

Microalgae Hydrothermal Liquefaction and Biocrude Upgrading: 2022 State of Technology

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Summary

A preliminary techno-economic analysis (TEA) was developed for the fiscal year (FY) 2022 state of technology (SOT) assessment to evaluate the benefits and risks of using demonstrated, high-productivity algae strains for fuels generation, including sustainable aviation fuel (SAF). In 2022, the marine algal strain, *Picochlorum celeri*, which demonstrated the highest outdoor biomass productivities reported to date in the DOE-funded open-pond raceway testbed at the Arizona Center for Algae Technology and Innovation (AzCATI) (Krishnan et al. 2021), was tested for continuous hydrothermal liquefaction (HTL) processing at PNNL. HTL testing results demonstrated a biocrude yield of 0.33 g/g algae on an ash-free dry weight (AFDW) basis from *P. celeri*. The hydrotreatment testing of the HTL biocrude from *P. celeri* was also conducted to investigate the production of jet fuel from marine algal biomass. To the best of our knowledge, this is the first report of jet fuel production from autotrophically grown marine algal biomass. The current hydrotreating testing demonstrated approximately 22.7 wt% of the hydrotreated oil within the typical boiling-point range of jet fuel (150–250 °C). Initial testing of the jet fuel cut (JFC) showed that the physical properties under investigation were within typical ranges for petroleum-based jet fuels. The experimental work of this study closes the gap between outdoor algae cultivation and algae conversion to critical transportation fuels using the same algae strain for both cultivation and conversion testing. The continuous HTL and the upgrading testing described herein demonstrate the potential of producing sustainable aviation fuel (SAF) from algae cultivated in open-pond systems using the primary inputs of sunlight and carbon dioxide.

Considering that the 2020 SOT focused on the investigation of a two-stage HTL conversion process and the 2021 SOT emphasized using wastewater-grown algae as the HTL feedstock, the results of the 2022 SOT were instead compared to the results of the 2019 SOT since both cases assumed open-pond cultivated algae and a single-stage HTL conversion process.

Table 1 summarizes the major changes of the 2022 SOT compared to the 2019 SOT case. Algae biomass with forest residue supplement during the lower algae productivity seasons (winter, fall, and spring) to match the algae production rate in the summer season was employed in the 2022 SOT to maintain consistent plant capacity in all seasons. The same assumption was also employed in the 2019 SOT. For the 2022 SOT, the SAF production was estimated based on experimental results for JFC generation in the upgrading process. Nitrogen level in the JFC from the hydrotreated oil was significantly higher than that of conventional petroleum-based jet fuel, which is reported to contain near zero to 20 ppm nitrogen (Hemighaus et al. 2007). Therefore, a hydrodenitrogenation (HDN) step was assumed to remove over 99% nitrogen in the JFC from the hydrotreated oil. The fuel after the HDN processing was assumed to be the final SAF product.

The total fuel yield of the 2022 SOT is estimated to be 83 GGE/ton feedstock at ash free dry weight (AFDW) basis, which includes a SAF yield at 25 GGE/ton. The minimum fuel selling price (MFSP) for the 2022 SOT is \$5.42/GGE, which is approximately 9% higher than the value reported in the 2019 SOT because of the lower biocrude yield and associated lower yields of the final fuel. The conversion cost (not including feedstock cost) of the 2022 SOT is \$0.36/GGE, which is 59% lower than that was reported in the 2019 SOT, \$0.88/GGE. The major reason for cost reduction is the high value of nutrient recycle credits. The cost results demonstrated great potential of using demonstrated high-productivity marine algae strains for SAF production.

Table 1. Major changes of the 2022 SOT case compared to 2019 SOT case

Changes	2022 SOT	2019 SOT	Effects and Reasons
Feedstock	Algal strain: <i>Picochlorum celeri</i> . The conversion throughput is 662 tons/d ash-free dry weight (AFDW) for algae with wood supplement; the algae feedstock flow rate is 423 tons/d (annual average). The algae feedstock cost is \$602/ton AFDW.	Algae strain: <i>Chlorella</i> sp. The conversion throughput is 598 tons/d AFDW for algae with wood supplement; the algae only flow rate is 350 tons/d (annual average). The algae feedstock cost is \$670/ton AFDW.	Higher algae production rate of the 2022 SOT leads to higher conversion plant throughput and thus cost reduction through economies of scale; Lower algae feedstock cost leads to lower blended feedstock cost and thus lower variable operating cost.
Biocrude yield	0.33 g/g feedstock AFDW	0.41 g/g feedstock AFDW	Algae assumed in the 2022 SOT has high protein and low lipid contents, which lead to lower biocrude yield and thus lower final fuel production rates than previous SOT reports. Lower fuel yield leads to higher production cost per unit of fuel.
Biocrude upgrading	0.07 g H ₂ /g dry biocrude; SAF production is tested and evaluated	0.05 g H ₂ /g dry biocrude; Production of diesel and naphtha only, no SAF production	The high-protein algal feedstock assumed in the 2022 SOT leads to high nitrogen content in the biocrude and thus higher hydrogen consumption for biocrude upgrading; The cost impact of isolating JFC and adding HDN unit for SAF production is insignificant.

Acronyms and Abbreviations

AFDW	ash free dry weight
DCN	derived cetane number
FY	fiscal year
GGE	gasoline gallon equivalent
GHG	greenhouse gas
HOC	heat of combustion
HDN	hydrodenitrogenation
HHV	higher heating value
HTL	hydrothermal liquefaction
JFC	Jet fuel cut
LCI	life-cycle inventory
LHSV	liquid hourly space velocity
MBSP	minimum biomass selling price
MFSP	minimum fuel selling price
NREL	National Renewable Energy Laboratory
PNNL	Pacific Northwest National Laboratory
SOT	state of technology
SAF	sustainable aviation fuel
TEA	techno-economic analysis
WHSV	weight hourly space velocity

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1.0 Introduction

The goal of the U.S. Department of Energy (DOE) Bioenergy Technologies Office (BETO) is to develop commercially viable bioenergy and bioproduct technologies to enable sustainable, nationwide production of biofuels that are compatible with today's transportation infrastructure, can reduce greenhouse gas emissions relative to petroleum-derived fuels, and can displace a share of petroleum-derived fuels to reduce U.S. dependence on foreign oil (DOE 2020). To meet this goal, techno-economic analyses (TEAs) have been developed to evaluate the impacts of the research and development progresses on sustainable production of renewable fuels from biomass conversion through the annual state of technology (SOT) assessment.

In 2021, the U.S. DOE, the U.S. Department of Transportation (DOT), the U.S. Department of Agriculture (USDA), and other federal government agencies issued a Memorandum of Understanding to develop a comprehensive strategy for scaling up new technologies to produce sustainable aviation fuel (SAF) on a commercial scale (DOE 2021). The proposed strategy resulted in the SAF Grand Challenge, which includes: (1) Achieving a minimum of a 50% reduction in life cycle greenhouse gas (GHG) emissions compared to conventional fuel; (2) meeting a goal of supplying sufficient SAF to meet 100% of aviation fuel demand or 35 billion gallons per year, by 2050. A near-term goal of 3 billion gallons per year is established as a milestone for 2030. To meet these goals, research work needs to be done to explore renewable feedstock sources and conversion technologies for SAF production.

