of nutrients will have to be found if algae cultivation is to reach the scale required for significant impact on domestic biofuel production targets. In the following sections of the paper, we discuss some of the potential N, P and K resources that could be repurposed for algae cultivation, and utilize current data and research findings to estimate the likely scales of these resources

3.0 Alternative Sources of N and P for algae cultivation

This section describes the methodology for determining the amount of nutrient that could be recycled and the amount of nutrients that are available from alternative sources.

3.1 Recycling within the algae fuels value chain

Conservation and re-use of the nutrient resources required for algae cultivation is the simplest method of reducing the net requirement for commodity fertilizers for algal biofuels. Fortunately, there appears to be significant potential for recycling of nutrients from waste streams from other

industries, and also within the algae production system. Algal fuel production is most often concerned with the oil fraction of the algae biomass, which can vary based on growing conditions and algal species [1,11]. After harvesting and drying, oil is extracted from the algae biomass and retained for further processing, while the leftover lipid-extracted algae (LEA) portion of the biomass is available for use in co-products to enhance the economic value chain or meet some of the energy demands during conversion [22]. The LEA can also be used in processes along with a sugar factory wastewater to provide an economic growth media that will enhance lipid production of some microalgae strains [23]. The algae LEA fraction contains a large majority of the N, P, and other nutrients [24] required by growing algae cells, and the nutrient content within the LEA can be captured and returned to the algae cultivation system using several potential technologies, as discussed next. The major methods investigated in the literature for nutrient recycling include anaerobic digestion, catalytic hydrothermal gasification and hydrothermal liquefaction.

3.1.1 Anaerobic digestion

Anaerobic digestion (AD) is a well-studied technology that has matured in areas like wastewater treatment. Carbon-containing feedstocks are metabolized in an anaerobic environment, releasing CH_4 , CO_2 and other gases, while retaining the non-volatile fractions of N and P in solid and liquid forms that can be re-purposed. Peer-reviewed literature on unique AD feedstocks has become more prevalent in recent years, and a limited set of data is available concerning AD of algae biomass. Frank, et al., reviewed relevant studies for the GREET LCA model of algae biofuels and concluded that a reasonable approximation for current AD technology would return 75% and 50% for N and P, respectively, for recycling back to the cultivation system [25].

3.1.2 Catalytic Hydrothermal Gasification

Catalytic hydrothermal gasification (CHG) converts biomass with any liquid water content into energy products like methane and hydrogen [26], also producing side products like carbon dioxide and carbon monoxide [27]. The process operates at, or above, the supercritical point of water, or can operate at subcritical conditions if catalysts are used [28]. Catalysts can also speed up gasification, eliminate char formation, or in the case of subcritical conditions, cause gas formation [28]. Changing the temperature affects the product distribution, with higher temperatures producing more hydrogen, while methane is formed at lower temperatures [26]. CHG can be used on algal biomass [27,29,30] on either the whole cell or the lipid extracted algae (LEA) portion. Algae can grow on the process waters produced from CHG, which contain NH_4^+ , K^+ , PO_4^{3-} , NO_3^- , and many other micronutrients [27]. Phosphorus can also be recovered as a separated mineral [30]. For this analysis, the estimations of Frank et. al., [31] indicate that 95% of N and 90% of P can be recycled using CHG when processed LEA is used. Data shows that potassium is found in the process water [27], but there is no information on the percent recovery of this nutrient, therefore, potassium recycling will be neglected for CHG because any value for percent recovery would currently be speculation.

3.1.3 Hydrothermal Liquefaction

Hydrothermal liquefaction (HTL) can use biomass with any liquid water content [32] to produce oil, solid, gas and aqueous phases [33]. Phosphorus goes into the aqueous phase [29,33–36], as well as potassium [34,36]. No reports were found of either going into the oil or gas phases, so it

is assumed that additional nutrients would be part of the solid residue. Without information on possible recoveries, the analysis of potassium and phosphorus will be neglected for HTL and it will be assumed that these elements are lost, as a worst case scenario. For nitrogen, Frank, et. al. [31] reported that nitrogen present in the oil after HTL would probably be lost during processing due to uncertainties in scaling up equipment and the upgrading processes. Recovery of nitrogen from the oil is only economical at high concentrations [31], but the concentration would likely be low due to co-processing with petroleum products. Due to this, the nitrogen in the oil is considered to be lost. Microalgae are capable of growing on nitrogen in the aqueous phase [33,35] and it is assumed that any nitrogen in the solid or gas phases could be recovered. Concentrations of nitrogen in both *Chlorella* [34,35] and *Nannochloropsis* [35,37] were reported in prior work. Values are averaged, showing possible nitrogen recovery at $63 \pm 12\%$ in *Nannochloropsis* and $86 \pm 5\%$ in *Chlorella*.

