Pacific Northwest
NATIONAL LABORATORY

PNNL-32695

Hydrothermal Liquefaction and Upgrading of Wastewater-Grown Microalgae: 2021 State of Technology

March 2021

Yunhua Zhu
Andrew Schmidt
Peter Valdez
Lesley Snowden-Swan
Scott Edmundson

U.S. DEPARTMENT OF ENERGY
Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes **any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights**. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <https://www.ntis.gov/about>
Online ordering: http://www.ntis.gov

PNNL-32695

Hydrothermal Liquefaction and Upgrading of Wastewater-Grown Microalgae: 2021 State of Technology

March 2021

Yunhua Zhu
Andrew Schmidt
Peter Valdez
Lesley Snowden-Swan
Scott Edmundson

Prepared for
the U.S. Department of Energy
under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory
Richland, Washington 99354

Acknowledgment

We gratefully acknowledge inputs from and discussions with Dr. Martin Gross and Jens Dancer of Gross-Wen Technologies and Dr. Kuldip Kumar of MWRD of Greater Chicago for very helpful information about using algae growth for nutrients removal from wastewater streams.

Summary

The fiscal year (FY) 2021 State of Technology (SOT) Assessment for the hydrothermal liquefaction (HTL) of wastewater (WW)-grown microalgae and biocrude upgrading system was completed and reported here. An industrial partner, Gross-Wen Technologies (GWT), provided algae feedstock cultivated on a revolving algal biofilm (RAB) system by using the primary effluent from a water resource recovery facility (WRRF). This provided algae was tested at PNNL for HTL processing. The experimental results provided the major design basis of the HTL process of the SOT baseline case. The primary effluent of the Metropolitan Water Reclamation District (MWRD) of Greater Chicago was assumed to be the nutrients source for algae growth and the algae yield data per gallon wastewater provided by GWT were used to estimate the total algae production rate and thus the HTL conversion plant scale. Considering different cultivation technologies and wastewater streams with different flow rates and nutrients contents can be used to produce algae, this SOT assessment just provided an example case study for WW-grown algae based HTL conversion to fuels systems. A preliminary economic analysis was developed based on process simulation results. Sensitivity analysis was implemented to evaluate cost impacts of plant scales, potential cost improvements and other key factors.

Table 1 summarizes the major changes of the 2021 SOT case compared to the 2017 to 2020 SOT cases and describes the reasons and effects of the changes. Compared to previous SOTs which assumed algae cultivated in natural water and external nutrient and CO_2 sources, the 2021 SOT assumes algae cultivated in wastewater with nutrients in wastewater and CO_2 in air as the nutrient and carbon sources. The WW-grown algae remove nutrients in wastewater and absorb carbon dioxide from air via algae growth. Therefore, the cultivation cost of WW-grown algae can be balanced or exceeded by the value of nutrient removal and the carbon capture credits. In addition, as a solid waste from a WRRF, the WW-grown algae needs to be removed and the conversion plant can serve as an algae disposal unit. Therefore, the feedstock cost of the WW-grown algae is assumed to be zero in this study. Compared to previous SOT cases assuming algae from a 5000-acre algae farm, the 2021 SOT with WW-grown algae as feedstock has a much lower production rate and thus a smaller plant scale because of the low nutrient content of the WRRF effluents. Other major differences between this year’s case and previous SOTs includes a lower biocrude yield of WW-grown algae based on HTL testing results, co-product generation of struvite fertilizer, and anaerobic digestion of the HTL aqueous phase for biogas generation. The smaller plant scale and lower biocrude yield for the FY21 SOT result in a higher conversion cost (not including feedstock cost) of \$2.61/GGE relative to the 2017-2020 SOTs, which ranged from \$-0.33 to 1.39/GGE. However, with the assumed zero cost of feedstock for WW-grown algae, the minimum fuel selling price (MFSP) for the FY21 SOT (\$2.61/GGE) is much lower than previous SOT cases, which ranged from \$4.48 to 8.05/GGE.

Table 1. Major changes of 2021 SOT compared to 2017 to FY20 SOT cases

Category	2021 SOT	2017 to 2020 SOTs	Reason	Effects
Feedstock	• Source: algae grown for wastewater treatment purpose via uptake of nutrients in wastewater and CO_2 in air; • Production rate: 139 ton/d ash free dry basis (annual average); • Feedstock cost: zero	• Source: algae grown in large-scale farms for producing a feedstock used for biofuel and chemical generation; external nutrients and CO_2 as nutrients and carbon sources; • Production rate: 228 to 598 ton/d AFDW; • Feedstock cost: $590 to 909/ton AFDW	Algae feedstock cost has the most significant impacts on algae HTL system cost and therefore, FY21 SOT shift to low-cost algae or algae waste. WW-grown algae is a solid waste from nutrient removal processes in a WRRF and needs to be treated, reduced, and disposed, therefore, it is chosen as a low-cost feedstock for FY21 SOT. WW-grown algae use wastewater as the only nutrients sources. The low nutrients availability of wastewater lead to low production rate of algae compared to large scale algae cultivation farm with external nutrients addition.	Lower algae production rate or smaller HTL conversion plant scale of FY21 leads to higher production cost per unit of fuel generated. Zero feedstock cost reduces the overall conversion system production cost
HTL conversion	Single-stage HTL	Single-stage HTL (FY17 to FY19) and two-stage sequential HTL (FY20)	Considering the high-ash content of WW-grown algae and the complexity of sequential HTL system, single-stage HTL was selected as initial research choice for WW-grown algae;	Compared to sequential HTL, single-stage HTL has no carbohydrate extraction and bioprocessing, which leads to less capital cost and operating cost
Biocrude yield	0.29 g/g algae AFDW	0.41 to 0.45 g/g algae AFDW for single-stage HTL	Low lipid content of WW-grown algae leads to low biocrude yield	Low biocrude yields leads to high conversion cost only on the basis of per unit final fuel
Aqueous phase treatment	Struvite synthesis and anaerobic digestion (AD)	Direct recycle of aqueous phase from single-stage HTL	Struvite generation recovers the majority of P and partial N from the algal solids disposed from the WRRF; AD is used to recover part of carbon from aqueous phase as biogas	Extra capital and operating cost for aqueous phase treatment; extra co-product credits from struvite generation and biogas generation via AD compared to direct recycle; resource sink for N/P to avoid nutrients accumulation in the cultivation to conversion loop; less external natural gas consumption and lower greenhouse gas (GHG) emissions resulting from biogas generation via AD

Acronyms and Abbreviations

AD	anaerobic generation
AFDW	ash free dry weight
ANL	Argonne National Laboratory
BETO	Bioenergy Technologies Office
DOE	U.S. Department of Energy
FY	Fiscal year
GWT	Gross-Wen Technologies
HTL	hydrothermal liquefaction
HRT	hydraulic retention time
LCI	life-cycle inventory
LHSV	liquid hourly space velocity
MFSP	minimum fuel selling price
MGD	million gallon per day
MWRD	metropolitan water reclamation district
NREL	National Renewable Energy Laboratory
PNNL	Pacific Northwest National Laboratory
RAB	Revolving algal biofilm
SAF	Sustainable aviation fuel
SOT	state of technology
TEA	techno-economic analysis
WW	wastewater
WRRF	water resource recovery facility

Contents

Figures

Tables

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.
- Encourage the creation of a new domestic bioenergy and bioproduct industry (US DOE 2016).

To meet national goals to increase the production of renewable fuels, products, and power from biomass, techno-economic analyses (TEAs) have been developed for both biological and thermochemical pathways for converting biomass to fuels and co-products.

Microalgal feedstocks are expected to contribute significantly to BETO’s strategy for sustainable and nationwide production of biofuels (US DOE 2016). The 2020 SOT for algal biomass production via open pond farm cultivation estimates the current and future costs for algal feedstocks (Davis and Klein 2021). The goal of the report is to benchmark the minimum biomass selling price for farmed microalgae, which is estimated at $603/ton AFDW. With technological advances, the projected sale price in 2030 is $488/ton AFDW. At the benchmarked price, microalgae cultivated solely as a feedstock for biofuel production is not cost-effective and other salable products or services must be considered to create an economically feasible solution for biofuel production from algae. One of the first TEAs for the HTL of an algal feedstock evaluated the current state of technology in 2014. The resulting estimation for minimum fuel selling price (MFSP) was $15.57/gge, with approximately 85% of the fuel cost attributed to the feedstock cost for farmed microalgae (Jones et al. 2014).

One option for reducing the cost of algal feedstock is to integrate cultivation with wastewater treatment. The synergistic coupling of microalgae cultivation and wastewater treatment services can significantly reduce the economic burden of farmed microalgae by providing nutrient removal services in addition to generating biomass feedstock (Clippinger and Davis 2021). The cultivation of microalgae in facultative lagoons is a common method for nutrient removal in municipal wastewater treatment (Craggs et al. 2013). However, a facultative lagoon is a low-cost and low-energy system for nutrient removal that generates minimal amounts of biomass. Research has optimized cultivation systems to yield high rates of both biomass growth and nutrient removal, utilizing different streams from within the wastewater treatment plant (Pittman et al. 2011). Process arrangements for high-productivity of biomass and effective nutrient uptake could include high-rate ponds (Craggs et al. 2013), floating soft-sided photobioreactors (Novoveská et al. 2016), and fixed biofilms (Kesaano and Sims 2014, Gross et al. 2015). Energy analysis has also shown algal systems for wastewater treatment to be less energy intensive than conventional systems, such as anaerobic digestion, with comparable rates for nutrient removal (Sturm and Lamer 2011).

Microalgae cultivated from wastewater effluent has proven to be a viable feedstock when coupled with HTL to produce a biocrude comparable to biocrudes derived from farm-cultivated algal feedstocks (Roberts et al. 2013, Zhou et al. 2013, Chen et al. 2014, Aida et al. 2016, Couto et al. 2018, Cheng et al. 2019). In several cases, the HTL biocrude can be readily upgraded to a finished distillate blend of saleable fuel (Wang et al. 2016, Lundquist and Spierling 2018, Arun et al. 2019). A detailed life-cycle assessment showed that, for an optimized system of wastewater algae coupled with HTL producing jet fuel, the net greenhouse gas emissions were 24% of the standard emissions for petroleum-derived jet (Fortier et al. 2014).