Microalgae have been demonstrated to be a promising renewable feedstock for biofuel production (Jones et al. 2014, Zhu et al. 2021). However, limited work has been reported for the production of jet fuel via microalgae conversion. A survey of available published work showed a focus on the hydroprocessing of microalgal lipids to produce jet fuel. Fortier et. al. (2014) implemented a life cycle assessment (LCA) of bio-jet fuel production from HTL conversion of wastewater-grown algae based on lab-scale batch HTL testing data and a simulated hydrotreating process. Their results showed a GHG emissions reduction by 76% compared to petroleum jet fuel based on a biocrude yield of 0.445 g/g algae AFDW and 90% conversion (assumed) of biocrude to bio-jet fuel. Bwapwa et. al. (2018) investigated the lab-scale cultivation of *Nannochloropsis sp.* and its conversion to jet fuel via oil extraction and then thermal cracking. Their work demonstrated that most physico-chemical parameters of the jet fuel product were within the range prescribed by ASTM standards, except freezing point and density. Gutiérrez-Antonio et. al. (2018) simulated a hydrotreating process of microalgae oil from a modified strain of *Chlorella sp.* to generate aviation fuel based on their experimental data. Their results showed 34% reduction in CO₂ emissions and 78% lower selling price compared to those of the petroleum jet fuel. Lim et al. (2021) reviewed the hydroprocessing method for converting microalgae oil into bio-jet fuel, as well as the gasification process with Fischer-Tropsch and sugar-to-jet technologies for directly converting microalgae to jet fuel. In 2021, Honeywell announced its UOP Ecofining™ technology supported the world's first jet flights using SAF produced from algal oil (Honeywell 2021). The Ecofining™ technology converts triglycerides via a pressurized catalytic hydrodeoxygenation reaction and a selective cracking step to reduce carbon chain lengths.

Although algae oil conversion to SAF has been commercialized, the conversion of whole algae to SAF is still in very early phase of development and implementation. Converting only algal oil part to SAF constrains the fuel production from other intracellular compounds, such as carbohydrates, which can also be converted to biocrude and then SAF with further treatment. In order to meet the goal of the SAF Grand Challenge, more research is needed to develop and scale up new technologies to produce SAF from whole algae on a commercial scale. Therefore, for the fiscal year 2022, the PNNL testing team conducted continuous HTL and biocrude upgrading experiments to investigate the possibility of generating SAF from outdoor-cultivated algae. The tested alga, *P. celeris*, was cultivated in open pond facilities and

consistently demonstrated the highest outdoor biomass productivities reported to date for the DOE DISCOVER consortium project (Klein and Davis, 2022). The open pond algae cultivation coupled with algae conversion testing demonstrated a realistic pathway for commercialization of algae-to-SAF production. Based on the testing work, the 2022 SOT case study is developed to evaluate the cost impacts of converting cultivated algae to biofuels, including SAF. In this report, the major experimental results are summarized, which provide the design basis for this study. The system evaluated in this study is overviewed and detailed information for feedstock and process assumptions are provided. Major cost results of this study are presented and compared to the previous SOT studies.

2.0 Experimental Work in Fiscal Year 2022

The fiscal year (FY) 2022 research efforts for HTL and hydrotreating are described in this section.

2.1 HTL

A frozen slurry of *Picochlorum celeri* was received from the Arizona Center for Algae Technology and Innovation (AzCATI) and characterized for solids and proximate content (Krishnan et al. 2021). Figure 1 shows the slurry as received (20.3 wt % biomass solids), the dried alga, and the algal ash. Table 2 includes the proximate analysis of *P. celeri*. The proximate analysis for a commercially grown *Spirulina* is reported in Table 2 for reference.

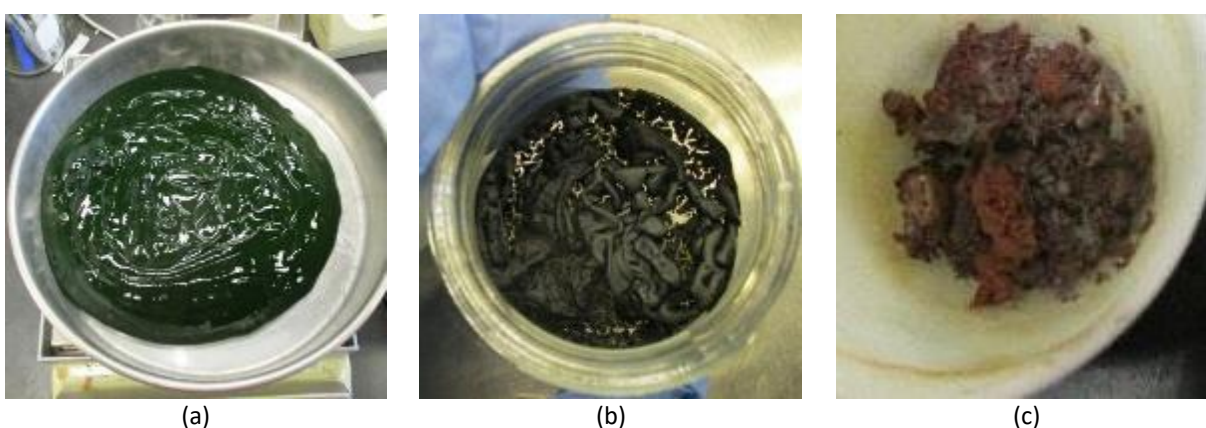


Figure 1. *Picochlorum celeri*, a) as-received, b) oven-dried, c) ash

Table 2. Proximate analysis of *Picochlorum celeri* and *Spirulina*

Composition (wt %)	<i>Picochlorum celeri</i>	<i>Spirulina</i>
Ash	17.3	12.3
Carbohydrate (ash-free)	20.4	20.1
Protein (ash-free)	72.6	71.2
Lipid (ash-free)	7.0	8.7

The *P. celeri* slurry (20.3 wt % total solids) was processed in the bench scale HTL system configured in a plug-flow reactor mode at a liquid hourly space velocity (LHSV) of 4 L/L/h. The reactor setpoint temperature was 350 °C. The design of the bench-scale system has been described previously (Elliott et al. 2013). The viscosity of the feed slurry required use of a pressurized feed tank to charge the high-pressure pumps, instead of feeding from a bucket with a diaphragm pump for typical slurries. Figure 2 shows the biocrude and water recovered from a product sample. The photo shows an ill-defined oil/water interface making the separation challenging.



Figure 2. Biocrude-water separation during HTL processing of *Picochlorum celeri*.

Mass and carbon yields, shown in Figure 3, are similar to those observed for *Spirulina* in a previous run, and typical for that of other high-protein algae. Biocrude compositions are provided in Table 3. With the exception of the higher moisture content in the *P. celeri* biocrude, the biocrudes are similar. The biocrude mass yield was 33% with a carbon recovery of 46% in the crude. These results are typical compared with other high-protein species of microalgae, such as *Spirulina*. High iron, potassium, sodium, and silicon are found in the *P. celeri* biocrude. These elements must be reduced or removed before hydrotreating by using a guard bed.

