Integrating direct air capture with algal biofuel production to reduce cost, energy, and GHG emissions

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ABSTRACT

We investigate the potential to reduce costs and greenhouse gas emissions of the utilization of direct air capture of CO_2 (DAC) for the production of algal biofuel. We examine four integrated designs for a DAC system comprised of solid amine monolith adsorbents delivering CO_2 at the required level for algae cultivation with a photo-bioreactor (PBR)-based fuel production facility. We show that the integration of DAC with this biofuel production facility provides cost and greenhouse gas emissions benefits. Heat integration decreases operating expenses by reducing energy demand for heating requirements. Mass integration, utilizing flue gas CO_2 as a carbon source for the PBRs, decreases the DAC system scale, resulting in both capital and operating cost savings. The most advantageous option depends on the interplay of heat and mass integration while matching the diurnal rhythm of algal growth with the inherently steady pace and energy requirements of the DAC system and fuel production. For these technologies, the DAC-PBR mass and energy integration provides an 18 % cost reduction and a 50 % reduction in greenhouse gas emissions for the current state of the technology.

1. Introduction

To meet goals to address and mitigate the impacts of climate change, a transition to low-carbon fuels and the removal of carbon dioxide from the atmosphere are required. The development of technologies for capturing carbon dioxide directly from the air (DAC) is currently underway [18,32]. However, the costs and energy requirements of DAC systems still remain high. According to a committee of the U.S. National Academies of Sciences [27], the costs for liquid and solid sorbent DAC systems are significantly more than $100 per ton.

The thermal energy requirements for the liquid sorbent systems are roughly 9–12 GJ per ton of CO_2 captured and 3.4–4.8 GJ/ton for solid sorbent systems; both types of systems have an electricity requirement in the range of 0.55 – 1.7 kWh/ton [27]. Using a learning curve approach, Fasihi et al. [10] forecast that costs could fall below about $50/ton by 2050 if the deployment of DAC could reach 15 Gt per year. This rate of DAC is consistent with, although at the high end, of estimated requirements of 2–15 Gt/year for carbon dioxide removal of all types to stabilize climate change to less than 1.5°C [34]. The International Energy Agency [13] found that DAC deployment of about 1 Gt CO_2 per year

is needed by 2050 both to meet climate goals and to scale down costs; it projects costs of less than $100/ton CO_2 and as low as $50/ton by 2050 in regions with low-cost renewable energy. As of 2024, only three plants have a capacity of more than a thousand tons of CO_2 per year [14]. The US Department of Energy is supporting the construction of two projects, in Texas and Louisiana, that aim to capture a total of two million tons of CO_2/year [39].

Direct air capture is energy intensive. Madhu et al. [24] found that with low-carbon energy supplies, a net carbon removal of 73 % and 86 % can be achieved for liquid and solid sorbent systems respectively. The US National Energy Technology Laboratory has evaluated greenhouse gas emissions from DAC systems that store CO_2 in saline aquifers, and DAC systems that use CO_2 for enhanced oil recovery and algal biofuel production, over a range of energy system carbon intensities [29]. For both enhanced oil recovery and algal biofuel production, the systems are carbon positive, meaning there is a net emission of CO_2 to the atmosphere, but can be carbon reducing, meaning that the emissions are lower than the systems replaced: baseline petroleum extraction in the case of enhanced oil recovery, and fossil-derived gasoline or diesel in the case of algal biofuels.

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To be able to contribute to climate mitigation, DAC technologies need to have lower costs and lower net greenhouse gas emissions. Improved sorbents that can lower costs and energy requirements are an ongoing research priority [35]. From a systems perspective, lower costs and low emissions can be achieved by siting carbon capture at low-cost and low-carbon energy sources. However, given the vast scale needed for direct air capture to make a meaningful contribution to reducing atmospheric greenhouse gas concentrations, all possible savings in both cost and energy requirements are needed.

Direct air capture systems must either sequester the captured CO_2 or utilize the CO_2 in a way that could displace a source of CO_2 emissions. Options for utilizing DAC CO_2 include the production of fuels with low greenhouse gas emissions [23]. DAC processes require electricity and heat. Making fuel from the CO_2 also generally requires electricity and heat. Here, we consider the proposition that there may be ways to combine the DAC process and the CO_2 utilization process that will reduce electricity, heat, or CO_2 requirements and thereby reduce costs and reduce lifecycle greenhouse gas emissions.

Techno-economic analysis (TEA) and life cycle assessment (LCA) are now widely used approaches to evaluate the cost and environmental impacts of new technologies. It is common to consider several scenarios in the analysis for different energy sources, different feedstocks, or different utilization of co-products [37,38]. Here, we go beyond combinations of different energy systems with DAC systems to create integrated process designs, using both techno-economic analysis and life cycle assessment to optimize the design for an integrated DAC-biofuels system.

Utilization of waste from one industrial process as raw material for another industrial process is a core concept of industrial ecology [21]. The potential for integrating electricity, heating, and carbon dioxide infrastructures extends beyond the utilization of waste toward co-design for multiple services [31]. The utilization of waste CO_2 is particularly attractive if it can replace the use of fossil carbon. Industrial waste heat is employed in district heating and combined heat and power systems. The value of waste heat as an energy source for direct air capture of carbon dioxide is well recognized because it is a low-cost and low-carbon source of energy [8]. Yet, the utilization of heat in integrated systems remains challenging, with, in some cases, multiple trade-offs determining whether the heat utilization is a net benefit compared to less integrated designs [5]. Analysis and retrospectives of industrial symbiosis emphasize the complexity and challenges [11].

The production of algal biofuels has advantages over other biofuel systems in that arable land is not required, and there is great flexibility regarding location. Additionally, its areal productivity is relatively high compared to other biofuel options. A challenge, however, is that CO_2 must be provided. CO_2 can be provided by capturing the emissions from fossil fuel power plants. This effectively requires locating the algal biofuel facility near the fossil fuel plant, where land availability is typically low, and it also puts biofuel production at risk if fossil fuel use declines. CO_2 can also be provided by utilizing onsite natural gas combined heat and power, yet this type of setup tends to have relatively high greenhouse gas emissions [1,2]. In principle, it would be highly advantageous to source the CO_2 from systems that can be located anywhere. Thus, Direct Air Capture (DAC) may be well suited as a CO_2 source for algal biofuel production.

A significant challenge in the production of algal biofuels is cost. The Bioenergy Technologies Office of the US Department of Energy set a goal of reaching $2.50 per gasoline gallon equivalent (GGE) by 2030 [30]. Technology assessments have consistently concluded that this is not feasible if relying on fuel production advancements alone [19]. The biofuel production process considered here has ethanol production directly from blue-green algae in closed photobioreactors; the production cost using conventional sources of CO_2 was estimated to be $13/GGE ($8.4/gallon ethanol) [20], while systems that co-produce fuels and other products do show this potential [42,43]. For fuel production without co-products, current algal biofuel production

technologies have reported minimum fuel selling prices in the range of $9 to $15 per GGE.

