ACS Sustainable Chemistry & Engineering

pubs.acs.org/journal/ascecg

Research Article

Implications of CO_2 Sourcing on the Life-Cycle Greenhouse Gas Emissions and Costs of Algae Biofuels

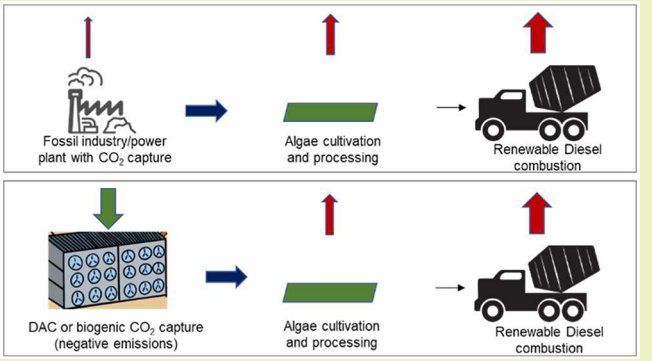
Udayan Singh, Sudhanya Banerjee, and Troy R. Hawkins*

Cite This: *ACS Sustainable Chem. Eng.* 2023, 11, 14435−14444

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: Production of algal biomass and its conversion to biofuels are important technological platforms within the larger umbrella of CO_2 capture and utilization. This analysis incorporates a life-cycle assessment (LCA) with respect to global warming potential and techno-economic assessment (TEA) of algae biofuels, focusing on the sourcing and delivery of CO_2. This analysis evolves past work in this area to include high-purity biogenic CO_2, industrial fossil fuel use, fossil power plants, and direct air capture, and uses a Sherwood plot approach to estimate the CO_2 capture energy penalty. We also show that allocation or displacement facilitates a more intuitive distinction between biogenic and fossil sources of carbon. Thus, the LCA better reflects the influence of coproduct handling strategies as compared to previous works. The TEA is also strongly influenced by the CO_2 concentration in the flue gas. Currently, when CO_2 is sourced from large-point sources, the price of biofuels ($4.5−6.5/GGE) may become comparable to fossil diesel. However, as DAC systems become more economical, they may deliver competitive CO_2 sources for biofuels in 2050 with a total cost of <$7/GGE. Based on the net emissions and costs, algae biofuels with CO_2 sourced from biogenic sources are consistent with a decarbonized economy as of now, with substantial potential for DAC with decreasing costs.



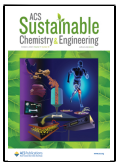
KEYWORDS: *CO_2 capture, CO_2 utilization, algae biofuels, LCA/TEA, allocation, displacement*

INTRODUCTION

Carbon capture and utilization (CCU) platforms have been evolving significantly over the past decade, and the recent report of the Intergovernmental Panel on Climate Change finds that they may contribute to mitigation of 100−200 Mt-CO_2 until 2030.[1] Several advantages of CCU have been noted in the literature over carbon capture and storage (CCS) in geologic formations. First, CCU results in one or more marketable products, thus, being potentially profitable even without government incentives. Second, CCU is generally perceived to be less risky by the public.[2] Third, the monitoring and verification requirements for CCU do not require an infrastructure lasting over several decades. That said, some concerns also accompany the CCU development. For instance, several CCU configurations may not provide appreciable emission reductions, especially for CO_2-derived fuels that involve combustion.[3] Moreover, if large-scale CCU infrastructure is developed for large fossil fuel point source emissions, then it may lock-in long-term CO_2 emissions. Thus, it is imperative to evaluate prospective CCU platforms from the point of view of their life-cycle greenhouse gas (GHG) emissions. Moreover, influential position pieces in the CCU domain point to the large variability in costs, ranging from −$100/t-$CO_2$ to $1000/t-$CO_2$.[4−6]

Because CCU platforms are diverse, it is essential that life-cycle assessment (LCA) and techno-economic assessment (TEA) are carried out with greater context about the CO_2 end use. In this study, we focus on the utilization of CO_2 to produce algae biofuel. This pathway has multiple advantages. Most notably, the pressure and purity requirements associated with it are substantially lower than other liquid fuel producing pathways, such as enhanced oil recovery and methanol production, respectively.[7] Microalgae is considered important because it has high carbon utilization efficiency (ratio of carbon fixed in biomass form to the amount of CO_2 supplied) compared to other biomass.[4] That said, past LCAs and TEAs have provided a wide spectrum of results regarding the feasibility of this pathway. Several meta-analyses have been published that try to harmonize production parameters to arrive at consensus results. These studies concluded that differences in net GHG emissions could be primarily attributed to methodological

Received: May 24, 2023
Revised: August 15, 2023
Published: September 18, 2023



https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

considerations and assumptions around CO_2 sourcing and delivery.[8,9]

Rationale and Scope of Study. This paper aims to evaluate the global warming potential (GWP) and economic costs of producing algae biofuels from a variety of CO_2 sources. This involves several previously unexplored methodological considerations, especially for LCAs of algae biofuels. The novelty of this work relies on its methodology around coproduct handling, use of pilot-scale data for CO_2 capture, and inclusion of a wider variety of CO_2 capture sources including direct air capture (DAC). Past work by Somers and Quinn[10] looked into these considerations. However, this study aims to evolve that work based on three important methodological considerations. First, Somers and Quinn[10] used an incremental or cutoff approach for estimating global warming potential. While useful, this approach is different from the approach suggested by the National Energy Technology Laboratory that relies upon system expansion. It is noteworthy that the system expansion approach is being used for awarding incentives under the 45Q tax credits by the U.S. Government, which necessitates evaluation of GWP using this approach. Second, the assumptions around CO_2 capture energy penalty in their study is based on minimum thermodynamic work, which leads to an underestimate compared to available pilot-scale data. For instance, the capture energy in their analysis for a coal-fired power plant is assumed as 0.5 MJ/kg-CO_2, which is about five times lower than published estimates for the state-of-the-art. Third, our analysis also includes a greater diversity of power sector and industrial sources, along with DAC, with the latter receiving much higher tax credits than point-source capture under the Inflation Reduction Act. Use of point sources alone and not DAC, as in the case of prior analyses, also limits the siting of algae ponds, as large areas in the U.S. do not have CO_2 provision at an industrially relevant scale.[11] This, in turn, also helps generalize our own past analysis through refined process parametrization of the CO_2 capture process that only considered high-purity sources of CO_2.[12]

One notable feature that is not within the scope of this Article is the influence of regional differences. Previous work from our group analyzed the effect of geography and found that this might affect GWP and costs.[12] However, these differences mostly arise in the algae growth stage and not in the CO_2 sourcing stage. Since the objective of this paper is to delineate the role of CO_2 sourcing and coproduct handling considerations around it, we have chosen to leave geographical differences outside the scope of this paper as they may countermand the insights obtained from detailed CO_2 sourcing analysis. That said, sensitivity shows the impact of varying geography on the costs via the metric of biomass productivity.

METHODS

System Boundary and Functional Unit. The system boundary considered in this study is illustrated in Figure 1. Based on the CO_2 sources and the demand for commercial scale algae cultivation, CO_2 provision from different sources is considered in the study. Following carbon capture from the different CO_2 sources, the CO_2 stream is compressed to the supercritical range and transported via pipelines for algae cultivation. Compression to a supercritical stage increases the density of the CO_2 by about 100 times, which makes it easier to transport and handle. Water is pumped and circulated to facilitate algae cultivation in the open raceways. Subsequent to microalgae growth and harvesting, harvested algae are then converted to biofuels in a colocated biorefinery via hydrothermal liquefaction (HTL) pathway and subsequent upgrading is carried out for conversion to renewable diesel. To facilitate hydrotreatment of the biofuels produced by HTL, natural

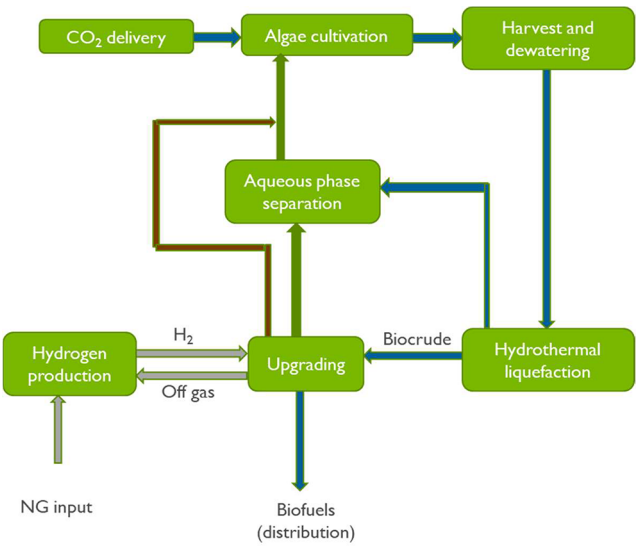


Figure 1. Illustrative system boundary evaluated in this study.

gas is used to provide necessary hydrogen by steam methane reforming. Biofuels produced in the process are transported, distributed to gas stations, and eventually used for vehicle combustion.