The TEA of the conversion process from algae to fuel product is critical to evaluate scalability and sustainability. The 2020 SOT report estimated the MFSP for algal HTL fuels at \$4.48/GGE (\$2016) (Zhu et al. 2021). TEAs specifically focused on the HTL of wastewater-grown algae coupled with upgrading and distillation steps have estimated the MFSP (\$2011) at \$7.14/GGE and \$4.30/GGE, respectively (Lundquist and Spierling 2018, Ranganathan and Savithri 2019). Another TEA for a process producing only renewable diesel calculated MFSP for the diesel product at \$6.62/gal (\$2015) (Juneja and Murthy 2017).

Technical challenges persist and will need to be addressed to support the use of wastewater-grown algae as a feedstock for HTL. The solid and aqueous phase co-products from HTL are rich in nitrogen (N) and phosphorus (P) and additional economic value can be generated by precipitating the nutrients as struvite ($NH_4MgPO_4 \cdot 6H_2O$) or other phosphorus-containing solid fertilizers. Struvite precipitation has been demonstrated using the HTL aqueous and solid products generated from microalgae as sources for N and P (Saravanan et al. 2017, Shanmugam et al. 2017, Ovsyannikova et al. 2020). Generation of struvite and other P-based fertilizers has also been demonstrated in wastewater treatment plants as a method for P removal and recovery (Hawthorne 2016). Another challenge is that the wastewater-grown algae is typically higher in ash content than farmed algae (Roberts et al. 2013). The high-ash content varies depending on cultivation and harvesting strategies, the microbial consortia present in the biomass (high concentration of diatoms), and the accumulation of dirt and debris in outdoor cultivation systems. High-ash feeds of algal biomass (up to 40 % w/w) can still produce upgradeable biocrude (Chen et al. 2017), but ash reduction strategies should be investigated to prevent equipment fouling and damage during HTL processing. Even though algae grown at wastewater treatment plants could be a viable cultivation strategy, successful and scaled implementation will be affected by constraints in available land for algae cultivation within large municipalities and optimizing the co-location of HTL facilities (Seiple et al. 2020). Additional investigation will be needed.

PNNL has led activities to develop HTL as a scalable processing solution in support of BETO’s goal to provide sustainable biofuels. The breadth of PNNL’s research with HTL includes investigation at different processing scales, with various feedstocks and feedstock combinations, with multi-step HTL, and with various methods to treat or valorize the aqueous phase from HTL (Elliott et al. 2015, Davidson et al. 2019, Snowden-Swan et al. 2020, Zhu et al. 2020). The HTL experiments are supported by expertise in hydrotreatment research to upgrade the HTL biocrude, by reducing heteroatom content (oxygen, nitrogen, and sulfur) to a product that could potentially be utilized as a crude blendstock in a petroleum refinery (Albrecht et al. 2016, Thorson et al. 2021). All the experimental work is synergistically supported by the TEA studies led by PNNL. The first benchmark TEA for HTL of microalgae was published in 2014 by PNNL (Jones et al. 2014). Since then, PNNL has published updates to that first TEA, incorporating the key advancements achieved through research and experimentation (Jiang et al. 2019, Zhu et al. 2019, Zhu et al. 2020, Zhu et al. 2020, Zhu et al. 2021).

This state of technology (SOT) report will examine the use of wastewater-grown microalgae as the feedstock for an HTL process. The assessment highlights and quantifies improvements over known technical barriers and challenges, facilitating the widespread adoption of new technologies for producing renewable biofuels from algal biomass (US DOE 2019). Two key features of the TEA presented herein that are different from previous SOTs are the incorporation of: (1) low-cost microalgae cultivated in municipal wastewater as the HTL feedstock and (2) struvite fertilizer as a saleable byproduct from the nutrient stripping of the HTL aqueous phase.

2.0 Experimental Work in Fiscal Year 2021

Due to the high costs associated with the cultivation of microalgae, low-cost alternatives were assessed as feedstocks for HTL. Several low-cost algal feedstocks were tested in FY21. The tested algae included *Alaria marginata* (macroalgae), astaxanthin-extracted *Haematococcus pluvialis* (microalgae residue) and wastewater-grown algae (microalgae) provided by Gross-Wen Technologies (see Figure 1).

Figure 1. WW-grown algae feedstock from FY2021 experimental work

Macroalgae was considered because of its current availability as a farm-cultivated feedstock. Nuisance macroalga are a potential feedstock due to their abundant availability as well. At present, additional investigation is needed to improve the viability of macroalgae as an HTL feedstock. Innovation is needed to format the material to a pumpable slurry, reduce the water content in the slurry for economical processing, reduce the ash in the biomass, and maximize biocrude yield. Residues from algae processing were also considered as HTL feedstock. For example, *Haematococcus pluvialis* can be cultivated to accumulate astaxanthin, a dietary supplement or food coloring agent. However, the scale-up potential for HTL processing of algal residues is relatively small to meet the national demands for renewable fuels. After consideration of several factors, such as scale-up potential, technology readiness, and initial experimental data, the WW-grown microalgae was selected for the 2021 SOT case.

For WW-grown algae, pilot scale cultivation has been successfully performed and its potential production rate from a large WRRF can be high. In addition, the WW-grown algae selling price can be zero or negative when the credits of N and P removal via algae cultivation from wastewater were considered (Clippinger and Davis 2021). The HTL conditions and configuration of experimental equipment are shown in **Error! Reference source not found.**.

Table 2. Single-stage HTL testing conditions for low-cost algae feedstock

Run ID		GWT-1
Reactor Configuration	Unit	Plug-flow
Sample Count	number	2
TOS	hour	0.86
Reactor Temperature	ºC	339
Pressure	psig	2893
Vol at Temp	mL	1150
Feed Rate	mL/h	2000
LHSV	L/L/h	1.7

The HTL testing results for the WW-grown algae, including mass and carbon yields of HTL products, are shown in **Error! Reference source not found.**, which are the major HTL design basis for the FY21 SOT case.

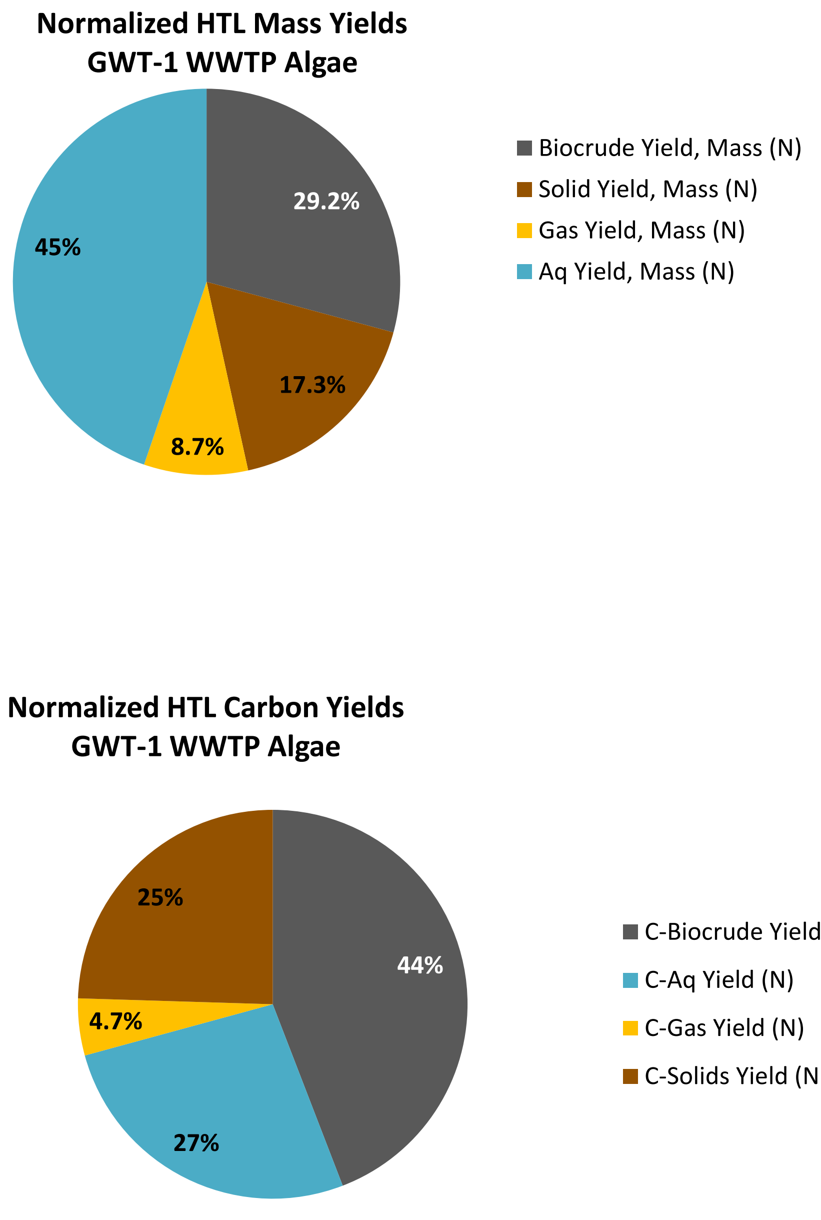

Figure 2. Mass and carbon yields of HTL testing of WW-grown algae

3.0 Process Design and Assumptions

In this section, the system of WW-grown algae HTL and upgrading system is overviewed. The details about plant scale, feedstock composition and process assumptions are described.

3.1 System Overview

Figure 3 shows the block flow diagram for the WW-grown algae HTL and biocrude upgrading system investigated in this study. The conversion system evaluated in this study comprises all processes inside of the dashed line boundary in

Figure 3. In this study, multiple wastewater treatment plants are assumed to use their primary effluents for algae cultivation. Each plant has an on-site algae cultivation farm. Cultivated algae was dewatered to 20 wt % dry solid at each farm and then sent to a centralized HTL and upgrading plant. The costs associated with cultivation, harvesting, dewatering, and transportation of the WW-grown algae are outside the scope of this analysis. The zero feedstock cost is based on the underlying assumption that the credits of N and P nutrients removal via algae cultivation in wastewater can balance the algae cultivation cost. The assumption also fosters the opportunity for a negative-cost feedstock if the nutrients removal credits exceed the algae cultivation cost in a WRRF (Clippinger and Davis 2021).

