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Full Length Article

One- or two-step processes: Which have a lower GHG emissions intensity for production of synthetic aviation fuel via indirect CO_2 electrolysis?

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ABSTRACT

The development of sustainable aviation fuel (SAF) could pave the way towards addressing the dual challenges faced by the aviation sector: meeting rising demand for air transport and achieving net-zero targets. In this study, the well-to-pump (WtP) and well-to-wake (WtW) greenhouse gas (GHG) emissions intensity (EI) of aviation fuel production via four CO_2-indirect pathways (intermediate products are required) is estimated and the WtW GHG EI compared to conventional fossil-based and bio-ethanol pathways. We aim to determine whether a one- or two-step electrochemical conversion is more likely to result in lower GHG intensity aviation fuel, under what conditions pathways incorporating these electrochemical processes have a lower GHG EI than conventional crude oil-based and biomass-based jet fuels, and whether these CO_2-derived sustainable aviation fuel (CO_2-SAF) pathways can approach "carbon neutrality." The key findings from this work are: (1) processes using ethylene as an intermediate tend to have a lower GHG EI, although there is not a meaningful difference between one- and two-step pathways; (2) all pathways could achieve a lower GHG EI than fossil and biomass based routes if the location is carefully selected to minimize the GHG EI of electricity supply and if the CO_2 source is strategically chosen; and (3) while these pathways have the potential to approach zero GHG emissions, emissions from fuel manufacturing will be challenging to eliminate entirely. Notably, the GHG EI of CO_2-based SAF is far more sensitive to background system parameters, such as the carbon intensity of electricity and CO_2 supply, than to technical parameters. Therefore, we suggest that background factors may play a greater role in determining GHG EI than technical innovation.

1. Introduction

The increasing concentration of carbon dioxide (CO_2) in the atmosphere, resulting largely from the combustion of fossil fuels, is the primary driver of climate change (IPCC 2021). According to the Intergovernmental Panel on Climate Change, global CO_2 emissions must be rapidly reduced to zero over the long-term to limit the global temperature increase to 1.5 °C (IPCC 2021). While a relatively small contributor to global energy-related greenhouse gas (GHG) emissions (2 % in 2022, 800 Mt CO_2), emissions from the aviation sector have grown rapidly due to the rising global demand for passenger travel and freight transport (IEA 2021; IEA 2023). As reported by the International Air Transport Association, air passenger continue to grow worldwide and have approached pre-pandemic levels of over 4.3 billion trips in 2018 and are expected to double in the next two decades (IATA 2024). The aviation industry is aiming to cut its GHG emissions by 50 % by 2050

Abbreviations: ATJ, Alcohol to jet fuel; BAF, Biomass-based aviation fuel in general; BEAF, Bioethanol-based aviation fuels; CCU, Carbon capture and utilization; CO_2-SAF, CO_2 conversion-based aviation fuel; DAC, Direct air capture; FAF, Fossil-based aviation fuel; FE, Faradaic efficiency; FT, Fischer-Tropsch; GHG, Greenhouse gas; GREET, Greenhouse Gases, Regulated Emissions, and Energy use in Technologies; GWP, Global warming potential; GHG EI, Greenhouse gas emission intensity; LCA, Life cycle assessment; PCC, Post-combustion capture; PSA, Pressure Swing Adsorption; RWGS, Reverse water gas shift; SAF, Sustainable aviation fuel; WtP, Well-to-pump; WtW, Well-to-wake.

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compared to its 2005 emissions level (IATA 2024), but hydrocarbon-based aviation fuel cannot be easily replaced by electric power or hydrogen for long-haul aviation (Bergero et al., 2023). Instead, Dray et al. conclude that synthetic aviation fuels from green hydrogen and atmospheric CO_2 could lead the aviation industry towards net-zero (Dray et al., 2022), and some scenarios (e.g., International Energy Agency Net Zero Emissions scenario), feature an increasing demand for such fuels (IEA 2022). Thus, developing energy-efficient technologies to produce sustainable aviation fuel (SAF) is one way to address CO_2 emissions from a growing aviation industry (Morganti et al., 2024, Yoo et al., 2022).

The main component of aviation fuel is kerosene, which are linear and branched alkanes and cycloalkanes with a carbon chain length from C_8 to C_{16} (Kallio et al., 2014). Most conventional kerosene is a "straight-run" product, obtained from fractionation and mild hydrotreating (to remove sulfur and metals) of crude oil, and some may also come from hydrocracking or catalytic cracking of heavier fractions (Liu et al., 2013). In recent years, driven by the challenges of energy security and greenhouse gas emissions, efforts have been made to commercialize biomass-based substitutes for kerosene (Bergero et al., 2023). Today, SAF certified for use in aviation is produced from a range of biomass feedstocks and associated conversion pathways (Wei et al., 2019).

