Pacific Northwest NATIONAL LABORATORY

PNNL-37209

Whole Algae Hydrothermal Liquefaction and Upgrading

A review of progress and challenges and insight into the future

January 2025

Jacob Watkins
Abhishek Kumar
Peter Valdez

U.S. DEPARTMENT OF ENERGY

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

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

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Summary

This report summarizes the research at Pacific Northwest National Laboratory (PNNL) to evaluate the economic viability and environmental impact of using microalgae to produce fuels and other products via hydrothermal liquefaction (HTL). Over the past several years, PNNL has examined key aspects of feedstock cost and availability, formatting and conversion techniques, and the utilization of all HTL products. Investigations of feedstock cost led to opportunities to work with cost-advantaged algal feedstocks that can be provided at minimal cost for HTL processing. Cost-advantaged algae include wastewater-grown algae and harvested algal blooms. Although farm-cultivated algae offer the best possible biomass composition and scalability for HTL processing, the cost of the feedstock is too high to yield an economically competitive biofuel. Processing cost-advantaged feedstocks creates other unique challenges in adapting HTL to upgrade biomass with higher than typical ash content and less preferred composition (low lipid). Despite the challenges, HTL of cost-advantaged algae results in economically competitive pricing scenarios and significant advantages in reducing net emissions below 70% of the petroleum baseline. The utilization of a variety of potential non-fuel products from algal HTL, such as the use of HTL solids as a cement additive, provides a significant reduction in net emissions by offsetting emissions from other carbon-intense products. This report presents an analysis of the research conducted at PNNL to develop an economically and environmentally beneficial process for algae HTL.

Acknowledgments

We thank Hao Cai and Longwen Ou from Argonne National Laboratory (ANL) for compiling results from previous reports of the Supply Chain Sustainability Assessment, adding life-cycle assessment (LCA) outcomes to this report.

Acronyms and Abbreviations

ABB	Algal bloom biomass
ANL	Argonne National Laboratory
BETO	Bioenergy Technologies Office
FY	fiscal year
GGE	gallon gasoline equivalent
GHG	greenhouse gas
HTL	hydrothermal liquefaction
LCA	life cycle analysis
LEA	lipid extracted microalgae
LHSV	liquid hourly space velocity
MFSP	minimum fuel selling price
MHTLS	modular hydrothermal liquefaction system
NAABB	National Alliance for Advanced Biofuels and Bio-Products
NREL	National Renewable Energy Laboratory
PNNL	Pacific Northwest National Laboratory
SCM	supplementary cementitious material
SEQHTL	sequential hydrothermal liquefaction
SOT	state of technology
TEA	technoeconomic assessment

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1.0 Introduction

The purpose of this document is to summarize advancements made to improve the economic viability and technical deployability of the hydrothermal liquefaction (HTL) of algal biomass to produce fuels and other products. The deployment of algal-based fuels and products presents an opportunity to reduce net carbon emissions compared to traditional fossil-fuel based products.

Presented herein is a review of previous techno-economic assessments (TEA) from PNNL-published results. Since 2013, reported experimental outcomes were used to inform a conceptual design for an industry-scale HTL facility. The design assumed n^{th}-plant economics, meaning that the designed facility was assumed to be capital and operations optimized, with little to no redundancies or inefficiencies. In other words, the best possible outcome was projected based on scale-up of the current state of technology (SOT). TEA investigations parallel to experimental work informed PNNL researchers of critical topics for technological improvements to achieve more favorable economic outcomes.

As early as the 1980’s, PNNL had investigated the processing of wet biomass to produce fuels (Elliott, 1980; Elliott and Sealock, 1985). At the time, residues from agriculture and silviculture specific to the Pacific Northwest were the feedstocks of interest, including some kelp biomass. Interest in wet biomass processing grew in the late 2000’s with the initiation of the National Alliance for Advanced Biofuels and Bio-products (NAABB), which emphasized development of the HTL pathway for algae as shown in Figure 1 (Olivares et al., 2014). The objective of NAABB was to examine the entire pathway for algal biofuels, encompassing the major process steps of cultivation, harvesting, and conversion. The reported achievement of NAABB was the reduction of the estimated minimum fuel selling price (MFSP) of algal-based fuels from $250 to $7.50 per gallon.

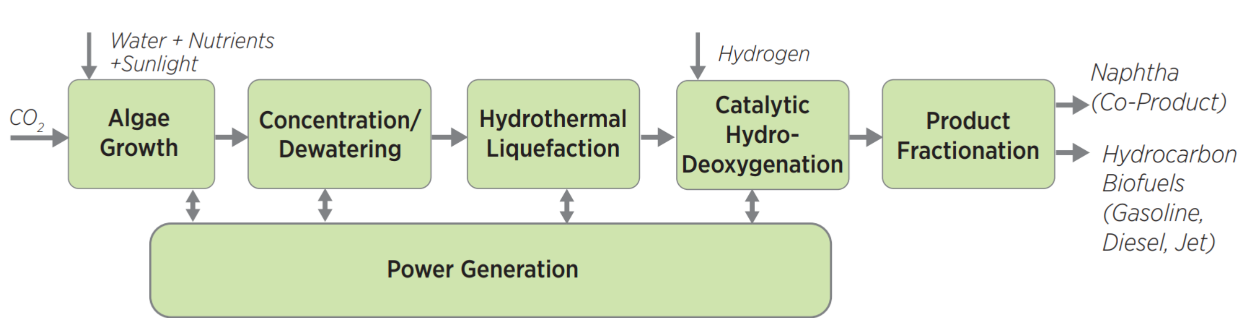

Figure 1. Process Block Diagram for Whole Algae Hydrothermal Liquefaction (BETO, 2012)

In 2014, the first TEA and SOT report for whole algae hydrothermal liquefaction was published by PNNL (Jones et al., 2014). Since 2014, multiple SOT and design case reports have been published to track the overall progress and development of the algal HTL pathway. Advancements in process technology, process configuration, feedstock cultivation and preparations, and co-product and waste disposition have all contributed to improving the potential scale-up and deployment of HTL. Advancements and outcomes have been shared across projects and institutions, enabling HTL pathways for other wet feedstocks such as sewage solids, food wastes, and animal manures.

