

Supply Chain Sustainability Analysis of Whole Algae Hydrothermal Liquefaction and Upgrading

Energy Systems Division

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1. Introduction

The Department of Energy's Bioenergy Technology Office (BETO) collaborates with a wide range of institutions towards the development and deployment of biofuels and bioproducts. To facilitate this effort, BETO and its partner national laboratories develop detailed techno-economic assessments (TEA) of biofuel production technologies as part of the development of design cases and state of technology (SOT) analyses. A design case is a TEA that outlines a target case for a particular biofuel pathway. It enables preliminary identification of data gaps and research and development needs and provides goals and targets against which technology progress is assessed. On the other hand, an SOT analysis assesses progress within and across relevant technology areas based on actual experimental results relative to technical targets and cost goals from design cases and includes technical, economic, and environmental criteria as available.

BETO also develops supply chain sustainability analyses (SCSA) for key biofuel production technologies that are the subject of design case or SOT analyses (Dunn et al. 2013). The SCSA utilizes a life-cycle analysis to estimate the energy use and greenhouse gas (GHG) emissions associated with biofuel production and assists in comparing several biofuel pathways. This report documents an SCSA of whole algae hydrothermal liquefaction (AHTL) as the conversion technology to produce renewable diesel (RD). Jones et al. (2014) developed the design case process model that provides the material and energy intensity of the feedstock conversion step in the SCSA.

The SCSA production stages for microalgae-derived RD are presented in Figure 1. Various inputs (red boxes) can be considered for each supply chain step (green boxes). These inputs can include energy, fertilizers for biomass growth, and any materials that may be needed during the conversion process. The major environmental output from the system is GHG emissions, which come from direct sources like fuel combustion during a processing step or indirect sources like fertilizer production. Another common output is coproducts, which can be used to displace materials or energy from other production processes. There can be difficulties in allocating emissions to these co-products (Wang et al., 2011), so care is needed during their consideration.

The SCSA for RD produced via AHTL starts with feedstock production, which requires nutrients (fertilizers), water (not considered in this study), and energy in the form of electricity and other fuels, e.g., natural gas. After production, the feedstock is transported to the conversion facility, or biorefinery, using energy in the form of a transportation fuel. In the case of microalgae, cultivation ponds are assumed to be co-located with the conversion facility (Davis et al., 2012; Frank et al., 2011) meaning a transportation fuel is not required. However, energy is needed for pumping the biomass from the harvesting units to the biorefinery. For the algae-to-RD production reported here, the harvested feedstock goes to a thermal conversion process, which includes material inputs like catalysts and sulfuric acid. A small amount of naphtha, which was treated as a liquid fuel, is produced along with RD in the AHTL pathway. No other co-products are produced in the fully integrated AHTL algae-to- RD pathway. The total supply chain emissions burdens were allocated to total fuel produced, including naphtha and RD.

The renewable fuel, after the conversion process, is transported to a fueling station by train, barge, and truck. The biogenic CO₂ released when the fuel is combusted balance out with the atmospheric CO₂ that the algae incorporated when it was growing (Frank et al., 2011). The emissions described above are the so-called, "fuel cycle" emissions. Emissions are also associated with the construction of the plant (Canter et al., 2014). These "infrastructure cycle" emissions were estimated in this study.

The supply chain sustainability impacts of the AHTL algae-to-RD pathway are analyzed using Argonne National Laboratory's Greenhouse gases, Regulated Emissions, and Energy use in Transportation model

(GREET). The version of GREET used in this analysis was GREET1_2013 (Argonne National Laboratory, 2013).