The objective here is to evaluate the potential to reduce costs and greenhouse gas emissions of using carbon dioxide from a DAC system to produce algal biofuel by integrating the DAC process and the algal biofuel production process. Specifically, we design several system options in which the CO_2 utilization process – algal biofuel production in this case - is the source of the heat for the CO_2 capture process, and in which carbon dioxide from heat and electricity production is utilized to reduce DAC requirements. Creating the system designs required detailed physical process modeling in order to evaluate sub-process energy use, heat integration capabilities, and cost. Cost and emissions reduction through system integration have not been previously examined. Previous studies have examined the siting of DAC facilities at sources of waste heat [8,38]. We explore the potential for efficiencies in DAC applications by thoroughly examining process design options for algal biofuels, combining the electricity and heat requirements of both systems to produce a low-carbon biofuel. One approach we consider is the use of multiple small DAC systems operating amidst the field of photobioreactors that are growing the blue-green algae; we compare this with the approach of a single large DAC system that serves the entire facility. Another approach that we consider is to run the DAC system only during the day, when the blue-green algae require CO_2; we compare this with running the DAC system 24/7 and compressing and storing the CO_2 at night. The latter option requires energy, capital expenditure, and operational expenditure to operate the CO_2 compression and storage, yet reduces the energy, capital expenditure, and operational expenditure to operate the DAC system. Combining these approaches provides four integrated engineering design options. More generally, this study brings forward opportunities for system-level improvements in systems that include DAC. These broader opportunities are discussed in the discussion section.

2. Methods

2.1. Techno-economic analysis

The techno-economic analysis presented here assumes standard n^{th}-plant economics. The standard n^{th}-plant economy indicates mature plant operation associated with low-risk, full-scale functionality. These assumptions are collected in Table 1 and are consistent with other analyses completed for DOE's Bioenergy Technologies Office and Industrial

Table 1
Summary of nth-Plant Assumptions for Techno-Economic Analysis.

Description of Assumption	Assumed Value
Internal Rate of Return (IRR)	10 %
Plant Financing by Equity/Debt	40 %/60 % of total capital investment
Plant Life	30 years
Income Tax Rate	21 %
Interest Rate for Debt Financing	8 % annually
Term for Debt Financing	10 years
Working Capital Cost	5 % of fixed capital cost (excluding land purchase cost)
Depreciation Schedule	
General Plant	7-years MACRS schedule
Steam Plant*	20-years MACRS schedule
Construction Period (Spending Schedule)	3 years (8 % Y1, 60 % Y2, 32 % Y3)
Plant Salvage Value	No value
Start-Up Time	0.5 years
Revenue and Costs During Startup	Revenue = 50 % of normal
	Variable costs = 75 % of normal
	Fixed costs = 100 % of normal
On-Stream Percentage After Startup	90 % (7884 operating hours per year)

* Steam plant depreciation schedule will apply only if a process produces excess electricity as a co-product.

Efficiency and Decarbonization Office [7,15].(Fig. 1).

2.2. Life cycle assessment

The goal of the life cycle greenhouse gas assessment developed here is to evaluate the effect of mass and energy integration of DAC with algal biofuel production on lifecycle greenhouse gas emissions. An attributional LCA approach is appropriate for this type of assessment of engineering process choices [28]. The scope of the study is to address the production of algal biofuel using direct air capture as a source of carbon. The system boundary includes the production facility configurations shown in Figs. 2, 3, 4, and 5, as well as the sourcing of natural gas, the combustion of the bioethanol at the end of its lifecycle, and the production of the DAC module systems and the PBRs. IPCC AR6 global warming potentials in the 100-year time horizon are used [17]. A process matrix life cycle assessment (LCA) approach, implemented in Python, is utilized to obtain the resulting greenhouse gas emissions for different model configurations in the biorefinery design. The functional unit is 1 MJ of bioethanol. As recommended by Terlouw et al. [37] and general best practice in LCA, we recognize the distinction between avoided emissions and negative emissions; avoided emissions are not included in this analysis.

The system is modeled with natural gas as the energy source. The supply chain of natural gas is assumed to involve leaks. In a review of life cycle assessment (LCA) studies of natural gas, a supply chain central estimate of fugitive emission of 10.6 g CO_2e/MJ_{HHV} has been reported [3]. Littlefield et al. [22] reported the average cradle-to-delivery methane leak rate is 0.97 %, with a 95-percentile mean confidence interval ranging from 0.61 % to 1.43 %. That is, for every 1 kg delivered, 0.98 %, ranging from 0.62 % to 1.44 %, is leaked. The global warming potential for fossil methane is 29.8±11 CO_2e/kg CH_4 in the 100-year time horizon [17]. Accordingly, the emissions associated with leakage are 0.29±0.26 kg CO_2e for each delivered kg of methane in the 100-year time horizon.

The DAC and PBR process models, described above and summarized in Tables 1 and 2, were used to evaluate energy inputs, CO_2 flows, and biofuel production. In addition, the lifecycle greenhouse gas emissions from the production of the monolith structure used to capture CO_2 are included, as described in the Supporting Information.

3. Description of the biofuel and direct air capture systems

3.1. Biofuel and direct air capture system designs

The production facility utilizes cyanobacteria growing in brackish water supplemented with nitrogen and phosphorus contained in photobioreactors (PBR). The system modeled here is a 2000-acre facility biorefinery [6,20]. Of the carbon consumed by the cyanobacteria, 66 % ends up in ethanol, and the rest in biomass. An additional 15 % CO_2 is supplied above the stoichiometric requirements [1,20]. The target ethanol production rate is 73,000 L/(ha-year).

The DAC and PBR process models were developed in Aspen Plus V10. The PBR system process model was based on previously published design information [20]. Versions of the PBR system process model have been previously published [1]. The DAC system process model is based on Eisenberger and Chichilnisky [9] and design information provided by Global Thermostat; see also Steutermann and Sakwa-Nowak (2022). The Aspen Plus models provided scaled, steady-state mass and energy flows required for this technology's operation. The underlying performance assumptions for each case were held constant between cases. High-level summaries of key assumptions for the algal biofuel and DAC systems are provided in Tables 2 and 3. Fig. 1 shows the simplified block flow diagram of the integrated DAC and PBR system, and the process components are described in the paragraphs below.

Fig. 1 Area 100 (Adsorption) encapsulates the capital and operating expenses for the adsorption step of the direct air capture process. In this section, ambient air is blown through a container, and CO_2 is adsorbed onto the polymeric material polyethyleneimine (PEI) contained in a monolith panel. A single container in this design consists of nine panels in the adsorption phase and simultaneously one panel in the regeneration phase. The panels are physically cycled around the container at desired time intervals to be regenerated [9,36]). These dynamics were modeled using time-average calculations of experimental and pilot plant data.