The continued development of the algal HTL pathway is needed as unique sources of readily available algae and potential product pathways are investigated.

2.0 Summary of Published Reports

This section presents a summary of year-to-year changes and progress described in the PNNL SOT and design case study reports for algae HTL from 2014 through 2024. The changes summarized here include modifications to the design of HTL process technology, improvements in biocrude upgrading techniques, development of co-product pathways, and new scenarios for feedstock acquisition. Highlights of the changes and advancements, in addition to scenarios investigated for potential commercial-scale embodiments of algae HTL, are summarized in Table 1. The net effect of the changes on the net greenhouse gas (GHG) emissions and the MFSP of biofuels produced through HTL of algae biomass are also presented in this section. The cost data in this and subsequent sections of the report have been normalized to the 2020 cost year.

Table 1 . Summary of year-to-year process improvements and scenarios

Year	Process improvements and scenarios	Reference
2014	The first PNNL report on algae HTL is published	(Jones et al., 2014)
2015	Capital costs are reduced by increasing the liquid hourly space velocity of the HTL reactor	Unpublished PNNL-internal document
2016	The aqueous co-products from HTL are recycled to algae cultivation ponds, rather than processed through hydrothermal gasification	Unpublished PNNL-internal document
2017	Blendstocks (mainly wood) are introduced to manage seasonal variations in the availability of algae biomass	(Zhu et al., 2020)
2018	Improvements to experimental methods and equipment for HTL improve outcomes for the quality and yield of HTL biocrude from algae	(Zhu et al., 2020)
2019	Capital costs are reduced through economies of scale by considering larger HTL systems	(Zhu et al., 2020)
2020	A two-stage sequential HTL system (SEQHTL) is evaluated using corn stover as a blendstock for HTL and producing lactic acid as a co-product	(Zhu et al., 2021)
2021	Wastewater-grown algae is used as a zero-cost feedstock with struvite fertilizer as a co-product	(Zhu et al., 2022)
2022	A pathway to sustainable aviation fuel via HTL is emphasized with proteins from algae as a co-product	(Zhu et al., 2023)
2023a	Steam flash vessels replace heat exchangers to improve the reliability of HTL equipment, using algal bloom biomass as a zero-cost feedstock, blended with farm-grown algae and woody biomass	(Xu et al., 2024)
2023b	100% algal bloom biomass (ABB) is used as the HTL feedstock	(Xu et al., 2024)
2023c	ABB blended with farm-grown algae is the HTL feedstock	(Xu et al., 2024)
2024a,b	Wastewater-grown algae is the feedstock, blended with wastewater sludge	(Kumar et al., 2024)
2024c	Wastewater-grown algae is the feedstock, blended with biosolids from anaerobic digestion	(Kumar et al., 2024)

Figure 2 presents a cost breakdown of the scenarios for algae HTL examined since 2014. In the earlier years of research (prior to 2020), major progress was made in developing an HTL framework which produced useable biofuel products from mass-cultivated or farmed algae at minimum cost. Improvements made during the initial development period include updates to the process design of HTL, supported by experimental design and results, directly recycling HTL products to cultivation ponds, and selecting more robust and effective catalysts for the upgrading of the biocrude to fuel products. These improvements reduced the cost of conversion from

$3.67/gallon gasoline equivalent (GGE) in 2015 to $0.98/GGE in 2019, corresponding to a total MFSP of $5.41/GGE. Concurrent with the development of the HTL processing, experimental improvements were being investigated and applied to the cost estimations for cultivation algae biomass. Estimates from the National Renewable Energy Laboratory (NREL) showed a reduction in the cost of feedstock preparation from $15.95/GGE in 2015 to $4.44/GGE in 2019 (Klein et al., 2024). The net effect of these improvements was a year-over-year reduction in the calculated MFSP, as shown in Figure 2.