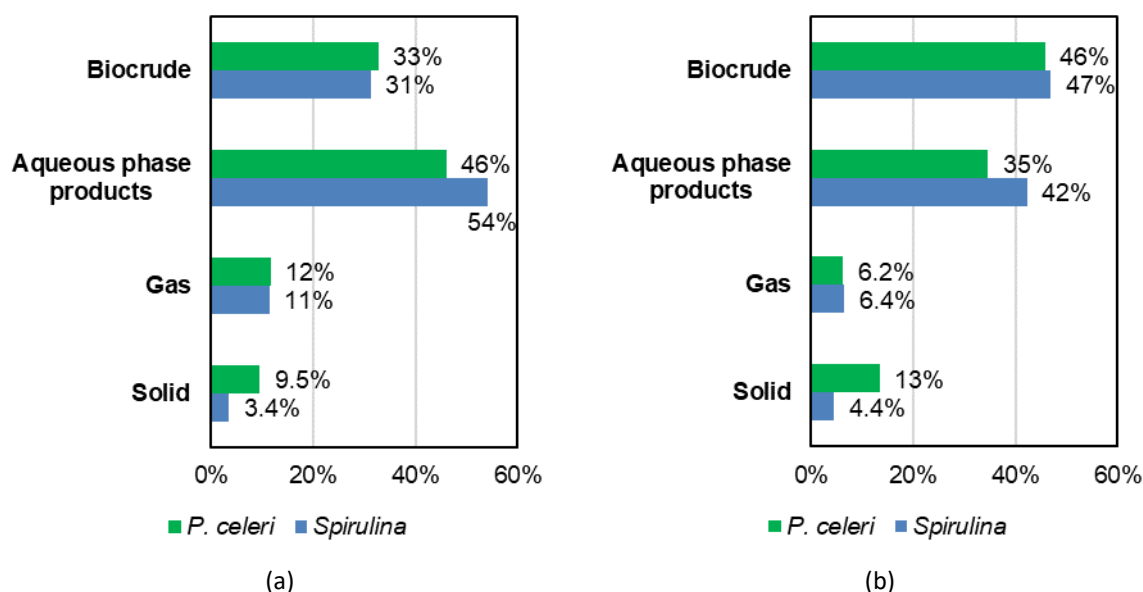


Figure 3. a) Yields from hydrothermal liquefaction of *Picochlorum celeri* and *Spirulina* a) mass yields (AFDW basis) and b) carbon yields (AFDW basis)

Table 3. Biocrude composition and physical properties

Parameter	Unit	<i>Picochlorum celeri</i>	<i>Spirulina</i>
Carbon	wt %	77%	75%
Hydrogen	wt %	9.6%	9.5%
H:C	mol ratio	1.49	1.52
HHV	MJ/kg	31.8	36.8
Oxygen	wt %	4.6%	5.0%
Nitrogen	wt %	7.1%	8.6%
Sulfur	wt %	0.88%	1.04%
TAN	mg _{KOH} /g _{oil}	44	47
Density	g/ml	0.99	0.99
Viscosity	cSt@40C	281	279
Moisture	wt %	14.1%	5.6%
Ash	wt %	1.07%	0.90%
Filterable Solids	wt %	0.91%	0.54%

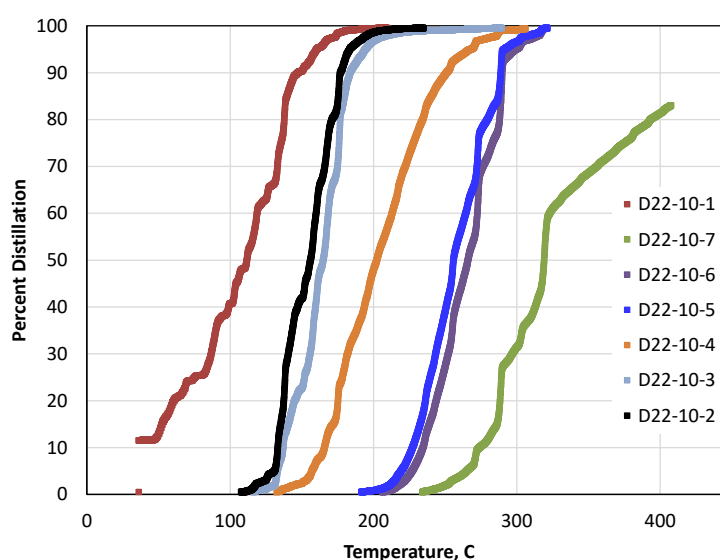
2.2 Hydrotreating

The hydrotreating testing in FY 2022 was focused on generation and evaluation of hydrocarbon fractions that are within range for SAF from the hydrotreated HTL biocrude. The biocrude from *P. celeri* was upgraded via hydrotreatment in a continuous trickle-bed reactor (PNNL's pilot scale hydrotreating unit). The hydrotreater was configured as a single pass, co-current, continuous, down-flow reactor. The system was operated at 1500 psi, with two zones, held at 350 and 400 °C. The system consists of a gas and liquid feed system, heated reactor system, and a gas-liquid separation system. The tubular fixed-bed catalytic hydrotreater was made of 316 stainless steel ($1/2$ in., outer diameter by 64 cm long with 40 mL capacity) and heated by a two-zone heater for two-stage hydrotreatment. The liquid feedstock and hydrogen gas entered the top of the catalyst bed and passed downward through the bed, assumed to be in a trickle flow. After exiting the catalytic reactor, the liquid products were separated from the gaseous products in one of two pressurized and cooled traps placed in parallel flow, downstream of the reactor system. Periodic liquid samples were collected when switching collection vessels and venting/draining the trap. The recovered liquid products were phase-separated, weighed, and sampled for further analysis. The sample was hydrotreated at a WHSV of 0.5 hr^{-1} with a NiMo catalyst. A separate guard bed of CoMo was placed at the front of the hydrotreatment reactor to remove and reduce metal impurities, protecting the catalyst bed. All catalysts were whole extrudates to mimic scalable and industrially relevant catalyst manufacturing practices. The reactor also included SiC inert packing to ensure plug flow and isothermal behavior.

The upgraded fuel sample was distilled at atmosphere by using PNNL's bench distillation unit and the results were shown in Table 4. The boiling point range of each distillation cut was verified via simulated distillation. Figure 4 shows the simulated distillation curves of the cuts 1 to 7 (D22-10-N; N is 1 to 7 corresponding to the cut numbers in Table 4). The simulated distillation verified that 22.7 wt% of the upgraded biocrude (cuts 2 to 4) was within the jet fuel range (150-250 °C) based on Olsen (2014). Based on the distillation results, 18.3 wt% of the upgraded biocrude is naphtha cut (< 150 °C, cut 1), about 39 wt% is in diesel range (250 to 340 °C, cuts 5, 6 and part of 7) and 20 wt% is the heavy oil (> 340 °C, part of cut 7).

Table 4. Distillation ranges and the corresponding fractions of the cuts

Cut	Temperature	wt %
1	25 – 150 °C	18.3
2	150 – 160 °C	2.9
3	160 – 165 °C	1.9
4	165 – 250 °C	17.9
5	250 – 260 °C	2.1
6	260 – 265 °C	1.3
7	>265 °C	55.5

Figure 4. SIMDIS of the upgraded biocrude distillation cuts from *Picochlorum celeri*.

Distilled cuts 3 and 4 were combined to represent jet range fuel and sent to the University of Dayton for Tier α and β testing (combined for 19.8% of the fuel sample). Tier α and β testing are a low-volume testing methodology for assessing the key characteristics of jet fuel samples and providing an initial screening for SAF candidacy (Heyne et al. 2021). Figure 5 shows characterization results of the JFC sample compared to the average results for petroleum-based jet fuel. Shaded green portions show the ranges for conventional jet fuel. The red lines denote the specification limit and shaded red portions fall outside the allowable limits for the specification. Figure 5a shows the mass distribution of molecules classified by carbon number and hydrocarbon classification (e.g., aromatics, n-alkanes, isoalkanes). The distributed profile of the algae-based JFC was within the average range for the petroleum-based jet fuel, and its average carbon number (11.5) is very close to the one for the petroleum-based jet fuel (11.4). Figure 5b shows the distillation profile of the sample. The profile was well within the specification limits and falls within the typical range for petroleum-based jet fuel.