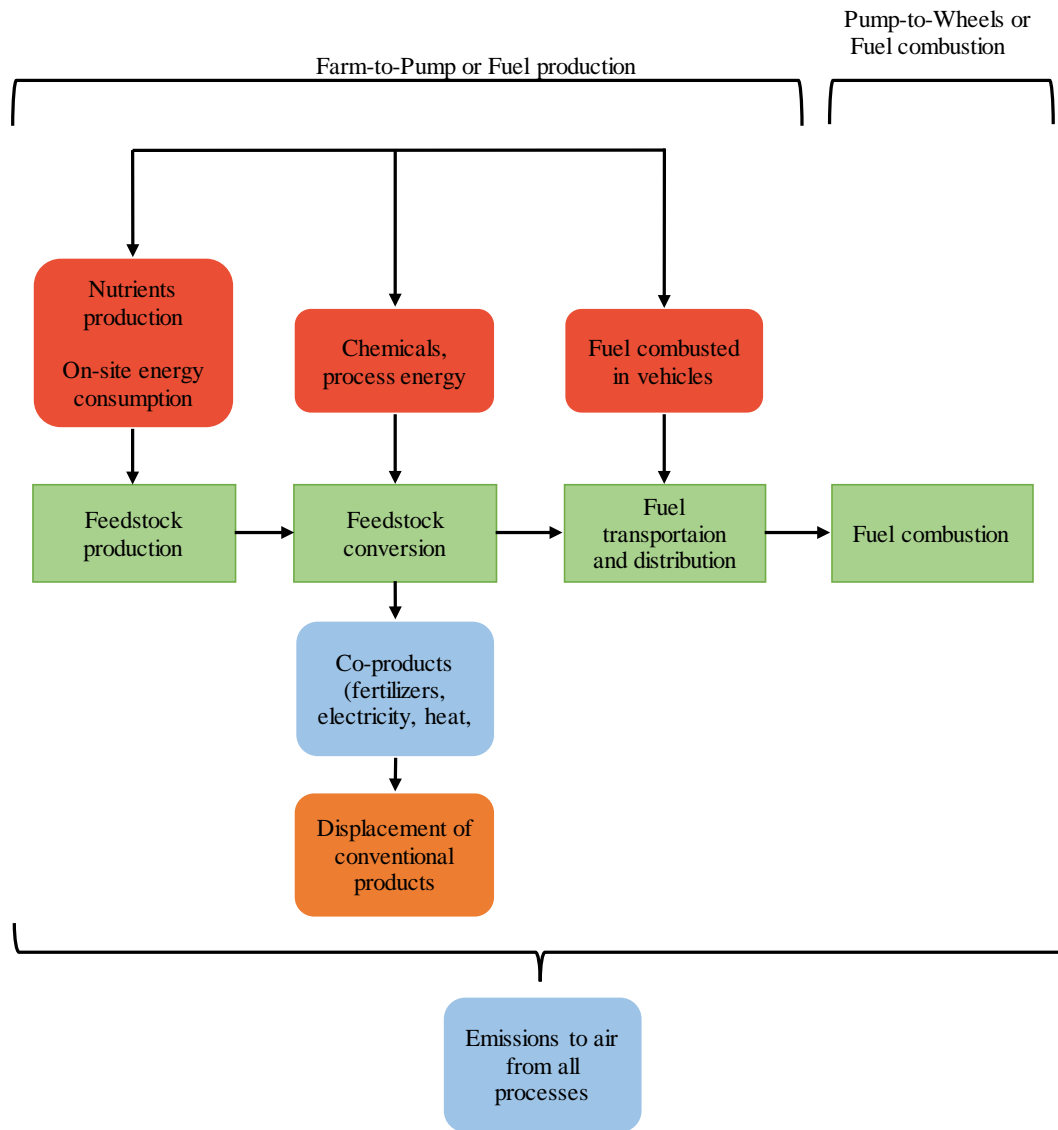


Figure 1. Stages considered in the SCSA of renewable fuels. Box colors indicate: green – supply chain process step, red – inputs to the supply chain, blue – impacts on the supply chain, orange - conventional products displaced by co-products (not applicable in this study).

2. Thermal Conversion of Algal Biomass to Renewable Diesel (RD)

In this study, conversion of algal biomass to RD is achieved by whole AHTL, as described in the design report by Pacific Northwest National Laboratory (Jones et al., 2014). Wet algal biomass, as undisturbed cells, is converted into a liquid fuel with pressurized water in a condensed phase. The SCSA for this pathway is described in the following sections.

2.1 Algal Biomass Production and Harvesting

The algal biomass production and harvesting model used in this SCSSA is the one developed previously in the baseline harmonization report (Davis et al., 2012), except with regard to nutrient recycling from the AHTL conversion process. The nutrient recycling was modified based upon (Jones et al., 2014). The microalgae are assumed to grow in lined, paddlewheel driven, open raceway ponds. After growth, algal biomass is dewatered in three stages up to a final solids content of 20% (on an ash free basis), as shown in Figure 2. The thickened solids then go to the AHTL conversion process. Previous reports (Davis et al., 2012; Davis et al., 2014) provide a detailed description of the dewatering processes, including parameters like efficiency and process input energy.