Recently, literature has suggested that CO_2 captured from industrial facilities (e.g., power generation, cement manufacturing) or removed from the atmosphere (Hepburn et al., 2019, Nishikawa et al., 2023) could be converted into aviation fuel. Conversion of CO_2 to aviation fuel can be achieved through both direct and indirect routes (Colelli et al., 2023). The direct route is based on the reverse water gas shift (RWGS) reaction to reduce CO_2 to CO and Fischer-Tropsch (FT) synthesis to convert CO to long-chain hydrocarbons with the help of hydrogenation and catalysts (Yao et al., 2020). In the indirect route, CO_2 is converted to intermediate products such as CO or methanol and subsequently to kerosene through a series of chemical reactions, such as in mature alcohol-to-jet (ATJ) fuel routes (Yao et al., 2020). The indirect route is attractive if coupled with electrochemical conversion, in which there has been rapid progress in one-step (CO_2→products) and two-step (CO_2→CO→products) conversion of CO_2 to intermediate products (Nabil et al., 2021). The electrochemical route offers multiple potential benefits, including cost reductions through large-scale manufacturing of modular plants, the ability to produce at small-scales and in remote locations, and to store variable renewable electricity (e.g., wind or solar energy) in the form of energy dense hydrocarbons (Segets et al., 2023). Additionally, this approach enables the co-production of hydrogen that can be used downstream in SAF production and further enhances its sustainability (Pipitone et al., 2023, Sadhukhan and Sen, 2021). However, electrochemical CO_2 conversion to chemical products is at a low technology readiness level (TRL 2–4) and tends to have low product selectivity (Grim et al., 2022). In this study, we refer to fossil-based aviation fuel as FAF, bioethanol-based aviation fuels as BEAF, and CO_2 conversion-based aviation fuel as CO_2-SAF.

Life cycle assessment (LCA) is an approach that is increasingly used to quantitatively and holistically evaluate the environmental impacts of a product or process over its full life cycle (Bergerson et al., 2020; Bergerson et al., 2019; ISO14040 2006). The LCA methodology is also frequently applied to more narrowly compare the climate change impact of different production systems and products. As such, LCA can provide insights for developers of electrochemical CO_2 conversion processes that could be used to reduce the GHG emissions intensity (GHG EI) of their products (Nabil et al., 2021; Nishikawa et al., 2023; Sternberg et al., 2017). Many recent studies have focused on the climate impact of aviation fuel production from varied processes sources of biomass, comparing them to the conventional pathway (Capaz et al., 2018; Geleynse et al., 2018; de Jong et al., 2017; Han et al., 2017; Li et al., 2019; Morganti et al., 2024; Obnamia et al., 2020; Ringsred et al., 2021; van der Giesen et al., 2014; Yoo et al., 2022). Some studies have also examined the feasibility of producing aviation fuel from bioethanol via commercial scale technologies, including alcohol dehydration, olefin oligomerization and hydrogenation (Cooney et al., 2017; Han et al., 2017, Geleynse et al., 2018; Lee et al., 2021). Other research has utilized the LCA method to evaluate climate impact of hydrocarbon fuels produced both from post-combustion capture (PCC) (Nabil et al., 2021; Liu et al., 2020) and direct air capture (DAC) (McQueen et al., 2021; Mohsin et al., 2020; Vunnava and Singh, 2021), concluding that the results are highly dependent on system configurations and technological routes (Ballal et al., 2023; Grim et al., 2022).

In this work, we seek to answer three research questions that are natural extensions of past work evaluating the climate change impact of CO_2-SAF production: (1) Is a one-step or two-step electrochemical conversion likely to be the more efficient process to convert CO_2 into jet fuel? And, if so, by which intermediate: ethanol or ethylene? (2) Under what conditions would pathways containing these electrochemical processes have a lower GHG EI than conventional crude oil-based and biomass-based jet fuels? (3) What is the lowest GHG EI that we could reasonably expect from the CO_2-SAF pathways described here?