The functional unit for this study is 1 MJ of algae biofuel produced. This functional unit is chosen to facilitate comparison with previously published algae biofuels LCAs and TEAs. It also is chosen to align with the HTL pathway, which prioritizes liquid fuel production. That said, other units are also reported to facilitate an easy comparison. Notably, TEA results are presented per gallon of gasoline equivalent (GGE).

CO_2 Sources, Capture and Compression. The energy requirement for CO_2 capture primarily depends on the concentration of CO_2 in the flue gas. While some past work also correlates this parameter with the rate of CO_2 capture, this change is largely considered to be statistically insignificant within a range of capture rates across the fraction of CO_2 captured from the flue gas (60−90%). Past work[13] estimates CO_2 capture costs based on regression relationships between costs and CO_2 concentrations. The input data for such regressions were obtained from results obtained in the Integrated Environmental Control Model (IECM), developed at the Carnegie Mellon University.[14,15] We utilized a similar approach to estimate the CO_2 capture energy. IECM provides the results for three power plant technologies with different CO_2 concentrations: natural gas combined cycle plants (3−4% CO_2), pulverized coal plants (12−15% CO_2), and integrated gasification combined cycle plants (40−50% CO_2). Once these results are used to develop a regression fit, the capture energy for other large point sources was estimated.

To obtain the current range of energy penalty for CO_2 capture with state-of-the-art capture (ammonia based capture for postcombustion plants and selexol capture for IGCC plants), we simulated three typical new power plants for the aforementioned technologies. The logarithmic regression of the analysis revealed the following relationship for current capture technologies, which is analogous to a Sherwood plot:

$$\text{Current energy for capture}\left(\frac{\text{MJ}}{\text{kg CO}_2}\right) = 10^{(0.7438-0.3639\log\beta)},\ R^2 = 0.97 \quad (1)$$

Here, β represents the concentration of CO_2 in the flue gas and the assumed 90% of CO_2 being captured from the flue gas. The minimum concentration of 3.5% for NGCC requires 3.5 MJ/kg-CO_2 for capture. The corresponding capture energy from the regression for pulverized coal plants is estimated to be 2.4 MJ/kg-CO_2, which is comparable to the results obtained for other analyses simulating new plant conditions

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

using advanced supercritical cycles[16,17] and ensemble values obtained from previously published literature reviews.[18]

Several analyses have found that future energy for CO_2 capture (in terms of the parasitic load (MJ/kg-CO_2) for the power plant) is related to the developments in capture technologies. IECM currently allows simulation of chemical looping capture, which is widely considered as a benchmark for an improved technology.[19,20] IECM models chemical looping capture based on the design provided by their publications, which involves a heat recovery steam generator. These processes are found to be thermodynamically much more efficient than post-combustion incumbents. Thus, we performed another regression between the future capture energy and the CO_2 concentration at 90% capture and obtained the following relationship:

$$\text{Future energy for capture}\left(\frac{\text{MJ}}{\text{kg CO}_2}\right) = 10^{(0.2183-0.3524\log\beta)},\ R^2 = 0.96 \quad (2)$$

The estimates from the above regression were comparable to past work using Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) within 5%[21] (with points of divergence mentioned in the SI adjoining Figure S2).

There were a few exceptions to the capture energy that we considered. For instance, two types of point sources considered here (ethanol and RNG processing) result in a nearly pure CO_2 stream.[22,23] These two sources were therefore assigned a capture energy burden of ∼0 MJ/kg-CO_2. Moreover, technology improvements for direct air capture are occurring due to the development of modular units in both high- and low-heat configurations. The high-heat configuration developed by Carbon Engineering currently runs at 1200 °C. Due to the high-grade heat requirement, the heat supply may come only from natural gas currently. The high-temperature configuration uses 8.81 MJ/kg-CO_2.[24] As the developers incorporate electrification in a future configuration, this is anticipated to reduce to 5.51 MJ/kg-CO_2 natural gas, with an additional electricity requirement of 0.077 kWh/kg-CO_2. The low-temperature DAC configuration developed by Climeworks currently uses 6 MJ/kg-CO_2 from low-carbon source (waste heat or geothermal) and 0.42 kWh/kg-CO_2.[25] In the future, the electricity requirement is anticipated to remain the same while the heat requirement may reduce to 5.4 MJ/kg-CO_2.[26]

CO_2 Delivery and Distribution for Algae Cultivation. An additional CO_2 compression energy requirement of 0.36 MJ/kg-CO_2 (electrical) is assumed for all sources to ensure compatibility with pipeline transport.[14] To ensure efficient CO_2 delivery and to counter pressure drop for long distance pipeline transportation, recompression is necessary. Hence, booster compression stations are required at every 150 km.[10,27] The energy demand for recompression obtained from previous studies is 0.01 MJ/kg-CO_2.[10,12,27] Following the CO_2 transportation, the next step is the efficient distribution of CO_2 for algae utilization. Energy requirement for CO_2 distribution is 0.14 MJ/kg- CO_2.[10] Algae CO_2 utilization efficiency is assumed to be 70% following a previous study.[28]

Algae Cultivation. Microalgae cultivation and harvest were modeled based on previous literature.[29,30] It was assumed that algae was cultivated in open raceway ponds in a 5000 acre facility with average pond depth of 20 cm.[29] To optimize algae growth and productivity, pond circulation using paddle wheels is a critical step that consumes a significant amount of energy which in turn impacts the LCA and TEA results. It was assumed that pond circulation occurs for 12 h per day and consumed 1.2 kW/ha.[29] Net energy requirements for algae cultivation, including pond operation, water circulation, and makeup water delivery were estimated based on our previous study to be 0.26 MJ/MJ biofuel.[12] The assumed algae yield was 25 g/m^2-day. Biomass harvesting was assumed to occur when algae concentration reached a density of 500 mg/L which was followed by algae dewatering before algae was routed to biorefineries for HTL pathway and subsequent upgrading.[29] Dewatering and harvesting of microalgae was assumed to be facilitated in three steps primarily by gravity settlers, hollow fiber membranes and finally by centrifugation which resulted in a biomass concentration of 200 g/L.[29] This concentrated algae solution was deemed suitable for HTL conversion and upgrading to biofuels.[29,30] Overall efficiency of algae harvesting and drying was estimated to be 87% following previous work.[12] It was also assumed there was 1% loss on an ash free dry weight (AFDW) basis when algae was subjected to temporary storage due to operational inefficiencies. After algae cultivation, harvesting, and storage, algal biomass was routed to a colocated biorefinery for conversion to renewable diesel via HTL pathway and upgrading.

Algae Conversion to Biofuel, Biofuel Distribution, and Fuel Combustion. Algal biomass is converted to hydrocarbon biofuels by HTL and upgrading in the biorefinery.[12] HTL produces biocrude, solids, and aqueous phase and off-gases of which the biocrude are hydro processed and upgraded to gasoline and diesel range fuels. The off-gas generated in the process is used to produce heat, power, and hydrogen. Supplemental hydrogen required for the process is produced from natural gas by steam reforming. Nutrients from HTL solids are recovered and recycled back to algae cultivation ponds. The overall fuel yield is evaluated to be 23.3 MJ fuel/kg-algae on AFDW basis. Transportation and distribution of biofuels to gas stations are evaluated using default parameters in the GREET model following previous work.[12]

LCA Considerations. The mass and energy balances of the pathway are used to perform environmental life-cycle assessment for the advanced freshwater-algae-based HTL biorefinery. The GREET model, a publicly available environmental assessment tool developed at Argonne National Laboratory is used to conduct the LCA.[31] For life-cycle environmental impacts, we differentiate between "avoided emissions" and "negative emissions". Avoided emissions refer to CO_2 captured from fossil sources. Negative emissions occur when CO_2 is captured from DAC and other facilities producing biogenic CO_2.[32,33]

We report results using four different coproduct handling approaches:

- Incremental or cutoff approach:[34] Here, the CO_2 capture benefits are allocated entirely to the algae biofuels. Thus, emissions resulting from combustion of the fuel is not accounted within the life cycle of the biofuel as these emissions would have otherwise been emitted into the atmosphere.
- Allocation of all emissions: In this approach, CO_2 capture benefits are allocated to product facilities from which the CO_2 is being captured as well as the algae biofuel. Here, fuel combustion emissions are accounted within the biofuel life cycle. Energy-based allocation is followed for facilities producing energy-based products and economic allocation is followed for other facilities. The allocation parameters are shown in Table S2.
- Allocation of captured CO_2 benefits: This approach is a combination of the two above approaches. Here, the CO_2 capture benefits are partly allocated to the algae biofuels. That said, the combustion emissions are allocated to both the algae biofuels and the point source from which CO_2 is captured based on the allocation factors shown in Table S2.
- System expansion: Some past works have suggested that displacement and systems expansion may be more suited to CCU applications.[35] Based on the recommendations of the NETL CO2U guidance document, we also report the results based on the system expansion approach. System expansion parameters are shown in Table S3 and the adjoining paragraph.