The process design of Area 200 (Regeneration & Harvesting) includes the regeneration of a single monolith panel per container and the condensation and separation of process steam from the product CO_2. Some capital expenses for this step were captured in Area 100. During this phase, a panel enters the regeneration box, and a vacuum is applied to remove excess air. Low-pressure steam is then injected into the regeneration box to pressurize the system, and the heat of the condensing steam provides the desorption energy for CO_2 recovery. Some fraction of that steam passes through the monolith panel with the desorbed CO_2. This steam is condensed in a heat exchanger with cooling water, and CO_2 is recovered with a flash drum. The condensed steam in the monolith is then flashed from the panel by applying a vacuum to the system. For model simplicity, the entire regeneration phase takes place at the pressure of the vacuum steam, and additional vacuum pumping requirements are approximated.

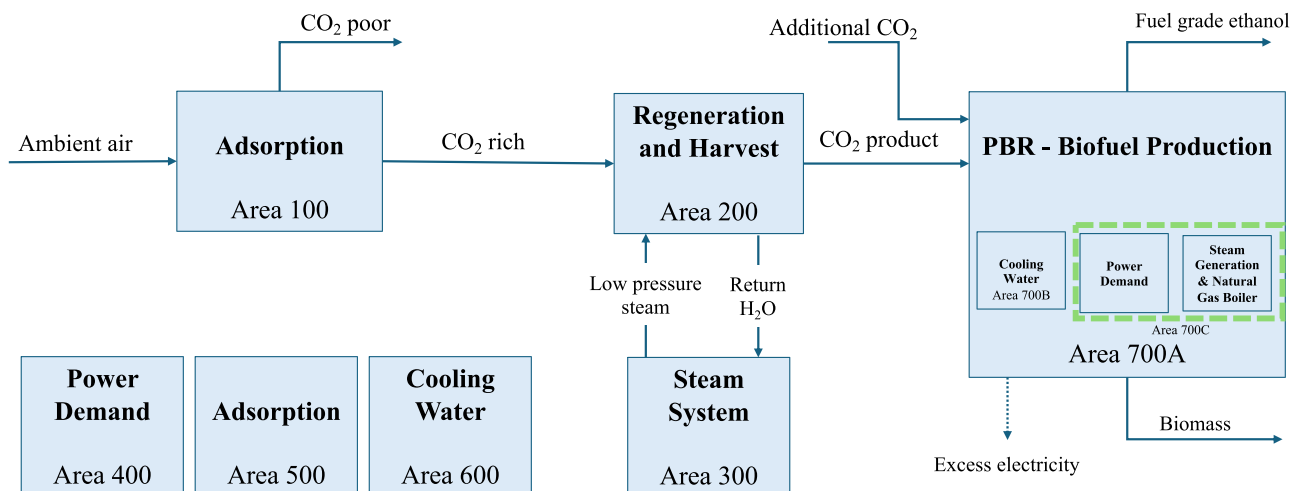Heat is provided to the DAC system by low pressure steam (Area 300,

Fig. 1. Simplified block flow diagram of the integrated DAC and PBR system.
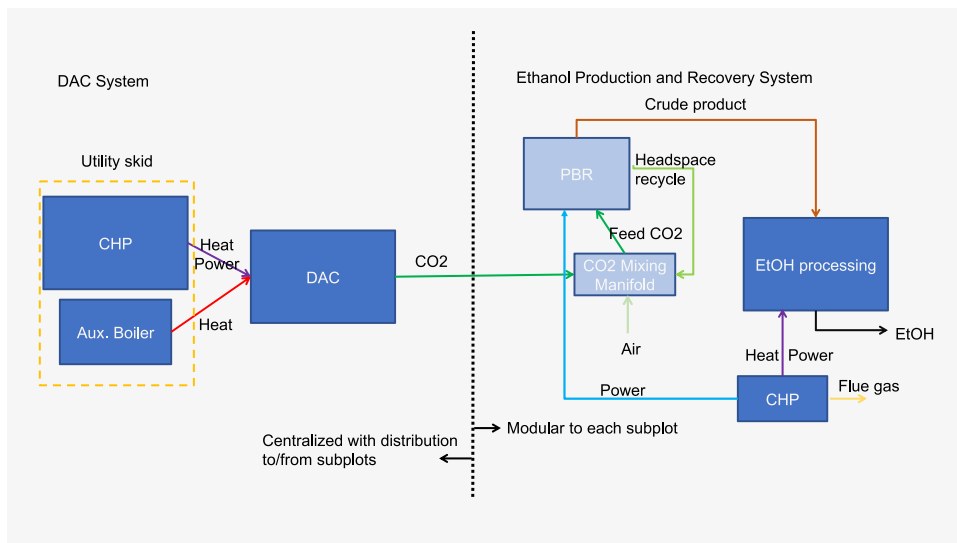
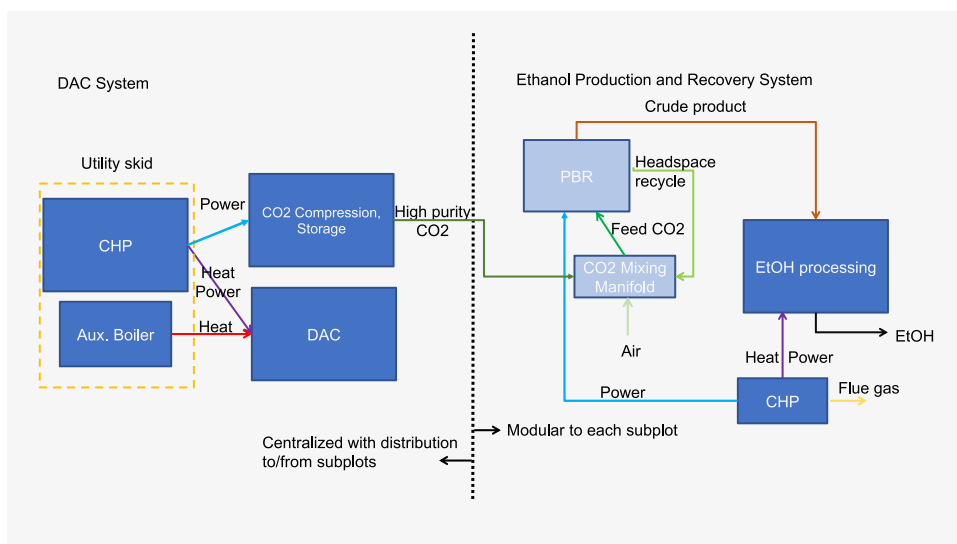**Fig. 2.** Diagrammatic representation of the configuration *BASELINE-12*.