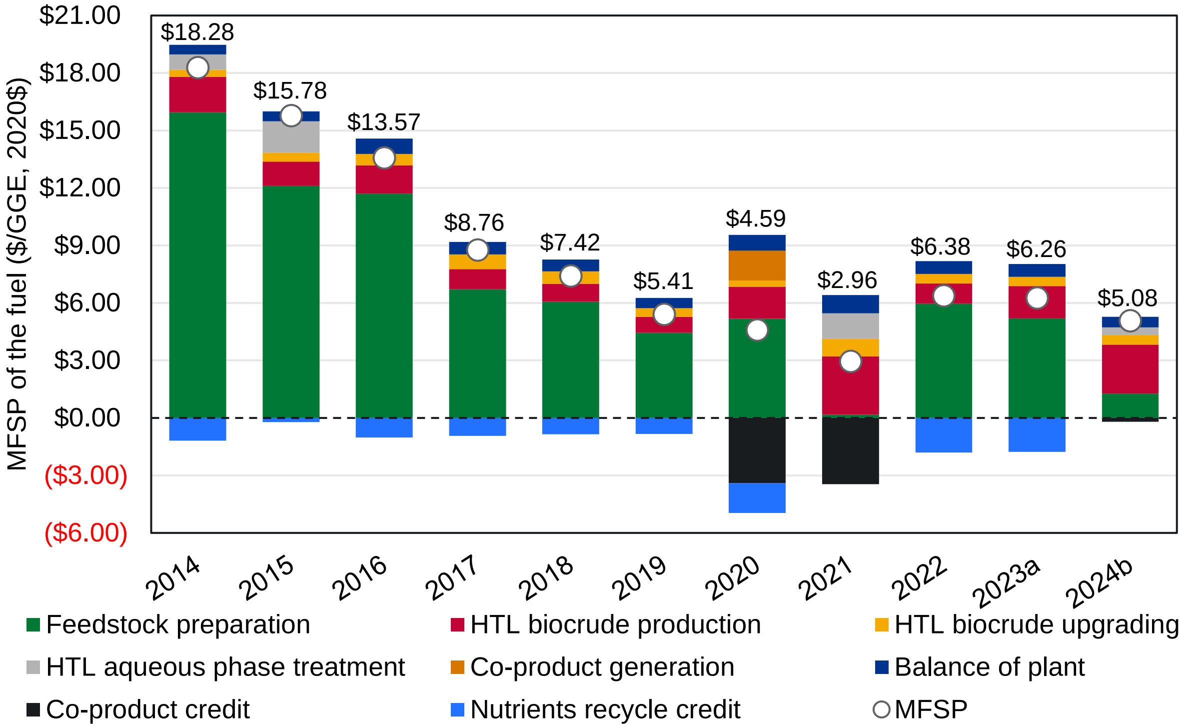

Figure 2. All contribution categories for MFSP

From 2014 to 2020 and in 2022 and 2023, the cost of the algal feedstock was the primary contributor to the cost of the algal fuel product. To offset the cost of feedstock, research efforts since 2020 have emphasized the use of low- and zero-cost algal feedstocks (a.k.a. cost-advantaged feedstocks) and/or the production and sale of co-products. Examples of more recently examined feedstocks include protein-rich farmed algae, algal residues from nutraceutical processes, wastewater-grown algae, and algal bloom biomass. Potential co-products include supplementary cementitious materials (SCMs; 2023, 2024), food-grade proteins, struvite fertilizer (2021), and lactic acid (2020).

Improvements to HTL technology include significant updates to the design of the HTL system in 2020 and 2023. The recent improvements in 2023 include replacing heat exchangers with steam flash vessels to improve process reliability, transporting untreated biocrude to existing refineries for hydrotreating instead of in-situ upgrading, and optimizing the scale of the HTL facility to best match the feedstock scenario (Li et al., 2024).

A summary of HTL processing scales, feedstocks with blendstocks, and corresponding MFSP is presented in Figure 3. Purchased feedstocks include wood, corn stover, and farm-cultivated algae, with algae prices based on the NREL algae cultivation models for each respective year (Klein et al., 2024). Zero-cost feedstocks include solid waste products from water reclamation facilities, biomass from harvested algal blooms, and algae cultivated using nutrients from wastewater. Zero-cost feedstocks are readily available cost-advantaged feedstocks. In general, the calculated MFSP for HTL processes using zero-cost feedstocks is lower than the MFSP for processes using purchased feedstocks. The scale of production for zero-cost feedstocks is significantly lower than the scale of production for the farmed feedstocks. The two lowest values for MFSP are reported in 2021 and 2024, each using wastewater-grown algae as the primary feedstock. In 2023 and 2024, multiple feedstock combinations and processing scales were modeled within the respective analysis year.

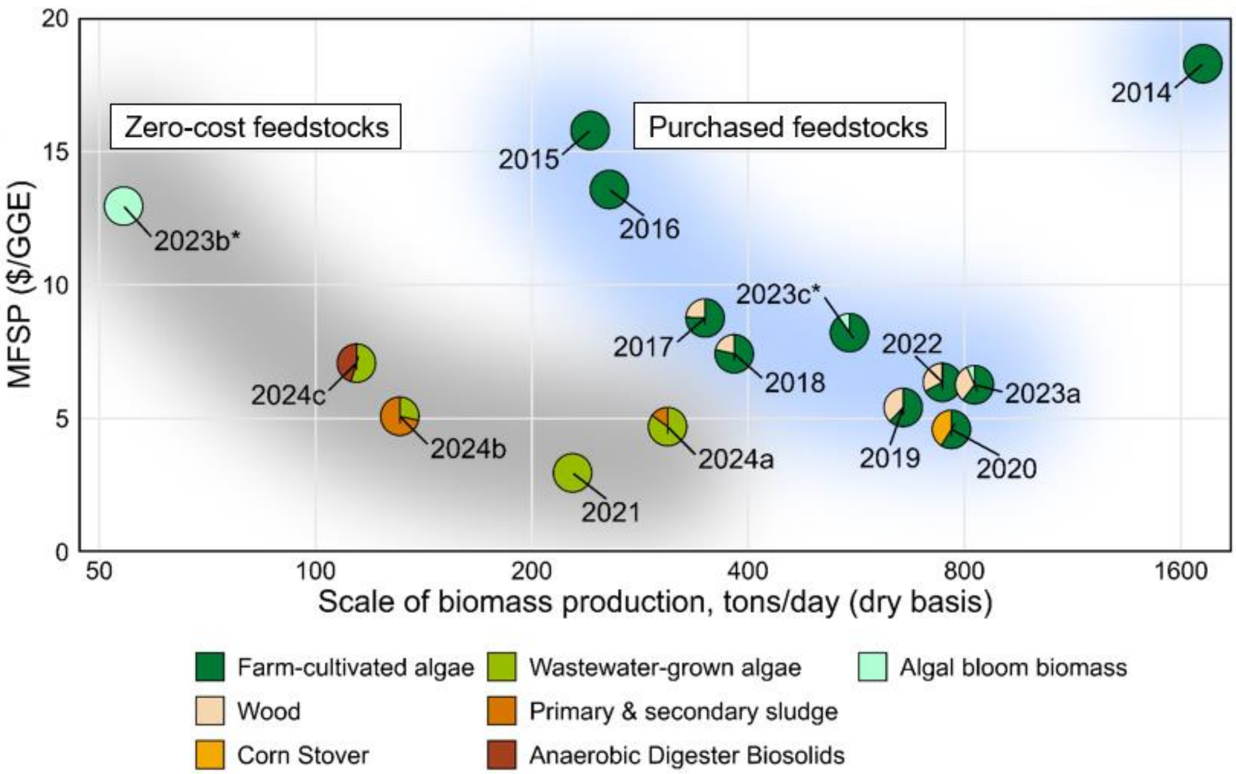

Figure 3. Biomass conversion scale, minimum product selling price, and feedstock source for HTL scenarios modeled between 2014 and 2024. Scenarios marked with an asterisk (*) exclude the cost of hydrotreating biocrude to finished fuel products.