TEA Assumptions. The formula for the costs for CO_2 capture from large point sources is adapted from Pilorge et al.[13] This formula is used because its use of the logarithmic form improves the goodness of fit, as compared to the linear version used in Psarras et al.[36]

$$\text{Current cost of capture}\left(\frac{\$}{\text{t-CO}_2}\right) = 10^{(2.18-0.426\log\alpha-0.391\log\beta-0.028\log\gamma)} \quad (3)$$

Here, α stands for the rate of CO_2 capture and γ represents the volumetric CO_2 capture per year. Again, the cost of CO_2 capture is the

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

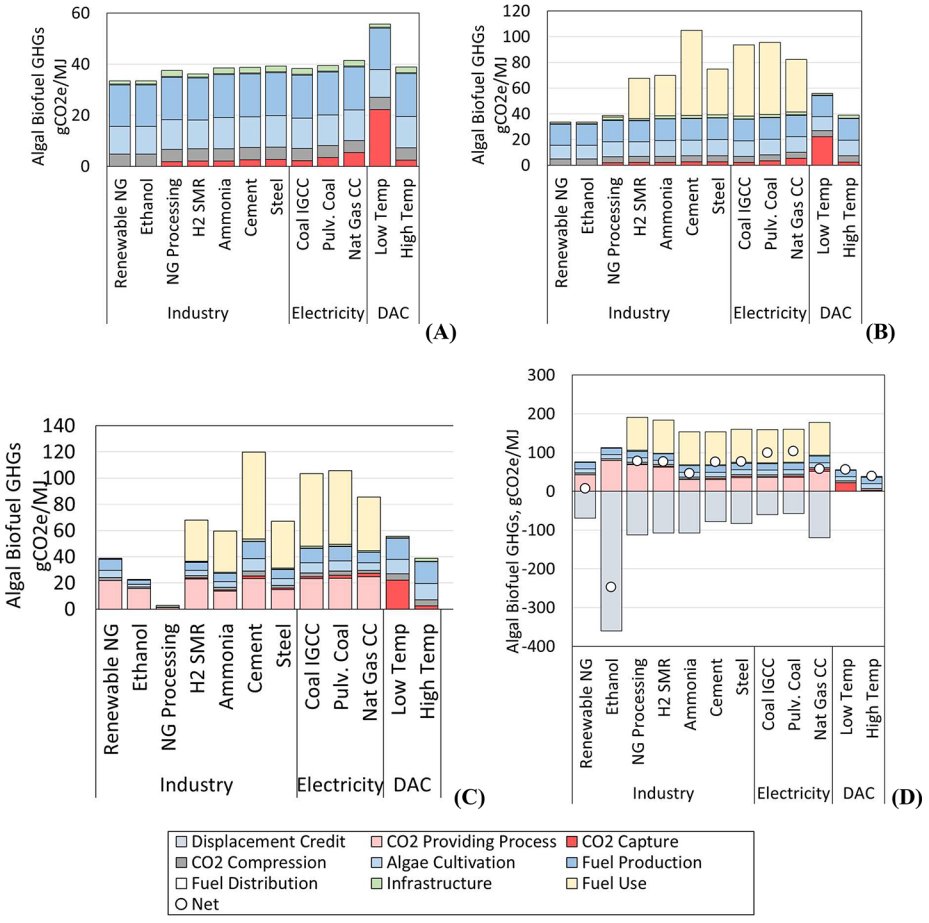


Figure 2. Net emissions allocated to algae biofuels in the current state-of-the-art for different coproduct handling strategies. (A) CO_2 capture benefits allocated to the algae biofuel. (B) CO_2 capture benefits assigned jointly to the point source and algae biofuels. (C) CO_2 capture benefits and emissions from the CO_2 providing process assigned jointly to the point source and algae biofuels. (D) System expansion.

highest for NGCC power plants ($77/t-$CO_2$) and gradually decreases as the concentration of the CO_2 in the flue gas increases. RNG and bioethanol facilities yield a near pure stream of CO_2 and the cost of capture associated with them is assumed to be close to zero.

The cost of CO_2 capture in the future is speculative, as it is dependent on the rate of technological learning. To simplify this assumption, we incorporate a capture cost reduction of 33% by 2050, as assumed by Muratori et al.[17] in their "moderate" scenario.

The cost of high-temperature DAC is currently quoted at $232/t-$CO_2$ at scale.[37] With process improvements and reduced risks (lower discount factor), these costs are anticipated to reduce to $97/t-$CO_2$.[37] Similarly, the current costs of low-temperature DAC are $223/t-$CO_2$ and are anticipated to drop to $100/t-$CO_2$.[38] Other works quote a much lower cost ($40−60/t-$CO_2$). However, these costs are based on a technological learning curve assuming cumulative DAC capacity addition of 15 Gt-CO_2/year by 2050.[39,40] More recent ensemble integrated assessment modeling shows that this estimate would be less than 5 Gt-CO_2 in scenarios limiting warming to 1.5 °C without overshoot.[1,41]

Table S1 shows the summarized estimates of the current and projected energy and cost estimates for CO_2 capture from various sources.

For other parameters, a process model developed in Aspen Plus was used to determine capital and operating costs of commercial scale algae cultivation and conversion to biofuels via the HTL pathway and subsequent upgrading. These costs were then used by a discounted case flow rate of return analysis to estimate a minimum biomass selling price (MBSP) that produces a net present value of zero for the nth plant at a 10% internal rate of return (IRR). Capital equipment subjected to an exponential scaling parameter for sizing and installation factors yielded total capital investment for the facility. Percentage allocations for direct and indirect costs for cultivation, dewatering, and outside boundary limits were derived from the algae farm design report.[42] Following determination of algae MBSP, algae conversion to biofuels costs was modeled based on previous studies.[43] Table S4 shows detailed costing parameters for the HTL system.

RESULTS AND DISCUSSION

GHG Emissions. The primary LCA metric of interest in evaluating algal biofuels is the net GHG emissions per MJ of biofuels produced. The entire supply chain of carbon capture, compression, delivery, algae cultivation, harvesting, and algae conversion to biofuels is considered to determine the net GHG emissions of the pathways. Net GHG emissions corresponding to different CO_2 sources are shown in Figure 2.