In the HTL plant for this analysis, condensed phase liquefaction takes place through the effects of time, heat and pressure. The resulting HTL products (oil, solid, aqueous, gas) are separated and the HTL biocrude is hydrotreated to produce diesel and some naphtha range fuels. The solid product is digested by dilute acid to recover most of phosphorus (P) from algae feedstock and then the acid digestate is mixed with the HTL aqueous phase to generate mainly struvite fertilizer. The residual solid after acid digestate is assumed to be disposed as solid waste. The treated aqueous phase after struvite generation is further treated in an AD process to generate biogas. The effluent from AD is assumed to be sent to a nearby WRRF algae cultivation unit. Process offgas is used to generate hydrogen, heat and power. A hydrogen plant is included for hydrotreating, which is assumed to be co-located with HTL conversion.

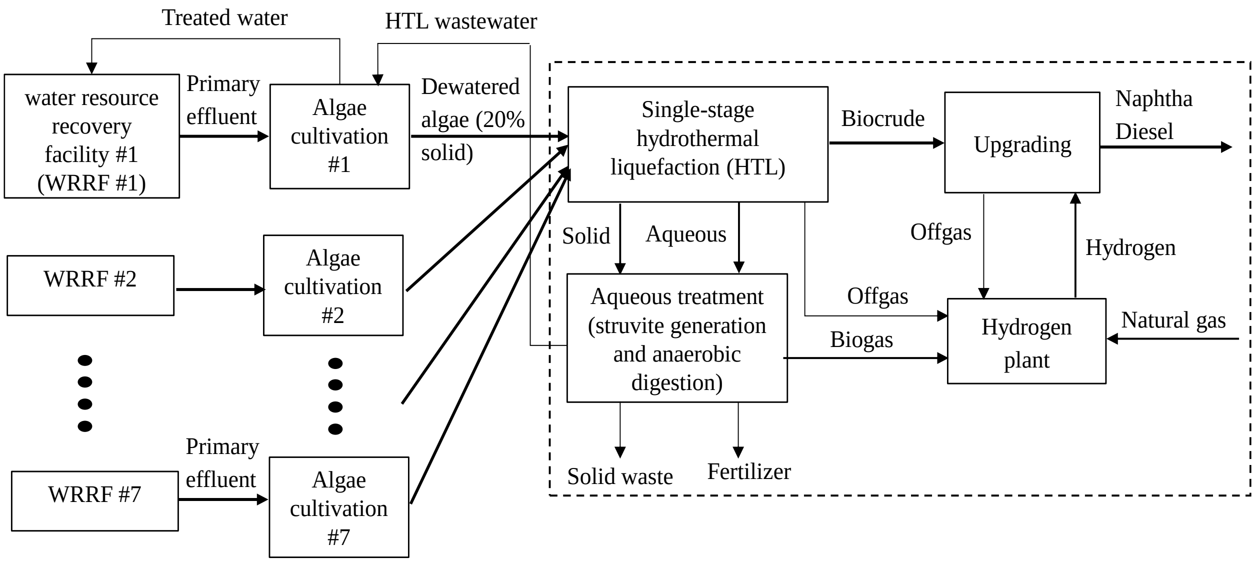

Figure 3. Simplified block diagram of WW-grown algae HTL and biocrude upgrading system.

3.2 Plant Scale

The WW-grown algae, which is assumed to be the feedstock of the FY21 SOT case, was grown in the primary effluent from a WRRF. The production rate of algae per gallon wastewater, with seasonal variations, and the flow rates of primary effluents are used to specify the plant scale for the HTL conversion process. Based on inputs from experts at GWT, the algae production yield of their RAB system with primary effluent from a WRRF as the nutrient source and at a short hydraulic residence time (HRT) is observed to be 0.1 (winter) to 0.2 g/gallon (summer) with an average of 0.15 g/gallon (or 1.65 ton/MMgal) on a dry basis. This algae production yield is assumed in the SOT as an example study for WW-algae HTL conversion and is subject to change when different cultivation technologies and wastewater streams with different flow rates and nutrients contents are used for WW-algae growth. Clippinger and Davis (2021) estimated the production rates of a high-protein algae grown by using primary effluent of WRRF ranging from 2 to 5 ton/MM gal wastewater. Wet storage was assumed in this study to store part of algae in summer/spring seasons with high algae production rates and used later in winter/fall to eliminate the seasonal algae productivity variation impacts on the conversion plant (Davis et al. 2020).

To specify the plant scale, we also need to assume the primary effluent flow rates used for the algae cultivation. In this study, the primary effluent for algae cultivation is assumed to be from the water reclamation plants in the greater Chicago area. The Metropolitan Water Reclamation District (MWRD) of Greater Chicago owns and operates one of the world's largest water reclamation plants (Stickney plant, located in Cicero, IL) and six other plants, with a combined treatment capacity of over 1 billion gallons of wastewater per day (MWRD 2021). As shown in Figure 4, based on the WRRFs locations, the HTL plant is assumed to be a centralized plant and located closest to the largest WRRF in Cicero (see Figure 4, WRRF1). The algae from each WRRF in the greater Chicago area is transported to the HTL plant for processing. The average transportation distance is about 50 miles based on the radius of the circle with HTL in the center as shown in Figure 4. The proposed arrangement facilitates the transportation of algae from the largest WRRF to the conversion facility and also the recycle of the aqueous stream from HTL to a nearby WRRF algae cultivation unit.

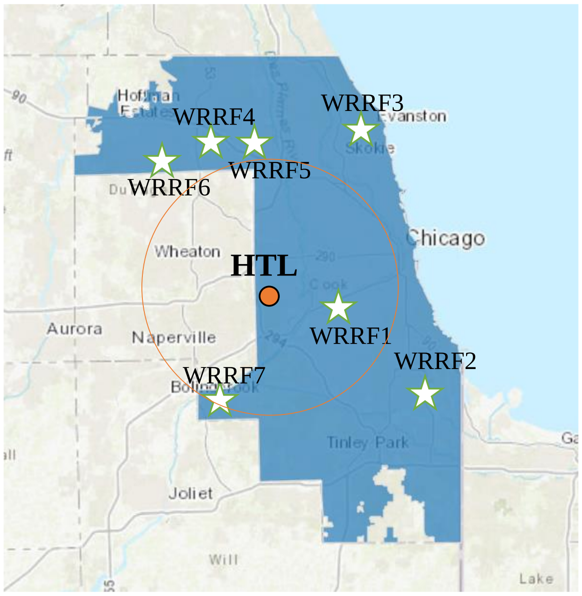

Figure 4. WW-grown algae HTL conversion plant scenario assumed for the SOT case

The detailed information for primary effluent flow rate and algae production rate for each WRRF are listed in Table 3. For the process model, the seasonal and average production rates of algae cultivation are estimated from the assumed algal productivity in primary effluent (0.15 g/gallon) and the flow rate of primary effluent from each WRRF. For the overall greater Chicago area, the total annual average algae production rate is 228 dry ton/d or 139 ton/d AFDW.

This study is to provide an example case for using WW-grown algae as a conversion feedstock for biofuels generation. To decide if algae cultivation is an appropriate technology for nutrient removal in a WRRF, the land availability/constraints for different algae cultivation technologies for large scale application need to be considered. If only a part of primary effluents or side streams from WRRFs are considered for algae cultivation, plant scales will be smaller than the baseline assumption of this study. To investigate the cost impacts of different plant scales on a WW-grown algae HTL system, sensitivity analysis for smaller plant scales was conducted and the results are described in Section 4.

Table 3. Wastewater-grown algae production assumptions

	MWRD of greater Chicago	Primary effluent flow rate (MGD)	Algae biomass to the conversion facility, dry ton/d Annual average
1	Stickney Water Reclamation Plant (WRRF)	700	116
2	Calumet WRRF	350	59
3	Terrence J. O'Brien WRRF	230	38
4	John E. Egan WRRF	30	5.0
5	James C. Kirie WRRF	50	8.6
6	Hanover Park WRRF	10	2.0
7	Lemont WRRF	2	0.38
	Total	1,400	228

3.3 Feedstock

The elemental and biochemical compositions of the selected WW-grown algae are listed in Table 4. The compositional data was input to the process simulation to specify the feedstock compositions and was also used to estimate the elemental balance of inlet/outlet streams of the HTL process. The HHV of the algae is estimated by using the Boie equation (Annamalai et al. 1987) and is reported in Table 4.

In the process of treating wastewater with algae, bacteria are also present in the system. Bacteria break down some organic waste components, which are then available for uptake by the algae. Algae, in turn, produce the oxygen necessary for the survival of aerobic bacteria (US EPA 2011). Therefore, the WW-grown algae is composed of both microalgae and bacteria. The algae feedstock is assumed to be dewatered to 20 wt % dry basis by each WRRF. The price of algae feedstock at the HTL plant gate is assumed to be zero. That is, the WRRD assumes all costs to deliver the feedstock to the plant gate for the HTL facility. In this study, the algae cultivation is assumed to be used to remove part of nitrogen (N) and phosphorus (P) and thus to reduce the nutrients removal cost by the WRRF. In addition, algae growth in this case uses CO2 in air as the only carbon source and thus helps to reduce the overall greenhouse gas emissions (GHG) of the WRRF system. The N removal cost from WRRF is about $0.36 to $3.85 per pound of nitrogen removed for the 20-year term (JJ Environmental. 2015). The P removal cost ranges from $42.22 to $60.88 per pound of P removed (Bashar et al. 2018). Considering the credits from nutrients removal and air carbon fixing, it is reasonable and may actually be quite conservative to assume the algae feedstock price at the HTL plant gate is zero as the HTL plant serves as a solid waste removal unit for WRRFs (similar to the cost WRRFs pay for disposal of sludge).