Figure 4 displays the calculated GHG emissions for algal HTL scenarios modeled between 2019 and 2023. LCA was completed by colleagues at ANL. Analyses from previous years are omitted as the scenario for 2019 represents the lowest achievable emissions using farm-cultivated algae. The displacement method is used to quantify the impact of replacing fuels and HTL co-products in existing markets. As shown in Figure 4, the major contributors to emissions from biofuel production are cultivation and conversion of the algal feedstock. The lowest GHG emissions were achieved in 2021 and 2023, which used zero-cost feedstocks, wastewater-grown algae and algal bloom biomass, respectively, with no GHG emissions attributed to feedstock production. Co-product and carbon sequestration credits offset GHG emissions and contribute to the negative net GHG emissions achieved in 2021 and 2023. The highest net GHG emissions occurred in 2020, which used a two-stage sequential HTL system (SEQHTL) to produce fuels and fermentable sugars. The three keys to successfully reducing the emission impact of algal HTL are the use of non-farm algal feedstock, the displacement of emission-intense products like fertilizers or SCMs, and the opportunity to sequester a portion of the algal carbon.

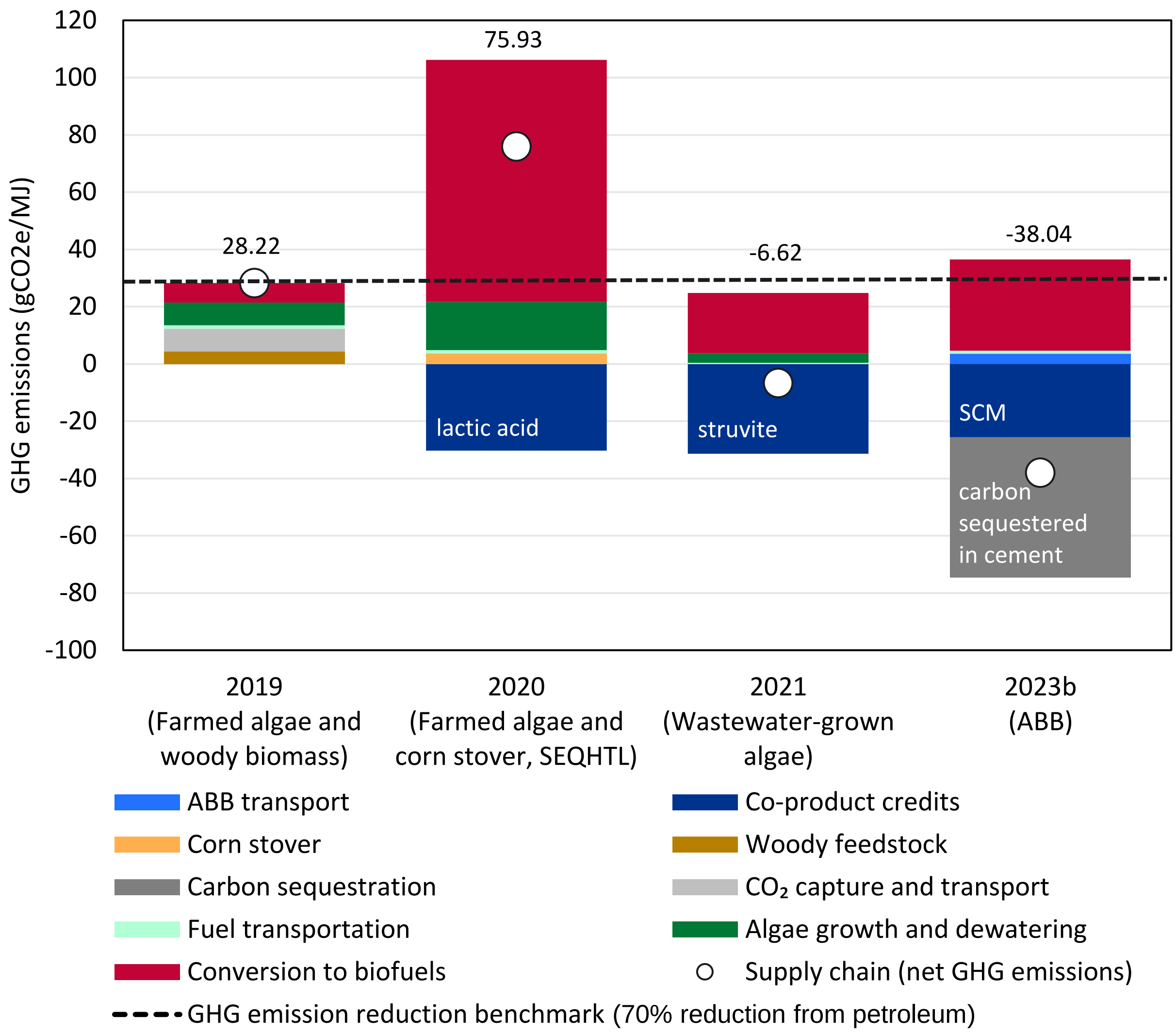

Figure 4. Supply chain GHG emission contributions for algae fuels via HTL

3.0 Key Cost/Process Drivers

Reports from PNNL identified the cost of the feedstock and the cost of conversion as key contributors to the MFSP. This section will discuss the research accomplishments and process assumptions that reduced the MFSP of algal-derived biofuels via HTL. The total capital cost for conversion, the sourcing of alternative feedstocks and blendstocks, and the value of co-products are discussed.