In the incremental approach, all of the CO_2 capture benefits are allocated to the algal biofuels (Figure 2A). This results in less variance in the carbon intensity of biofuels since the nature of the captured CO_2 (biogenic versus fossil) does not influence the emissions. In other words, this is a "cut off" approach where the system boundary is not affected by anything before the CO_2 capture stage. The results for this stage are similar to past studies carried out by our group and others.[12,44] However, this strategy leads to low emissions for carbon-intensive infrastructure powered by fossil fuels. For instance, the GHG intensity for when CO_2 is captured from a NGCC power plant (41 gCO_2e/

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

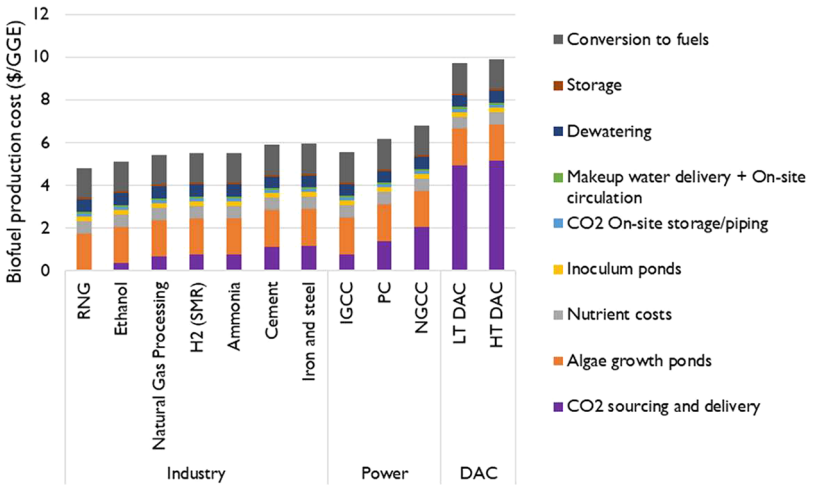


Figure 3. Estimated costs of algae biofuel based on different CO_2 sources in the present-day context. RNG: renewable natural gas, SMR: steam methane reforming, IGCC: integrated gasification combined cycle, PC: pulverized coal, NGCC: natural gas combined cycle, DAC: direct air capture, LT/HT: low/high temperature.

MJ) is only marginally higher than the ethanol fermentation counterpart (33 gCO_2e/MJ).

Accordingly, we report additional allocation strategies, where there may be a clearer differentiation between biogenic and fossil sources of carbon. In this approach, the CO_2 capture benefits are allocated based on energy content when the CO_2 is captured from an energy-related facility and based on economic value when the CO_2 is captured from nonenergy producing facilities (ammonia, cement, and steel), as shown in Figure 2B. Here, the sources involving CO_2 capture from biogenic sources or the ambient air do not result in any additional emissions compared to the cutoff approach as they do not represent any additional net flux of the CO_2 to the atmosphere (shown in Figure S6). Thus, their emissions are identical to the "cut off approach".[45] However, the emissions when biofuels are sourced from fossil sources are higher. Depending on the allocation factor, these differences may be substantial or trivial. For instance, NG processing is associated with a lower emission factor compared to those of other point sources (in terms of gCO_2e/MJ) because the allocation factor estimated for algae biofuels is low (1.5%). As such, the emissions are 38 gCO_2e/MJ in this case. When the CO_2 is sourced from SMR, ammonia, steel, and NGCC plants, the algae biofuel has a carbon intensity of 65, 75, 73, and 77 gCO_2e/MJ. As such, these emissions do represent a marginal reduction below the current diesel baseline. The emissions from biofuels produced with CO_2 sourced from coal-fired (both IGCC and PC) are above 90 gCO_2e/MJ, which may not represent any sizable benefit below the fossil diesel baseline. The highest emissions are seen when the CO_2 is sourced from a cement plant (103 gCO_2e/MJ), where the cumulative emissions actually exceed the fossil diesel baseline. This is because the relatively lower economic value of cement results in a higher allocation factor for the algae biofuels. This allocation strategy shows a significant difference between the biogenic and fossil sources of carbon. This is because this approach not only considers the energy penalty for CO_2 sourcing and delivery but also the positive or negative emissions associated with the original carbon source.

Another approach involves allocation of all the supply chain emissions (Figure 2C), including those from the CO_2 providing process and the emissions not sequestered by the algal biomass (which is 30% of the captured CO_2 as the default assumption of the CO_2 utilization efficiency of the algae is 70%). Here, the net emissions for the configurations vary widely. Because the NG processing configuration is associated with a low allocation factor, it results in the lowest emissions (2 gCO_2e/MJ). When CO_2 is sourced from RNG processing and bioethanol, the net emissions still represent a sizable reduction with a carbon intensity of 39 and 22 gCO_2e/MJ. Within the case of biogenic capture, bioethanol involves the lowest net emissions (22 gCO_2e/MJ). When CO_2 is captured from RNG processing facilities, the CO_2 capture penalty is low. This is because of the near pure CO_2 stream being emitted from the pressure swing adsorption system at a pressure of 17 bar, which is suitable for algae cultivation. Thus, the additional pressurization or purification requirements are less for this source. However, the upstream emissions associated with this case are high, some of which get allocated to the algae biofuels. CO_2 sourced from other industrial sources (SMR, ammonia, and cement) and NGCC plants also results in lower emissions than the fossil diesel baseline in this case. However, when CO_2 is sourced from coal-fired power plants or cement plants, the resulting carbon intensity is higher than the fossil diesel baseline.

Finally, we also discuss the results for the system expansion approach (Figure 2D). These results show the highest variance, because the displaced emissions vary highly by context. For instance, bioethanol is assumed to replace gasoline. However, because the process CO_2 emissions associated with bioethanol production are lower, it involves 4 MJ bioethanol production per MJ of algae biofuel. Thus, the corresponding benefits are close to 360 gCO_2e/MJ, resulting in net emissions of −247 gCO_2e/MJ. Emissions attributed to the RNG processing case are also close to carbon-neutral, i.e., 6 gCO_2e/MJ. High-purity CO_2 industrial sources result in net emissions of 48 gCO_2e/MJ to the algae biofuel, which is comparable but slightly higher than the "cut off approach" because the displacement approach also accounts for the emissions from the CO_2 providing process. Interestingly, the emissions for the algae biofuel are correspondingly lower (53 gCO_2e/MJ) when CO_2 is captured from the NGCC power plant because of higher efficiency and similar emission factors of such plants compared to the grid average. On the other hand, CO_2 captured from a coal-fired power plant results in net emissions of 97−99 gCO_2e/MJ, which is comparable or slightly higher than the fossil diesel baseline. While system expansion avoids some

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

allocation ambiguities, it is also evident that it has some shortcomings because the net benefits are influenced by the actual market share of incumbent products (discussed in the sensitivity analysis section).

The emissions associated with DAC-sourced CO_2 remain identical irrespective of the coproduct handling strategy as the algae biofuels are the only saleable product. In all other approaches other than the cut-approach, this results in emissions (38−55 gCO_2e/MJ) that are lower than those of CO_2 sourced from most fossil sources. Particularly, the LT-DAC case is associated with higher emissions (55 gCO_2e/MJ) than the HT-DAC case (38 gCO_2/MJ) as the former uses electricity that is currently sourced from the US average grid. This may be anticipated to come down further as process electrification and grid decarbonization occur (discussed later).

We emphasize here that the net emissions are strongly influenced by the coproduct handling strategy. As seen in Figure 2, the coproduct handling strategy strongly influences the net emissions, and some ambiguities are likely to remain. The "cut off" approach does not appreciably distinguish between biogenic and fossil sources of CO_2. The system expansion approach might exaggerate the CO_2 displacement due to the high carbon intensity of several products in the *status quo*.[46] Further, the carbon intensity of several products and electricity is projected to decline substantially in scenarios converging to 1.5 °C (see sensitivity analysis). The allocation strategy may also inflate the benefits associated with cases such as NG processing which may have lower emission factors as they depict only a part of the supply chain.[47]

Economic Costs. A detailed economic analysis was performed for the entire value chain of carbon capture and delivery, algae cultivation, harvest, and conversion to biofuels via the HTL pathway. Figure 3 shows that biofuel costs in the present day vary between \$4.8−9.9/GGE. The algae production costs range from \$440 to \$1100 per dry tonne of algae. Because other conversion parameters are assumed to be constant, this variation in the costs can primarily be attributed to the differences in upstream costs of carbon capture and delivery from various sources. As shown in Table S1, RNG and bioethanol have near-zero costs of CO_2 capture. They are therefore associated with the lowest cost of biofuel production. The price of diesel is \$4.8/GGE during the writing of this paper,[48] with the 2021 year average being \$3.3/GGE. The cost of producing fuel from the CO_2 portion of RNG biogas is \$0.3/GGE less than using the high-purity CO_2 from ethanol production because it requires less compression than ethanol CO_2. The two biogenic sources considered in this study are below this viability threshold in the present day. Most other industrial sources and IGCC power plants may also form affordable linkage of CO_2 as the costs of biofuel production remains as <\$5.5/GGE when CO_2 is sourced from them. Coal and gas combustion power plants have higher prices of biofuel production (\$6.2 and \$6.8/GGE) because of the comparatively lower CO_2 concentrations in the flue gas and high compression energy requirements, as opposed to gasification plants. Finally, biofuel costs with CO_2 sourced from DAC facilities are close to ∼\$10/GGE. Despite the advantages of DAC facilities in achieving carbon neutrality goals, these costs are currently too high to compete with conventional diesel without significant incentives for greenhouse gas reductions. We note here that the comparison with fossil diesel may not be a completely *apples-to-apples* comparison since the diesel price may be inclusive of other taxes, retail/wholesale margins, and delivery costs at scale. We have not compared the as-delivered price because biofuels may be incentivized as a result of CCU tax credits, the value of which are under consideration by the United States Senate at the time of the study period.[49]