To address these questions, we estimate the well-to-wake (i.e., cradle-to-grave) GHG EI of aviation fuel produced from CO_2 indirect conversion routes and compare the results with the biofuel and incumbent conventional crude oil pathways. We consider eight indirect pathways based on two CO_2 electrolysis routes using (1) ethanol or (2) ethylene as the intermediate products, as both can be derived from CO_2 electrolysis (Nabil et al., 2021), where CO_2 is captured from either flue gas or atmosphere. The intermediate products are selected because of the possibility of subsequent conversion at the commercial scale (Geleynse et al., 2018; Grim et al., 2022; Han et al., 2017). The CO_2 capture process is included in the system by using a GHG EI of CO_2 supply. Key variables influencing the resulting GHG EI are identified through a sensitivity analysis, and recommendations for future technology improvements in the industry are proposed accordingly. We anticipate that the findings of this study will guide technology development for CO_2-SAF and contribute to the broader discussion on technology and policy strategies for achieving net-zero goals in the aviation sector.

This paper is structured as follows: In Section 2, we describe the technologies we model in more detail, define the systems in which they are incorporated, and our choices for system boundaries and functional units; in Section 3, we discuss the results from the perspective of four plausible, near-term futures; identify the parameters that most strongly influence the results; and the parameters that could be adjusted to minimize GHG EI of the resulting fuels.

2. Methods

In this work, we seek to assess the life cycle GHG EI of early-stage (or emerging) technologies. As such, we follow the general approach to LCA outlined in ISO 14,040/44 but develop an inventory based on extrapolation of BEAF and CO_2-SAF technology performance from its current level of technological maturity to commercial scale focused on GHG emissions. We vary the parameters characterizing technology performance to explore the impact of uncertainty on the resulting GHG EI. Furthermore, we assess the GHG EI of these technologies in the context of a future energy system (i.e., background system) that could reasonably be expected to exist in the near-to-medium term future if we are to achieve net-zero targets.

2.1. Product system and boundaries

Fig. 1 shows the CO_2-SAF production system is composed of three major components: a CO_2 supply system, intermediate product production using CO_2 electrolysis, and jet fuel production. The different CO_2 supply options—i.e., from the atmosphere (using DAC) and industrial point sources (using post-combustion capture, PCC)—and intermediate products—i.e., ethanol and ethylene—are treated as being

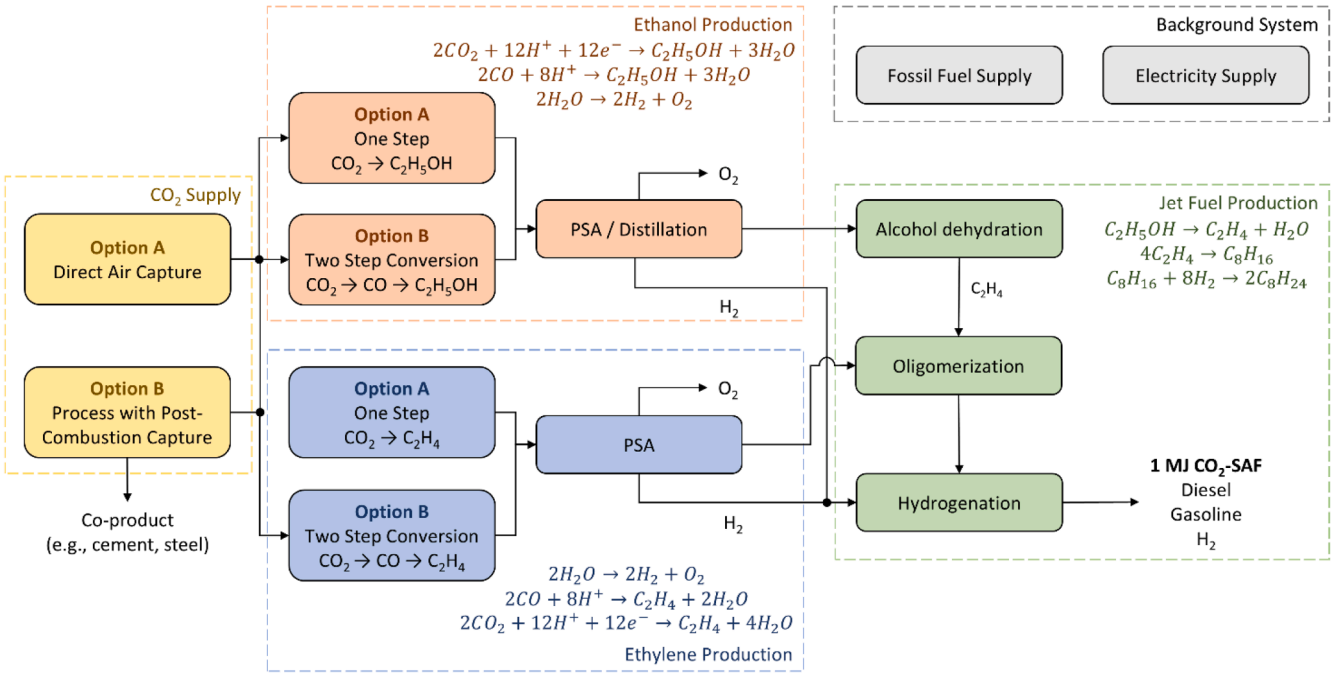

Fig. 1. CO_2-SAF product system with ethanol or ethylene as intermediate products, showing all the processes included as part of the WtP life cycle.