Capital Cost for Conversion

The cost of capital equipment contributes 50% to 76% of the total cost of conversion for single-stage HTL. As the process is scaled to less than 110 dry tons per day of processing capacity, as calculated in the 2023 and 2024 reports, sensitivity analysis demonstrates that the capital cost and plant scale were the most influential factors impacting MFSP. The capital cost can be reduced through economies of scale, as shown in Figure 3. There are limitations in scaling, for example, a cost-advantaged feedstock like algal bloom biomass cannot be scaled up and is reliant on the seasonal availability of biomass. Although wastewater-grown algae have significant potential, only a small number (~30/15,000) of wastewater treatment facilities are large enough to produce sufficient algal biomass (>100 dry tons per day) for HTL to reach economies of scale with existing technology (Seiple et al., 2017). One potential opportunity is to consider smaller scale (<100 dry tons per day), more capital-efficient HTL systems to increase the adoption of the technology.

Since 2013, experimental findings have been used to inform design and process assumptions resulting in improvements to the HTL design and cost. Unique designs such as two-stage SEQHTL were introduced as means to produce fermentable sugars, therefore creating a co-product stream to be monetized. Ultimately, SEQHTL resulted in negative outcomes, increasing cost, complexity, and GHG emissions that could not be offset by a co-product of fermentable sugars and fermentation products (i.e., lactic acid) (Zhu et al., 2021).

Additional innovations were introduced to improve throughput and reliability of HTL processing. In 2020, the heat recovery system for HTL was split into 2 sections to improve the effectiveness of heat transfer in the viscous slurry of biomass feedstock (Snowden-Swan et al., 2021). In 2023, additional changes were made to the HTL design (Li et al., 2024). In the previous design, the heat recovery system was implemented to improve energy efficiency. Based on laboratory operations, the heat recovery system was prone to fouling and clogging. It was proposed to replace the heat exchanger system with a steam flashing and recovery system. The use of flash vessels to transfer heat from products to the reactants prevents fouling and is envisioned to improve the process reliability at a commercial scale. Figure 5 and Figure 6 show the change in process configurations in 2020 and 2023, respectively. Additional improvements to the HTL design have been proposed, including solvent extraction to improve the recovery of biocrude (Kilgore et al., 2024) and autothermal processing to reduce energy requirements for heating (Thorson et al., 2024). The proposed improvements are still being investigated at the laboratory scale.

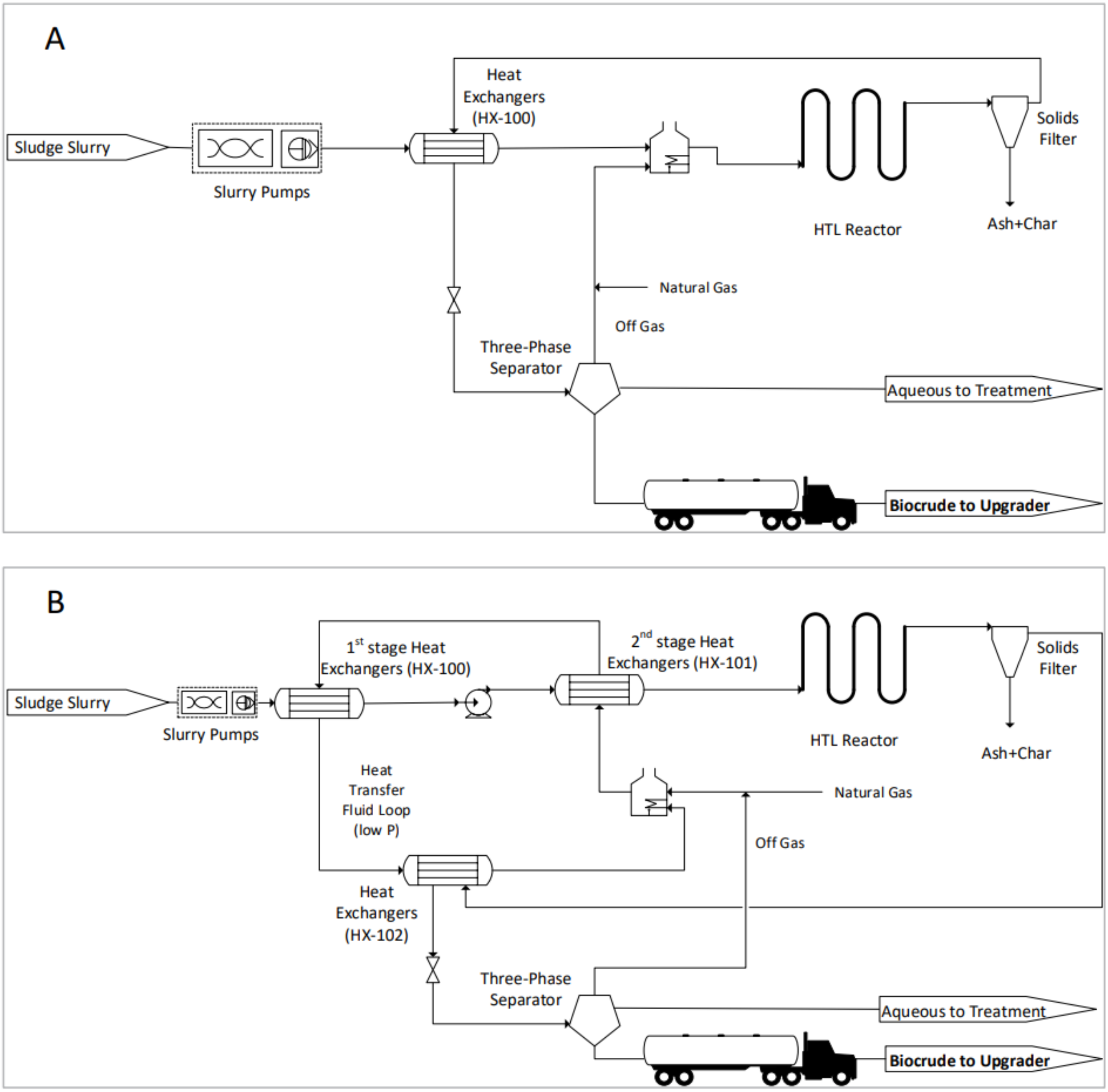

Figure 5. Updates made in 2020 to create 2 zones for heat recovery, showing the previous (a) and updated (b) configurations (Snowden-Swan et al., 2021)