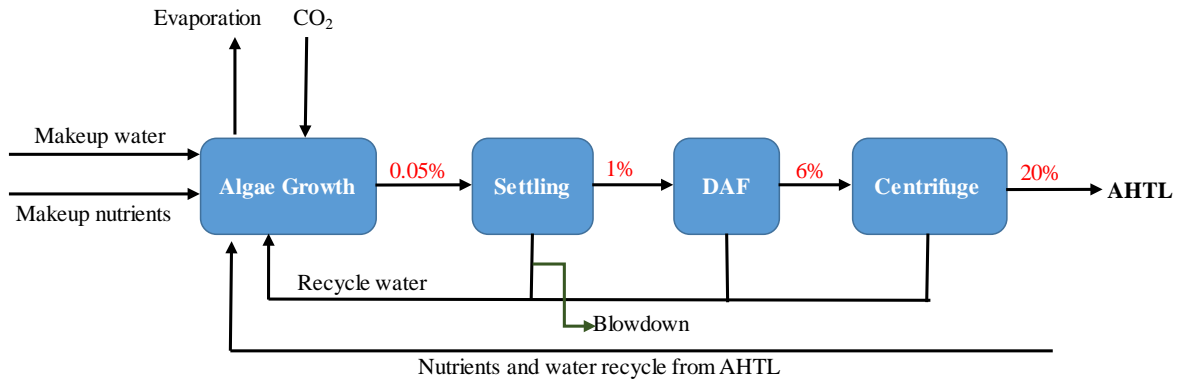


Figure 2. Process flow diagram for algal biomass production. Percentages, in red, indicate the weight percentages of algae in the flow on an ash free dry weight basis

Energy inputs for feedstock production include manufacturing energy for nutrients (Johnson et al., 2013), and process energy for algal growth, pumping, dewatering, and harvesting. The nitrogen (N) and phosphorus (P) demands for growth are estimated from the elemental composition of the algal biomass and the nutrient recycle flows from the AHTL process. The elemental composition of the algal biomass used in this analysis is displayed in Table 1 (from Table 2 of (Jones et al., 2014)).

Table 1. Elemental composition of algal biomass. From (Jones et al., 2014)

Aspen Design Case for Algae	
Component	Weight (%)
C	52.0
H	7.5
O	22.0
N	4.8
S	0.61
Ash	13
P	0.6

The elemental composition for microalgae from the design case (Jones et al., 2014) was used to calculate the molar ratio at 223:17.6:1 (C:N:P). The expected recycle rates of N in the AHTL process are described in Figure 3 (taken from Figure 9,(Jones et al., 2014)). Although 95% of the input N can be recovered from the process, off-gassing losses at the pond will reduce the percent of N that is recycled to 89%. The net N demand is estimated from the mass balance of N based on the algae composition (Table 1) and recycle

flows (Figure 3) from the design report (Jones et al., 2014). Similarly, the design report estimates that 90% of incoming P to the AHTL process can be recovered from the solids stream and recycled back to the algae ponds after acid treatment. The net P demand is calculated as the difference between the stoichiometric P requirement (Table 1) and recycled P from AHTL process (Jones et al., 2014).