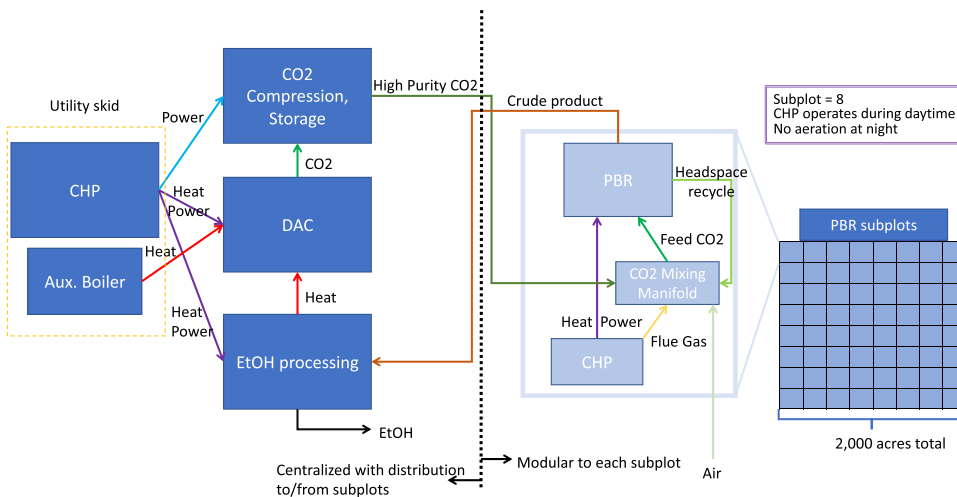**Fig. 3.** Diagrammatic representation of the configuration *BASELINE-24*.

Fig. 4. Diagrammatic representation of the configuration *OPTION-24–1*.
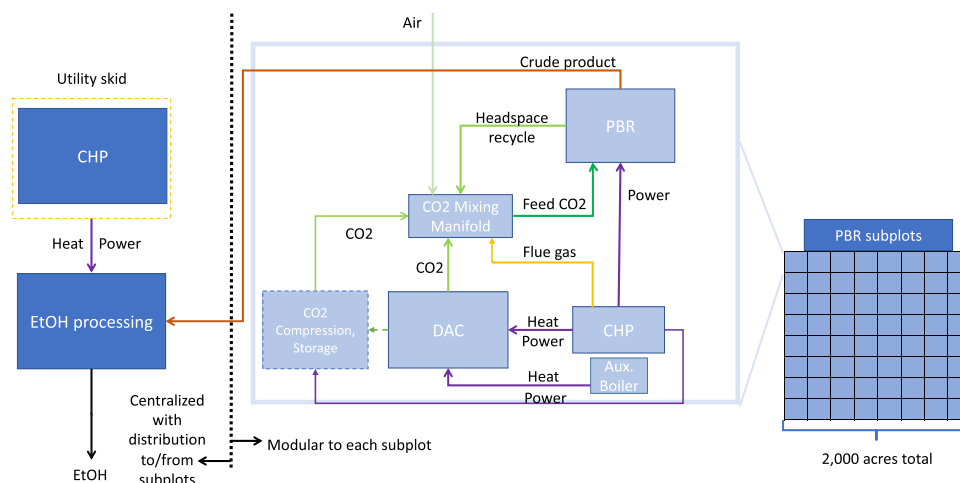
Fig. 5. Diagrammatic representation of the configuration *OPTION-24 2a and OPTION-12 2b* (with and without the inclusion of CO_2 compression and storage, respectively).

Table 2
Design Basis for Algal Biofuel Production.

Process Parameter	Design Information
Feed information	
Total CO_2 Fed (24 hr average), tonne/hr	20
CO_2 to PBR, wt%	12.00 %
H_3PO_4 Feed Rate, tonne/hr	0.1
Urea Feed Rate, tonne/hr	0.5
Algae (Biomass) Composition	$C_{106}H_{181}O_{45}N_{15}P$
vWeight Percent Carbon, %	52.68 %
Seed Biomass (expense calculation only), tonne/hr	0
Seawater Feed Rate, kgal/hr	165.56
Salt (NaCl) Concentration, %	3.50 %
Operating Conditions	
PBR	
Temperature, C	30
Pressure, atm	1
Excess CO_2 to PBR, %	16 %
Ethanol Yield, kg/hr	6856
Concentration in Seawater, wt%	1.00 %
Biomass Yield, kg/hr	2044
Concentration in Seawater, wt%	0.32 %
Vapor Compression Steam Stripper (VCSS)	
Temperature, C	97.3
Pressure, atm	1.03
Ethanol Product Concentration, wt%	9.16 %

Table 3
Design Basis for Direct Air Capture.

Process Parameter	Design Information
Area 100: Adsorption	
Monolith Pressure Drop, Pa	200
CO_2 Selectivity %	100 %
Maximum Container Capacity, tonne/yr	25,000
Area 200: Regeneration & Harvesting	
Regeneration Pressure, bar	0.8
Cooling Fluid in Condenser	Cooling Water
Regeneration Time, seconds	100
Recovered CO_2 Purity, mole %	92 %
Area 300: Steam System	
Steam Generation Pressure, bar	5.86
Steam to Pressurize Regeneration Module, bar	2
Steam to Thermal Capacitance, bar	1
Steam to Harvesting, bar	0.8
Net Steam Delivered, kg H_2O/kg CO_2	3.83
Area 500: Heat & Natural Gas Boiler (CHP unit)	
Natural Gas Boiler Efficiency %	80 %
Energy Required per Tonne CO_2, GJ/tonne	12.77
Excess Air to Boiler %	20 %
Area 600: Cooling Water	
Cooling Water Feed Temperature, F	90
Cooling Water Return Temperature, F	100

Steam System). This section calculates the steam demand as a function of CO_2 captured and models the generation and distribution of that steam. A fraction of this steam is lost to the atmosphere; however, a fraction is recycled as condensed steam from Area 200, and a fraction is recycled as condensed steam from the vacuum ballast.

The power demand area (Area 400) calculates the power required for the operation of the entire plant. This includes the ambient air blowers, pumps, and vacuum pumps required for each area. Electricity is generated on-site via a combined heat and power (CHP) unit. The base case investigates the impacts of only meeting the plant's power demand. The capital expenses for the CHP unit are included in Areas 400 and 500.

Area 500 (Heat & Natural Gas Boiler) calculates the heat required for the plant as a function of CO_2 captured. A CHP natural gas boiler was modeled to provide heat for steam generation. Cooling water (Area 600) is utilized in steam and vacuum ballast condensers. Area 600 models a cooling tower system and calculates cooling water demand and make-up required.

Ethanol Production and Separation (Area 700 A) models the production of ethanol and algal biomass. Algal biomass was assumed to be sequestered via deep well injection. The base case assumes that 20 % of

the 2,000-acre production facility's CO_2 demand is provided by direct air capture, and the remainder is purchased as pure CO_2. Ethanol and biomass are produced in photobioreactors; the ethanol and water are separated by a vapor compression steam stripper (VCSS) technology and traditional ethanol purification methods to produce a fuel-grade ethanol (99.7 wt%) with minimal loss. A similar model for algal ethanol production was developed in Arora et al. [1] and Arora et al. [2], all drawing on Legere [20].