Table 4. Elemental and biochemical compositions of algae feedstock for 2021 SOT

Elemental composition, wt % ash free dry weight (AFDW)	Algae
Carbon	51.3
Hydrogen	6.8
Oxygen	31.5
Nitrogen	7.8
Sulfur	2.5
Total	100
Ash, wt % dry basis	39
Phosphorus (in ash)	3.3
Biochemical composition, wt % AFDW	
Carbohydrates (balance)	28.8
Fat	16.9
Protein	54.3
Total	100
Higher heating value, Btu/lb (MJ/kg), AFDW	9,811 (22.8)

3.4 Process Assumptions

The major process inputs and assumptions for the HTL and upgrading system for the 2021 SOT are listed in Table 5. The HTL process parameter assumptions are based on the most recent HTL testing results for WW-grown algae, which was grown by using primary effluent from a WRRF. The yield of biocrude is reduced using the WW-grown algae when compared to farm-grown microalgae. The HTL solid acid digestion process parameters assumptions are based on a lab-scale testing done by Ovsyannikova et al. (2020) for P recovery from microalgae HTL solid product. The process design for the biocrude upgrading process is similar to the previous SOT cases for algae HTL and the details are published in Zhu et al. (2020).

The process design assumptions for aqueous phase treatment, including struvite synthesis and anaerobic digestion are listed in Table 6. After acid digestion of the solid product, 90% P is extracted as mainly phosphate to the liquid part. The acid digestate was filtered and the insoluble solid was assumed to be disposed. MgO was assumed to be added to adjust the pH value to 8.5. The original Mg in the acid digestate was from the algae feedstock and its mass amount was estimated by using the solid ICP analysis results from the HTL testing. The filtered acid digestate was mixed with the HTL aqueous phase and $MgCl_2$ is supplemented to the mixture to reach a molar ratio of Mg^{2+}: PO_4^{3-} of 1:1. The ammonia content in the HTL and upgrading aqueous phase already meet a molar ratio NH_4^+ : PO_4^{3-} over 1 and thus no extra ammonia is needed to supplement the stream. The chemical reaction for struvite synthesis is shown in Eq. (1) (Shanmugam et al. 2016):

$$Mg^{2+} + NH_4^+ + H_nPO_4^{(3-n)} + 6H_2O \longleftrightarrow MgNH_4PO_4 \cdot 6H_2O \text{ (struvite)} + n\,H^+ \quad (n = 0, 1, 2) \qquad (1)$$

With pH adjusted and $MgCl_2$ supplemented to meet the required molar ratio, the struvite crystal will be formed. The design of the struvite synthesis process is mainly based on a lab scale testing by Shanmugam et al. (2016) for algae HTL phosphorus recovery and a pilot scale testing by Park et al. (2020) for struvite crystallization from the side stream of WRRFs. The elemental and mass balance of the struvite synthesis was estimated in an Excel-based model. The struvite generation testing for algae HTL solid and aqueous phase is also ongoing at PNNL and the testing results will be used in our future TEA work.

Table 5. Major parameter assumptions for HTL process

Processes	Assumptions
HTL conversion	
Feed slurry solid wt %, AFDW	20
Temperature, °C	350
Pressure, psia	3000
LHSV, L/L/h	4
Products yields, g/g feedstock, AFDW	
Biocrude	0.29
Aqueous	0.45
Gas	0.09
Solid	0.17
Elemental analysis of biocrude, wt % dry basis	
Carbon	77.6
Hydrogen	10.7
Oxygen	4.9
Nitrogen	5.5
Sulfur	1.3
Moisture, wt % of biocrude	22
HTL solid acid digestion	
Temperature, °C	20

Pressure, psia	14.7
H_2SO_4 concentration, mol/L	1
Residence time, mins	120
Acid: dry solid ratio, ml/g-dry solid	2
P recovery, % of P in feedstock	90

The anaerobic digestion process design was based on the testing conducted by Shanmugam et al. (2016). With ammonia removed from the aqueous phase via struvite formation, biogas production from AD of the aqueous is much higher than the original aqueous phase without ammonia removal via struvite generation. The COD removal from the original aqueous phase without ammonia removal is only 13% based on Shanmugam et al. (2016). The generated biogas is assumed to be used for process heating and hydrogen generation. The digestate from the AD process is assumed to be combusted for process heat.

Table 6. Major parameter assumptions for aqueous phase treatment process

Processes	Assumptions
Struvite synthesis	
Temperature, °C	15
Pressure, psia	14.7
Reaction time, mins	60
pH	8.5
pH adjustment	MgO
Mg supplement	$MgCl_2$
Mg^{2+} :$PO_4{}^{3+}$: NH_4^+ molar ratio	1: 1: 1
$PO_4{}^{3-}$ removal, %	> 99%
Anaerobic digestion	
Temperature, °C	35
Pressure, psia	14.7
COD removal, %	59

4.0 Results and Discussions

In this section, the major cost results are described and discussed. A sensitivity analysis and assessment of sustainability metrics are also described.

4.1 Cost Results

A cost comparison between of previous SOT cases since 2017 is shown in Table 7. The detailed cost contributions for each processing area and key technical parameters for all cases are listed in Appendix A. The total production cost in the 2021 SOT case is lower than other cases mainly because of the assumption of a zero-cost feedstock. The feedstock cost is about 55 to 75% of the total production cost (without including nutrients and co-product credits) of other SOT cases. Compared to other cases, the 2021 SOT has much higher cost for HTL biocrude production due to a much smaller plant scale (139 versus over 300 ton/d AFDW, see Table 8) and lower biocrude yield. Other SOT cases typically have biocrude yields at or above 0.4 g/g feedstock AFDW, which is higher than that of the assumed value for the 2021 SOT of 0.29 g biocrude/g feedstock AFDW. The 2021 SOT also has extra cost for HTL aqueous phase treatment. The credits from co-product struvite generation exceeds the extra cost from the aqueous phase treatment. A median selling price for struvite fertilizer at $0.30/lb is assumed based on Kim et al. (2021). Combining the zero-feedstock cost and the co-product credit for the 2021 SOT case results in a MFSP much lower than the previous cases despite of its smaller plant scale and reduced biocrude yield, which increase capital cost and reduce product revenue, respectively.

Table 7. Algae HTL SOT costs FY17 to FY21

Production cost breakdown, $/GGE ($2016)	2017 SOT	2018 SOT	2019 SOT	2020 SOT	2021 SOT
Feedstock	6.66	5.61	4.10	4.81	0.00
Algae storage	0.00	0.00	0.00	0.00	0.14
HTL biocrude production	0.95	0.84	0.75	1.54	2.75
HTL biocrude upgrading to finished fuels	0.69	0.59	0.42	0.30	0.83
HTL aqueous phase treatment	0.00	0.00	0.00	0.00	1.21
Bioprocessing for co-product generation	0.00	0.00	0.00	1.43	0.00
Balance of plant	0.61	0.57	0.49	0.74	0.87
Co-product credit	0.00	0.00	0.00	-2.92	-3.19
Nutrients recycle credit	-0.86	-0.78	-0.78	-1.43	0.00
Minimum fuel selling price (MFSP)	8.05	6.83	4.98	4.48	2.61

Figure 5 graphically shows the total cost breakouts for each case shown in Table 7. The minimum fuel selling price (MFSP) for the FY21 SOT is approximately 43% lower than that of the FY20 SOT resulting mainly from zero-cost of feedstock. Figure 6 shows the conversion cost only for each SOT case. Without the benefits from zero feedstock, FY21 SOT has higher conversion cost than other cases due to much smaller plant scale and lower biocrude yields. Compared to other cases, FY21 SOT case has no nutrient recycle credits because there are no costs for nutrients for algae cultivation since the carbon and N/P nutrients needed for algae growth are assumed to be from air and wastewater effluent, without any extra nutrients needed. Although the wastewater streams from the HTL plant are assumed to be recycled back to algae cultivation or a WRRF, there are no recycle credits for nutrients recovered internally to the system.

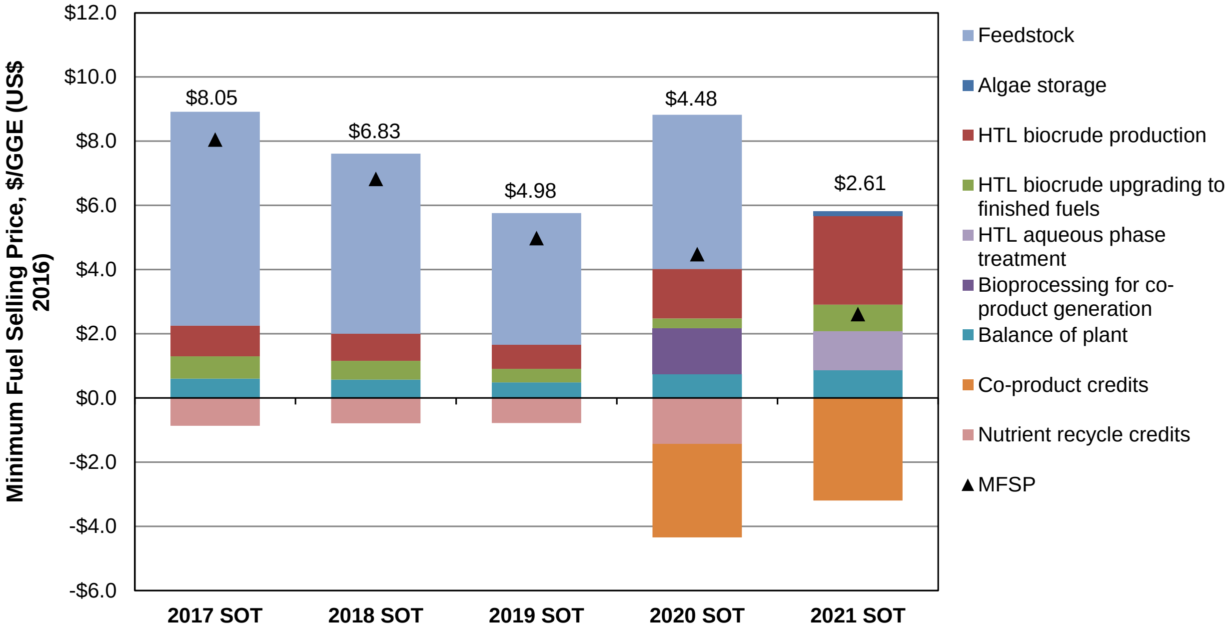

Figure 5. Algae HTL and biocrude upgrading system cost allocations.