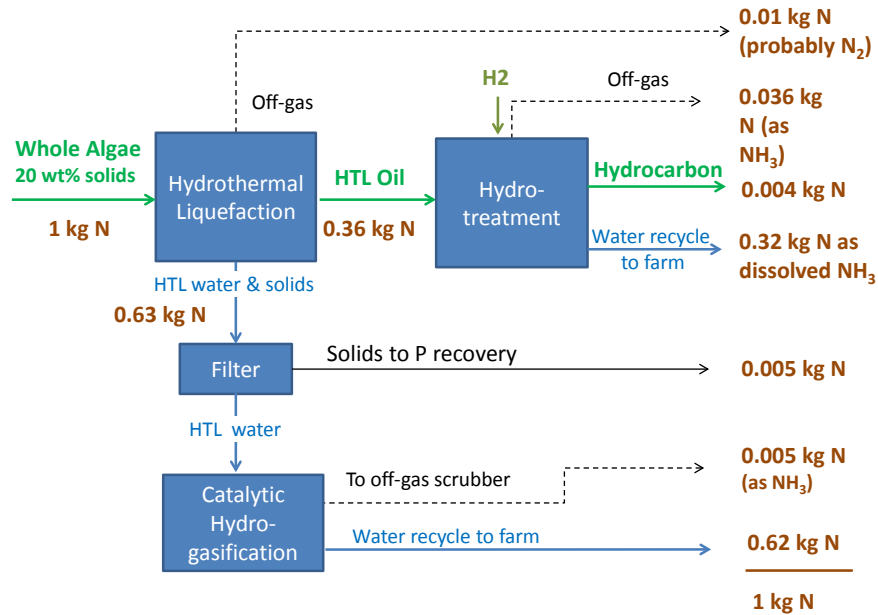


Figure 3. Expected nitrogen recycle flows in AHTL process (From Figure 9, (Jones et al., 2014))

Figure 4 (Figure ES-1, (Jones et al., 2014)) displays the carbon flows in the AHTL process, which results in a dissolved and gaseous CO₂ recycle back to the growth ponds. The net CO₂ demand for growth is calculated from the mass balance of CO₂ in the recycle stream from the AHTL process, the stoichiometric CO₂ requirement, and the CO₂ utilization efficiency. This net amount is assumed to be delivered to the site via low-pressure pipelines from a central source, as described in previous studies (Davis et al., 2012; Frank et al., 2011). The key process parameters and energy input values for algal biomass production are given in Table 2.

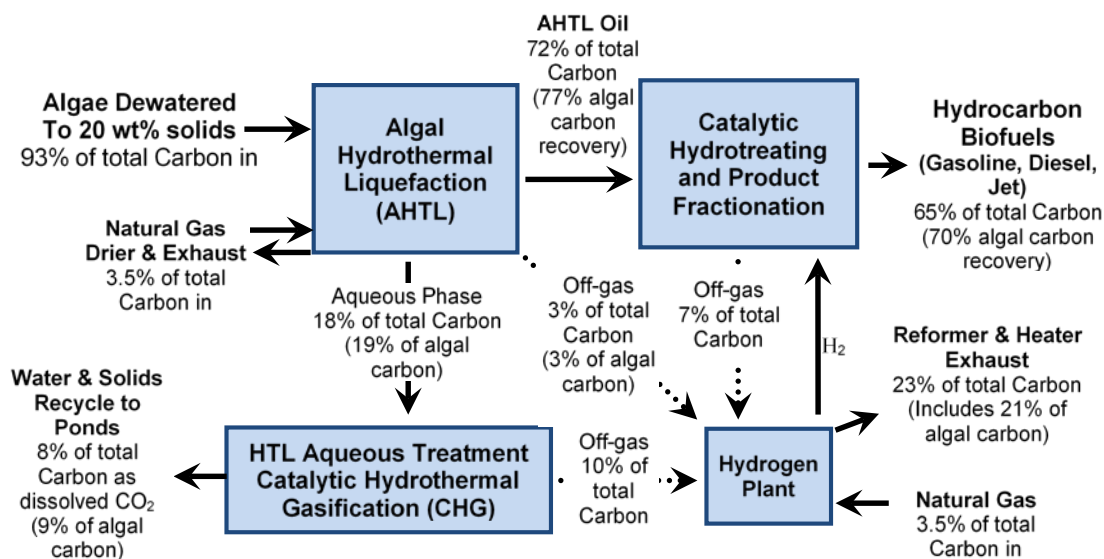


Figure 4. Process block diagram with carbon flow in AHTL process (Figure ES-1, Jones et al., (2014))