3.2 Alternative Sources of Nutrients

In this section, alternative sources of N and P are examined for their ability to provide nutrients on a scale required for 19 BLPY of algal biofuels production levels. Although we are not attempting a detailed logistical assessment of procurement and transport for any of these nutrients, we recognize that alternate sources of N and P would need to be economically sourced and shipped to algae cultivation sites. For the purposes of this report, algae cultivation is likely to take place in the US Southeast, following the recommendations of Davis, et al., [17], although cultivation in the US Southwest may also be feasible if water source limitations are overcome.

3.2.1 Municipal wastewater

Municipal wastewater is seen as a viable source to provide nitrogen and phosphorus for microalgae growth [19,21]. In municipal wastewater treatment plants, incoming waters undergo a variety of treatments to make the water safe for release into the environment. Preliminary and primary treatments remove large objects and suspended solids [38]. Secondary treatments remove organic matter and also nutrients like nitrogen and phosphorus [38]. The potential nutrients available from wastewater depend on the total flow of wastewater and which stages of treatment have occurred in the wastewater treatment chain. The 19 BLPY target from the Davis, et. al., report [17] was modeled for the Gulf Coast, including the states of Texas, Louisiana, Mississippi, Alabama and Florida. Wastewater flow rates for these states were 18.5 billion liters per day (BLPD) in 2008 [39]. For comparison, the wastewater availability of the entire United States was also evaluated at 121 BLPD [39].

Wastewater nutrient concentrations were found for a variety of treatment options [19,21,40,41], including primary and secondary treatments. All concentrations, for both primary and secondary treatments, were averaged to account for the various types of treatment plants, giving nitrogen and phosphate concentrations of 20 ± 15 mg-N/L and 6.8 ± 6.6 mg-P₂O₅/L, respectively. The yearly nitrogen available is 0.14 ± 0.10 billion kg of N in the Gulf Coast and 0.9 ± 0.6 billion kg of N in the entire United States. Compared to the amount of nitrogen the algal species need for growth, 0.05 ± 0.03 kg-N/kg-*Chlorella* and 0.06 ± 0.01 kg-N/kg-*Nannochloropsis*, municipal wastewater would only make a small contribution. For phosphorus, there is 0.046 ± 0.044 billion kg of P₂O₅ available in the gulf coast and 0.31 ± 0.30 billion kg of P₂O₅ in the entire United States. Once again, this is a small contribution to the overall phosphorus requirements. These values assume that algae could use all of the nutrients available in the wastewater, which would

represent the most ideal scenario. Further details of the wastewater analysis can be found in Section S.3 of the Supplementary Data.

3.2.2 Spent Coffee Grounds

In 2011, the United States consumed 1.5 million tons of coffee beans or 4.24 kg per person [42]. After brewing, 10 – 15% [43] of the coffee oil can be extracted from the residual grounds depending on the type of coffee. The oil can be converted into biodiesel [43–46], which could help meet the 79 BLPY advanced biofuels requirement by 2022 [47]. Spent coffee grounds also contain nutrients [48–50], like nitrogen and phosphorus, which could be used to lower the amount of synthetic fertilizers required for microalgae. The grounds also contain other nutrients like Cu, Zn, and Mn [48,50] that are trace nutrients used for algae growth [13]. For this analysis, it is assumed that the entire mass of nutrients can be utilized from the spent coffee grounds for N, P, and K. Nutrient amounts were averaged for both nitrogen [48,49], phosphorus [48–50] and potassium [48–50], and multiplied by all of the coffee consumed in the United States. For the entire country, there are 0.0327 billion kg of N, 0.004 billion kg of P_2O_5 and 0.008 billion kg of K_2O available annually.