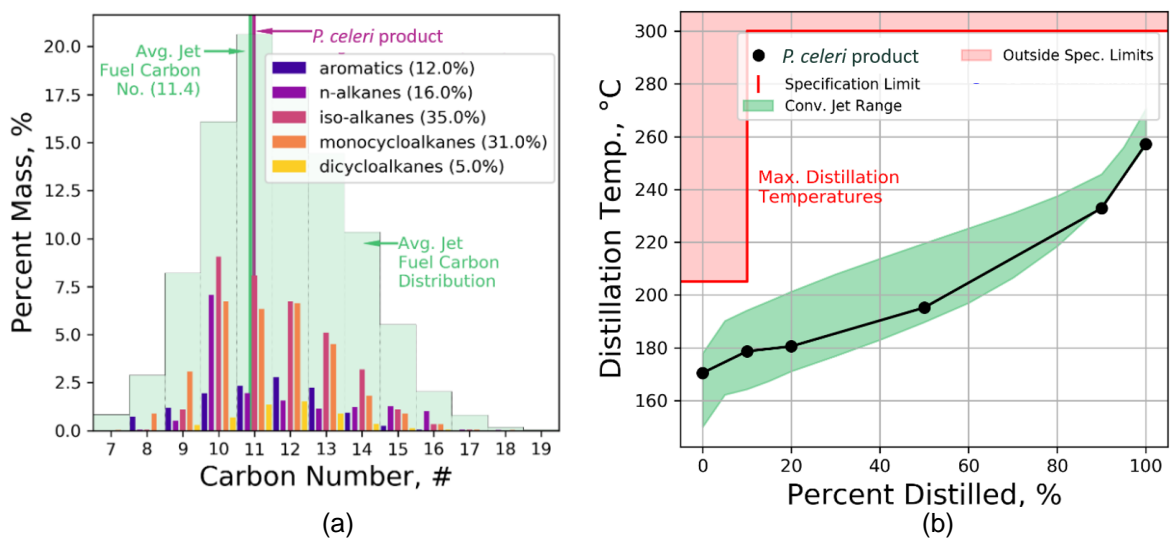


Figure 5. Characterization results from JFC sample derived upgraded biocrude from *Picochlorum celeri* showing a) carbon number and hydrocarbon type distribution and b) distillation profile.

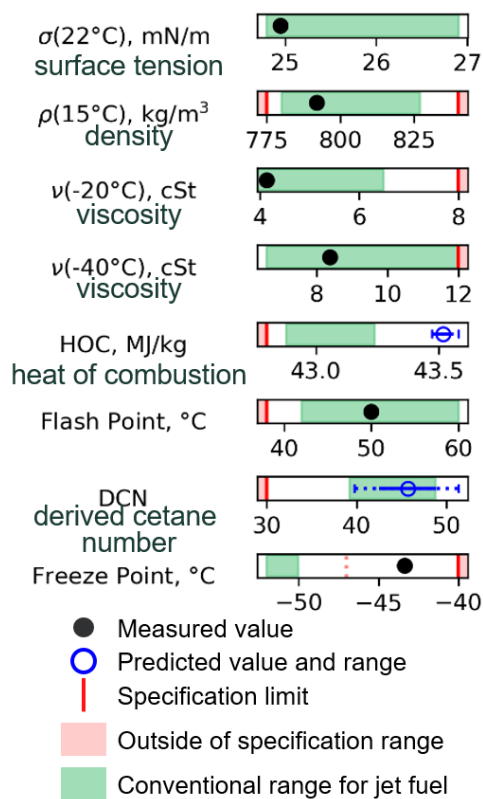


Figure 6. Thermophysical and compositional properties of the JFC sample derived from upgraded biocrude from *P. celeri*. Note: HOC and DCN are estimated from data and not directly measured.

Figure 6 shows measurements of key thermophysical and compositional parameters that are relevant to the assessment of jet fuel. The number bars are color coded similarly to the graphs in Figure 5. Values for surface tension (σ), density (ρ), viscosity (ν), flash point, and derived cetane number (DCN) fall within the range for conventional jet fuel. Heat of combustion (HOC) and freeze point are within specification but just outside the typical range. Adjusting the properties of the algal SAF in this study to be within the typical ranges for jet fuel may be achieved by optimizing the processing parameters for biocrude upgrading and distillation or blending the algal SAF with another jet fuel (conventional or SAF). Overall, the results show the potential of a high-protein algae yielding a potentially viable SAF product via HTL. Preliminary results for Tier α and β testing show that physical properties are within specification for jet fuel qualification.

3.0 Process Inputs and Assumptions

In this section, the major design inputs and assumptions for the algae HTL and biocrude upgrading system for the 2022 SOT study are described.

3.1 System Overview

Figure 7 shows the block flow diagram for an algae/wood blend feedstock conversion via HTL and a biocrude upgrading system investigated in this study. In the modeled commercial-scale plant, algae blended with woody biomass slurry is pumped to the HTL reactor. In the HTL reactor, condensed phase liquefaction takes place through the effects of time, heat and pressure. The resulting HTL products (biocrude, solid, aqueous, gas) are separated. The biocrude is upgraded to generate diesel, jet fuel and naphtha range fuels. The jet cut is further sent to an HDN unit to reduce nitrogen to trace levels to produce a SAF quality product. The HTL aqueous phase is assumed to be recycled directly to the algae farm. The gas stream is used for process heating and hydrogen generation. A hydrogen plant is included for hydrotreating, which is assumed to be co-located with the HTL conversion. Nutrients recovered by acid extraction of the HTL solids are recycled to the pond along with the HTL and the hydrotreating processes aqueous streams. Flue gas containing carbon dioxide is also assumed to be recycled to the farm to provide carbon elements for algae growth.

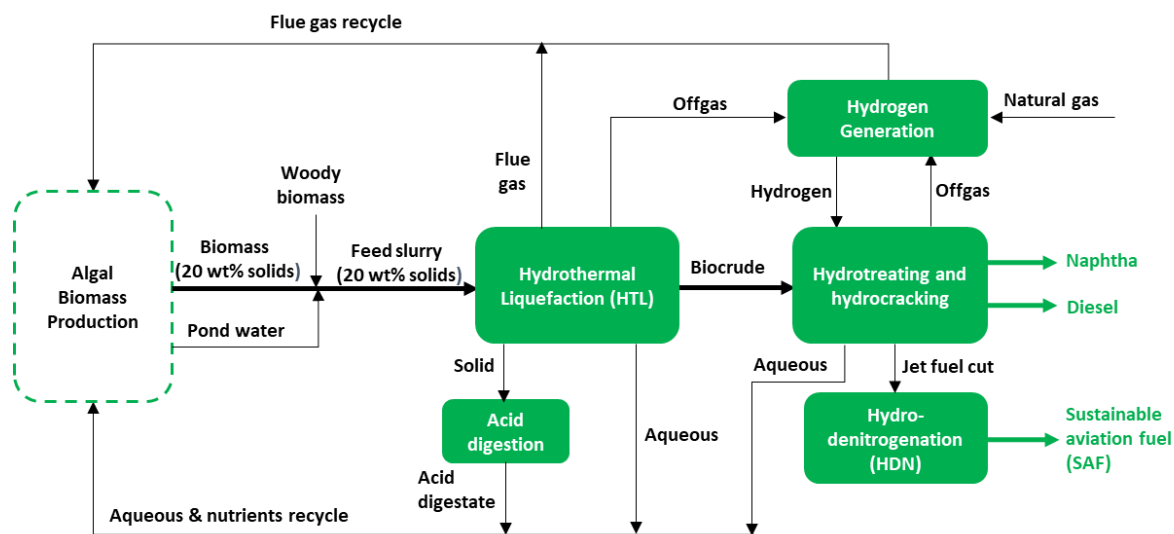


Figure 7. Simplified block diagram of algae/wood blend feedstock HTL and biocrude upgrading system

3.2 Feedstock

Algae biomass with woody biomass supplement is employed in the 2022 SOT to maintain a constant plant capacity in all the seasons. During the lower algae productivity seasons (winter, fall, and spring), the woody biomass provides supplemental feed to match the peak production rate of algae in the summer. The same assumption has been employed in the 2017 to 2020 SOT studies. The conversion plant throughput in this study matches the maximum algae production rate in summer. Algae supplemented with non-algae feedstock in lower productivity seasons have the following advantages: elimination of the extra cost for algae drying and storage in high productivity (spring/summer) seasons, increased annual plant throughput, and reduced overall feedstock cost by blending a lower cost feedstock. The details have

been discussed in previous TEA studies (Zhu et al. 2020a&b). The feed flowrates of algae and supplemental woody biomass to the conversion plant at seasonal and annual average basis are calculated and shown in Figure 8. The algae seasonal flow rates to the conversion plant and the associated minimum biomass selling price (MBSP) for dewatered algae (80 wt% moisture) are provided from the NREL 2022 SOT algae cultivation model for a 5000-acre open pond. The annual average flowrate for the blended feedstock is 662 US tons/d at AFDW basis. The annual average mass ratio for algae/wood feedstock is 64/36. The MBSP for algae without storage is 602 \$/ton AFDW (2016 US\$). The woody biomass feedstock cost is assumed to be \$70.31/dry ton based on data from Hartley et al. (2021).