Carrying out the TEA and LCA with identical system boundaries and assumptions allows us to estimate the cost of GHG avoidance, as well. This is defined as

$$\text{Cost of GHG avoidance} = \frac{\left(\text{Cost of new product}, \frac{\$}{MJ}\right) - \left(\text{Cost of reference product}, \frac{\$}{MJ}\right)}{\left(\text{GHG intensity of reference product}, \frac{gCO_2e}{MJ}\right) - \left(\text{GHG intensity of new product}, \frac{gCO_2e}{MJ}\right)} \quad (4)$$

We assume that the reference product here is fossil diesel with a market price of \$4.8/GGE and carbon intensity of 91 gCO_2e/MJ. Given the wide range of GHG intensities for algae biofuels, we discuss the results for the system expansion approach suggested by NETL for CO_2 utilization projects. In the current context, biogenic CO_2 capture is quite cost-effective, with the cost of GHG avoidance being \$2/t-CO_2e and \$10/t-CO_2e when the CO_2 is sourced from RNG processing and ethanol. Among fossil sources, the costs are cheapest (\$185/t-CO_2) when the CO_2 is sourced from ammonia facilities. For all other sources, the cost of avoidance exceeds \$500/t-CO_2. When CO_2 is sourced from DAC, the costs of avoidance are \$1100−1500/t-CO_2. However, there is substantial potential for cost reductions here, and a future context involving reduction in capital costs of DAC and carbon intensity of electricity (Figure S4) brings down the cost of avoidance below \$500/t-CO_2e, while the costs for fossil sources remain as >\$500/t-CO_2. This again demonstrates the need to decouple algae biofuel production from fossil carbon and energy sources.

Sensitivity Analysis. *GHG Emissions and Costs in a Future Context.* Kleijne et al.[3] indicates that products made using CCU are compatible with the Paris agreement only when their net emissions are at least half of the reference product they are replacing for a 2030 timeline. When the CO_2 is sourced from coal-fired power plants, the biofuels have emissions higher than the threshold of 49 gCO_2e/MJ (50% of fossil diesel). As such, they may not produce biofuel compatible with the Paris agreement targets, barring efficiency improvements in other components of the production chain. The future analysis assumes a time frame of 2050. We discuss these results for the cutoff approach and the system expansion approach (Figure S3).

The cutoff approach accounts for reduction in the capture energy penalty and grid decarbonization (Figure S3A). Here, the emissions for point sources (29−33 gCO_2e/MJ) and DAC (29−34 gCO_2e/MJ) fortuitously become similar in the future time frame. The reductions, particularly for DAC, occur primarily due to reductions in compression of CO_2 as the electricity supply becomes fully decarbonized.

The results are, however, more nuanced for the system expansion approach (Figure S3B). In addition to the aforementioned factors, the reduction in benefits for the displaced products. Ensemble modeling analyses suggest that both fossil natural gas and gasoline will remain a part of the future energy mix, even though their share may decline. As such, the emissions offset from the algae biofuel when CO_2 is captured from RNG processing, ethanol, and NG processing do not change as they will displace key fossil products from the market. However, for most other point sources, the benefits decline considerably. Both the electricity grid and hydrogen are assumed to reach net-zero carbon intensity in line with future projections.[50,51] Similarly, the carbon intensity of other marketable products is also assumed to decline by 75−80% in

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

deep decarbonization scenarios (Table S4). As such, the emissions for algae biofuels sources from all fossil point sources barring NG processing increase to 126−177 gCO_2e/MJ. Thus, in the long term, algae biofuels could be compatible with decarbonization goals only if they are produced with CO_2 sourced from biogenic sources or DAC.

It is also anticipated that the economic costs of algae biofuel production will decline in the future (Figure S4). Based on the projected CO_2 capture cost decreases from large-point sources (33%) and for DAC (40%) decrease (Table S1), several configurations appear to be more economically viable. The cheapest sources of CO_2 do not show any major difference because of the near-zero cost of capture presently. However, industrial sources such as NG processing and SMR could deliver CO_2 with a corresponding biofuel cost of $5.2/GGE. This is very close to the current diesel prices. Even when CO_2 is sourced from combustion power plants, the biofuel cost is <$6/t-$CO_2$ as CO_2 capture costs fall below $50/t-$CO_2$. The most notable change here is with DAC facilities, which can deliver CO_2 to produce biofuels at $6.8−7/GGE. While these costs may not be competitive with current diesel prices, they could be competitive, especially considering potential incentives associated with GHG reduction in the future. Diesel prices themselves are projected to rise with a compounded annual growth rate of 3% between 2021 and 2050 as per the EIA projections in the "reference" scenario.[52] In such a scenario, conventional diesel prices in 2050 would cost $7.6/GGE. This would mean that algae biofuels could deliver an economic alternative, irrespective of the CO_2 source.

Influence of Biomass Productivity. As noted in the Introduction, spatial and temporal differences may have a significant impact on the metrics estimated in this paper. While Ou et al.[12] have focused specifically on these differences, we carry out a sensitivity analysis around biomass productivity as a variable that is most influenced by spatial and temporal factors. The impact of productivity changes on the GWP is within 10% of the nominal value estimated in Figure S5. As the biomass productivity is reduced from 25 to 12 g/m^2-day, the total circulation energy required increases from 1.92×10^{-4} kWh/g-algae (AFDW) to 4×10^{-4} kWh/g-algae (AFDW). This leads to an increase in the GWP by 3.4 gCO_2e/MJ. On the other hand, an increase in productivity up to 50 g/m^2/day decreases the circulation energy further to 9.6×10^{-5} kWh/g-algae, with GWP decreasing by 1.6 gCO_2e/MJ. While nontrivial, these differences are relatively small compared to the nominal value. This is because the main contributor to GHG emissions in the biofuel production stage is the HTL process, which involves production of hydrogen from SMR. Thus, the influence of algae pond in the overall life cycle is limited. Productivity changes markedly affect the costs of biofuel production (Figure S5). At 12 g/m^2-day productivity, the costs are $7.6 and $9.9/GGE for ethanol- and DAC-sourced CO_2. As productivity increases to 50 g/m^2-day, these costs reduce to $4.0 and $5.9/GGE. As Davis et al. have noted, these changes occur because a higher productivity entails lower capital costs for the land as well as equipment.[42] These reductions are exponential in the first part of the graph as productivity increases up to 25 g/m^2-day, and subsequently becomes linear.

Influence of CO_2 Utilization Efficiency of Algae. Another key parameter in the LCA and TEA of algae biofuels is the CO_2 utilization efficiency (CUE) of the algae. A higher CO_2 utilization efficiency entails a lower capture energy and costs for CO_2 capture. Furthermore, the system expansion approach also accounts for the unused CO_2 within the GHG inventory of algae biofuels. Currently, the literature suggests that the CUE is close to 30% for existing systems. However, many *n*th-of-a-kind plant studies also assume this parameter at 90% based on projected improvements.

When the CUE is assumed at 30% instead of the default value of 70% assumed in this study, the volume of CO_2 to be captured and compressed increases 2.3 times. As the CO_2 sourcing and delivery cost component is low for high-purity sources, the cost increase is negligible for RNG/ethanol and below $1/GGE for NG processing, SMR, ammonia, and IGCC plants. However, this increase is $2.7/GGE for NGCC plants and more than $6/GGE for DAC plants. Contrastingly, if the CO_2 capture benefit increases to 90%, then the benefits for point sources is <$0.5/GGE, while that for DAC is above $1/GGE.