3.2.3 Animal husbandry waste products

Rearing of animals using modern agricultural practices results in significant accumulation of nutrient-laden animal wastes [51], which are treated as potential pollutants with the ability to damage ecosystems if releases are not controlled [52]. These situations present synergistic opportunities to remove nutrient wastes from animal husbandry facilities while providing algal

cultivation facilities with desired nutrients. While there may be hurdles to overcome in transport and processing of these waste products into a form that is suitable for algae utilization, we present a high-level analysis of the availability of important waste products from different agricultural practices.

3.2.3.1 Concentrated Animal Feeding Operations (CAFOs)

In 2007, the United States contained over one million livestock farms containing hogs, dairy cows, beef cattle, broiler chickens and layer hens [53]. There were over 9.5 billion animals on these farms, of which 94% were broiler chickens used for human consumption [53]. Animal waste can be sold or given away, in the case of hogs and broiler chickens, for crop fertilizer use [54]. In 2006, animal manure was used as a fertilizer on only five percent of the total cropland area in the United States [54]. Animal manure on hog and dairy farms can also be converted to energy in anaerobic digesters. However, less than one percent of dairy farms use this energy conversion technology [54].

The potential contribution of CAFOs was taken from an analysis of the 1997 agriculture census data [55]. This data is over ten years old, but the number of livestock in the United States has not changed dramatically, so it was assumed the reported CAFOs potential would provide adequate estimation. In their analysis, excess nutrients were found through using the maximum amount of nutrients for a farm and subtracting what would be used as a crop nutrient at the same location. Results were presented based on farm size and, for this paper; all of the excess nutrients were considered. In the entire United States, 0.1, 0.06, 0.1 and 0.5 billion kg of N are available as excess from feedlot beef, dairy cows, swine and poultry, respectively. For phosphorus, 0.2, 0.1, 0.3 and 0.6 billion kg of P_2O_5 is available from feedlot beef, dairy cows, swine and poultry

respectively. To determine the amount of nutrients located in the gulf region, the percentage of each animal type was determined from the 2007 Census of Agriculture Report [53]. In the gulf region, 24%, 6%, 2% and 30% of the feedlot beef, dairy cows, swine and poultry live in this area, respectively.

3.2.3.2 Aquaculture

Aquaculture is a significant industry in the Gulf Coast region of the United States, with catfish culturing in open ponds being the primary species and method of cultivation [56]. These large-scale aquaculture operations can often produce discharges of water polluted with high levels of nitrogen and phosphorus, necessitating federal regulations concerning best management practices [57]. To estimate total annual N and P discharges from catfish aquaculture in the Gulf Coast, two separate methods were used. The total hectares of catfish ponds in the region, over 29,500 ha [58], was multiplied by the average kg of N and P per hectare of catfish pond [59] to develop an area-based estimate of N and P production from catfish aquaculture effluent. Similarly, total annual catfish production in the region, over 0.11 billion kg [58], was multiplied by the average kg of N and P released from catfish ponds per ton of fish produced [60] to arrive at a mass-based estimate of N and P production from catfish aquaculture. The amount of potentially available fertilizers was 0.00330 ± 0.00008 billion kg N and 0.00013 ± 0.00004 billion kg P_2O_5 .

3.2.3.3 Slaughterhouses

Animals are killed in slaughterhouses to provide meat primarily for human consumption. Water is used during animal processing and equipment cleaning, and provides a wastewater containing

nitrogen and phosphorus [61,62]. To determine the amount of nutrients available from poultry, data was found for three poultry slaughterhouses [63]. Concentrations were averaged and multiplied by the total number of broiler hens sold in the gulf coast and the entire United States, Table S.8 of the Supplementary Data. The source [63] also contained data for third processing, but these values were not used because information was not found on the amount of chickens processed in this step. Analysis for cook plants and rendering was also neglected. The amount of N available from the gulf coast and entire United States is 0.0086 ± 0.0006 and 0.028 ± 0.002 billion kg of N per year respectively. The amount of phosphorus available from the Gulf Coast and the entire United States is 0.005 ± 0.002 and 0.015 ± 0.006 billion kg of P_2O_5 per year, respectively. The full analysis is available in Section S.4 of the Supplementary Data.