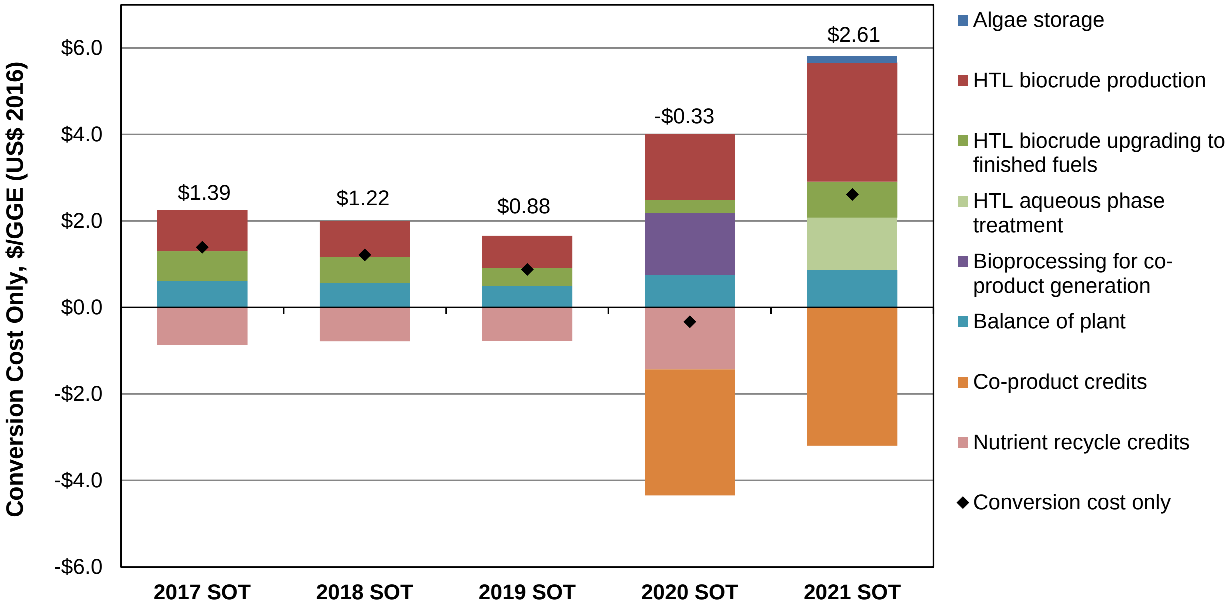

Figure 6. Algae HTL and biocrude upgrading conversion cost only allocations.

4.2 Sensitivity Analysis

Sensitivity analysis was implemented for selected parameters for the WW-grown algae HTL and biocrude upgrading system. Different effluent source streams from a WRRF were evaluated for use for algae cultivation. Potential improvement in capital and operating cost for different wastewater sources were evaluated. The impacts of other selected process and cost parameters were also investigated.

4.2.1 Plant Scale Impacts and Potential Cost Improvement

Although the tested WW-algae was cultivated by using primary effluent, using side streams for algae cultivation could be more feasible for existing WRRF. Using different nutrients sources WRRF for algae cultivation will affect algae production rate and the scale of the conversion plant. Using solely the primary effluent stream for algae cultivation would require significant modification WRRF and thus a substantial cost investment. Therefore, based on suggestions of industrial experts, other side streams within the WRRF that have been tested for algae cultivation, such as the anaerobic digestion (AD) centrate from sludge AD units, are considered as alternative nutrient sources for WW-algae cultivation in this study. Using AD centrate for algae growth almost has no seasonal variations in algae productivities since the AD centrate water temperature remains constant in all seasons and thus leads to constant seasonal algae production rate, which eliminate the need for algae drying or storage in high productivity seasons. The AD centrate also has higher N and P concentrations than the primary effluent, which leads to higher algae production rate per gallon wastewater. The algae production rate is 0.8 g/gallon AD centrate at a short hydraulic retention time (HRT) based on inputs from industrial experts, while this value is 0.15 g/gallon primary effluent. Based on the scenario of the MWRD of Greater Chicago, the flow rate of AD centrate is assumed to be about 1% of the flow rate of primary effluent (14 MGD). Therefore, the total algae production rate is estimated to be 12 dry ton/d (or 7.4 ton/d AFDW) by using only AD centrate from the WRRF system, which is much lower than the 139 ton/d AFDW by using the primary effluent from the same system. As shown in Figure 7, the MFSP for the conversion system with algae cultivated by using AD centrate is much higher than the baseline case because of much smaller plant scale of the AD centrate case.

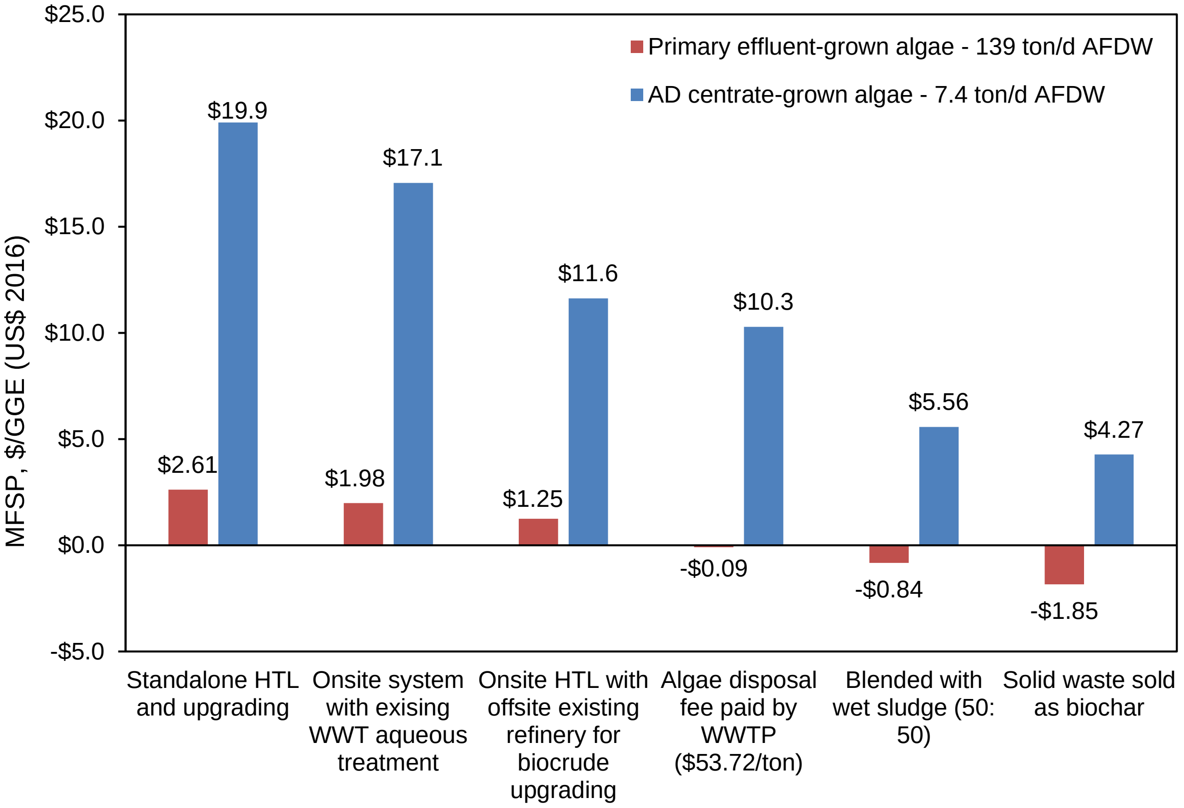

Figure 7. Aggregated cost impacts of potential improvements on the primary effluent and AD centrate-grown algae HTL and biocrude upgrading systems

The calculated MFSP of several other operational scenarios and assumptions are shown in Figure 7. For the primary effluent and AD centrate-grown algae HTL plants, the flow rate of the by-product aqueous streams are 200,000 and 9,000 gallons per day, respectively. Both of them are much smaller than the AD effluent flow rate of the Stickney WRRF with 700 MGD raw wastewater. In addition, as reported by Harthorne (2016), the Stickney WRRF operates a P-recovery process that yields a solid fertilizer product. Therefore, a potential cost improvement for the conversion system is to use existing AD and struvite equipment in the nearby WRRF to treat HTL aqueous stream. It has the potential to save both capital and labor cost, while the related variable operating cost (chemicals and utilities) is still included in the cost of the conversion plant. The option to utilize existing infrastructure for aqueous treatment leads to 24% and 14% reductions in the MFSP for the primary effluent and AD centrate cases, respectively. Another opportunity for cost reduction is to use an existing offsite refinery for biocrude upgrading. This option reduces the upgrading capital cost and labor cost. The variable operating cost for biocrude upgrading, including catalysts, hydrogen cost and biocrude shipping, is still included in the system cost. This adjustment leads to about 27% reduction in the MFSP for both cases. A potential credit to be included in the conversion system is the algae disposal fee. Since the produced algae would be considered a solid waste for the WRRFs, it needs to be disposed or further treated to avoid solid accumulation in the WRRF. If the WRRF pays a disposal fee to the HTL conversion plant for algae removal, it will further reduce the conversion system cost. It is assumed that a high-end fee for algae disposal that a WRRF would be willing to pay is the same as the national average municipal solid waste (MSW) tipping fee, which is $53.72/ton (BioCycle 2021). Assuming the disposed algae has a 90% dry matter, the tipping fee of $53.72/ton at as received basis is equal to $98.40/ton algae at AFDW basis (assuming 39% ash content as shown in Table 4). This fee paid to the conversion plant leads to about $1.3/GGE reduction in the MFSP for both cases. Considering wet sludge has been tested in HTL process with effective biocrude conversion

(Snowden-Swan et al. 2021), another option is blending WW-grown algae with sludge from the WRRF to have a 50/50 algae/sludge blended feedstock. This option leads to larger plant scales and thus leads to over 20% reduction in MFSP for both cases. The solid waste after acid digestion contains over 50% C based on the HTL testing and the elemental balance calculation of the acid digestion process. This solid can be sold as biochar for soil amendment. The biochar selling price is assumed to be $300/ton in average based on its cost information in literatures (Yost et al. 2021, Sorensen and Lamb 2018). But the algae biochar application needs to be further investigated for its feasibility. This potential credit leads to $1.0 and $1.3/GGE cost reductions in the MFSP for the baseline and AD centrate case, respectively.