The LCA results are influenced by the CO_2 utilization efficiency depending on the coproduct handling approach used. In the incremental approach, a CUE of 30% entails an increase in emissions resulting from the CO_2 capture and compression stages by 2.3 times, which results in net emissions of 40 gCO_2e/MJ when CO_2 is derived from biogenic sources and 55 gCO_2 when the NGCC plant is treated as the source of the CO_2. With an increase in the CUE to 90%, each of the sources sees a decline in 1 gCO_2e/MJ in the net emissions. However, a much more marked change is observed in the system expansion approach, where the unutilized emissions for fossil facilities are attributed to the algae biofuels. This corresponds to an increase in net emissions by 168 gCO_2e/MJ when the CUE is 30%, compared to the baseline 70% (i.e., the results shown in Figure 2) for all fossil sources. Contrastingly, a reduction of 27 gCO_2e is seen compared to the baseline when the CUE increases to 90%. The difference between the incremental approach and the systems expansion approach exists because the former does not differentiate between avoided and negative emissions. This shows that the coproduct handling approach becomes increasingly important in estimating life-cycle results for emerging technologies. As Cooney et al. have pointed out, the incremental approach is simpler but is more poised to handle results in a context where non-CO_2 capture facilities continue to exist. On the other hand, the system expansion approach avoids double-counting but has greater data requirements.[45]

DISCUSSION

The analysis shows that several high-purity sources of CO_2 already show readiness, as most existing CO_2 capture facilities globally belong to this sector. Ou et al.[12] have discussed the integration potential of such sources with algae production at limited costs of pressurization and purification. It is notable that despite the recent reductions in DAC costs in the past decade, it is still significantly more costly than point-source capture. That said, DAC costs are anticipated to decline more sharply in the next three decades, especially with recent investments by the US government and private investors.[53]

While DAC costs are higher, it does offer carbon dioxide removal from the atmosphere or so-called negative emissions that are deemed necessary in energy transitions compatible with the 1.5 °C constraints.[54] While the metrics reported in Figure 2 do not show those effects, we have provided a synthesis of the carbon lock-in effects if different sources are relied upon for algae cultivation (Table S1). It is important to note that biogenic sources offer low, near-zero capture costs and energy while also delivering a net carbon intensity of close to zero. While these processes are important, the overall potential associated with

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444

ethanol sources is limited to <100 Mt-CO_2/year and is regionally concentrated in some states.[12] This translates to 40 million dry tonnes of algal biomass and 17 TJ biofuel. As such, DAC is an important technology because it can provide nonfossil CO_2 availability at scale and with greater regional flexibility, allowing operators to benefit from advantageous high-algae-yield, low-water-stress locations such as northern Florida and other Gulf coast states.[55,56] Currently, DAC systems are assumed to be powered by natural gas. About 70 DAC such facilities of up to 1 Mt-CO_2/year are planned by 2035, for example, by 1Point5. Other power and industrial sources, however, are characterized by a large fossil fuel use. If algae facilities are constructed by prioritizing these sources, then they would lead to potential prolonging of fossil infrastructure and inhibit their phasing down. Note that some fossil fuel infrastructure could be repurposed to integrate with biogenic carbon sources. For instance, IGCC power plants could be used to cofire biomass.[57]

The methodological uncertainty from the four coproduct handling approaches and systematic evaluation of capture energy requirements can help in future policy framing of algae biofuels and other CCU platforms. In terms of the costing results, harmonizing the LCA and TEA with a uniform system boundary helps in estimating the costs of GHG avoidance, which are used in developing CO_2 supply curves by multiple stakeholders.[58,59]

Future analysis may compare configurations studied in this paper with other approaches to direct carbon uptake during algae fixation. Such approaches can use flue gas directly, which would eliminate the CO_2 capture energy requirements and costs. Another potential route is via high pH cultivation to enhance CO_2 exchange between the algae pond and the atmosphere as an alternative to all of the modeled pathways and to decouple dependence of algae on CO_2. As the efficiencies of green hydrogen are improving, its use instead of gray hydrogen from SMR could reduce the GHG intensity of biofuels by another 7−9 gCO_2e/MJ.[60] However, the costs of electrolysis are currently high,[61,62] and the radiative forcing induced by hydrogen is itself uncertain,[63] which future analyses could resolve. The HTL configuration studied in this analysis can be tuned to prioritize other liquid fuels or products. These uncertainties can also be evaluated in future analyses.[64−66]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.3c02082.

Detailed system boundary diagram for CO_2 utilization by algae; regression plots for CO_2 capture energy versus concentration, sensitivity analysis results for future context and changed productivity; key process, allocation and system expansion parameters for the analysis; paragraph comparing this analysis and prior GREET work (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Troy R. Hawkins − *Energy Systems Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0001-6897-175X; Email: thawkins@anl.gov

Authors

Udayan Singh − *Energy Systems Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0003-3359-6296

Sudhanya Banerjee − *Energy Systems Division, Argonne National Laboratory, Lemont, Illinois 60439, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acssuschemeng.3c02082

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The research effort at Argonne National Laboratory was supported by the Bioenergy Technologies Office of the Office of Energy Efficiency and Renewable Energy of the U.S. Department of Energy (DOE) under contract DE-AC02-06CH11357. The authors would like to thank Dan Fishman for his support and insights. The views and opinions of the authors expressed herein do not necessarily state or reflect those of the U.S. Government or any agency thereof. Neither the U.S. Government nor any agency thereof, nor any of their employees, makes any warranty, expressed 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.

■ REFERENCES

(1) IPCC. Technical Summary. In *Working Group III Contribution to the Sixth Assessment Report*; Intergovernmental Panel on Climate Change, 2022.

(2) Arning, K.; Linzenich, A.; Engelmann, L.; Ziefle, M. More Green or Less Black ? How Benefit Perceptions of CO2 Reductions vs. Fossil Resource Savings Shape the Acceptance of CO2-Based Fuels and Their Conversion Technology. *Energy and Climate Change* **2021**, *2*, 100025.

(3) de Kleijne, K.; Hanssen, S. V.; van Dinteren, L.; Huijbregts, M. A. J.; van Zelm, R.; de Coninck, H. Limits to Paris Compatibility of CO2 Capture and Utilization. *One Earth* **2022**, *5* (2), 168−185.

(4) Hepburn, C.; Adlen, E.; Beddington, J.; Carter, E. A.; Fuss, S.; Mac Dowell, N.; Minx, J. C.; Smith, P.; Williams, C. K. The Technological and Economic Prospects for CO2 Utilization and Removal. *Nature* **2019**, *575* (7781), 87−97.

(5) Majumdar, A.; Deutch, J. Research Opportunities for CO2 Utilization and Negative Emissions at the Gigatonne Scale. *Joule* **2018**, *2* (5), 805−809.

(6) Centi, G.; Perathoner, S.; Salladini, A.; Iaquaniello, G. Economics of CO2 Utilization: A Critical Analysis. *Frontiers in Energy Research* **2020**, *8*, 567986.

(7) Ho, H. J.; Iizuka, A.; Shibata, E. Carbon Capture and Utilization Technology without Carbon Dioxide Purification and Pressurization: A Review on Its Necessity and Available Technologies. *Ind. Eng. Chem. Res.* **2019**, *58* (21), 8941−8954.

(8) Tu, Q.; Eckelman, M.; Zimmerman, J. Meta-Analysis and Harmonization of Life Cycle Assessment Studies for Algae Biofuels. *Environ. Sci. Technol.* **2017**, *51* (17), 9419−9432.

(9) Garcia, R.; Figueiredo, F.; Brandão, M.; Hegg, M.; Castanheira, E.; Malça, J.; Nilsson, A.; Freire, F. A Meta-Analysis of the Life Cycle Greenhouse Gas Balances of Microalgae Biodiesel. *International Journal of Life Cycle Assessment* **2020**, *25* (9), 1737−1748.

(10) Somers, M. D.; Quinn, J. C. Sustainability of Carbon Delivery to an Algal Biorefinery: A Techno-Economic and Life-Cycle Assessment. *Journal of CO2 Utilization* **2019**, *30*, 193−204.