In beef cattle processing [64], slaughterhouses only kill and dress cattle, while meat processing facilities do those same tasks, plus make additional products from the meat, like steak, sausages and ground meat. Further processing steps also include rendering, which turns scraps into edible (lard) and inedible products. The majority of the wastewater produced at these facilities comes from cleaning the facility after slaughter [64,65]. The nitrogen in the wastewater comes mostly from blood and manure, and is mostly organic [64]. Phosphorus also comes from blood, manure, stomach contents and cleaning products [64]. Nutrient concentrations from the wastewater effluent were found as a mass per mass of animal [65]. Those results were multiplied by the total animal weight in either the gulf region or the entire United States. The amount of N available from the gulf coast and entire United States is 0.0046 ± 0.0002 and 0.0191 ± 0.0008 billion kg N per year respectively. The amount of phosphorus available from the gulf coast and the entire United States is 0.00066 ± 0.00003 and 0.003 ± 0.001 billion kg P_2O_5 per year respectively. The full analysis is available in Section S.4.2 of the Supplementary Data.

Swine slaughter wastewater is similar to cattle wastewater, with the nitrogen and phosphorus coming from the same source [65]. However, nitrate concentrations will be larger in swine wastewater as the curing of bacon and ham are conducted at the plant [65]. Nutrient concentrations were found for six hog slaughterhouses [66], which varied in capacity from 1,900 to 45,000 animals slaughtered per week. Potassium wastewater concentrations were also reported, and are analyzed here. Nutrient concentrations, presented as mass per volume, were multiplied by the volumetric flow rate in each plant. Yearly nutrient masses were found by multiplying by the number of animals in the gulf region and the entire United States. In the gulf region, 0.0004 ± 0.0002 billion kg as N, 0.00030 ± 0.00027 billion kg as P_2O_5 and 0.00024 ± 0.00027 billion kg as K_2O was available. For the entire United States, 0.018 ± 0.009 billion kg as N, 0.013 ± 0.012 billion kg as P_2O_5 and 0.010 ± 0.012 billion kg as K_2O was available. The full analysis is available in Section S.4.3 of the Supplementary Data.

3.2.4 Industrial wastewater sources

3.2.3.1 Carpet Wastewater

Industrial wastewaters from the carpet production industry, which is a significant industry in the U.S. Southeast [67], contain nitrogen and phosphorus that can be used for algal growth [68]. The reported nutrient concentrations available in the wastewater were 32.6 mg-N/L and 12.6 mg- P_2O_5 /L. To determine the potential contribution from this source, the amount of wastewater produced by this industry was found [67], which, for 2008, there was 29 billion liters of wastewater produced. Concentrations and the wastewater flow rate were multiplied together to arrive at the potential contribution from carpet production wastewater. Annually there are 0.00094 billion kg of N and 0.00037 billion kg of P_2O_5 available.

3.2.3.2 Brewery Wastewater

In 2011, United States breweries produced over 23 billion liters of beer [69]. Wastewaters from these breweries contain nitrogen and phosphorus that algae can use for growth [70]. Wastewater concentrations, in mg/L, were found for nitrogen [71–73] and phosphorus [71,73], and then averaged. To convert concentrations into mass flows, it was assumed the annual volume of wastewater produced is five times the annual beer production rate [74]. Wastewater flows for individual states was not found, so results are presented for the entire United States. The amount of nitrogen and phosphorus available is 0.006 ± 0.005 billion kg N and 0.010 ± 0.013 billion kg of P_2O_5 . Complete analysis for brewery wastewater can be found in Section S.5 of the Supplementary Data.

5.0 Results and Discussion

5.1 Nutrient Requirements for 19 BLPY Biofuel Production Target

The 2013 projected world surplus of fertilizers and the stoichiometric requirement of *Chlorella* and *Nannochloropsis* are presented in Table 1. To satisfy a 19 BLPY biofuel production target, both *Chlorella* and *Nannochloropsis* would require almost half of the nitrogen available. With phosphorus, *Chlorella* would use 34% while *Nannochloropsis* would need approximately 49%. Potassium has a smaller requirement than the other nutrients with both algal species requiring less than one percent of the surplus.