Table 2. Key parameters for algal biomass production

Parameter	Value	Source
Algal productivity		
Summer	49.9 g / m ² ·d	Davis (2014)
Fall	28.3 g / m ² ·d	Davis (2014)
Winter	9.3 g / m ² ·d	Davis (2014)
Spring	32.5 g / m ² ·d	Davis (2014)
Average	30 g / m ² ·d	Jones et al. (2014)
Pond Mixing/Circulation energy	48 kWh/ha·d	Frank et al. (2011)
Retention efficiency of settling tank	90%	Frank et al. (2011)
Efficiency of DAF unit	90%	Frank et al. (2011)
Centrifuge efficiency	95%	Frank et al. (2011)
Overall harvesting efficiency ^a	>99%	Davis et al. (2014)
Energy to pump water to site	4.37e-4 kWh/L	Davis et al. (2014)
Energy to pump culture to site	2.5e-5 kWh/L	Frank et al. (2011)
Energy for DAF	1.33e-4 kWh /dry g algae ^b	Davis et al. (2014)
Centrifuging energy	1.93e-5 kWh/g-afdw out ^c	Davis et al. (2014)
Solids concentration from centrifuge	20%	Frank et al. (2011); (Jones et al., 2014)
Nutrient N demand	0.0064 g/g afdw algae	From mass balance ^d
Anhydrous ammonia for N makeup	7.04e-3 g/g afdw algae	From mass balance
Nutrient P demand	0.0007 g/ g afdw algae	From mass balance
Diammonium phosphate for P and N makeup	2.96e-3 g/g afdw algae	From mass balance
Sulfuric acid	3.2 g/g-P recovered	Davis et al. (2014)

^a Assuming biomass lost from the dewatering stage is returned to the pond; ^b whole biomass basis; ^c afdw-ash free dry weight basis; ^d mass balance of nutrients from recycle and biomass composition

The TEA of the AHTL conversion process sized the conversion facility based on an average algal productivity of 30 g/m²·d for four seasons. These seasonal algal productivity projections (Table 2) were made by NREL for highly productive microalgae (Jones et al. 2014; Davis 2014). The conversion unit was sized to process biomass throughout the year without idling during the slower growing seasons. The biomass flows vary by approximately by 5:1 (Jones et al. 2014) from summer to winter. To address this disparity, Jones et al. (2014) assumed that part of the dewatered biomass from summer (approximately 30%) is dried and stored, then processed in winter along with the winter produced biomass. Thus, biomass processed in winter includes two flows, one from winter and the other from summer, which has been dried. The average supply chain GHG emissions from the annual operation are estimated from fuel-weighted average of the GHG emissions from each season.

2.2 Algal Biomass to RD Conversion

Conversion from algal biomass to RD via the AHTL process, as reported by Jones et al. (2014), is shown in Figure 5. Biomass conversion occurs with the following integrated processes: hydrothermal liquefaction (HTL), hydrotreating (HT) and hydrocracking, and hydrogen generation. Algal biomass is converted into liquids (bio-oil and aqueous phases), solid, and gas streams via the HTL process. The aqueous portion of the liquid product from HTL is treated by catalytic hydrothermal gasification (CHG). The organic phase (bio-oil) of the liquid product is sent to catalytic upgrading by HT to remove excess oxygen and nitrogen. Heavier portions of the hydrotreated oil are converted into diesel components with hydrocracking. Upgrading and cracking/fractionation requires hydrogen, which is provided on site by steam reforming using off-gases from HTL, HT, and CHG, along with supplemental natural gas. The solid phase from HTL is filtered and treated for phosphorus recovery. Nutrients recovered from the conversion process are sent back to the growth ponds (Frank et al., 2013).