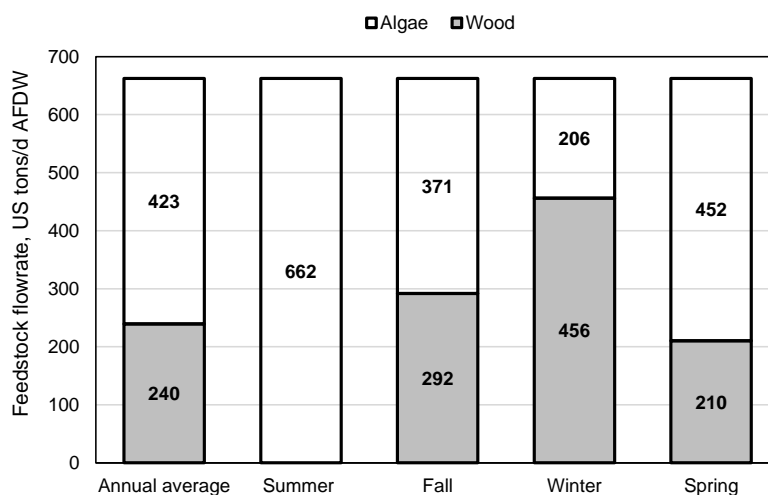


Figure 8. Feedstock seasonal and annual average flow rates.

In this study, *P. celeris* is assumed to be the algae feedstock to match the HTL testing work and the non-algal supplemental feedstock is assumed to be forest residue based on our previous algae/wood blended feedstock HTL testing (Zhu et al. 2020a). Forest residue is a byproduct from forest harvesting, consisting of leaves, barks, trunk, and branches (Rudra and Jayathilake 2022). Table 5 lists the elemental compositions for the algae and woody biomass assumed in this study.

Table 5. Elemental composition for algal and woody feedstocks.

Elements, wt% ash free dry weight (AFDW) ¹	Algae (<i>P. celeris</i>)	Woody biomass (forest residue)
Carbon	53.7	50.0
Hydrogen	7.2	6.2
Oxygen	26.5	43.6
Nitrogen	11.3	0.2
Sulfur	1.3	0
Total	100.0	100.0
Ash, wt% dry basis	15.9	1.0 ²
Phosphorus (in ash)	1.6	0

Notes: ¹ Elemental compositions reported on a dry ash free basis were calculated from dry basis data from PNNL measurements. ² Ash content of the woody biomass is assumed to be 1% based on 2016 MYPP (U.S. DOE 2016).

Based on the NREL 2021 SOT algae cultivation report (Klein and Davis 2022), maximum algae biomass generation is based on measurements for seasonally rotated strains (*Picochlorum celeri*, *Tetraselmis striata*, and *Monoraphidium minutum*), grown in September–October (fall) and June–August (summer), November–February (fall through winter), and March–May (spring), respectively. The above algae strains demonstrated high productivities in the specified seasons. However, HTL conversion testing for *Tetraselmis striata*, and *Monoraphidium minutum* have not been conducted yet and thus *P. celeri* is assumed to be the only algae feedstock for the 2022 algae HTL SOT. To close the gap between the inputs from the NREL algae cultivation model and the feedstock assumptions for the algae HTL conversion model, *Tetraselmis striata* and/or *Monoraphidium minutum* biomass should be tested for HTL conversion and the results can be integrated into future SOT evaluations.

Based on varying research direction and purpose for each annual SOT report, different feedstocks and processes were evaluated from 2017 to 2022, as shown in Table 6. The 2017 to 2019 SOT reports evaluated the freshwater alga (*Chlorella* sp.) blended with wood for single-stage HTL conversion and the 2020 SOT tested another freshwater alga (*Scenedesmus obliquus*) blended with corn stover for two-stage HTL conversion. The 2021 SOT is based on the single-stage HTL testing of wastewater-grown algae.

Table 6. Algal elemental and biochemical compositions assumed in the SOT and target cases.

SOT Time Period	2017 to 2019	2020	2021	2022
Elemental composition, wt% AFDW	<i>Chlorella</i> sp.	<i>Scenedesmus obliquus</i>	Wastewater-grown algae	<i>Picochlorum celeri</i>
Carbon	53.8	53.5	51.3	53.7
Hydrogen	7.5	7.1	6.8	7.2
Oxygen	30.8	28.6	31.5	26.5
Nitrogen	7.2	8.4	7.8	11.3
Sulfur	0.6	2.38	2.5	1.3
Total	100	100	100	100
Ash, wt% dry basis	13.9	10.5	39	15.9
Phosphorus (in ash), wt% of dry feed	0.3	1.1	3.3	1.6
Biochemical composition, wt% AFDW				
Lipid	27.6	7.4	16.9	7.0
Protein	44.9	72.9	54.3	72.6
Carbohydrates (balance)	27.6	19.8	28.8	20.4
Total	100	100	100	100

3.3 Process Assumptions

The major process inputs and assumptions for the 2022 SOT report are listed in Table 7. Similar to the 2017 to 2019 SOT reports, there are seasonal variations in algae/wood blend ratios and the blended feedstock compositions because of different algae production rates in each season. To facilitate analysis of such a system, a key consideration is to specify input parameters accommodating these seasonal variations. The blend ratios for each season are specified based on the algae seasonal production rates and their differences between summer and non-summer seasons. The feedstock compositions in different seasons are calculated based on the blend ratios of algae/wood and the compositions for algae only and wood only. With the parameters for each season specified, the parameter assumptions on an annual average basis are calculated and input to the process simulation to estimate the system mass and energy balance.

Table 7. Major parameter assumptions for the algae HTL and upgrading system

Processes	Value
HTL	
Temperature, °C	350
Pressure, psia	3000
LHSV, L/L/h	4
Products yields, g/g feedstock, AFDW	
Biocrude	0.33
Aqueous	0.46
Solid	0.09
Gas	0.12
Elemental of biocrude, wt % dry basis	
Carbon	78%
Hydrogen	9.3%
Oxygen	3.7%
Nitrogen	7.3%
Sulfur	0.9%
Ash	0.9%
Moisture	14%
Upgrading	
<i>Hydrotreating</i>	
Temperature, °C (outlet)	400 (main bed); 325 (guard bed)
Pressure, psia (outlet)	1500
Weight hourly space velocity (WHSV), h ⁻¹	0.5 (main bed); 0.72 (guard bed)
H ₂ consumption, g/g dry feed	0.07
Hydrotreated oil yields, g/g dry feed	0.79
Hydrotreated oil distillation fraction, wt%	
Naphtha (B.P. < 150 °C)	18.3
JFC (150 to 250 °C)	22.7
Diesel (250 to 340 °C)	39.0
Heavy (> 340 °C)	20.0
<i>Hydrocracking (for heavy cut)</i>	
Temperature, °C (inlet)	390
Pressure, psia (inlet)	1,000
Liquid hourly space velocity (LHSV), h ⁻¹	1
H ₂ consumption, kg/kg dry feed	0.006
<i>Hydrodenitrogenation (HDN, for JFC)</i>	
Nitrogen in JFC, ppm	3200
Temperature, °C (outlet)	400
Pressure, psia (outlet)	1,500
WHSV, h ⁻¹	0.5
Nitrogen removal, % of feed nitrogen	> 99

The HTL process parameter assumptions are based on the most recent HTL testing results for *P. celerii*. The biocrude yield is 0.33 g/g AFDW feed based on the testing results, which is lower than previously reported yields from the 2017 to 2019 SOT reports, ranging from 0.41 to 0.45 g/g feedstock (DOE 2020). The primary reason for the reduced yield is that the tested *P. celerii* strain has higher protein and lower lipid content than the algae assumed in previous SOT studies. The shift in biochemical content leads to lower biocrude yield based on its compositions (Jiang et al. 2019).