The estimated nutrient requirements in this evaluation were for the stoichiometric requirement of each algal species. In commercial-scale cultivation systems, algae would compete with grazers

for nutrients, and increase the overall nutrient requirement. An estimation of additional nitrogen and phosphorus to account for the grazers could be approximately two times the stoichiometric requirement [21], meaning double the values of Table 1. Potassium values may have the same increase. Another possibility would be if biofuels from microalgae made up the entire 2022 advanced biofuel requirement [16] of 79 BLPY. In this case, the nutrient requirements would be 4.2 times the values in Table 1. World surplus values would not be able to handle the nitrogen and phosphorus requirements under these scenarios.

5.2 Nutrients Provided by Production Recycling and Alternative Sources

Recycling options and alternative sources of nitrogen, phosphorus and potassium are available in Tables 2 – 4. All of the recycling options reduce the nitrogen synthetic fertilizer requirement, with CHG providing the largest reduction at 95% for both algal species. AD and CHG are technology choices that produce energy from the lipid extracted algae (LEA), and can reduce the overall energy requirement for microalgae production. The nutrient recycling benefits make them an attractive technology choice for LEA as opposed to using it as an animal feed or for other co-products. Similar results are seen for phosphorus, Table 3, with CHG providing the largest recycling percentage at 90% for both algal species.

Municipal wastewater provides both nitrogen and phosphorus, Tables 1 – 2, with only a minor contribution of both nutrients for the gulf region. Nitrogen was less than five percent of the stoichiometric requirement, and phosphorus less than two. Wastewater from the entire United States provided a maximum of $31\pm 23\%$ of the nitrogen for *Chlorella*. Phosphorous for the same species was $11\pm 10\%$. The wastewater from the entire country would not be a reasonable source

for algae grown in the gulf region due to the energy required to transport the wastewater from around the country to this region.

CAFOs in both the gulf region and the entire United States cannot provide all of the nitrogen and phosphorus required for microalgae growth. For nitrogen, Table 2, waste from all animals (dairy cattle, poultry, feedlot beef and swine) in the gulf region provides 5% or less of the nutrients for both species. Of all the animal wastes, poultry provides the most nitrogen at 0.5 billion kg-N and dairy cattle the least at 0.004 billion kg-N. Nutrient availability estimates for CAFOs for the entire United States show that there would not be a major contribution from this one source.

Poultry can provide 17% and 16% of the nitrogen for *Chlorella* and *Nannochloropsis* respectively. Feedlot beef, dairy cattle and swine can only contribute less than 5% for both algal species if using all of the excess nitrogen in the United States. No CAFOs sources from the gulf region can provide all of the phosphorus, Table 3, but the percentages are higher than for nitrogen. There are similar trends seen as for nitrogen, with broiler chickens providing the most in the gulf region at 28% for *Chlorella* and 20% for *Nannochloropsis*. Excess CAFOs phosphorus for the entire country can provide up to 94% of the phosphorus for *Chlorella*, though. All other animal sources can provide at least 10% of the phosphorus for both algal species. Results were not found for CAFO's potassium availability, but there is potassium available in animal manure [51]. It is expected that all of these sources would provide some of the nutrient needs.

CAFOs are an area that would benefit from additional research on the ability to provide nutrients for microalgae growth. One area of investigation is the potential nutrient uptake percentages. This analysis assumes that microalgae would take up the entire nutrient available in each source. It is expected uptake percentages would be less than 100%, and would be different for each

nutrient type for each species. Another area of further investigation is whether or not the waste could be used directly without pretreatment, or if energy and materials are required for pretreatments prior to use, which would reduce environmental benefits of recycle. Animal waste contains pathogens that may be harmful to a microalgae production process. It may be more reasonable to extract nutrients from the waste, in which case the increased energy demands would need to be evaluated to see their effect on the energy balance of the entire process. Also, nutrients may be lost during extraction, further lowering the nutrient potential from CAFOs.