3.2. Integration configurations

For the integrated algal biorefinery and DAC facility, we consider two baseline configurations and four integrated options, each varying by operating schedule, facility design, and heat and mass integration design. These six models are described in Table 4, and the diagrammatic representations of the respective configurations are depicted in Figs. 2–5. The *BASELINE-12* configuration (Fig. 1) has a DAC system that operates for 12 hr/day, while *BASELINE-24* (Fig. 2) has a DAC system that operates for 24 hr/day. The PBR system can only operate during the daytime, in the presence of sunlight, so the 12 hr/day configuration

Table 4
Process Configurations and Operational Descriptions.

Scenario	Description
Baseline−12	**No heat and mass integration** between DAC and PBR/EtOH processing facilities. **DAC unit operates 12 hours/day**, capturing 40 tonnes/hr (total PBR demand over a 12-hour period), as depicted in Fig. 2.
Baseline−24	**No heat and mass integration** between DAC and PBR/EtOH processing facilities. **DAC unit operates 24 hours/day**, capturing 20 tonnes/hr. At night, CO_2 is compressed, liquified, and stored. A total of 40 tonnes/hr is then distributed to the PBRs during the day (total PBR demand over a 12-hour period), as represented in Fig. 3.
Option−24 1 use sub-plot flue gas CO2	**Heat and mass integration.** Centralized DAC and EtOH processing facility equipped with liquefaction unit, CHP, auxiliary boiler and cooling water unit. Excess heat from ethanol production provides heat to the DAC system **DAC operates 24 hr/day** and captures ~17.9 tonnes/hr CO_2, slightly lower than 20 tonne/hr due to sub-plot flue gas utilization during daytime. DAC CO_2 is captured and stored at night. A total of 40 tonnes/hr (DAC_{day} + DAC_{Stored} + flue gas CO_2) is then distributed to the PBRs during the day (total PBR demand over a 12-hour period). Sub-plots (8) consist of separate CHP and PBRs. See Fig. 4.
Option−24 2a DAC distributed to subplots	**Heat and mass integration.** Centralized EtOH processing (op 24 hr) equipped with CHP. Sub-plots (8) contain a liquefaction unit, PBRs, DAC, CHP, auxiliary boiler, and cooling water unit. In option 2a, **DAC operates 24 hr/day**. PBR utilizes flue gases from auxiliary boilers and subplot combined heat and power (CHP) units during the day. At night, DAC CO_2 is captured and stored, but flue gases go to the atmosphere. See Fig. 5.
Option−12 2b DAC distributed to subplots	**Heat and mass integration.** Centralized EtOH processing (op 24 hr) equipped with CHP. Sub-plots (8) contain PBRs, DAC, CHP, auxiliary boiler, and cooling water unit. In option 2b, **DAC operates 12 hr/day**. PBR utilizes flue gases from auxiliary boilers and subplot CHPs during the day. No sub-plot operation at night. See Fig. 5.
Option−24 2a (nth plant) DAC distributed to subplots	**Heat and mass integration.** Centralized EtOH processing (op 24 hr) equipped with CHP. Sub-plots (8) contain a liquefaction unit, PBRs, DAC, CHP, auxiliary boiler, and cooling water unit. In option 2a (n^{th}+ plant), **DAC operates 24 hr/day**. PBR utilizes flue gases from auxiliary boilers and subplot combined heat and power (CHP) units during the day. At night DAC CO_2 is captured and stored, but flue gases go to the atmosphere. N^{th} plant improvements entail increased adsorption efficiency, percentage steam bypass, and overall energy requirements, from the development of multiple iterations of existing technology. See Fig. 5.

feeds a CO_2 stream directly to the PBR, while a 24 hr/day configuration uses compressed CO_2 storage, a liquefaction system, at night. The operating capacity of the ethanol production facility requires the provision of 20 tonnes/hr of CO2 to produce 6116 kg/hr of ethanol product on a 24-hour basis. The DAC system in these non-integrated setups (baseline configurations) would need to operate at different capacities, with *BASELINE-12* operating at 40 tonnes/hr for 12 hours and *BASE-LINE-24* operating at 20 tonnes/hr for 24 hours. The baseline configurations have no heat and mass integrations to develop a basis of comparison between these and the integration improvements with the additional options.

The baseline configurations are separated into two segments within the overall facility: the *DAC system* and the *Ethanol Production and Recovery System*. The integrated options are separated into two different segments, namely *Centralized Facility* and *Modular Facility*. The

centralized facility operates on a single large-scale unit size, while the modular facility is segmented into eight subplots.

OPTION-24–1 (Fig. 4) is composed of a centralized facility that contains the main CHP and auxiliary boiler system, DAC system, CO2 compressed storage system, and ethanol processing system. *It also includes eight modular components* that contain the PBR system and smaller CHP systems. *OPTION-24 1* differs from *BASELINE-24* due to heat and mass integration. The excess heat from the ethanol production system is used to supply the DAC system. Additionally, the combustion gas from the modular facility CHP units is incorporated into the feedstock CO_2 delivered to the PBR systems. This heat and mass integration reduces the carbon capture and system heat required.

An improved model design, '*OPTION 2*' (Fig. 5), explores the placement of the DAC system and compression storage system within the modular facility. The centralized facility contains the ethanol production system and the external CHP that supplies heat and electricity. The modular facility in *OPTION 2* now includes the DAC and compression storage systems, in addition to the PBR that is present in scenario *OPTION 1* (Fig. 4). This design incorporates a larger CHP system within the modular facility, alongside an auxiliary boiler, to provide for the heat and power requirements of the DAC system. For the purposes of cost minimization, a scenario design only allows for the use of heat and mass integration within the sub-facility, i.e., the modular facility can be heat and mass integrated only within the modules. The *OPTION 1* configuration is thereby limited by the presence of a smaller CHP for mass integration within the modular facility. *OPTION 2* improves upon this limitation by configuring the DAC system and the larger CHP/auxiliary boiler system within the modular facility. The presence of a greater quantity of flue gas within the modular facility allows for a larger mass integration with the PBR system, which results in reducing the DAC operational capacity required to meet the total supply of 20 tonnes/hr of CO_2. Accordingly, *OPTION 2* reduces the DAC operational energy and greenhouse gas emissions compared to *OPTION 1*.

Option 2 is further split into two sub-categories, namely *OPTION-24 2 A* and *OPTION-12 2B* (see Fig. 5), with the time of operation being the only difference between the two. *OPTION-24 2 A* parallels the 24-hour DAC operation that is represented by *BASELINE-24*, while *OPTION-12 2B* parallels the 12-hour DAC operation delineated in the *BASELINE-12* assumption. Both these options have the same modular structure as described above, with the DAC and liquefaction storage system within the modules. The distinguishing feature is the varied time scale of DAC operation, which affects the life cycle greenhouse gas emissions from each option.