Although HTL testing has not been conducted for *P. celerii* blended with forest residue, the biocrude yield for the blended feedstock should be equal or higher than that of *P. celerii* alone based on previous HTL testing (Zhu et al. 2020b). Therefore, for the 2022 SOT case study, the biocrude yield for the HTL conversion process with blended feedstock is conservatively assumed to be equal to that of *P. celerii*. Additional testing is needed to validate this assumption.

For the upgrading process, the design assumptions and hydrotreated oil yield are based on the hydrotreating testing of the HTL biocrude from *P. celerii*. As described in Section 2.2, the jet fuel cut fraction in the hydrotreated oil is 22.7%. The heavy cut of the upgraded biocrude is assumed to be further treated in a hydrocracking unit to generate additional naphtha, jet, and diesel fuels. The total JFC from both hydrotreating and hydrocracking steps is about 28% of the total fuel product. Based on the hydrotreating testing results, the JFC generated from the algae HTL biocrude has a nitrogen content of 3200 ppm. An initial HDN testing was conducted for the JFC sample from wet sludge HTL conversion and a 99% nitrogen removal efficiency was achieved (Snowden-Swan et al. 2022). The same HDN testing conditions and nitrogen removal efficiency are assumed in this SOT study considering the compositional similarity of the hydrotreated biocrude from algae and wet sludge. Ideally, HDN testing for the JFC from the upgraded algal biocrude should be done to verify this assumption. The SAF is assumed to be sold together with naphtha and diesel cuts as blendstock products and the yields for each cut are estimated based on simulation results for the hydrotreating and hydrocracking processes.

4.0 Results and Discussion

In this section, the major cost results are described and discussed.

4.1 Cost Results

The cost results for the 2017 to 2022 SOTs are shown in Table 8. The detailed cost contributions for each processing area and key technical parameters for all cases are listed in Appendix A. The 2022 SOT assumes a single-stage HTL process and open-pond cultivated algae. Compared to previous SOTs using a similar process design (2017 to 2019 SOTs), the 2022 SOT includes algae cultivated in fully saline, nutrient-replete conditions and SAF production, leading to differences in the cost results. The 2021 SOT included a two-stage sequential HTL process and the fermentation of lactic acid from extracted carbohydrates to increase revenues. The 2021 SOT included wastewater-grown algal feedstock with \$0 feedstock cost. The 2022 SOT is compared to the 2017 to 2019 SOTs because of similarities in feedstock selection (cultivated algae) and process design (single-stage HTL). Compared to previous SOTs, the major differences in the 2022 SOT include using a different alga strain for feedstock, which leads to lower biocrude yield, and adding SAF production, which leads to higher natural gas usage and extra costs related to the HDN process.

The feedstock cost on a per GGE basis is affected by the algae feedstock cost, blend ratio with non-algae feedstock and final fuel production rates. The 2022 SOT has about 24% higher feedstock cost than the 2019 SOT on a per unit GGE fuel basis because it has much lower biocrude yield than that of the 2019 SOT, which are 0.33 and 0.41 g/g feedstock AFDW, respectively. The lower biocrude yield of the 2022 SOT results from the specific algae strain used, which has high protein and low lipid contents. Based on previous work (Jiang et al. 2019), lower lipid content leads to lower biocrude yield. However, nutrient-replete conditions generally lead to higher productivities and thus lower algae biomass feedstock cost (Huesemann et al. 2021). Therefore, for specific algae strains, analysis of trade-offs between low biocrude yield and low algae feedstock cost is needed to evaluate the impacts of nutrient-replete cultivation conditions on the conversion system.

For the HTL process cost, the 2022 SOT has higher cost than the 2019 SOT at per GGE fuel basis due to its lower biocrude yield and thus lower final fuel yields. The biocrude upgrading cost for the 2022 SOT is only 7% higher than that of 2019 SOT. The lower biocrude yield of the 2022 SOT leads to lower equipment cost and operating cost for the upgrading process compared to the 2019 SOT. However, the high-protein algae feedstock of the 2022 SOT leads to higher nitrogen content of the HTL biocrude and thus leads to higher hydrogen consumption for the hydrotreating process than that of the 2017 to 2019 SOTs. Combining with the lower final fuel yield, the 2022 SOT has slightly higher upgrading cost than the 2019 SOT at per GGE fuel basis. The cost contribution from adding a HDN unit for SAF production is only \$0.02/GGE based on the cost estimation. The 2022 SOT has a larger hydrogen plant and more natural gas consumption at per GGE fuel basis resulting from lower fuel yields and higher hydrogen consumption. It leads to a higher cost for the balance of plant than those of the 2017 to 2019 SOTs. As a result of the higher protein content (more N and P) the 2022 SOT has increased nutrient recycle credits compared to the 2017 to 2019 SOT studies. In addition, because of the lower biocrude and thus final fuel yields of the 2022 SOT, more carbon remained in the recycled aqueous phase and thus it also has higher carbon recycle credit.

The minimum fuel selling price (MFSP) for the 2022 SOT is approximately 9% higher than the 2019 SOT mainly because of the lower biocrude yield and thus lower final fuel yields. Comparing to the 2017 and 2018 SOT cases, the 2022 SOT has lower MFSP resulting from larger plant scale, lower feedstock cost, improvement in hydrotreating process and higher nutrient recycle credits.

Table 8. Algae HTL system 2017 to 2022 SOT costs

Production cost breakdown, \$/gge (\$2016)	2017 SOT	2018 SOT	2019 SOT	2020 SOT	2021 SOT	2022 SOT
Feedstock	\$6.66	\$5.61	\$4.10	\$4.81	\$0.00	\$5.07
HTL biocrude production	\$0.95	\$0.84	\$0.75	\$1.54	\$2.90	\$0.94
HTL biocrude upgrading to finished fuels	\$0.69	\$0.59	\$0.42	\$0.30	\$0.83	\$0.45
HTL aqueous phase treatment	\$0.00	\$0.00	\$0.00	\$0.00	\$1.21	\$0.00
Co-product generation	\$0.00	\$0.00	\$0.00	\$1.43	\$0.00	\$0.00
Balance of plant	\$0.61	\$0.57	\$0.49	\$0.74	\$0.87	\$0.62
Co-product credit	0.00	0.00	0.00	-2.92	-\$3.19	\$0.00
Nutrients recycle credit	-0.86	-0.78	-0.78	-1.43	\$0.00	-\$1.66
Minimum fuel selling price (MFSP)	\$8.05	\$6.83	\$4.98	\$4.48	\$2.61	\$5.42

Figure 9 depicts the product cost contributions for each SOT case from 2017 to 2022. Figure 10 shows the conversion cost only (without feedstock cost) for each SOT case. The net conversion cost for 2022 SOT is 59% lower than that of 2019 SOT primarily due to higher nutrient recycle credits.