Another sensitivity analysis for plant scale impacts was implemented to investigate a minimum algae tipping fee to be paid by each WRRF to the conversion plant to meet the same baseline MFSP at $2.61/GGE. As shown in Figure 8, if algae as a solid waste from WRRFs needs to be disposed or treated, then a tipping fee can be paid by the WRRFs to the landfill or the treatment plants. To meet the baseline case MFSP, the required algae tipping fee varies for different plant scale. If the upper limit of the tipping fee that a WRRF is willing to pay is equal to the national average landfill tipping fee of $53.72/ton algae (assuming 90% dry matter for algae at as received basis), the plant scale must be larger than 90 dry ton/d (or 55 ton/d AFDW) with baseline process design. The required wastewater flowrate for this algae production rate is 550 MGD if assuming the wastewater used for algae cultivation is primary effluent and its algae production rate is 0.15 g/gallon dry algae.

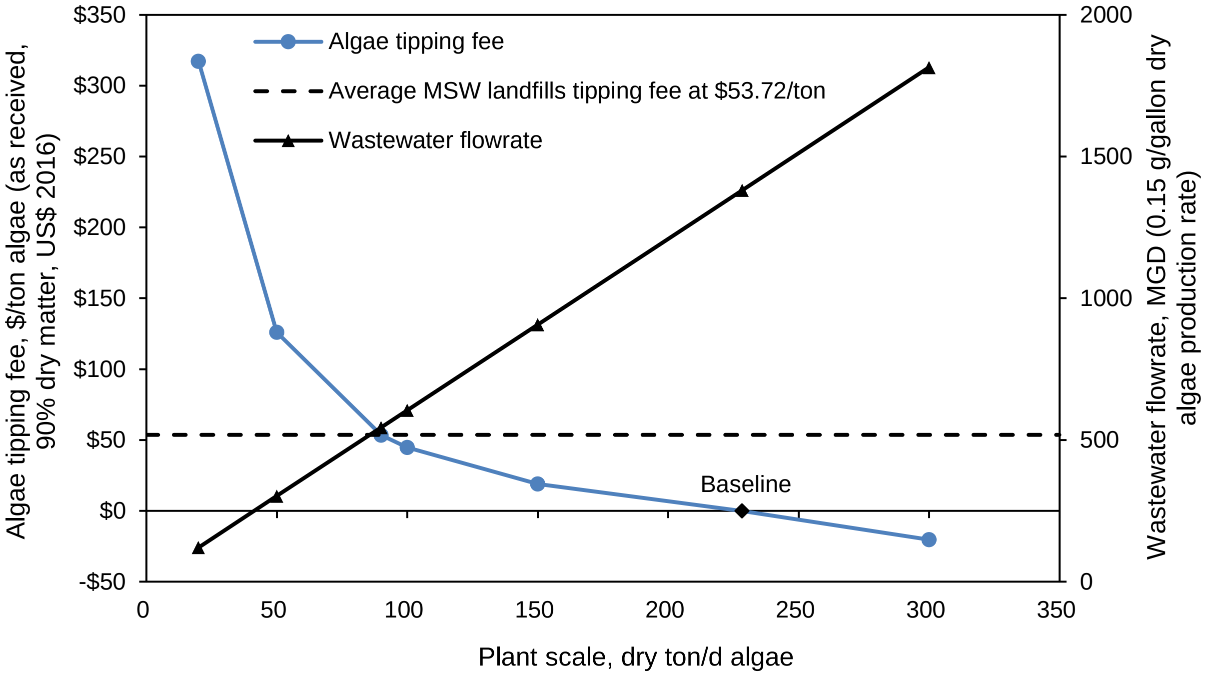

Figure 8. Algae tipping fee based on different plant scales to meet baseline MFSP

4.2.2 Selected Process and Cost Parameters

The cost impacts of other selected parameters are depicted in Figure 9. The algal feedstock cost has significant impacts of the MFSP. When algae generated from the WRRF has other market applications than use as a feedstock for biofuel generation, it is possible for the WRRF to market and sell the material for revenue rather than paying another service to dispose the algae. The struvite price range is based on information in Kim et al. (2021). Based on this range, the MFSP increases 50% when the struvite selling price is at its low end ($0.19/lb) compared to the baseline case. The P content also has important impacts on the MFSP. Lower P content of the same amount of algae feedstock leads to lower co-product credits and thus higher MFSP. When the biocrude yield reduces to 0.2 g/g feedstock AFDW, the MFSP increases 30% compared to the baseline case. For a fixed algae flow rate at AFDW basis and fixed dry solid wt % of the feed slurry, lower ash content leads to lower wet slurry flow rate and thus lower capital and operating cost for HTL process and solid treatment. Assuming the decrease in ash content does not affect the total P amount in the feedstock, the lower ash content at 20% lead to a 16% reduction in the MFSP. Lower ash content has also other impacts on the system based on HTL testing results. The lower ash content enables higher biomass content in the wet slurry for HTL processing, which leads to lower operating cost and higher biocrude yield. Lower ash content in the algae reduces the frequency of blowdown operations within the HTL process, which minimizes the accumulation of ash in the process equipment. Less frequent blowdown operations also reduce biocrude losses to the ash solids. However, the quantitative relationship between ash content and biocrude yield has not been developed and therefore, for this analysis, the impacts of lower ash content on biocrude changes are not considered. More HTL testing data are needed to investigate the impacts of ash content on biocrude yields.

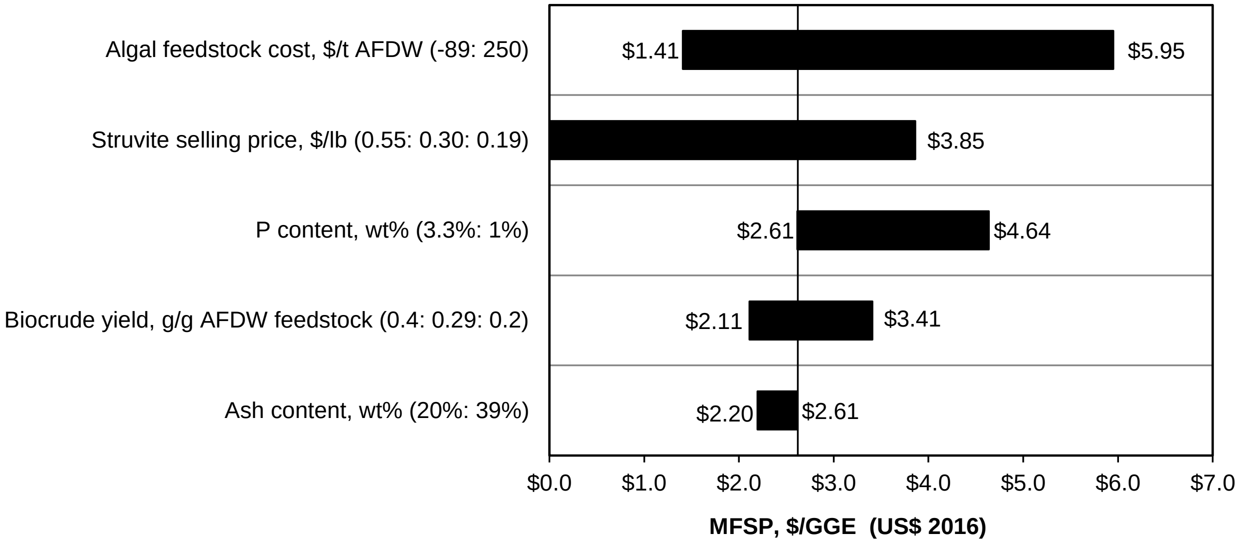

Figure 9. Cost impacts of selected process parameters for FY21 SOT algae HTL and upgrading system.

4.3 Sustainability Metrics

Table 8 lists the conversion sustainability metrics for the SOT cases from 2017 to 2021. The SOT cases from 2017 to 2019 assumed single-stage HTL systems and the recent 2020 case assumed a two-stage sequential HTL system. As the current SOT also assumes a single-stage HTL system, the comparison focuses on differences between the current case and the SOT cases from 2017 to 2019. Because of the lower biocrude yield from algae HTL conversion, the FY21 SOT has a lower final fuel yield. The natural gas consumption for the FY21 is much lower than other cases primarily because part of the carbon contained in the aqueous phase stream is converted to biogas which reduces the natural gas consumption of the system, while other cases assumed direct recycle of aqueous streams to algae ponds. The makeup water usage of the 2021 case on a per unit fuel basis is higher than other cases because of the extra cooling water usage of the AD process and lower final fuel yields. The same reason applies to electricity consumption compared to other single-stage HTL cases (2017 to 2019). The reduction in natural gas and electricity consumption, along with the credits associated with production of struvite fertilizer coproduct should result in significantly lower GHG emissions for the current SOT compared to previous cases. Because of lower fuel yields, FY21 SOT also has lower carbon efficiency and energy efficiency than other single-stage HTL SOT cases.

Table 8. Conversion sustainability metrics.