(11) Middleton, R. S.; Clarens, A. F.; Liu, X.; Bielicki, J. M.; Levine, J. S. *CO2 Deserts: Implications of Existing CO2 Supply Limitations for*

Carbon Management. Environmental science & technology **2014**, *48* (19), 11713−11720.
(12) Ou, L.; Banerjee, S.; Xu, H.; Coleman, A. M.; Cai, H.; Lee, U.; Wigmosta, M. S.; Hawkins, T. R. Utilizing High-Purity Carbon Dioxide Sources for Algae Cultivation and Biofuel Production in the United States: Opportunities and Challenges. *Journal of Cleaner Production* **2021**, *321*, 128779.
(13) Pilorgé, H.; McQueen, N.; Maynard, D.; Psarras, P.; He, J.; Rufael, T.; Wilcox, J. Cost Analysis of Carbon Capture and Sequestration of Process Emissions from the U.S. Industrial Sector. *Environ. Sci. Technol.* **2020**, *54* (12), 7524−7532.
(14) Rao, A. B.; Rubin, E. S. A Technical, Economic, and Environmental Assessment of Amine-Based CO2 Capture Technology for Power Plant Greenhouse Gas Control. *Environ. Sci. Technol.* **2002**, *36* (20), 4467−4475.
(15) Rao, A. B.; Rubin, E. S. Identifying Cost-Effective CO 2 Control Levels for Amine-Based CO 2 Capture Systems. *Ind. Eng. Chem. Res.* **2006**, *45* (8), 2421−2429.
(16) Hu, B.; Zhai, H. The Cost of Carbon Capture and Storage for Coal-Fired Power Plants in China. *International Journal of Greenhouse Gas Control* **2017**, *65*, 23−31.
(17) Muratori, M.; Kheshgi, H.; Mignone, B.; Clarke, L.; McJeon, H.; Edmonds, J. Carbon Capture and Storage across Fuels and Sectors in Energy System Transformation Pathways. *International Journal of Greenhouse Gas Control* **2017**, *57*, 34−41.
(18) Bui, M.; Adjiman, C. S.; Bardow, A.; Anthony, E. J.; Boston, A.; Brown, S.; Fennell, P. S.; Fuss, S.; Galindo, A.; Hackett, L. A.; et al. Carbon Capture and Storage (CCS): The Way Forward. *Energy Environ. Sci.* **2018**, *11* (5), 1062−1176.
(19) Mantripragada, H. C.; Veser, G. Hydrogen Production via Chemical Looping Dry Reforming of Methane: Process Modeling and Systems Analysis. *AIChE J.* **2022**, *68* (5), No. e17612.
(20) Mantripragada, H. C.; Veser, G. Intensifying Chemical Looping Dry Reforming: Process Modeling and Systems Analysis. *Journal of CO2 Utilization* **2021**, *49*, 101555.
(21) Zang, G.; Sun, P.; Yoo, E.; Elgowainy, A.; Bafana, A.; Lee, U.; Wang, M.; Supekar, S. Synthetic Methanol/Fischer−Tropsch Fuel Production Capacity, Cost, and Carbon Intensity Utilizing CO2 from Industrial and Power Plants in the United States. *Environ. Sci. Technol.* **2021**, *55* (11), 7595−7604.
(22) Wong, J.; Santoso, J.; Went, M.; Sanchez, D. Market Potential for CO2 Removal and Sequestration from Renewable Natural Gas Production in California. *Environ. Sci. Technol.* **2022**, *56* (7), 4305−4316.
(23) Sanchez, D. L.; Johnson, N.; McCoy, S. T.; Turner, P. A.; Mach, K. Correction for Sanchez et al., Near-term deployment of carbon capture and sequestration from biorefineries in the United States. *Proc. Natl. Acad. Sci. U. S. A.* **2018**, *115* (42), E9991.
(24) Keith, D. W.; Holmes, G.; St. Angelo, D.; Heidel, K. A Process for Capturing CO2 from the Atmosphere. *Joule* **2018**, *2* (8), 1573−1594.
(25) McQueen, N.; Psarras, P.; Pilorgé, H.; Liguori, S.; He, J.; Yuan, M.; Woodall, C. M.; Kian, K.; Pierpoint, L.; Jurewicz, J.; Lucas, J. M.; Jacobson, R.; Deich, N.; Wilcox, J. Cost Analysis of Direct Air Capture and Sequestration Coupled to Low-Carbon Thermal Energy in the United States. *Environ. Sci. Technol.* **2020**, *54* (12), 7542−7551.
(26) Viebahn, P.; Scholz, A.; Zelt, O. The Potential Role of Direct Air Capture in the German Energy Research Program—Results of a Multi-Dimensional Analysis. *Energies* **2019**, *12* (18), 3443.
(27) Knoope, M. M. J.; Ramírez, A.; Faaij, A. P. C. A State-of-the-Art Review of Techno-Economic Models Predicting the Costs of CO2 Pipeline Transport. *International Journal of Greenhouse Gas Control* **2013**, *16*, 241−270.
(28) Brune, D. E.; Lundquist, T. J.; Benemann, J. R. Microalgal Biomass for Greenhouse Gas Reductions: Potential for Replacement of Fossil Fuels and Animal Feeds. *J. Environ. Eng.* **2009**, *135* (11), 1136−1144.
(29) Davis, R.; Markham, J.; Kinchin, C.; Grundl, N.; Tan, E.; Humbird, D. In *Process Design and Economics for the Production of Algal Biomass: Algal Biomass Production in Open Pond Systems and Processing Through Dewatering for Downstream Conversion*; National Renewable Energy Lab. (NREL): Golden, CO, 2016; pp 1−19.
(30) Biddy, M.; Davis, R.; Jones, S.; Zhu, Y. *Whole Algae Hydrothermal Liquefaction Technology Pathway. Technical Report Efficiency & Renewable Energy*; NREL/TP-5100-58051 PNNL-22314; Alliance for Sustainable Energy, LLC, 2013.
(31) *The Greenhouse Gases, Regulated Emissions, and Energy Use in Technologies Model*; Argonne National Lab. (ANL): Lemont, IL, 2022. https://greet.es.anl.gov (accessed 2023−04−07).
(32) Singh, U.; Colosi, L. M. The Case for Estimating Carbon Return on Investment (CROI) for CCUS Platforms. *Applied Energy* **2021**, *285*, 116394.
(33) Terlouw, T.; Bauer, C.; Rosa, L.; Mazzotti, M. Life Cycle Assessment of Carbon Dioxide Removal Technologies: A Critical Review. *Energy Environ. Sci.* **2021**, *14* (4), 1701−1721.
(34) Yoo, E.; Lee, U.; Zang, G.; Sun, P.; Elgowainy, A.; Wang, M. Incremental Approach for the Life-Cycle Greenhouse Gas Analysis of Carbon Capture and Utilization. *Journal of CO2 Utilization* **2022**, *65*, 102212.
(35) Garcia-Garcia, G.; Fernandez, M. C.; Armstrong, K.; Woolass, S.; Styring, P. Analytical Review of Life-Cycle Environmental Impacts of Carbon Capture and Utilization Technologies. *ChemSusChem* **2021**, *14* (4), 995−1015.
(36) Psarras, P. C.; Comello, S.; Bains, P.; Charoensawadpong, P.; Reichelstein, S.; Wilcox, J. Carbon Capture and Utilization in the Industrial Sector. *Environ. Sci. Technol.* **2017**, *51* (19), 11440−11449.
(37) Keith, D. W.; Holmes, G.; St. Angelo, D.; Heidel, K. A Process for Capturing CO2 from the Atmosphere. *Joule* **2018**, *2* (8), 1573−1594.
(38) McQueen, N.; Gomes, K. V.; McCormick, C.; Blumanthal, K.; Pisciotta, M.; Wilcox, J. A Review of Direct Air Capture (DAC): Scaling up Commercial Technologies and Innovating for the Future. *Progress in Energy* **2021**, *3* (3), 032001.
(39) Fasihi, M.; Efimova, O.; Breyer, C. Techno-Economic Assessment of CO 2 Direct Air Capture Plants. *Journal of Cleaner Production* **2019**, *224*, 957−980.
(40) Breyer, C.; Fasihi, M.; Aghahosseini, A. Carbon Dioxide Direct Air Capture for Effective Climate Change Mitigation Based on Renewable Electricity: A New Type of Energy System Sector Coupling. *Mitigation and Adaptation Strategies for Global Change* **2020**, *25* (1), 43−65.
(41) Clarke, L.; Wei, Y.-M.; de la Vega Navarro, A.; Garg, A.; Hahmann, A. N.; Khennas, S.; Azevedo, I. M.; Löschel, A.; Singh, A. K.; Steg, L. Energy Systems. In *Climate Change 2022: Mitigation of Climate Change, Working Group III Contribution to the IPCC Sixth Assessment Report*; Cambridge University Press, 2022.
(42) Davis, R.; Markham, J.; Kinchin, C.; Grundl, N.; Tan, E. C. D.; Humbird, D.; Davis, R.; Markham, J.; Kinchin, C.; Grundl, N.; Tan, E. C. D.; Humbird, D. *Process Design and Economics for the Production of Algal Biomass: Algal Biomass Production in Open Pond Systems and Processing Through Dewatering for Downstream Conversion*; Technical Report NREL/TP-5100-64772; National Renewable Energy Lab. (NREL), 2016.
(43) Jones, S.; Zhu, Y.; Anderson, D.; Hallen, R. T.; Elliott, D. C. *Process Design and Economics for the Conversion of Algal Biomass to Hydrocarbons*; Technical Report PNNL-23227; Pacific Northwest National Lab. (PNNL), 2014.
(44) Cai, H.; Ou, L.; Wang, M.; Davis, R.; Dutta, A.; Harris, K.; Wiatrowski, M. R.; Tan, E.; Bartling, A.; Bruno, K. *Supply Chain Sustainability Analysis of Renewable Hydrocarbon Fuels via Indirect Liquefaction, Ex Situ Catalytic Fast Pyrolysis, Hydrothermal Liquefaction, Combined Algal Processing, and Biochemical Conversion: Update of the 2020 State-of-Technology Cases*; Report No. ANL/ESD-20/2; Argonne National Lab. (ANL): Argonne, IL, 2021.
(45) Cooney, G.; Benitez, J.; Lee, U.; Wang, M. *Clarification to Recent Publication - Incremental Approach for the Life-Cycle Greenhouse Gas Analysis of Carbon Capture and Utilization*; Technical Publications; Argonne National Laboratory: Lemont, IL, 2022. https://greet.es.anl.gov/files/ccu_lca_memo (accessed 2023−04−07).