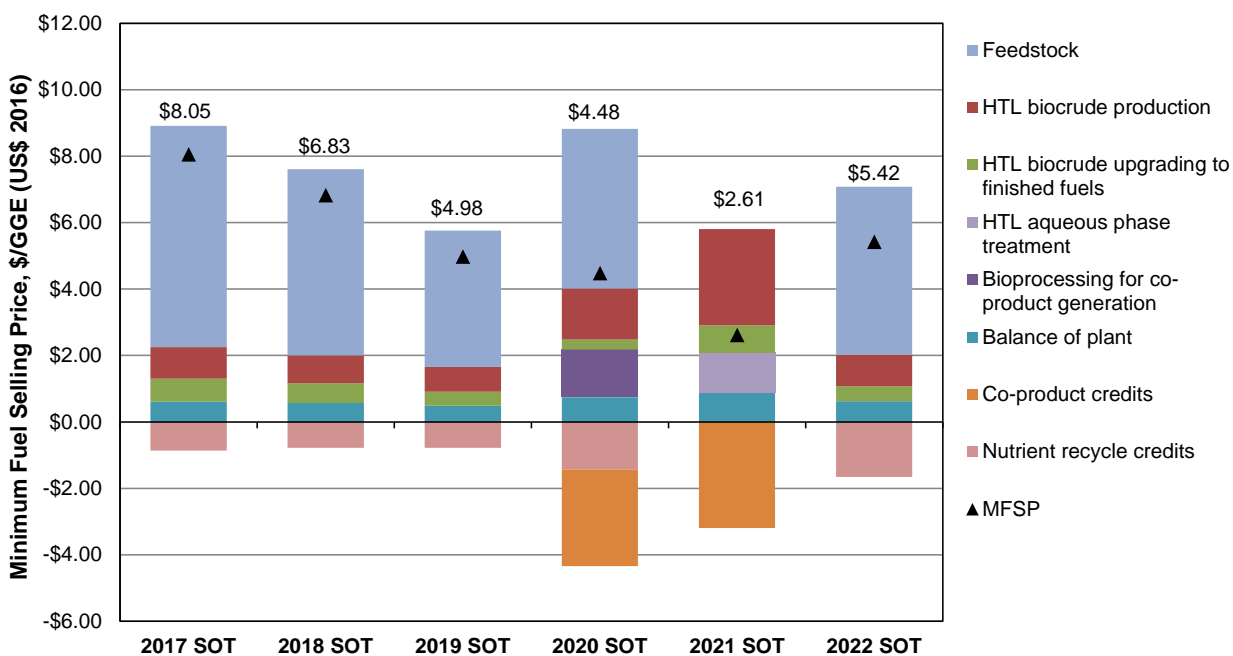


Figure 9. Cost contribution for the microalgae HTL system SOT cases.

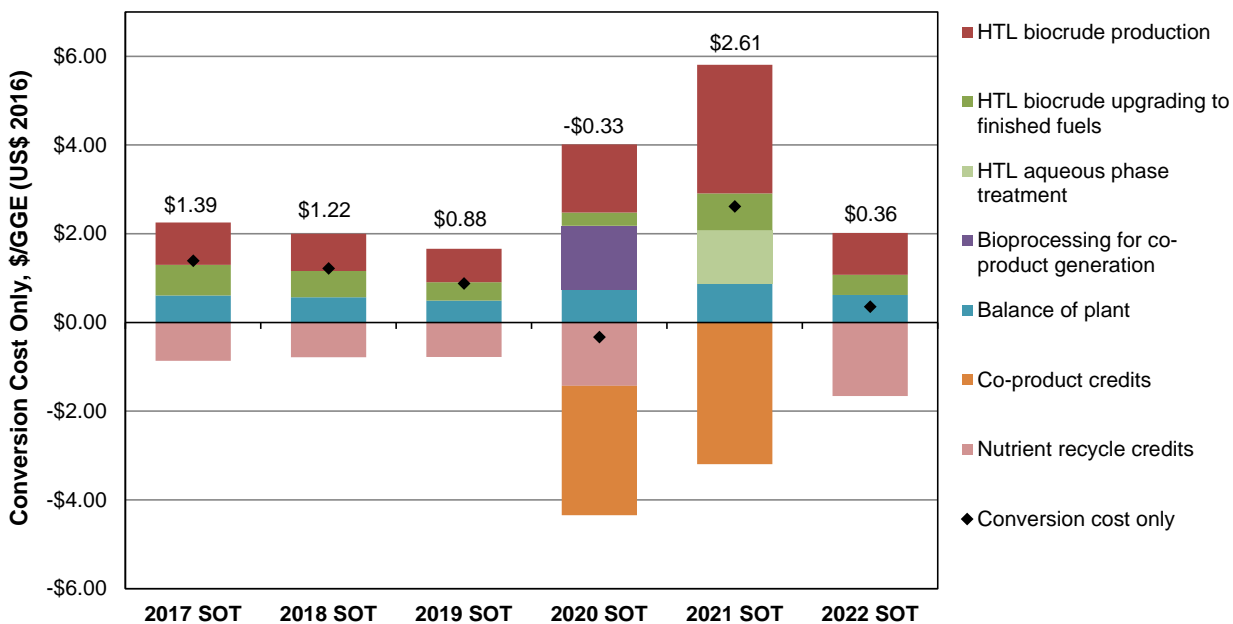


Figure 10. Conversion cost only (without feedstock cost) allocation for the microalgae HTL system SOT cases.

4.2 Sustainability Metrics

Table 9 lists the conversion sustainability metrics for 2017 to 2022 SOT cases. Because of the lower biocrude yield, the 2022 SOT has 21% lower fuel yields than that of the 2019 SOT at per ton feedstock basis. Also, compared to previous SOTs, the natural gas consumption at per GGE fuel basis is higher because of the lower biocrude yield and increased hydrogen consumption due to the high-protein algae feedstock assumed for the 2022 SOT. For the similar reasons, the makeup water and net electricity use of the 2022 SOT are also higher than those of 2017 to 2019 SOT cases. The 2022 SOT also has lower carbon and energy efficiencies than the 2017 to 2019 SOTs due to lower fuel yields and higher natural gas use.

Conversion plant sustainability metrics are not useful by themselves and need to be coupled to the life-cycle inventory (LCI) of the algae farm, to account for aqueous recycle from the conversion plant back to the farm. An LCI for the conversion plant will be delivered to Argonne National Laboratory, to complete a well-to-wheels life-cycle analysis using the farm inputs from NREL.

Table 9. Conversion sustainability metrics.

Input	Units	2017 SOT	2018 SOT	2019 SOT	2020 SOT	2021 SOT	2022 SOT
Feedstock flow rate							
Algae	ton/d AFDW	228	258	350	405	139	423
Non-algae biomass	ton/d AFDW	83	82	248	293	0	240
Total	ton/d AFDW	311	340	598	698	139	662
Fuel yield	GGE fuel/ton feedstock AFDW	104	115	106	78.7	73.4	83.2
Co-product yield	lb/ton feedstock AFDW	0	0	0	238	771	0
Natural gas consumption							
For fuel production	MMscf/y	419	475	822	1,069	72	923
For co-product generation	MMscf/y	0	0	0	631	0	0
Total	MMscf/y	419	475	822	1,701	72	923
	scf/ton feedstock AFDW	4,078	4,228	4,160	7,387	1,574	4,343
	scf/GGE fuel	39.2	36.9	39.4	93.8	21.5	52.2
Makeup water	kg/GGE fuel	5.16	4.70	5.23	2.99	28.6	10.2
Electricity	kwh/GGE fuel	0.76	0.70	0.73	3.44	1.71	0.83
Carbon efficiency							
Fuel C/feedstock C	%	54	58	53	41	38	42
Fuel + co-product C/feedstock C	%	--	--	--	50	38	--
Overall carbon efficiency	%	48	51	47	32	36	37
Energy efficiency							
Fuel products/feedstock only	% HHV basis	65	70	64	55	46	50
Overall energy efficiency	% HHV basis	54	57	52	44	42	41