OPTION-24 2 A has a DAC unit that captures CO_2 for 24 hrs/day, with 12 hours of operation entailing direct feed to the PBR system and 12 hours of compressed storage. The main combined heat and power (CHP) system specific to the DAC operates for 24hrs/day, but the flue gas from the main CHP is only utilized for 12hrs/day since a dilute stream of CO_2 cannot be liquefied as the DAC output CO_2 can be. As a result, half the flue gas from the main CHP is used as feedstock for the PBR process, while the other half is emitted to the atmosphere. Additionally, *OPTION-24 2 A* benefits from operating the DAC at a lower capacity of 20 tonnes/hr, offsetting the greenhouse gas emissions expended by the loss of flue gas during the nighttime. On the other hand, *OPTION-12 2B* operates the DAC for 12 hrs/day, utilizing all the flue gas from the main CHP that results from powering the DAC operation. As the modular facility in *OPTION-12 2B* does not operate at night, the DAC system operates at twice the capacity (40 tonnes/hr) to account for the lost time at night. These modeling nuances highlight the importance of considering sub-categories A and B within *OPTION 2*.

OPTION-24 2 A nth+ has the same configurations as *OPTION-24 2 A* but has additional improvements to the technology, beyond the standard nth plant economics assumed for the entire assessment. The nth+ for this option denotes additional foreseeable improvements such as better adsorption efficiency (from 38.1 % to 45 %), a lower percentage steam bypass (from 50 % to 20 %), which thereby lowers the mass of steam

delivered to harvest (from 1.79 to 0.57 kg H_2O/kg CO_2), and lower energy required per tonne CO_2 capture (12.77–5.7 GJ/tonne).

4. Results

4.1. Cost

The results of the techno-economic analysis are shown in Table 6. The baseline unintegrated case (Baseline-12) utilizes no heat and mass integration, and the DAC system provides 100 % of the CO_2 required by the photobioreactors (20 tonnes/hr). The DAC system in the baseline scenario operates for 12 hours/day and captures 40 tonnes CO_2/operating hour. The minimum fuel selling price (MFSP) of ethanol calculated from the baseline case is $10.68/gal ethanol. Increasing the operating time from 12 hours/day to 24 hours/day (i.e., capturing 20 tonnes CO_2/operating hour), Baseline-24 leads to a 12.6 % improvement in the MFSP ($9.33/gal ethanol).

The heat and mass integration of DAC and PBR and the system integration configuration exhibit noticeable economic benefits. The MFSP and the percent improvement relative to Baseline-12 increase in the order: Options 24–1 ($9.10/gal; 14.8 %), Option 12–2b ($8.93/gal; 16.4 %), and Option 24–2a ($8.78/gal; 17.8 %). The integrated cases also significantly reduce the Fixed Capital Investment (FCI). With the projected advanced "nth+-plant" DAC performance (i.e., including the improved adsorption efficiency for CO_2 capture, improved % steam bypass, and improved overall energy requirement), the MFSP of $8.25/gal (a 22.8 % improvement compared to Baseline-12) for Option 24–2a can be achieved.

Reductions in cost are attributed to two primary process considerations. First, CO_2 storage at night significantly reduces the capital expenses associated with the direct air capture system. Direct air capture on-stream time is one of the largest contributors to MFSP reduction. Operating the DAC system for 24 hours and storing CO_2 overnight reduces capital expenses. The savings are more than enough to offset the additional capital expenses associated with CO_2 liquefaction and storage. Additional cost savings can be achieved through greater flue gas utilization. For example, the distributed DAC scenarios (2a and 2b) localize high-emitting processes, such as the boiler and DAC CHP unit, with the PBR system allowing more low-cost (flue gas) CO_2 to be utilized. This decreases the weighted average cost of CO_2 to the PBRs, thus decreasing the overall MFSP of ethanol (Table 5).

4.2. Life cycle inventory

4.2.1. Energy consumption

The on-site energy consumption from the natural gas used for heat and power in each configuration is summarized in Table 6. We highlight option 24–2a, which provides a 36 % reduction in energy consumption, and option 12–2b, which provides a 50 % reduction in energy consumption, compared to the respective baseline options. Option 2a (nth) shows a potential future energy savings of 52 % compared to the

Table 6
On-site Energy Consumption for Baseline and Integration Options (MJ/MJ EtOH).

MJ/MJ EtOH	DAC	PBR+EtOH	Total	Reduction (%)
Baseline−24	1.266	0.709	1.98	
Baseline−12	1.328	0.705	2.03	
Option−24 1	1.707	0.234	1.94	1.7
Option−24 2a	0.985	0.278	1.26	36
Option−24 2a (nth)	0.672	0.278	0.95	52
Option−12 2b	0.734	0.278	1.01	50

baseline, with future scale-up technological efficiency in adsorption and reduced DAC heat requirement.

4.2.2. Nitrogen oxides and particulates

We estimate NOx and PM2.5 particulate emissions, as these are among the best quantified of the non-greenhouse air pollutants.

The on-site emissions from the combustion of natural gas have been widely studied and reported [40,41]. For on-site emissions from natural gas combustion, we assume emissions factors of 5.5 g/GJ for NOx and 3.6 g/GJ for PM 2.5. For emissions from the supply chain of natural gas, we use Meng and Dillingham [25]. For fuel combustion, we use results from Huo et al. [12] and Iodice et al. [16]. The life cycle impact of the production of polyethyleneimine (PEI) has been modeled by Wu et al. [44] and Barjoveanu et al. [4]. These studies have results of the same order of magnitude; we use the average. Emissions from the periodic calcination of the PEI are not estimated due to lack of data, which would contribute additional emissions.

As is shown in Tables 7 and 8, using the available data, PEI production is the largest source of NOx and particulate emissions for all scenarios. As there has been limited work on the lifecycle emissions of PEI, we consider these results to be a preliminary indication that PEI production may be a hotspot for some air pollutant emissions; further research is needed to confirm the PEI lifecycle emissions inventories.

4.3. Greenhouse gas emissions

The life cycle greenhouse gas emissions are summarized in Table 9 and Fig. 6. These emissions include several components. The largest component of the emissions is the sourcing and combustion of natural gas, which provides all the on-site energy for the DAC and biofuel system. The first row of data shows the total CO_2 emitted by the on-site natural gas combustion. The next row shows that some of this CO_2 is absorbed in the integrated scenarios; this is shown as a negative value. The third data row shows the CO_2 absorbed from the air by the DAC system; these values are also negative because the DAC system is removing CO_2 from the air. The fourth data row is the CO_2 that is vented from the PBRs through their normal operation. The fifth row is the CO_2 that is emitted when the ethanol fuel is combusted in a vehicle. The sixth row is the upstream leakage of methane during its extraction and transport to the site. The ± values in the table represent the combination

Table 5
Economic Summary for Baseline and Integration Options.