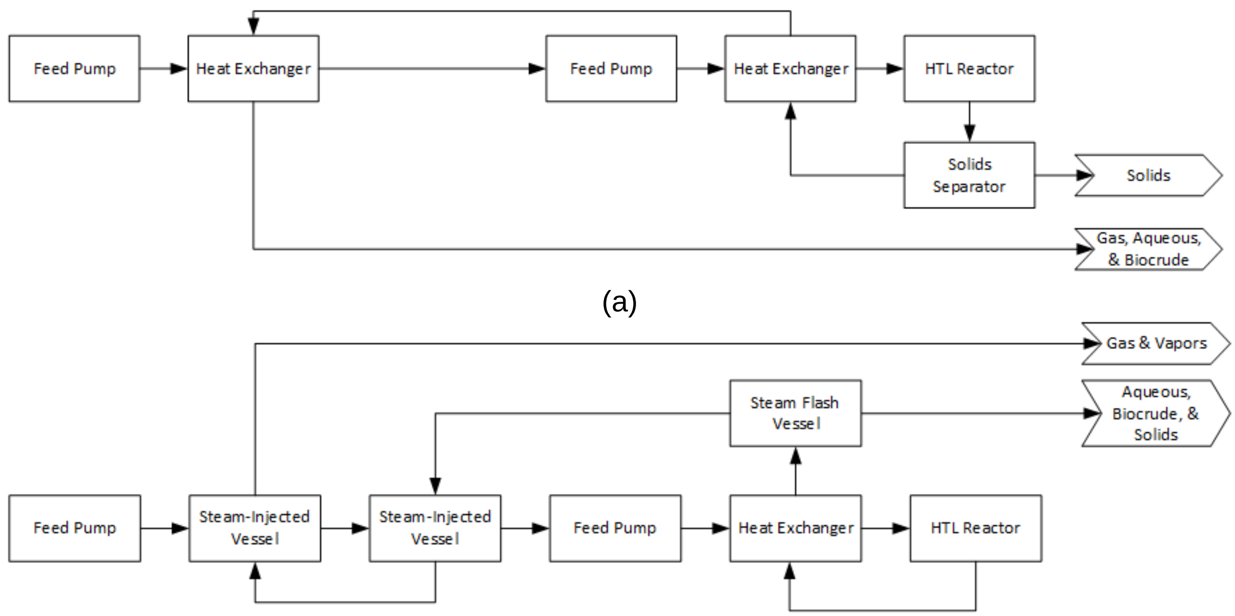

Figure 6. Updates made in 2023 to incorporate steam flashing into the HTL design, showing the previous (a) and updated (b) configurations (Xu et al., 2024)

Alternatively, if HTL cannot be scaled down effectively, centralizing HTL processing can enable economies of scale. In general, larger HTL systems (>100 dry tons per day) are needed to benefit from economies of scale. Figure 3 shows that for HTL of cost-advantaged algae a system scale >100 tons per day is needed to approach the minimum efficient scale. For farm-cultivated algae, Figure 3 and Figure 7 show the minimally efficient processing scale for HTL is at least 500 dry tons per day as there are diminishing benefits in cost for processing at larger scale.

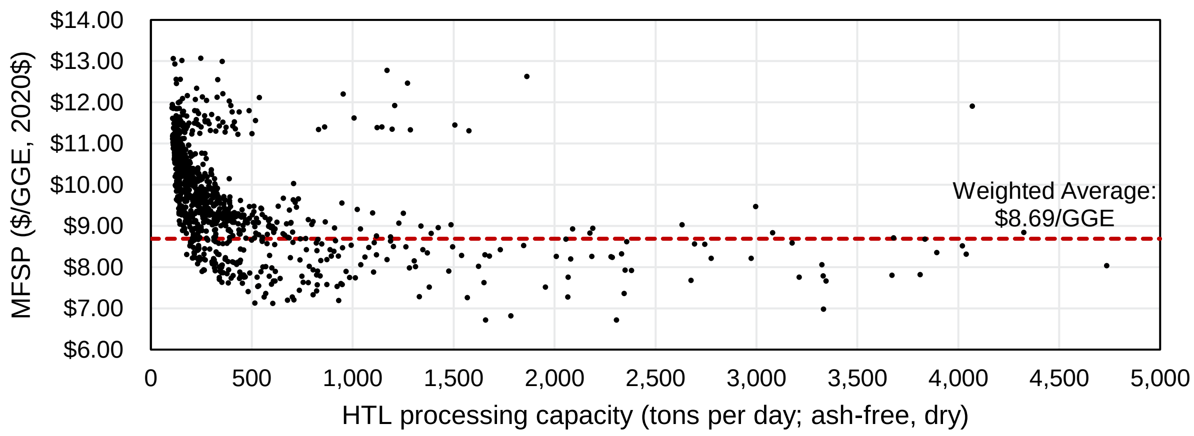

Figure 7. Estimated MFSP for fuel from algal HTL (Davis et al., 2024)

In 2021, a centralized system was introduced where the algae from several wastewater treatment facilities were consolidated before processing at a single facility for HTL (Zhu et al., 2022). The produced biocrude was then transported to a nearby refinery for upgrading. In other embodiments, it could be considered that several HTL facilities feed into a centralized refinery or biocrude-specific biorefinery (Snowden-Swan et al., 2020). When removing the refinery from within the battery limits for estimating the MFSP, the cost of upgrading is added on a per gallon basis of produced biocrude. The assumed cost reflects operational expenses only, such as the costs of transportation, hydrogen, and catalyst replacement and omits the capital contribution for upgrading assets.