None of the additional sources evaluated in this work provided any significant nitrogen or phosphorus to meet the target biofuel volume. However, swine slaughterhouse wastewater in the entire United States can provide $18\pm 20\%$ of the potassium for *Nannochloropsis* and the entire requirement for *Chlorella*. Spent coffee grounds cannot provide the entire amount for either species. Even though these additional sources cannot provide the entire requirement, their use would make sense in certain situations. If an algal farm was located near an aquaculture farm, a brewery or a carpet plant, it might be more cost effective and less energy intensive to use waste sources from these locations for nutrients than purchasing fertilizers on the open market.

In an ideal production scenario, nutrients could be recycled within the system (AD, CHG, and HTL) and nutrients could also be added from alternative sources. To determine if combining these practices would result in none of the world surplus nutrients being required for growth, the highest nutrient recycling option of CHG at 95% of N and 90% of P, was used to determine the new nutrient requirement for *Chlorella* and *Nannochloropsis*, Table 5. When compared to the 2013 world surplus, both algal species would require approximately 2% of the nitrogen, 5% of the phosphorus for *Nannochloropsis*, and 3% of the phosphorus for *Chlorella*. These new nitrogen and phosphorus requirements are compared to the alternative nutrient sources available

in the gulf region, Figures 1 and 2. Poultry from CAFOs can provide all of the nitrogen required for *Chlorella* and all of the phosphorus for both algal species. If all of the nitrogen, phosphorus and potassium from alternative sources in the gulf region are added together, there is 0.4 ± 0.1 billion kg of N, 0.29 ± 0.04 billion kg of P_2O_5 and 0.008 billion kg of K_2O available. Compared to the amount of nutrients required by the microalgae species after CHG, there is enough nitrogen and phosphorus available to produce 19 BLPY. There is not enough potassium as calculated by this study, but as mentioned before, there are unreported amounts of the nutrient in CAFOs, which may provide a significant amount.

5.3 Gulf Region Microalgae Biofuel Production Potential

To determine the biofuel production potential in the gulf region, without using any synthetic fertilizers, alternative nutrient amounts are divided by the new nutrients requirements including CHG, of each algal species. Results differ depending on the algal species and nutrient evaluated. This means that the amount of nitrogen in the region divided by the nitrogen requirement for *Chlorella* equals 50 ± 20 BLPY, but the amount of phosphorus divided by the requirement is 100 ± 32 BLPY. To determine the maximum amount of biofuels that could be produced in the region, with no impact on world surplus fertilizer amounts, the smallest value from the combined analyses for nitrogen and phosphorus is used. Potassium was not analyzed because both algal species would require less than 1% of the 2013 world surplus. In the United States gulf region, 50 ± 20 BLPY of biofuels from *Chlorella* and 45 ± 19 BLPY of biofuels from *Nannochloropsis* could be produced. Both of these values are less than the year 2022 advanced biofuels requirement of 79 BLPY, showing that if microalgae-based biofuels were solely used to meet this target, the process would have to include different regions or use part of the world fertilizer

surplus. If the same methodology is taken to add the nitrogen and phosphorus available from the alternative nutrient sources with United States nutrient amounts available (wastewater, CAFOs and meat processing wastewater), 238 ± 112 BLPY and 213 ± 104 BLPY of fuel could be produced from *Chlorella* or *Nannochloropsis*, respectively.

5.4 Energy Implications of Fertilizer Use

There have been concerns about decreasing the energy demand of producing biofuels from microalgae. Energy is used in every step of the cultivation process, from agitation during growth, dewatering, and conversion. Some processing steps like dewatering are seen as areas of concern due to their energy requirements [75,76]. Fossil fuels are used during fertilizer production [18] as either a feedstock or used for energy for production. Reducing the fertilizer requirements can reduce the cumulative energy demand of microalgae [76]. This can be accomplished through modifying growing conditions like nitrogen deprivation to increase lipid content for more biofuel production. Recycling and utilizing alternative nutrients would also require additional energy, such as pumping to recycle through the system or extraction of nutrients for microalgae use. However, a detailed analysis would need to be performed to determine what affect these scenarios would have on overall energy consumption.