	Baseline-12	Baseline-24	Option-24 1	Option-24 2a	Option-24 2a nth	Option-12 2b
MFSP ($/gal EtOH)	$10.68	$9.33	$9.10	$8.78	$8.25	$8.93
% MFSP reduction	-	12.60 %	14.80 %	17.80 %	22.8 %%	16.40 %
EtOH annual production (MMGal/yr)	16	16	16	16	16	16
FCI (MM$)	860.5	724.1	695.2	694.6	654.3	719.9
Total operating costs (MM$/yr)	53.6	51.5	51.8	47.2	44.8	45.7
CO_2 from DAC (tonne/hr)	40	20	17.9	12.9	14.9	18.9
DAC operating hours (hr/day)	12	24	24	24	24	12
Percent of total CO_2 demand from DAC	100 %	100 %	90 %	64 %	75 %	47 %
DAC CO_2 cost ($/tonne)	$407	$275	$258	$353		$585
Weighted CO_2 cost ($/tonne)*	$407	$275	$232	$226		$275

* Including DAC and flue-gas sourced CO_2.

Table 7
Estimated NOx emissions for baseline and integration options (g NOx/MJ EtOH).

g NOx/MJ EtOH	Baseline-12	Baseline-24	Option-24 1	Option-24 2a	Option-24 2a nth	Option-12 2b
NG combustion	0.01	0.01	0.01	0.01	0.01	0.01
NG supply	0.17	0.17	0.16	0.11	0.08	0.08
PEI synthesis	1.53	1.53	1.37	0.98	1.14	0.72
Fuel combustion	0.02	0.02	0.02	0.02	0.02	0.02
Total	1.72	1.73	1.56	1.12	1.25	0.83

Table 8
Estimated particulate emissions for baseline and integration options (g PM2.5/MJ EtOH).

g PM2.5/MJ EtOH	Baseline-12	Baseline-24	Option-24 1	Option-24 2a	Option-24 2a nth	Option-12 2b
NG combustion	0.007	0.007	0.007	0.005	0.003	0.004
NG supply	0.0005	0.0006	0.0005	0.0003	0.0003	0.0003
PEI synthesis	0.125	0.125	0.112	0.080	0.093	0.059
Fuel combustion	0.003	0.003	0.003	0.003	0.003	0.003
Total	0.136	0.136	0.123	0.089	0.100	0.066

Table 9
Summary of Greenhouse Gas Emissions for Selected Options (100-yr time horizon).

Lifecycle GHG Emissions (kg CO2e/kg EtOH)	Baseline-12	Baseline-24	Option-24 1	Option-24 2a	Option-24 2a nth	Option-12 2b
Natural Gas Combustion	2.21	2.23	2.12	1.40	0.92	1.29
Natural Gas CO_2 Absorbed	-	-	(0.33)	(1.16)	(0.82)	(1.73)
DAC Absorbed	(3.27)	(3.27)	(2.93)	(2.11)	(2.45)	(1.55)
CO2 from PBRs	0.49	0.49	0.49	0.49	0.49	0.48
EtOH Combustion in Vehicle	1.91	1.91	1.91	1.91	1.91	1.91
NG upstream life cycle	0.23±0.17	0.24±0.18	0.22±0.17	0.15±0.11	0.10±0.07	0.14±0.10
Ancillary	0.28	0.28	0.28	0.27	0.28	0.27
Total (kg CO2e/kg EtOH)	1.86±0.17	1.88±0.18	1.77±0.17	0.95±0.11	0.42±0.07	0.81±0.10
Total (g CO2e/MJ EtOH)	68.9±6.4	69.7±6.5	65.4±6.2	35.1±4.1	15.7±2.7	30.2±3.8

of the 37 % uncertainty in the radiative forcing impact of methane relative to carbon dioxide over 100 years and the 37 % uncertainty in the supply chain methane leaks. Ancillary emissions include the CO_2 emissions from the production of the DAC system monoliths, the production of the PBR bags, and the site preparation. There are uncertainties in the modeling of the operation of the direct air capture system, the biofuel production system, and their integration; these are not quantified here.

BASELINE-24, BASELINE-12, and *OPTION-24 1* encompass minimal integration and benefits obtained, with overall GHG emissions of 68, 69, and 65 gCO2e/MJ, respectively. In comparison, the EPA 2005 baseline gasoline carbon intensity is 93 gCO2e/MJ. *OPTION 2* improves upon the first three options, incorporating significant mass integrations that reduce the overall emissions by about 50 % to 35 gCO2e/MJ (in *OPTION-24 2 A)* and 30 gCO2e/MJ (in *OPTION-12 2B*). The mass integration of flue gas incorporates a secondary CO_2 source that significantly reduces the DAC operating capacity, thereby reducing the energy expended from carbon capture.

While the baseline options meet the CO_2 demand entirely through direct air capture, *OPTION-24 2 A* and *-12 2B* utilize the DAC to meet 64 % and 47 % of the total demand, respectively. The nth plant option (*OPTION-24 2 A nth*) reduces GHG emissions by 78 % from that of the non-integrated options and by a further 56 % from the original *OPTION-24 2 A*. As discussed previously, the improvements foreseen in scale-up technological efficiency in adsorption, as well as the reduced heat requirement of direct air capture, bring down the overall greenhouse gas impact of the extended n^{th} plant assumption. The US Renewable Fuel Standard requires that all advanced biofuels must achieve a 50 % reduction from the base petroleum equivalent (i.e., must achieve 45.7 gCO2e/MJ or less). *OPTION-24 2 A* qualifies, OPTION-12 2B does even a bit better, and *OPTION-24 2 A nth* has emissions well below the maximum allowed emissions to qualify. Further integrations and

technological developments can be foreseen.

The main greenhouse gas emitted from this system is carbon dioxide. The second largest contributor to the greenhouse gas emissions is methane; this is primarily from the supply chain leakage of methane in the natural gas production and delivery system. In addition, there may be some small emissions of N_2O from the photobioreactor system; we have estimated these as in Arora et al. [1]. Table 9 and Fig. 6 show greenhouse gas emission results in the 100-year time horizon. This is the most used time horizon; it provides a combined metric of the contribution of methane and carbon dioxide to provide an integrated measure of climate impact. The 20-year time horizon provides a measure of the climate impact over the next twenty years; methane carries more weight in this time horizon. Those results are shown in the supporting information.

5. Discussion

Direct air capture on-stream time is one of the most significant contributors to cost reduction. Operating the DAC system for 24 hours and storing CO_2 overnight reduced capital expenses; the decrease in DAC capital costs considerably offset liquefaction and storage costs. Additional cost savings were achieved with greater flue gas utilization. Options 2a and 2b localize high-emitting sources within the PBR system, reducing the demand for CO_2 from the DAC system while still providing 20 % of total CO_2 from the DAC system. This decreases the weighted average cost of CO_2 to the PBRs, thus decreasing the overall cost of ethanol.