Alternative Feedstocks and Blendstocks

The cost of farm-cultivated algal feedstock exceeds $600/ton (2020$), contributing >54% to the total cost of production of HTL biocrude (Klein et al., 2024). Since 2021, PNNL has investigated the use of cost-advantaged feedstocks which include algal residues from nutraceutical manufacturing, wastewater-grown algae, and algal bloom biomass. As shown in Figure 7, even with high production scales, the benefit of economy of scale is outweighed by the high cost of the feedstock. Figure 8 plots conversion cost against feedstock cost for single-stage HTL of algae. Even with more recent advances in algal productivity and cultivation system design, the feedstock alone contributes more than $5.19/GGE to the final MFSP.

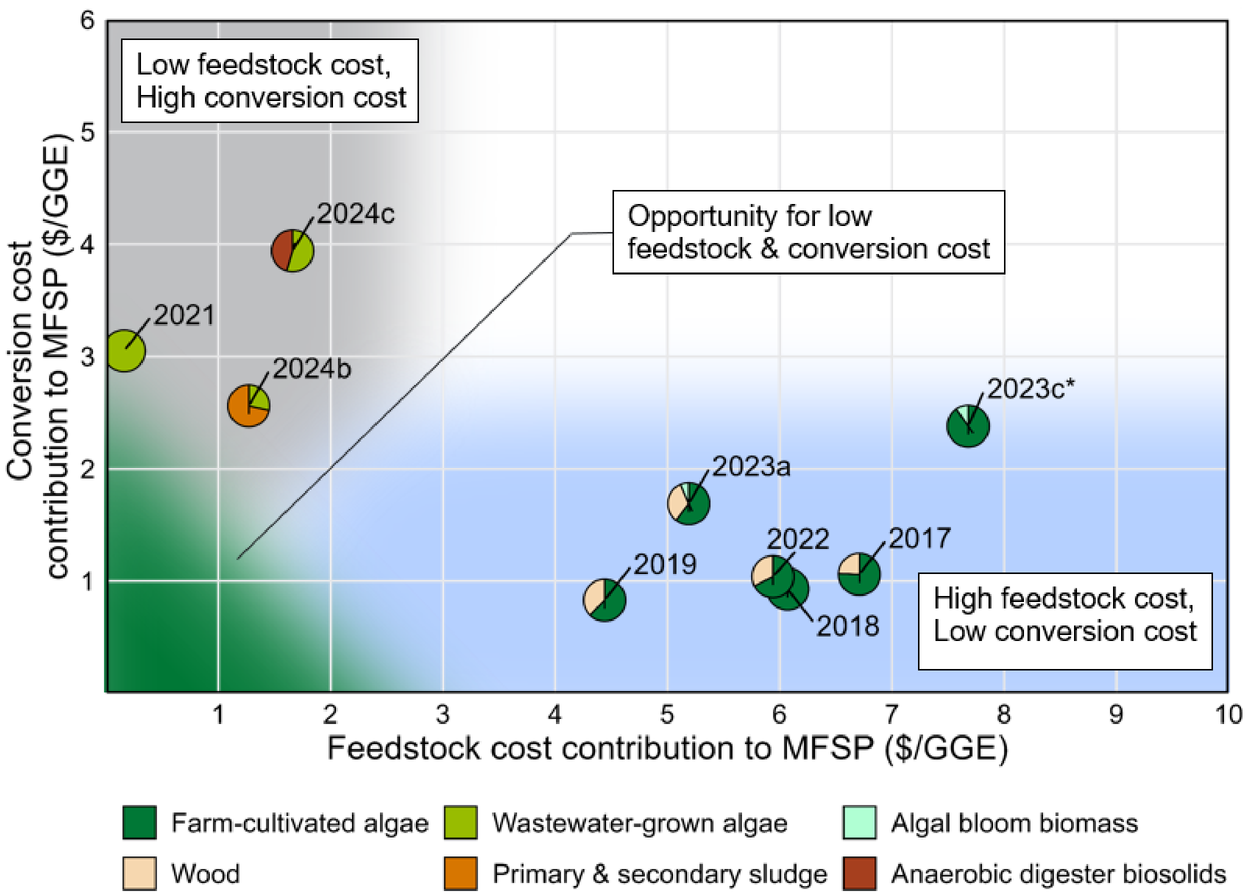

Figure 8. Conversion cost vs. feedstock cost for single-stage HTL scenarios

Cultivated algal biomass still offers the benefits of having a composition that can be tuned to favor HTL processing. High-lipid algae will boost biocrude yields. Low-protein algae will reduce the quantity of nitrogen in the final fuel product. As the technology and infrastructure of algae cultivation continues to develop, farm-cultivated algae should still be considered as a future feedstock for HTL.

Blendstock materials such as wood were introduced to offset seasonal variations in algae production and to maximize the capacity and scale of the conversion system. Using a blendstock removed the need and associated costs to store algae to balance the annual supply. Woody biomass also had a favorable cost at <$100/ton (dry, 2020$). The use of blendstock reduced costs by 48%.

When using cost-advantaged feedstocks, conversion cost contributions to MFSP are larger than feedstock cost contributions to MFSP, as illustrated in Figure 8. Although the feedstocks can be provided at little to no cost, they tend to contain more ash and store less energy per mass than purchased feedstocks. The less-preferred composition of cost-advantaged algae results in a reduced yield of biocrude and a higher cost of conversion for cost-advantaged feedstocks when compared to purchased feedstocks. For cost-advantaged feedstocks, the cost of conversion is the dominant cost contributor to MFSP. New innovations are needed to reduce the overall cost of conversion.

The 2024 and 2021 reports quantify the MFSP for fuel from wastewater-grown algae. Blending wastewater-grown algae with wastewater solids such as sewage sludge and solid digestate can increase the total amount of biomass available for HTL, therefore increasing the capacity of the HTL system and decreasing cost contributions from conversion due to economies of scale. Table 2 shows the potential availability of wastewater solids and wastewater-grown algae from the 31 largest water resource recovery facilities in the US (Seiple et al., 2017). The combined treatment capacity for the largest facilities is 7.5 billion gallons per day of wastewater. The methodology for estimating wastewater solids and algae potential is detailed in the 2024 design case report (Kumar et al., 2024). In Table 2, the scenarios 2024a and 2024b represent the blending of wastewater-grown algae with primary and secondary sludges. Scenario 2024c represents the blending of wastewater-grown algae with biosolids produced by the anaerobic digestion of the same primary and secondary sludges.