5.5 Benefits and Disadvantages of Recycling Nutrients and Alternative Sources

The solution to the nutrient problems for producing biofuel from microalgae may not be solved by one single source, but rather a multitude of sources that would be locally available to microalgae farms. To take advantage of these nutrients, the best location for a biofuel plant is next to the industries evaluated in this work. Production companies might save money from not

having to treat their waste and could either sell or receive credit for providing biomass to biofuel producers. Avoiding synthetic fertilizer use allows other industries to create partnerships with biofuel producers, which could be beneficial to both groups, depending on governmental policies and local economics.

Conclusion

Biofuels produced from microalgae would have a large impact on the world surplus of fertilizers if synthetic fertilizers are used entirely for their growth. Recycling nutrients within the production system with technologies like AD, HTL and CHG can significantly reduce the overall nutrient requirements. Alternative sources of nutrients can also reduce the synthetic fertilizer requirement, but there is not a single source that could provide all of the nutrients. There is a research need to determine if microalgae can grow off the types of nutrients discussed in this paper. Based on this study, biofuels from microalgae will not have a significant impact on the world surplus fertilizer values if recycling technologies are used, along with alternative nutrient sources.

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Figure 1: Mass of nitrogen that can be provided by the alternative nutrient sources compared the nitrogen required by *Nannochloropsis* and *Chlorella* with CHG recycling for the United States gulf region. Microalgae nutrient requirements are for 19 BLPY.

Figure 2: Mass of phosphorus that can be provided by the alternative nutrient sources compared the phosphorus required by *Nannochloropsis* and *Chlorella* with CHG recycling for the United States gulf region. Microalgae nutrient requirements are for 19 BLPY.

Table 1: Nitrogen and phosphorus synthetic fertilizer availability based on world surplus values [14], along with nutrient requirements to produce 19 BLPY of biofuels for two algal species.

	Nitrogen (as billion kg N)	Phosphorus (as billion kg P ₂ O ₅)	Potassium (as billion kg K ₂ O)
2013 World Surplus	6.91	1.75	10.7
<i>Chlorella</i>	2.8±0.9	0.6±0.2	0.009±0.002
<i>Nannochloropsis</i>	3.2±1.0	0.85±0.06	0.056±0.0497

Table 2: The amount of nitrogen provided by recycling scenarios and alternative sources to produce 19 BLPY of biofuel. Synthetic fertilizer reduction represents the percentage of fertilizers that could be replaced by each scenario, for *Chlorella* and *Nannochloropsis* compositions. C/N under recycling options represents the mass of nutrients that could be recycled for each species to produce the target fuel volume.

	Nitrogen (10 ⁹ kg N)	Synthetic Fertilizer Reduction (<i>Chlorella</i>)	Synthetic Fertilizer Reduction (<i>Nannochloropsis</i>)
<u>Recycling Options</u>			
AD (C/N)	2.1/2.4	75%	75%
CHG (C/N)	2.7/3.0	95%	95%
HTL (C/N)	2.4±0.1/2.0±0.4	86±5%	63±12%
<u>Municipal Wastewater</u>			
Gulf Region	0.13±0.10	5±3%	4±3%
U.S.	0.9±0.7	31±23%	28±20%
<u>CAFOs</u>			
Dairy Cattle – Gulf	0.004	0.1%	0.1%
Dairy Cattle – U.S.	0.06	2%	2%
Feedlot Beef – Gulf	0.03	1%	1%
Feedlot Beef – U.S.	0.1	5%	4%
Swine – Gulf	0.003	0.1%	0.1%
Swine – U.S.	0.1	4%	4%
Poultry – Gulf	0.2	5%	5%
Poultry – U.S.	0.5	17%	16%
<u>Slaughterhouse Wastewater</u>			
Poultry – Gulf	0.009±0.001	0.30±0.02%	0.27±0.02%
Poultry – U.S.	0.028±0.002	1.1±0.1%	0.89±0.07%
Feedlot Beef – Gulf	0.0046±0.0002	0.16±0.01%	0.144±0.006%
Feedlot Beef – U.S.	0.019±0.001	0.67±0.03%	0.60±0.03%
Swine – Gulf	0.0004±0.0002	0.013±0.008%	0.012±0.007%