Input	2017 SOT	2018 SOT	2019 SOT	2020 SOT	2021 SOT
Algae feed flow rate, ton/d AFDW	311	340	598	698	139
Fuel yield, GGE fuel/ton AFDW biomass	104	115	106	78.7	73.4
Co-product yield, lb/ton AFDW biomass	0	0	0	238	771
Natural gas, mmscf/y					
To fuel production (HTL and H_2 plant)	419	475	822	1,069	72.0
To bioprocessing	0	0	0	631	0
Total natural gas usage	419	475	822	1,701	72.0
SCF natural gas/ton AFDW feedstock	4,078	4,228	4,160	7,387	1,574
SCF natural gas/GGE final fuel	39.2	36.9	39.4	93.8	21.5
Makeup water, kg/GGE final fuel	5.16	4.70	5.23	2.99	28.6
Electricity, kwh/GGE final fuel	0.76	0.70	0.73	3.44	1.71
Carbon efficiency					
Fuel C/feedstock C, %	54	58	53	41	38
Fuel + co-product C/feedstock C, %	--	--	--	50	38
Overall products carbon efficiency, %	48	51	47	32	36
Energy efficiency					
Final products/feedstock only, % HHV basis	65	70	64	55	46
Overall efficiency, % HHV basis	54	57	52	44	42

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

5.0 Conclusions and Future Work

As demonstrated by the most recent experimental work, wastewater-grown algae can be effectively converted to biofuel via HTL processing but has lower biocrude yield than previously tested farm-cultivated algae because of its reduced fat and high ash content. Plant scale or algae feed flow rate is important for the production cost of WW-grown algae HTL systems. Using high-volume streams, such as primary effluent from WRRF, or side streams, such as AD centrate, for algae cultivation leads to significant differences in algae production rates and thus the plant scale of the HTL conversion plant. For new WRRFs, using primary effluent for algae cultivation is promising to achieve the BETO 2030 goal of $2.50/GGE. For existing and growing WRRFs, using side streams for algae cultivation can avoid substantial modification and investment of the WRRF system but leads to high HTL conversion cost due to small plant scale. Potential improvements in system design and consideration of nutrient removal credits lead to significant cost reductions for WW-grown algae HTL plants and thus can reduce the impacts of small plant scales on the system cost. When an algae tipping fee is paid by the WRRF to the conversion plant, even smaller plant scales (<139 tons/day) can still meet the baseline MFSP. Combining all the potential cost improvements, using WW-grown algae as feedstock for HTL is a promising pathway to generate market-competitive biofuels in near future.

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

- Larger scale and more testing of WW-grown algae HTL to underpin the design basis for commercial scale WW-grown algae HTL system
- Upgrading testing of biocrude from WW-grown algae HTL conversion, especially for sustainable aviation fuel (SAF) production
- Testing of blended feedstock of algae and wet sludge for HTL processing as an integrated method to treat solid wastes from WRRF and more importantly to increase the conversion plant scale
- Optimizing the struvite generation to maximize P recovery and minimum acid and base consumption.
- Testing and analysis of the elemental composition of insoluble solids after acid digestion of HTL solid product and evaluating its potential application as biochar
- Exploring other low-cost algae feedstock opportunities for the HTL conversion
- Investigating metal recovery opportunities from the residual solids from HTL of low-cost algae feedstocks
- Investigating ash content impacts on biocrude yields and developing quantitative relationship between ash content and biocrude yields if data is available

6.0 References

Aida TM, T Nonaka, S Fukuda, H Kujiraoka, Y Kumagai, R Maruta, M Ota, I Suzuki, MM Watanabe, H Inomata and RL Smith. 2016. "Nutrient recovery from municipal sludge for microalgae cultivation with two-step hydrothermal liquefaction." *Algal Research* 18: 61-68.

Albrecht KO, Y Zhu, AJ Schmidt, JM Billing, TR Hart, SB Jones, G Maupin, R Hallen, T Ahrens and D Anderson. 2016. "Impact of heterotrophically stressed algae for biofuel production via hydrothermal liquefaction and catalytic hydrotreating in continuous-flow reactors." *Algal Research* 14: 17-27.

Arun J, KP Gopinath, SJ Shreekanth, R Sahana, MS Raghavi and D Gnanaprakash. 2019. "Effects of Process Parameters on Hydrothermal Liquefaction of Microalgae Biomass Grown in Municipal Wastewater." *Petroleum Chemistry* 59(2): 194-200.

Annamalai K, JM Sweeten, and SC Ramalingam. 1987. “Estimation of gross heating values of biomass fuels.” *Transactions of the American Society of Agriculture Engineers* 30:1205-1208

Bashar R, Gungor K, Karthikeyan KG, Barak P. 2018. “Cost effectiveness of phosphorus removal processes in municipal wastewater treatment.” Chemosphere 2018 (197): 280–90.

BioCycle CONNECT, 2021. “Average U.S. MSW Landfill Tip Fee Is $53.72/Ton.” February 16, 2021. https://www.biocycle.net/average-u-s-msw-landfill-tip-fee-is-53-72-ton/ (accessed December 2021)

Chen WT, W Qian, Y Zhang, Z Mazur, CT Kuo, K Scheppe, LC Schideman and BK Sharma. 2017. "Effect of ash on hydrothermal liquefaction of high-ash content algal biomass." *Algal Research* 25: 297-306.

Chen WT, Y Zhang, J Zhang, G Yu, LC Schideman, P Zhang and M Minarick. 2014. "Hydrothermal liquefaction of mixed-culture algal biomass from wastewater treatment system into bio-crude oil." *Bioresource Technology* 152: 130-139.

Cheng F, K Mallick, SM Henkanatte Gedara, JM Jarvis, T Schaub, U Jena, N Nirmalakhandan and CE Brewer. 2019. "Hydrothermal liquefaction of Galdieria sulphuraria grown on municipal wastewater." *Bioresource Technology* 292: 121884.

Clippinger J and R Davis. 2021. *Techno-Economic Assessment for Opportunities to Integrate Algae Farming with Wastewater Treatment*. NREL/TP-5100-75237. National Renewable Energy Laboratory, Golden, CO. September 2021

Couto EA, F Pinto, F Varela, A Reis, P Costa and ML Calijuri. 2018. "Hydrothermal liquefaction of biomass produced from domestic sewage treatment in high-rate ponds." *Renewable Energy* 118: 644-653.

Craggs, RJ, TJ Lundquist and JR Benemann. 2013. *Wastewater Treatment and Algal Biofuel Production*. In: Borowitzka M., Moheimani N. (eds) Algae for Biofuels and Energy. Developments in Applied Phycology, vol 5. Springer, Dordrecht: 153-163.

Davidson SD, JA Lopez-Ruiz, Y Zhu, AR Cooper, KO Albrecht and RA Dagle. 2019. "Strategies to Valorize the Hydrothermal Liquefaction-Derived Aqueous Phase into Fuels and Chemicals." *ACS Sustainable Chemistry & Engineering* 7(24): 19889-19901

Davis R, M Wiatrowski, C Kinchin and D Humbird. 2020. *Conceptual Basis and Techno-Economic Modeling for Integrated Algal Biorefinery Conversion of Microalgae to Fuels and Products: 2019 NREL TEA Update: Highlighting Paths to Future Cost Goals via a New Pathway for Combined Algal Processing*. NREL/TP-5100-75168, National Renewable Energy Laboratory, Golden, CO. September 2020.

Davis R and B Klein. 2021. *Algal Biomass Production via Open Pond Algae Farm Cultivation: 2020 State of Technology and Future Research*. NREL/TP-5100-79931, National Renewable Energy Laboratory, Golden, CO. May 2021.

Elliott DC, P Biller, AB Ross, AJ Schmidt and SB Jones. 2015. "Hydrothermal liquefaction of biomass: Developments from batch to continuous process." *Bioresource Technology* 178: 147-156

Fortier MOP, GW Roberts, SM Stagg-Williams and BSM Sturm. 2014. "Life cycle assessment of bio-jet fuel from hydrothermal liquefaction of microalgae." *Applied Energy* 122: 73-82

Gross M, D Jarboe and Z Wen. 2015. "Biofilm-based algal cultivation systems." *Applied Microbiology and Biotechnology* 99(14): 5781-5789

Hawthorne M. 2016. “Chicago turning river pollutants into fertilizer.” *Chicago Tribune*. May 2016. https://www.chicagotribune.com/investigations/ct-chicago-phosphorus-pollution-met-20160525-story.html. (accessed in December 2021)

Jiang Y, SB Jones, Y Zhu, L Snowden-Swan, AJ Schmidt, JM Billing and D Anderson. 2019. "Techno-economic uncertainty quantification of algal-derived biocrude via hydrothermal liquefaction." *Algal Research* 39: 101450.

JJ Environmental. 2015. *Final Report—Low Cost Retrofits for Nitrogen Removal at Wastewater Treatment Plants in the Upper Long Island Sound Watershed*. Prepared for New England Interstate Water Pollution Control Commission. March 2015

Jones SB, Y Zhu, LJ Snowden-Swan, D Anderson, RT Hallen, AJ Schmidt, KO Albrecht and DC Elliott. 2014. *Whole Algae Hydrothermal Liquefaction: 2014 State of Technology*. PNNL-23867. Pacific Northwest National Laboratory, Richland, WA. June 2014.

Juneja A and GS Murthy. 2017. "Evaluating the potential of renewable diesel production from algae cultured on wastewater: techno-economic analysis and life cycle assessment." *AIMS Energy* 5(2): 239-257.

Kesaano M and RC Sims 2014. "Algal biofilm based technology for wastewater treatment." *Algal Research* 2014 (5): 231-240.

Kim AH, YC Anthony, SH El Abbadi, K Lu, D Chan, EA Appel and CS Criddle. 2021. “More than a fertilizer: wastewater-derived struvite as a high value, sustainable fire retardant.” *Green Chem.* 2021 (23): 4510–4523.

Lundquist T and R Spierling 2018. *Final Report: Scale-up of Algal Biofuel Production Using Waste Nutrients*, California Polytechnic State University, San Luis Obispo, CA.

Lowry PP and KA Wagner. 2015. *Hydrothermal Liquefaction Treatment Preliminary Hazard Analysis Report*, PNNL-24219. Pacific Northwest National Laboratory, Richland, WA.

McGinn PJ, KC Park, G Robertson, L Scoles, W Ma and D Singh. 2019. "Strategies for recovery and recycling of nutrients from municipal sewage treatment effluent and hydrothermal liquefaction wastewaters for the growth of the microalga Scenedesmus sp. AMDD." *Algal Research* 38: 101418

MWRD 2021. *Water Reclamation Plants Fact Sheets*. Metropolitan Water Reclamation District of Greater Chicago. Chicago, IL. https://mwrd.org/water-reclamation-plants (accessed in December 2021)

Novoveská L, AKM Zapata, JB Zabolotney, MC Atwood, ER Sundstrom. 2016. “Optimizing microalgae cultivation and wastewater treatment in large-scale offshore photobioreactors.” *Algal Research* 2016 (18): 86–94

Ovsyannikova E, A Kruse and GC Becker. 2020. “Feedstock-Dependent Phosphate Recovery in a Pilot-Scale Hydrothermal Liquefaction Bio-Crude Production”. *Energies* 2020 (13): 379.