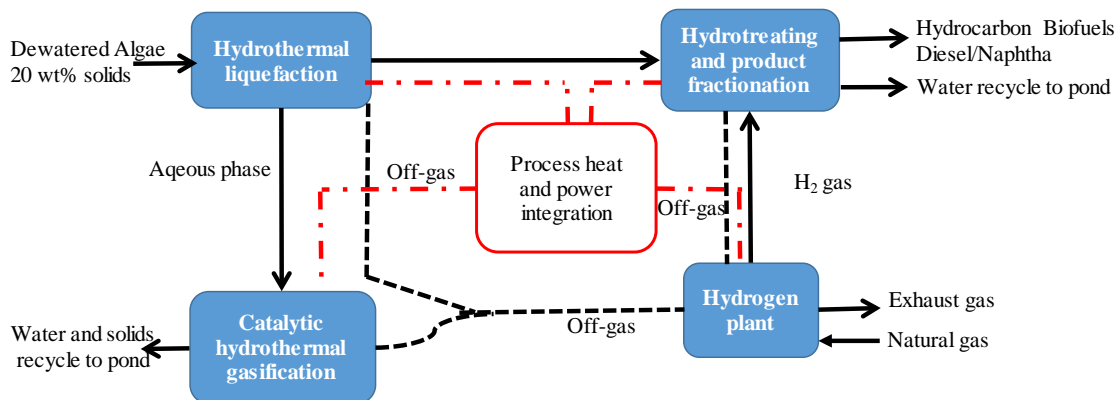


Figure 5. Process flow for the AHTL conversion process

The analysis presented here did not model Figure 5 explicitly. Instead, the rolled-up energy values from the Jones et al. (2014) Aspen model were used as the basis of the SCSA analysis. Table 3 shows the rolled-up values. Values, expressed per gram of equivalent RD produced (in Table 3), are obtained from the PNNL design report and email communications (Tables 11 & 14, Figure ES-1 (Jones et al., 2014; Jones, 2014)). The two products from the upgrading step are naphtha and renewable diesel. In this analysis, naphtha is treated as a fuel product. It is expressed in terms of its energy content via its lower heating value (LHV) and is combined with the RD LHV yield to represent the total fuel energy output. In cases where the product must be considered on a mass basis, the total fuel-energy is expressed as an

“equivalent RD mass” (RD_e) by dividing the total fuel energy (RD plus naphtha) by the RD density. Naphtha accounts for 16.3% of the total RD equivalent output on an energy basis.

Table 3. Parameters for algal biomass conversion to RD_e . See text for definition of RD_e .

Parameter	Value
Yield	146 gal RD_e / ton of ash free dry biomass
Mass of ash free dry biomass/ mass of RD_e	2.06
Net electricity consumed*	2.21e-4 kWh/g RD_e
Net natural gas consumed ^{*a}	8.20e-04 kWh/g RD_e
Net natural gas consumed ^{*b}	2.53e-3 kWh/g RD_e
Gaseous CO ₂ recovery	1.054 g CO ₂ / g RD_e
Dissolved CO ₂ recovery	0.498 g CO ₂ / g RD_e

*After using electricity and heat produced with in the conversion process; ^aWithout natural gas for drying of dewatered biomass in summer; ^bWith natural gas for drying of dewatered biomass in summer

Catalysts are used in hydrotreating, hydrocracking, steam reforming, and CHG processes, but have not been included in the SCSA because the catalyst lifetime, rates of recycle, regeneration, and net consumption are not yet known for this system. Information is also needed about material input and recovery, along with the energy used during catalyst regeneration and recycling. The addition of catalysts to the analysis may affect the SCSA results (Dunn et al., 2013).

2.3 RD Transportation, Distribution and Use

RD produced from microalgae utilizing the AHTL conversion process can be transported by pipeline, barge and rail. Table 4 shows the key parameters for this stage.

Table 4. GREET parameters for RD transportation and distribution

Parameter	Renewable Diesel		
	Barge	Pipeline	Rail
Transportation to terminal by mode and their share	8.0%	63%	29%
Transport distance for transportation to terminal (miles)	520	400	800
Payload (tons)	20,000	NA	NA
Energy intensity of pipeline transportation (BTU/ton-mile)		404	
Transportation distance between bulk terminal and fueling station, traveled by truck	30 miles		

3. AHTL Pathway Results

Recent work has demonstrated that algae analyses must consider seasonal variations in biomass production (Davis et al., 2014). Therefore, this analysis considered a seasonal estimation of the supply chain GHG emissions. Table 5 presents the supply chain total fossil (e.g., coal, natural gas, petroleum) and total petroleum energy use estimated in this analysis for each season. Two sets of results were generated for the summer productivity, one set with NG consumption for drying included, and one set without. The differences in fossil energy use, petroleum use, and GHG emissions between the two sets of

results are the consequence of consuming natural gas for drying and are added to the summer burdens. The breakdown of direct process energy demand (purchased energy) for each season is presented in Figure 6.