Swine – U.S.	0.016±0.009	0.6±0.3%	0.5±0.3%
<u>Other Sources</u>			
Aquaculture	0.0033±0.0008	0.12±0.03%	0.10±0.03%
Spent Coffee Grounds	0.0305±0.0001	1.071±0.002%	0.958±0.002%
Brewery Wastewater	0.006±0.005	0.21±0.16%	0.19±0.15%
Carpet Wastewater	0.00094	0.03%	0.03%

Table 3: The amount of phosphorus provided by recycling scenarios and alternative sources to produce 19 BLPY of biofuel. Synthetic fertilizer reduction represents the percentage of phosphorus that could be replaced by each scenario, for *Chlorella* and *Nannochloropsis* compositions. C/N under recycling options represents the mass of nutrients that could be recycled for each species to produce the target fuel volume.

	Phosphorus (10 ⁹ kg P ₂ O ₅)	Synthetic Fertilizer Reduction (<i>Chlorella</i>)	Synthetic Fertilizer Reduction (<i>Nannochloropsis</i>)
<u>Recycling Options</u>			
AD (C/N)	0.3/0.4	50%	50%
CHG (C/N)	0.5/0.8	90%	90%
<u>Municipal Wastewater</u>			
Gulf Region	0.046±0.044	1.61±1.56%	0.144±1.39%
U.S.	0.31±0.30	11±10%	10±9%
<u>CAFOs</u>			
Dairy Cattle – Gulf	0.05	1%	1%
Dairy Cattle – U.S.	0.1	14%	10%
Feedlot Beef – Gulf	0.05	9%	6%
Feedlot Beef – U.S.	0.2	36%	26%
Swine – Gulf	0.01	1%	1%
Swine – U.S.	0.3	47%	34%
Poultry – Gulf	0.2	28%	20%
Poultry – U.S.	0.6	94%	68%
<u>Slaughterhouse Wastewater</u>			
Poultry – Gulf	0.004±0.002	0.79±0.35%	0.5±0.2%
Poultry – U.S.	0.014±0.006	3±1%	1.7±0.8%
Feedlot Beef – Gulf	0.00066±0.00003	0.12±0.01%	0.077±0.003%
Feedlot Beef – U.S.	0.0027±0.0001	0.49±0.02%	0.32±0.01%
Swine – Gulf	0.00030±0.00027	0.053±0.048%	0.035±0.031%
Swine – U.S.	0.013±0.011	2.3±2.0%	1.5±1.3%
<u>Other Sources</u>			
Aquaculture	0.00029±0.00008	0.05±0.01%	0.009±0.003%

Spent Coffee Grounds	0.004±0.001	0.7±0.2%	0.12±0.04%
Brewery Wastewater	0.009±0.012	1.7±2.2%	1.1±1.4%
Carpet Wastewater	0.00037	0.07%	0.04%

Table 4: The amount of potassium provided by recycling scenarios and alternative sources to produce 19 BLPY of biofuel. Synthetic fertilizer reduction represents the percentage of potassium that could be replaced by each scenario, for *Chlorella* and *Nannochloropsis* compositions.

	Potassium (10 ⁹ kg K ₂ O)	Synthetic Fertilizer Reduction (<i>Chlorella</i>)	Synthetic Fertilizer Reduction (<i>Nannochloropsis</i>)
<u>Slaughterhouse Wastewater</u>			
Swine – Gulf	0.00024±0.00027	2.7±3.1%	0.4±0.48%
Swine – U.S.	0.010±0.011	113±131%	18±20%
<u>Other Sources</u>			
Spent Coffee Grounds	0.008±0.005	93±60%	14±9%

Table 5: Smallest nutrient requirement for the two algal species by using the CHG technology choice to recycle 95% of the nitrogen and 90% of the phosphorus. Microalgae nutrient requirements are for 19 BLPY.

Algal Species	Nitrogen (10 ⁹ kg as N)	Phosphorus (10 ⁹ kg as P ₂ O ₅)	Potassium (10 ⁹ kg as K ₂ O)
<i>Chlorella</i>	0.14±0.04	0.06±0.02	0.009±0.002
<i>Nannochloropsis</i>	0.16±0.05	0.09±0.01	0.06±0.05