(46) Singh, U.; Dunn, J. B. Shale Gas Decarbonization in the Permian Basin: Is It Possible? *ACS Engineering Au* **2022**, *2* (3), 248−256.
(47) Seto, K. C.; Davis, S. J.; Mitchell, R. B.; Stokes, E. C.; Unruh, G.; Ürge-Vorsatz, D. Carbon Lock-In: Types, Causes, and Policy Implications. *Annual Review of Environment and Resources* **2016**, *41*, 425−452.
(48) US EIA. Gasoline and Diesel Fuel Update. https://www.eia.gov/petroleum/gasdiesel/ (accessed 2022-04−27).
(49) SB 1383 Senate Bill - Bill Analysis. http://www.leginfo.ca.gov/pub/15-16/bill/sen/sb_1351-1400/sb_1383_cfa_20160831_214733_sen_comm.html (accessed 2022-10−11).
(50) Denholm, P.; Brown, P.; Cole, W.; Mai, T.; Sergi, B.; Brown, M.; Jadun, P.; Ho, J.; Mayernik, J.; McMillan, C. *Examining Supply-Side Options to Achieve 100% Clean Electricity by 2035*; National Renewable Energy Lab. (NREL): Golden, CO, 2022.
(51) Horowitz, R.; Binsted, M.; Browning, M.; Fawcett, A.; Henly, C.; Hultman, N.; McFarland, J.; McJeon, H. The Energy System Transformation Needed to Achieve the US Long-Term Strategy. *Joule* **2022**, *6* (7), 1357−1362.
(52) US EIA. 2021 Annual Energy Outlook. https://www.eia.gov/outlooks/aeo/tables_side.php (accessed 2022-01−01).
(53) Jackson, F. Major Technology Companies To Provide Nearly $1 Billion Building Market Demand For CO2 Removal *Forbes*, 2022. https://www.forbes.com/sites/feliciajackson/2022/04/13/major-technology-companies-provide-nearly-1-billion-backing-for-co2-removal/?sh=65d61ec82969.
(54) Fuss, S.; Lamb, W. F.; Callaghan, M. W.; Hilaire, J.; Creutzig, F.; Amann, T.; Beringer, T.; De Oliveira Garcia, W.; Hartmann, J.; Khanna, T.; Luderer, G.; Nemet, G. F.; Rogelj, J.; Smith, P.; Vicente, J. V.; Wilcox, J.; Del Mar Zamora Dominguez, M.; Minx, J. C. Negative Emissions - Part 2: Costs, Potentials and Side Effects. *Environ. Res. Lett.* **2018**, *13* (6), 063002.
(55) Xu, H.; Lee, U.; Coleman, A. M.; Wigmosta, M. S.; Wang, M. Assessment of Algal Biofuel Resource Potential in the United States with Consideration of Regional Water Stress. *Algal Research* **2019**, *37*, 30−39.
(56) Xu, H.; Lee, U.; Coleman, A. M.; Wigmosta, M. S.; Sun, N.; Hawkins, T.; Wang, M. Balancing Water Sustainability and Productivity Objectives in Microalgae Cultivation: Siting Open Ponds by Considering Seasonal Water-Stress Impact Using AWARE-US. *Environ. Sci. Technol.* **2020**, *54* (4), 2091−2102.
(57) Lu, X.; Cao, L.; Wang, H.; Peng, W.; Xing, J.; Wang, S.; Cai, S.; Shen, B.; Yang, Q.; Nielsen, C. P.; McElroy, M. B. Gasification of Coal and Biomass as a Net Carbon-Negative Power Source for Environment-Friendly Electricity Generation in China. *Proc. Natl. Acad. Sci. U. S. A.* **2019**, *116* (17), 8206−8213.
(58) Ogland-Hand, J. D.; Cohen, S. M.; Kammer, R. M.; Ellett, K. M.; Saar, M. O.; Bennett, J. A.; Middleton, R. S. The Importance of Modeling Carbon Dioxide Transportation and Geologic Storage in Energy System Planning Tools. *Frontiers in Energy Research* **2022**, *10*, 855105.
(59) Fuhrman, J.; Bergero, C.; Weber, M.; Monteith, S.; Wang, F. M.; Clarens, A. F.; Doney, S. C.; Shobe, W.; McJeon, H. Diverse Carbon Dioxide Removal Approaches Could Reduce Impacts on the Energy−Water−Land System. *Nature Climate Change* **2023**, *13* (4), 341−350.
(60) Elgowainy, A.; Vyawahare, P.; Ng, C.; Bafana, A.; Burnham, A.; Sun, P.; Cai, H.; Lee, U.; Reddi, K.; Wang, M. *Hydrogen Life Cycle Analysis in Support of Clean Hydrogen Production*; Argonne National Lab. (ANL): Argonne, IL, 2022.
(61) Pivovar, B.; Rustagi, N.; Satyapal, S. Hydrogen at Scale (H2@ Scale): Key to a Clean, Economic, and Sustainable Energy System. *Electrochemical Society Interface* **2018**, *27* (1), 47.
(62) Ruth, M. F.; Jadun, P.; Gilroy, N.; Connelly, E.; Boardman, R.; Simon, A. J.; Elgowainy, A.; Zuboy, J. *The Technical and Economic Potential of the H2@ Scale Hydrogen Concept within the United States*; National Renewable Energy Lab. (NREL): Golden, CO, 2020.
(63) Ocko, I. B.; Hamburg, S. P. Climate Consequences of Hydrogen Emissions. *Atmospheric Chemistry and Physics* **2022**, *22* (14), 9349−9368.
(64) Zhu, Y.; Jones, S. B.; Schmidt, A. J.; Billing, J. M.; Thorson, M. R.; Santosa, D. M.; Hallen, R. T.; Anderson, D. B. *Algae/Wood Blends Hydrothermal Liquefaction and Upgrading: 2019 State of Technology*; Pacific Northwest National Lab. (PNNL): Richland, WA (United States), 2020.
(65) Zhu, Y.; Xu, Y.; Schmidt, A. J.; Thorson, M. R.; Cronin, D. J.; Santosa, D. M.; Edmundson, S. J.; Li, S.; Snowden-Swan, L. J.; Valdez, P. J. *Microalgae Hydrothermal Liquefaction and Biocrude Upgrading: 2022 State of Technology*; Pacific Northwest National Lab. (PNNL): Richland, WA, 2023.
(66) Snowden-Swan, L. J.; Li, S.; Jiang, Y.; Thorson, M. R.; Schmidt, A. J.; Seiple, T. E.; Billing, J. M.; Santosa, D. M.; Hart, T. R.; Fox, S. P. *Wet Waste Hydrothermal Liquefaction and Biocrude Upgrading to Hydrocarbon Fuels: 2021 State of Technology*; Pacific Northwest National Lab. (PNNL): Richland, WA, 2022.

https://doi.org/10.1021/acssuschemeng.3c02082
ACS Sustainable Chem. Eng. 2023, 11, 14435−14444