When compared to biomass produced in other seasons, the winter-produced biomass required more purchased energy during the algae growth and 1st dewatering stage because of the higher mixing energy requirement per mass of algae, which is a result of the low winter biomass productivity. The energy consumption for biomass storage in the design case comes entirely from drying. Refrigeration was assumed to be unnecessary in the Aspen model and other warehousing energy demands were expected to be negligible (Jones et al., 2014). High protein *Spirulina* has been stored at PNNL for five years without spoilage (temperature range of 60 – 80°F). Refrigeration, if required in the Gulf region, would likely add significantly to the energy demand and GHG emissions (Jones et al., 2014; Jones, 2014).

The seasonal supply chain GHG emissions, along with corresponding seasonal productivities, are presented in Table 6. The GHG emissions for the yearly fuel-weighted averages are presented in Table 7, along with the results of the harmonization study using the AHTL conversion process (Davis et al., 2014) and LS petroleum diesel (Argonne National Laboratory, 2013).

The results can be compared with the harmonization study, Davis et al. (2014), which assumed a lower productivity at 14.6 g/m²·d based on today’s productivity performance estimates compared to future target projection of 30 g/m²·d used in this analysis. The natural gas consumption in the present analysis is 1.2 times higher than Davis et al. (2014) when not drying the algae and is 3.6 times higher when drying is included. On the other hand, the electricity demand is 21% lower in this analysis. In total, the GHG emissions are 11% lower in the present study than the harmonization study (Davis et al., 2014).

The results presented here are the fuel-cycle results. Infrastructure-cycle emissions will increase the total GHG emissions associated with RD. The infrastructure emissions are amortized over the biomass produced during the equipment lifetime and thus depend upon the season through the varying productivity. When materials for biomass growth and dewatering are included and the AHTL equipment and associated infrastructure are ignored, the infrastructure emissions in fall, spring, summer, and winter add 1810, 1640, 1230, and 2500 gCO₂e/MMBTU RD_e (including plastic pond liners for the cultivation step). Without pond liners, the infrastructure emissions add 1170, 1080, 869 and 1530 g CO₂e/MMBTU RD_e in fall, spring, summer and winter seasons. Table 7 shows the four-season averages when infrastructure cycle emissions are included.

Table 5. Seasonal supply chain results for RD_e production. Only fuel-cycle emissions are included. See Table 6 and Table 7 for infrastructure-cycle emissions.

Season	Fossil fuel use ^a (BTU/BTU RD _e)	Petroleum use ^a (BTU/BTU RD _e)
Fall	0.383	0.016
Spring	0.375	0.015
Summer	0.514 ^b	0.016 ^c
Winter biomass	0.505	0.019
Summer biomass processed during winter	0.357	0.015

^a Well-to-wheel energy use; ^b Of this, 0.157 BTU/BTU RD_e were required for drying; ^c Of this, 0.001 BTU/BTU RD_e were required for drying