The combined heat and power (CHP) system modeled here uses natural gas. Systems using low-emission fuels and waste heat [8,38] may have somewhat different process integration results. The most cost-effective and CO_2-efficient design found here has a natural gas combined heat and power (CHP) system powering the ethanol
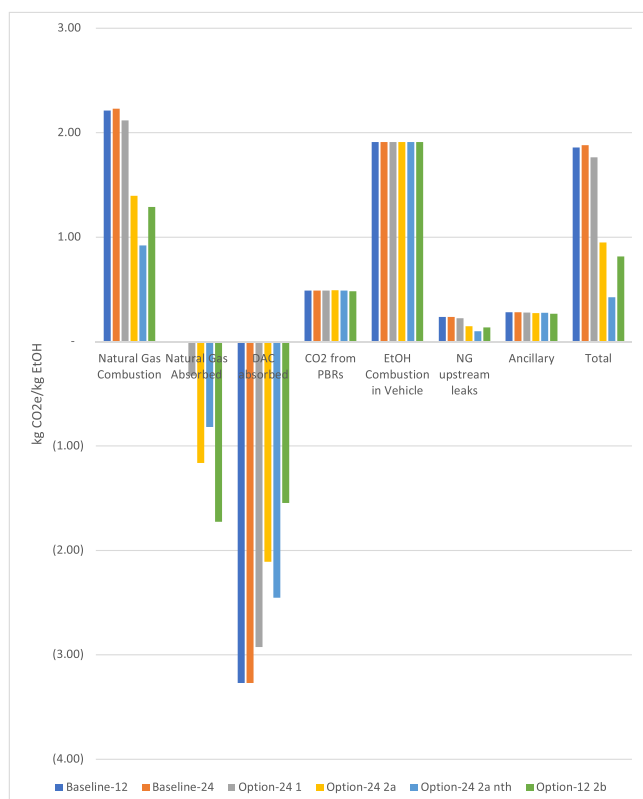
Fig. 6. Life cycle greenhouse gas emissions (kg CO_2e/kg ethanol, 100-year time horizon) for the baseline and integrated options for production of algal biofuel via direct air capture of carbon dioxide.

production and modular natural gas CHP systems in each sub-plot of the field of photobioreactors. Investigating the costs and benefits of using grid electricity or solar power either in the PBR sub-plots, or in the ethanol processing area, could provide additional insights on integration. In this regard, another aspect of the utilization of low-cost or waste heat is that it is a limited resource. Considering a partial application of waste heat, along with either electrification or some use of natural gas, could reveal pathways with both cost reduction and greater efficiency in carbon reduction.

Heat and mass integration of DAC and PBR is critical to reducing cost. The integration of DAC with the PBR and ethanol recovery processes poses several potential benefits. The heat integration can decrease operating expenses by reducing natural gas demand for heating requirements. Additional benefits can be gained from mass integration. The utilization of flue gas CO_2 as a carbon source for the PBRs decreases the DAC system's scale, resulting in both capital and operating cost savings.

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Reductions in cost (MFSP) are attributed to two primary process considerations: a) CO_2 storage at night reduces the capital expenses associated with DAC (i.e., increasing on-stream time), and b) distributed DAC scenarios (DAC-PBR integration Options 2a and 2b) make use of boiler and DAC CHP flue gas CO_2 (free).

This study evaluated the integration of direct air capture with algal biofuel production. The resulting minimum biofuel selling price, $8.25

per gallons of ethanol, equivalent to nearly $13/GGE. As discussed earlier, strategies for cost effective algal biofuel production involve co-production of fuel with other products. A US DOE assessment finds significant potential for cost reduction via co-production of protein and fuel from microalgae [30]. As the focus of this study is on integration of DAC with CO2 utilization, we did not include co-product analysis. By including co-production of protein or other products with fuel production, and evaluation of multiple integrated system designs as illustrated here, additional cost reductions may be found.

The design integrated of DAC with CO_2 could be developed for other applications. The use of CO_2 to grow crops in greenhouse is widespread; typical CO_2 sources include the combustion of kerosene and natural gas, and in some cases, CO_2 from industrial sources. Reinoso Moreno et al. [33] have evaluated the use of carbon capture using activated carbon for a greenhouse; they consider design aspects such as the varying requirements at night versus day and the potential combination with heating requirements. Further work for applications to greenhouses could, as illustrated here, evaluate multiple designs to identify options to reduce costs and environmental impacts. Monteiro and Roussanaly [26] have evaluated the breakeven costs of carbon dioxide utilization in a number of applications, including greenhouse agriculture, emphasizing the need for integration. Further work may identify opportunities for efficiency in linking the production of heat, electricity, and CO_2, as well as the benefits of local air sourcing of CO_2 to avoid transportation costs.

6. Conclusion

Large-scale removal of carbon dioxide from the air can help stabilize the climate. Direct air capture is a leading carbon dioxide removal technology, but its cost is currently too high for deployment at scale. Previous and ongoing research rightly focuses on improving the technology of DAC, and on understanding how costs may fall if DAC were deployed at large scale. This work brings forward an additional approach to reducing the costs of DAC: by co-designing DAC with the system that will use the CO_2, integrating process heat, CO_2 management, and the spatial and temporal arrangements of processes, there are opportunities to bring down costs and emissions.

The integration of carbon dioxide capture within the biofuel production facility reduces costs and greenhouse gas emissions significantly. The use of flue gas as a secondary feedstock enables the system to operate at a lower DAC capacity. With the improvement in carbon integration and operational efficiency within the facility, there are opportunities for further cost and greenhouse gas emission reductions. Specifically, in the DAC algal biofuel system studied here, we find that heat and mass are most effectively integrated using a modular approach, with a direct air capture system in each subplot of photobioreactors. With this modularization, we find additional savings by including a CO_2 liquefaction system with each DAC module, allowing the DAC system to run continuously and store CO_2 at night for use in algae cultivation during the day. With this fully modular design, mass and heat integration provide an 18 % cost reduction and a 50 % greenhouse gas emission reduction compared to the baseline design in which the DAC system supplies only CO_2 to the photobioreactor biofuel system. The amount of cost reduction (18 %) is smaller than the 50 % or more cost reduction that would make DAC a truly cost-effective technology. Yet the reduction of 18 % is a significant advance and notably is achieved with today's level of technology.

Moving forward, additional work is needed to find cost-effective approaches to capturing carbon dioxide from air and storing or utilizing it to provide substantial reductions in atmospheric carbon dioxide concentrations. Considering applications of direct air captured CO_2 for the production of biofuel, many of the general features of the integration are likely to remain the same even for different algal species or fuel systems. Key differences that could be considered within the algal biofuel application are to source electricity from renewable electricity, and to source process heat from an industrial or natural source.

CRediT authorship contribution statement

Shavonn D'Souza: Formal analysis, Data curation. **Jaden Johnston**: Software, Formal analysis. **Valerie M. Thomas**: Writing – original draft, Supervision, Methodology, Conceptualization. **Kylee Harris:** Methodology, Formal analysis. **Eric C. D. Tan**: Writing – original draft, Supervision, Methodology, Conceptualization. **Ronald R. Chance:** Writing – review & editing, Conceptualization. **Yanyui Yuan**: Methodology, Investigation, Data curation.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Valerie M. Thomas, Kylee Harris, Jaden Johnson, Shavonn D'Souza, Eric Tan reports financial support was provided by US Department of Energy. R. R. Chance and Yanhui Yuan reports a relationship with Global Thermostat that includes: employment. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jcou.2024.102911.

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