Park N, H Chang, Y Jang, H Lim, J Jung and W Kim. 2020. “Critical conditions of struvite growth and recovery using MgO in pilot scale crystallization plant.” *Water Sci Technol* (2020) 81 (12): 2511–2521

Pittman JK, AP Dean, O Osundeko. 2011. “The potential of sustainable algal biofuel production using wastewater resources.” *Bioresource Technology* 102 (1): 17-25

Preisner M, E Neverova-Dziopak, Z Kowalewski. 2020. “Analysis of eutrophication potential of municipal wastewater.” *Water Sci Technol* 81 (9): 1994-2003

Ranganathan P and S Savithri. 2019. "Techno-economic analysis of microalgae-based liquid fuels production from wastewater via hydrothermal liquefaction and hydroprocessing." *Bioresource Technology* 284: 256-265.

Roberts GW, MOP Fortier, BSM Sturm and SM Stagg-Williams. 2013. "Promising Pathway for Algal Biofuels through Wastewater Cultivation and Hydrothermal Conversion." *Energy & Fuels* 27(2): 857-867.

Seiple TE, RL Skaggs, L Fillmore and AM Coleman 2020. "Municipal wastewater sludge as a renewable, cost-effective feedstock for transportation biofuels using hydrothermal liquefaction." *Journal of Environmental Management* 270: 110852.

Shanmugam SR, S Adhikari and R Shakya 2017. "Nutrient removal and energy production from aqueous phase of bio-oil generated via hydrothermal liquefaction of algae." *Bioresource Technology* 230: 43-48

Snowden-Swan LJ, JM Billing, MR Thorson, AJ Schmidt, Y Jiang, DM Santosa, and TE Seiple, et al. 2021. *Wet Waste Hydrothermal Liquefaction and Biocrude Upgrading to Hydrocarbon Fuels: 2020 State of Technology*. PNNL-30982, Pacific Northwest National Laboratory, Richland, WA.

Sorensen RB and MC Lamb 2018. Return on investment from biochar application. *Crop, Forage & Turfgrass Management* 2018, 4(1): 1–6.

Sturm BSM and SL Lamer 2011. "An energy evaluation of coupling nutrient removal from wastewater with algal biomass production." *Applied Energy* 88(10): 3499-3506.

Thorson MR, DM Santosa, RT Hallen, I Kutnyakov, MV Olarte, M Flake, G Neuenschwander, L Middleton-Smith, AH Zacher, TR Hart, AJ Schmidt, T Lemmon and M Swita. 2021. "Scalable Hydrotreating of HTL Biocrude to Produce Fuel Blendstocks." *Energy & Fuels* 35(14): 11346-11352

U.S. Department of Energy. 2016. *Bioenergy Technologies Office Multi-Year Program Plan. Bioenergy Technologies Office*. Energy Efficiency and Renewable Energy, U.S. Department of Energy, Washington D.C. March 2016.

U.S. Department of Energy. 2019. *Bioenergy Technologies Office - FY19 Multi-Year Plan*. Bioenergy Technologies Office. Energy Efficiency and Renewable Energy, U.S. Department of Energy, Washington D.C. 2019.

Wang Z, S Adhikari, P Valdez, R Shakya and C Laird. 2016. "Upgrading of hydrothermal liquefaction biocrude from algae grown in municipal wastewater." *Fuel Processing Technology* 142: 147-156.

Yost M, J Holt, J Austin, E Creech, D McAvoy, D Winward and N Allen. 2021. *Biochar impacts on crop yield and soil water availability [Fact sheet].* Utah State University Extension. April 2021.

Zhou Y, L Schideman, G Yu and Y Zhang. 2013. "A synergistic combination of algal wastewater treatment and hydrothermal biofuel production maximized by nutrient and carbon recycling." *Energy & Environmental Science* 6(12): 3765-3779

Zhu Y, SB Jones, AJ Schmidt, KO Albrecht, SJ Edmundson and DB Anderson 2019. "Techno-economic analysis of alternative aqueous phase treatment methods for microalgae hydrothermal liquefaction and biocrude upgrading system." *Algal Research* 39: 101467.

Zhu Y, SB Jones, AJ Schmidt, JM Billing, DM Santosa and DB Anderson 2020a. "Economic impacts of feeding microalgae/wood blends to hydrothermal liquefaction and upgrading systems." *Algal Research* 51: 102053.

Zhu Y, SB Jones, AJ Schmidt, JM Billing, MR Thorson, DM Santosa, RT Hallen, and DB Anderson. 2020b. *Algae/Wood Blends Hydrothermal Liquefaction and Upgrading: 2019 State of Technology*. PNNL-29861, Pacific Northwest National Laboratory, Richland, WA.

Zhu Y, SB Jones, AJ Schmidt, HM Job, JM Billing, JR Collett, KR Pomraning, SP Fox, TR Hart, SJ Edmundson, MR Thorson, PA Meyer, LJ Snowden-Swan and DB Anderson 2021. *Microalgae Conversion to Biofuels and Biochemical via Sequential Hydrothermal Liquefaction (SEQHTL) and Bioprocessing: 2020 State of Technology*. PNNL-30124, Pacific Northwest National Laboratory, Richland, WA. April 2021

Appendix. Detailed SOT Costs

Processing Area Cost Contributions & Key Technical Parameters	**Metric**	**2017 SOT Florida - no liners**	**2018 SOT Florida - no liners**	**2019 SOT Florida - no liners**	**2020 SOT Florida - no liners**	**2021 SOT**
Fuel selling price	$/gge	$8.05	$6.83	$4.98	$4.48	$2.61
Conversion Contribution	$/gge	$1.39	$1.22	$0.88	($0.33)	$2.61
Production Diesel	mm gge/yr	7.1	8.9	13.7	12	2.4
Production Naphtha	mm gge/yr	3.6	4.0	6.6	6.3	1.0
Diesel Yield (AFDW feedstock basis)	gge/US ton feedstock	69	79	70	51	52
Naphtha Yield (AFDW feedstock basis)	gge/US ton feedstock	35	36	33	27	22
Diesel Yield (areal basis)	gge/acre-yr	1,416	1,771	2,746	2,365	6,705
Naphtha Yield (areal basis)	gge/acre-yr	724	800	1,310	1,261	2,804
Co-product Yield (AFDW feedstock basis)	lb /lb feedstock	0	0	0	0.12	0.39
Natural Gas Usage-drying (AFDW feedstock basis)	scf/US ton feedstock	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
Carbon from Biomass in Fuels	%	54%	58%	53%	41%	38%
Carbon from Biomass in Other Productsc	%	0%	0%	0%	10%	0%
Feedstock						
Total Cost Contribution	$/gge fuel	$6.66	$5.61	$4.10	$4.81	$0.00
Feedstock Type	Algae with non-algae feedstock supplement in non-summer seasons	*Algae with wood supplement*	*Algae with wood supplement*	*Algae with wood supplement*	*Algae with corn stover suppplement*	*Algae grown in wastewater*
Feedstock Cost (AFDW basis)	$/US ton feedstock	$694	$643	$421	$379	$0
Algae storage						
Total Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.00	$0.14
Capital Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.00	$0.09
Operating Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.00	$0.06
HTL Biocrude Production						
Total Cost Contribution	$/gge fuel	$0.95	$0.84	$0.75	$1.54	$2.75
Capital Cost Contribution	$/gge fuel	$0.56	$0.50	$0.47	$0.56	$1.39
Operating Cost Contribution	$/gge fuel	$0.39	$0.34	$0.28	$0.98	$1.36
Liquid Hourly Space Velocity (LHSV)	vol/h/vol	4.0	4.0	4.0	Stage I: 4; Stage II: 3.5	4.0
HTL Carbohydrate Extraction	%, extracted/carbohydrate in feedstock	0%	0%	0%	58%	0%
HTL Biocrude Yield (AFDW)	lb /lb feedstock	0.41	0.45	0.41	0.30	0.29
HTL Biocrude Hydrotreating to Finished Fuels						
Total Cost Contribution	$/gge fuel	$0.69	$0.59	$0.42	$0.30	$0.83
Capital Cost Contribution	$/gge fuel	$0.30	$0.27	$0.23	$0.17	$0.39
Operating Cost Contribution	$/gge fuel	$0.39	$0.32	$0.19	$0.13	$0.45
Mass Yield on dry HTL Biocrude	lb/lb biocrude	0.81	0.82	0.81	0.83	0.82
HTL Aqueous Phase Treatment						
Total Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.00	$1.21
Capital Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.00	$0.46
Operating Cost Contribution	$/gge fuel	$0.00	0.00	0.00	0.00	$0.75
Bioprocessing for Co-product Generation						
Total Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$1.43	$0.00
Capital Cost Contribution	$/gge fuel	$0.00	$0.00	$0.00	$0.64	$0.00
Operating Cost Contribution	$/gge fuel	$0.00	0.00	0.00	0.79	0.00
Fermentation Productivity	g/L-hr	0	0	0	0.46	0.00
Fermentation Process Yield	g product/g extracted carbohydrates	0	0	0	0.37	0.00
Balance of Plant						
Total Cost Contribution	$/gge fuel	$0.61	$0.57	$0.49	$0.74	$0.87
Capital Cost Contribution	$/gge fuel	$0.29	$0.28	$0.23	$0.41	$0.49
Operating Cost Contribution	$/gge fuel	$0.31	$0.29	$0.26	$0.34	$0.37
Co-product Credits	$/gge fuel	$0.00	0.00	0.00	(2.92)	(3.19)
Nutrient Recycle Credits	$/gge fuel	($0.86)	(0.78)	(0.78)	(1.43)	0.00
Models: Case References		Blend-111317-17SOT-16$-FL-NL-R2	Blend-092018-18SOT-16$-FL-NL	Blend-092019-19SOT-16$-FL-NL	Blend-020821-SEQHTL-FY20SOT-FL-NL.xlsm	WWT algae-120621-21SOT-16$

**Pacific Northwest
National Laboratory**

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99354
1-888-375-PNNL (7665)

www.pnnl.gov