Table 2. Theoretical potential for algae and wastewater solids from the largest 31 water resource recovery facilities in the US (cumulative 7.5 billion gallons per day wastewater flow)

Biomass Availability Scenario	2024a	2024b	2024c
Algae:Wastewater Solids	17:3	2:5	6:5
Biomass potential (dry tons per day)	2,700	980	830
Biocrude potential (million GGE per year)	77	32	22

The report in 2023 evaluated algal bloom biomass as a feedstock for HTL. The net availability of algal bloom biomass is limited. An analysis was completed of 15 lakes, within a 50-mile radius near Orlando, FL to estimate the potential of algal bloom biomass in a region. With conservative assumptions, the low-end estimation predicted a daily algae production rate of 30 tons per day when averaged throughout the year. The processing capacity is relatively small compared to other HTL systems, resulting in an above average estimation for MFSP ($12.95/GGE) (Xu et al., 2024). Although the harvesting of algal blooms has a meaningful significance for environmental remediation, it will be challenging to scale this source as a reliable feedstock. Algal bloom biomass should be considered as an episodic resource to boost other biomass feedstocks.

Value of Co-products

For farm-grown algae, it is a priority to return nitrogen, phosphorus, and inorganic nutrients recovered from the HTL co-products (aqueous and solid products) to the algae farm. About 99% of phosphorous in the HTL solids and 90% of nitrogen from the aqueous phase can be recycled for cultivation. The associated credits for the sale of nutrients back to the algae farm outweigh the cost of additional materials and equipment to recover the nutrients.

In the SEQHTL scenario, fermentable sugars were investigated as a co-product, producing lactic acid as a fermentation product. However, there were no significant improvements to the net cost, and there were increases to the net GHG emissions.

Controlled farm cultivation can also be used to produce high-quality co-products, such as proteins fit for animal and/or human consumption. The possibility for algal proteins was examined in 2022, but more development is needed to demonstrate protein extraction and monetization (Zhu et al., 2023). Additionally, national scale protein and fuel production from algae become constrained by market size for protein supplements, limiting the financial and environmental benefits (Davis et al., 2024).

In cost-advantaged scenarios, recycling nutrients is unnecessary but instead there is an opportunity to monetize the nutrients and carbon for co-products. In 2021, struvite fertilizer ($NH_4MgPO_4 \cdot 6H_2O$) was assumed as a potential co-product. The sale of struvite added significant value, reducing the MFSP by $3.46/GGE (2020$), while the displacement of traditional fertilizers with struvite resulted in a net negative estimation for GHG emissions (Cai et al., 2022; Zhu et al., 2022).

Another opportunity with cost-advantaged algae is to utilize the high-ash HTL solids as a supplementary cementitious material (SCM). The ash in the HTL solids is rich with Ca, Fe, Al, and Si, which is similar to fly ash, a common SCM (Xu et al., 2024). Although the direct use of the HTL solids is relatively simple, the economic value can be limited (<$0.40/GGE (2020$)). However, the use of HTL solids as an SCM offers significant reductions in GHG emissions by offsetting carbon-intensive cement processing steps and by sequestering some carbon permanently into the cement itself (see Figure 4). Additional development is in progress to formulate an SCM-cement mixture that matches all the necessary criteria for cement products.

4.0 Open Challenges and Future Outlook

Reducing capital intensity

Although the cost of feedstock has been a primary contributor to the cost of HTL, the shift to cost-advantaged feedstocks emphasizes the need to reduce the capital expenditures of HTL processing. Although high-temperature (350 °C) and high-pressure (>2,400 psig) processing conditions are readily achievable in industrial processing, additional cost will be required to ensure the equipment can operate reliably over the lifetime of the system. Pilot systems currently use thick-wall stainless-steel tubing for operations. The wear on certain systems, such as pressure control valves or pumps, can be considerable for slurry systems. One opportunity to reduce capital expenditures could be to reduce the severity of process conditions. Reducing operating temperature to 300 °C could reduce the required pressure to maintain liquefaction conditions to 1,200 psig, half of the current pressure requirement. Yield and heteroatom content of the biocrude would be adversely affected, but the opportunity to reduce costs is worth consideration to fully explore the trade-offs (Reddy et al., 2016). Other innovative approaches to reduce capital intensity should be ideated and investigated. Making smaller capacity HTL systems economically viable can create more opportunities for deployment.

Other co-products or alternatives to fuel

If capital intensity cannot be reduced, then the development and monetization of co-products will be necessary to improve the economic viability of HTL. Proteins, fertilizers, and SCMs have been discussed. These co-products each offer unique opportunities, but still require development to validate their technical and economic feasibility. Preliminary estimations and initial experiments are promising, showing opportunities for increased economic value or reduced GHG emissions with the utilization and sale of co-products (Zhu et al., 2022; Davis et al., 2024; Xu et al., 2024).

Although the production of fuels has been the primary target of algae HTL, there is an alternative opportunity to decarbonize asphalt by the direct use of HTL biocrude as an asphalt binder. Preliminary testing shows promising results and additional development work is needed to prove the technical feasibility of algae-derived asphalt binders (Pahlavan et al., 2024).

Scaling cost-advantaged feedstocks via blending opportunities

The use of cost-advantaged feedstocks provides a pathway toward economically competitive fuel prices, but the quality, accessibility, and scale of these feedstocks is limiting. One opportunity to improve the utilization of cost-advantaged algae feedstocks is through blending with other types of readily available biomass. HTL of wet wastes such as animal manures or sewage sludge has been well studied, with published results and analyses confirming economic viability and a reduction in GHG emissions when compared to fossil diesel (Li et al., 2024). Blending will create opportunities to utilize multiple feedstocks and benefit from economies of scales to produce economically viable fuels.

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