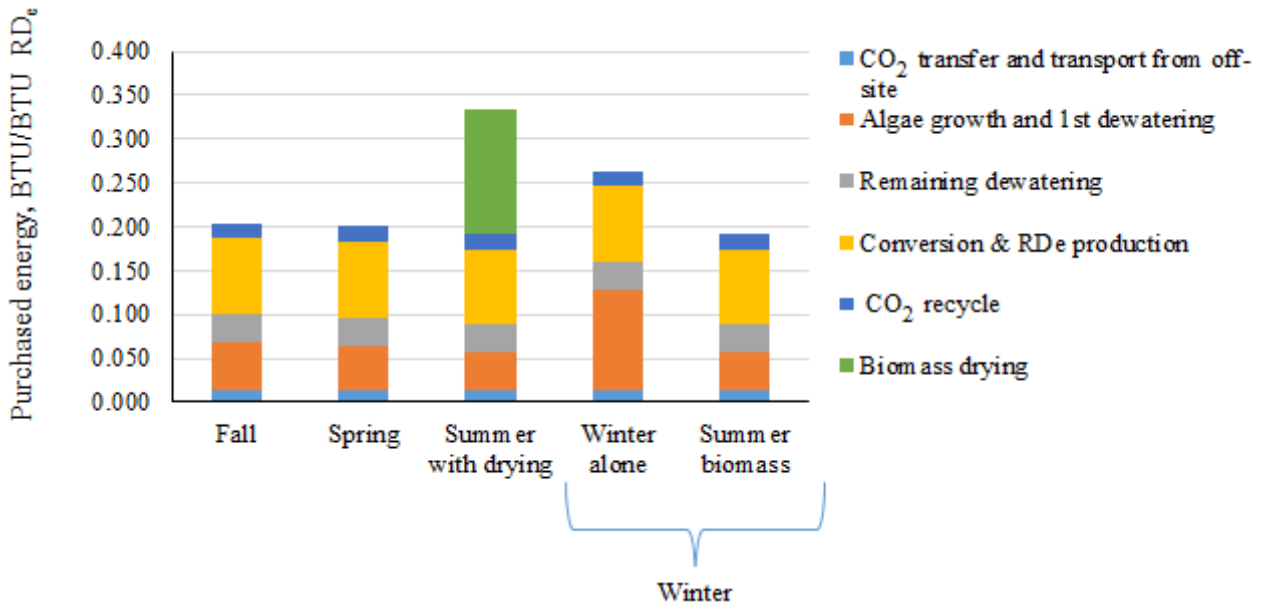


Figure 6. Direct energy use (purchased electricity and natural gas) in each step of the pathway

Table 6. Supply chain GHG emissions, biomass flows, and RD production. “Average winter” is the fuel weighted average of “Winter biomass” and “Summer biomass processed during winter”.

Season	Productivity (g/m ² ·d)	Biomass ^a (kg/hr)	RD _e ^b (MMBTU/season)	Fuel-cycle GHG emissions (gCO ₂ e / MMBTU)	Infrastructure GHG emissions ^c (gCO ₂ e / MMBTU)
Fall	28.3	47,756	2.06E+06	33,000	1,810
Spring	32.5	54,844	2.45E+06	32,200	1,640
Summer	49.9	59,989	2.68E+06	40,900 ^d	1,230
Winter					
Winter biomass	9.3	15,694	6.71E+05	44,300	4,550
Dried summer biomass ^e	49.9	24,218	1.08E+06	30,600	1,230
Fuel-weighted average				35,800	2,500

^a Estimated for 4,050 ha pond area; ^b Hours assumed in Fall, Spring, Summer and Winter are 2160, 2232, 2232 and 2136 hrs. Produced RD_e per season is expressed in MMBTU using LHV of RD; ^c Infrastructure emissions with lined ponds without AHTL equipment; ^d Of this, 10,300 gCO₂e/MMBTU were from drying; ^e ~30% of summer biomass is processed in winter

Table 7. Supply chain GHG emissions for RD production via the AHTL pathway

	Productivity (g/m ² ·d)	Fuel cycle GHG emissions (gCO ₂ e/MMBTU RD _e)	Total GHG emissions (fuel cycle plus infrastructure cycle) (gCO ₂ e/MMBTU RD _e)	Source
Four season average*	30	35,700	37,400	This work
LS petroleum diesel	NA ^a	101,000	NA	Argonne National Laboratory (2013)
Annual average, four seasons ^b	14.6	40,092	NR ^c	Davis et al. (2014)

* The fuel weighted average of emissions from all seasons; ^a NA-not applicable. ; ^bProductivity estimated based on the current performance of algal systems in harmonization study using AHTL process; ^cNot reported.

4. Conclusions

Algal RD has lower GHG emissions, lower fossil energy use, and lower petroleum use than does LS petroleum diesel on a well-to-wheels life cycle basis when the algal RD is produced via the AHTL pathway. The season-based analysis showed that details of the farm operation during winter is critical due to the high supply chain GHG emissions from low biomass production. The effect is the result of both the energy demand for cultivation and the productivity itself. Thus, further research should consider both factors and not just productivity, e.g., reducing energy consumption during biomass cultivation can reduce the winter productivity targets required to achieve good GHG emissions performance. The underlying AHTL model assumes that recovered N is available for recycling to the cultivation steps. This assumption should be explored in more detail. If upgrading is located off-site so that N incorporated in the AHTL oil is recovered off-site, then it may not be possible to recycle this N back to the algae cultivation step. This may lead to higher N demand for the AHTL pathway compared lipid extraction pathways. See Frank et al. (2013) for additional discussion of this issue. Similarly, the model assumes that P can be recycled in a biologically accessible form, but this has not been demonstrated.

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