5.0 Conclusions and Future Work

In the 2022 SOT study, a high-productivity marine alga strain was tested and evaluated for its HTL conversion and upgrading to SAF and other fuel blendstocks. The experimental work of this study is the first of its kind to show a direct pathway from outdoor-cultivated algae conversion to intermediate product (biocrude) to finished fuel. Continuous HTL coupled with the upgrading testing demonstrated clear potential for producing jet fuel cut from a demonstrated high-productivity algal strain. The results of the SOT assessment also demonstrated that the cost increase due to adding SAF production is marginal. Therefore, the results of this study indicate that the algae HTL conversion is a promising pathway to produce market competitive SAF. Although using high protein alga strain leads to lower biocrude yield, higher hydrogen consumption and thus lower energy/carbon efficiencies than the 2019 SOT, the results of this study provide more accurate and reliable information for commercialization of algae-based fuel (including SAF) production by using demonstrated outdoor alga strain and continuous HTL conversion testing condition as major design basis.

Future work needed for advancement of the technology and supporting analysis includes:

- Conduct HTL testing of high-protein algae blended with low-cost non-algal feedstock
- Conduct HTL testing of other high productivity algae strains for different cultivation seasons
- Conduct hydrotreating and hydrocracking testing to verify and optimize SAF generation from algae HTL biocrude
- Investigate and assess alternative pre-processing steps, such as protein extraction methods, allowing co-products generation from algae strains with high protein content.

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Appendix A – Detailed SOT Costs

Processing Area Cost Contributions & Key Technical Parameters	Metric	2017 SOT	2018 SOT	2019 SOT	2020 SOT	2021 SOT	2022 SOT
Fuel selling price	\$/GGE	\$8.05	\$6.83	\$4.98	\$4.48	\$2.61	\$5.42
Conversion Contribution	\$/GGE	\$1.39	\$1.22	\$0.88	-\$0.33	\$2.61	\$0.36
Production Diesel	mm GGE/yr	7.1	8.9	13.7	12	2.4	9.3
Production Sustainable Aviation Fuel (SAF)	mm GGE/yr	0.0	0.0	0.0	0.0	0.0	5.0
Production Naphtha	mm GGE/yr	3.6	4.0	6.6	6.3	1.0	3.4
Diesel Yield (AFDW feedstock basis)	GGE/US ton feedstock	69	79	70	51	52	42
SAF Yield (AFDW feedstock basis)	GGE/US ton feedstock	0	0	0	0	0	25
Naphtha Yield (AFDW feedstock basis)	GGE/US ton feedstock	35	36	33	27	22	16
Diesel Yield (areal basis)	GGE/acre-yr	1,416	1,771	2,746	2,365	6,705	1,850
SAF Yield (areal basis)	GGE/acre-yr	0	0	0	0	0	997
Naphtha Yield (areal basis)	GGE/acre-yr	724	800	1,310	1,261	2,804	687
Co-product Yield (AFDW feedstock basis)	lb /lb feedstock	0	0	0	0.12	0.39	0.00
Natural Gas Usage-drying (AFDW feedstock basis)	scf/US ton feedstock	0	0	0	0	0	0
Natural Gas Usage-HTL, H2 gen, bioprocessing (AFDW feedstock basis)	scf/US ton feedstock	4,078	4,228	4,085	7,387	1,574	4,343
Carbon efficiency, C in fuels/C in feedstock	%	54%	58%	53%	41%	38%	42%
Carbon efficiency, C in co-products/C in feedstock	%	0%	0%	0%	10%	0%	0%
Feedstock							
Total Cost Contribution	\$/GGE fuel	\$6.66	\$5.61	\$4.10	\$4.81	\$0.00	\$5.07
Feedstock Type		<i>Algae with wood supplement</i>	<i>Algae with wood supplement</i>	<i>Algae with wood supplement</i>	<i>Algae with corn stover supplement</i>	<i>Algae grown in wastewater</i>	<i>Algae with wood supplement</i>
Feedstock Cost (AFDW basis)	\$/US ton feedstock	\$694	\$643	\$421	\$379	\$0	\$410
HTL Biocrude Production							
Total Cost Contribution	\$/GGE fuel	\$0.95	\$0.84	\$0.75	\$1.54	\$2.90	\$0.94
Capital Cost Contribution	\$/GGE fuel	\$0.56	\$0.50	\$0.47	\$0.56	\$1.48	\$0.59
Operating Cost Contribution	\$/GGE fuel	\$0.39	\$0.34	\$0.28	\$0.98	\$1.42	\$0.35
Liquid Hourly Space Velocity (LHSV)	h ⁻¹	4.0	4.0	4.0	Stage I: 4; Stage II: 3.5	4.0	4.0
HTL Carbohydrate Extraction	% extracted/carbohydrate in feedstock	0%	0%	0%	58%	0%	0%
HTL Biocrude Yield (AFDW)	lb /lb feedstock	0.41	0.45	0.41	0.30	0.29	0.33
HTL Biocrude Hydrotreating to Finished Fuels							
Total Cost Contribution	\$/GGE fuel	\$0.69	\$0.59	\$0.42	\$0.30	\$0.83	\$0.45
Capital Cost Contribution	\$/GGE fuel	\$0.30	\$0.27	\$0.23	\$0.17	\$0.39	\$0.25
Operating Cost Contribution	\$/GGE fuel	\$0.39	\$0.32	\$0.19	\$0.13	\$0.45	\$0.20
Mass Yield on dry HTL Biocrude	lb/lb biocrude	0.81	0.82	0.81	0.83	0.82	0.79
HTL Aqueous Phase Treatment							
Total Cost Contribution	\$/GGE fuel	\$0.00	\$0.00	\$0.00	\$0.00	\$1.21	\$0.00
Capital Cost Contribution	\$/GGE fuel	\$0.00	\$0.00	\$0.00	\$0.00	\$0.46	\$0.00
Operating Cost Contribution	\$/GGE fuel	\$0.00	0.00	0.00	0.00	\$0.75	\$0.00
Bioprocessing for Co-product Generation							
Total Cost Contribution	\$/GGE fuel	\$0.00	\$0.00	\$0.00	\$1.43	\$0.00	\$0.00
Capital Cost Contribution	\$/GGE fuel	\$0.00	\$0.00	\$0.00	\$0.64	\$0.00	\$0.00
Operating Cost Contribution	\$/GGE fuel	\$0.00	0.00	0.00	0.79	0.00	0.00
Fermentation Productivity	g/L-hr	0	0	0	0.46	0.00	0.00
Fermentation Process Yield	g product/g extracted carbohydrates	0	0	0	0.37	0.00	0.00
Balance of Plant							
Total Cost Contribution	\$/GGE fuel	\$0.61	\$0.57	\$0.49	\$0.74	\$0.87	\$0.62
Capital Cost Contribution	\$/GGE fuel	\$0.29	\$0.28	\$0.23	\$0.41	\$0.49	\$0.30
Operating Cost Contribution	\$/GGE fuel	\$0.31	\$0.29	\$0.26	\$0.34	\$0.37	\$0.32
Co-product Credits							
Total Cost Contribution	\$/GGE fuel	\$0.00	\$0.00	\$0.00	-\$2.92	-\$3.19	\$0.00
Nutrient Recycle Credits							
Total Cost Contribution	\$/gge fuel	-\$0.86	-\$0.78	-\$0.78	-\$1.43	\$0.00	-\$1.66

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