mutually exclusive, which results in eight different possible production systems. Each of the relevant unit processes shown in Fig. 1 are for a particular production system are included within the well-to-pump (WtP) system boundary. Transportation of feedstocks and products between each unit process is not considered for all cases in this study due to its expected small contribution to the overall life cycle (as is the case for FAF). This figure shows multiple points in the life cycle where different products are being produced from the same unit process, making this a multi-functional system.

We explore the impact of DAC and PCC as possible CO_2 supply options, as they have different energy requirements and are treated differently in LCA. In comparison to PCC, a DAC system requires two- to three-times more energy to capture one ton of CO_2 due to the extremely low CO_2 concentration in the air (~426 ppm in 2025) (McQueen et al., 2021, 2020; Vaccarelli et al., 2014; Wang and Song, 2020). On the other hand, the sole function of DAC is to remove CO_2 from the atmosphere whereas PCC reduces emissions of CO_2 from fossil fuel combustion (or calcination of minerals) during production of a co-product (e.g., electricity, cement). Where CO_2 from PCC is geologically sequestered it is clearly a waste; while supplying it as a feedstock to a CO_2 conversion process makes it a product and quantification of its specific burden requires allocation or substitution (see Section 2.2).

A CO_2 electrolysis unit converts the captured CO_2 to an intermediate product through electrochemical reactions (Nabil et al., 2021). Electrolysis of CO_2 to a wide range of products has been demonstrated under ambient conditions (Nabil et al., 2021). Here, we chose ethanol and ethylene as the intermediate products for SAF synthesis. Electrochemical ethylene production is more developed than electrochemical ethanol production (Nabil et al., 2021; Wang et al., 2021), and producing aviation fuel from ethanol requires one more reaction step than from ethylene. However, because ethanol is produced in large amounts at low cost from biomass today (Dagle et al., 2020; Posen et al., 2015), many mature synthetic fuel technologies have been developed to use ethanol as a feedstock (Kallio et al., 2014).

Within both ethanol and ethylene-based pathways, we considered the one-step and two-step electrochemical reactions. The one-step reaction directly converts CO_2 to the desired products while the two-step reaction converts CO_2 to an intermediate product CO, and then CO to the desired products. The CO_2 electrolysis process generates hydrogen and oxygen as co-products, which is the second unit process with multi-functionality in the system. The half-cell reactions for both one-step and two-step conversions are presented as Eqs. (1) and (2), and Eqs. (3), (4), and (5), respectively.

One-Step Conversion:

$$Ethanol: \quad 2CO_2 + 12H^+ + 12e^- \rightarrow C_2H_5OH + 3H_2O \tag{1}$$

$$Ethylene: \quad 2CO_2 + 12H^+ + 12e^- \rightarrow C_2H_4 + 4H_2O \tag{2}$$

Two-Step Conversion:

$$Step1 - To\ CO: \quad 2CO_2 + 2H^+ + 2e^- \rightarrow CO + H_2O \tag{3}$$

$$Step\ 2 - To\ Ethanol: \quad 2CO + 8H^+ + 8e^- \rightarrow C_2H_5OH + H_2O \tag{4}$$

$$To\ Ethylene: \quad 2CO + 8H^+ + 8e^- \rightarrow C_2H_4 + 2H_2O \tag{5}$$

Conversion of ethylene and ethanol to aviation fuel requires two and three chemical steps, respectively. For an ethanol feedstock process, the three catalytic reactions are ethanol dehydration, olefin oligomerization, and hydrogenation (Geleynse et al., 2018; He et al., 2019; LiBretto et al., 2021; Van-Dal and Bouallou, 2013; Wei et al., 2016, 2017, 2018). An ethylene feedstock process only requires olefin oligomerization and hydrogenation. In both cases, the major constituent of the resulting product is aviation fuel, with diesel and gasoline as the co-products (Geleynse et al., 2018; Han et al., 2017). This is the third multifunctional unit process in the system. The processes to convert ethylene and ethanol to fuels are mature commercial technologies that have been successfully used in refining and synthetic fuels production for decades and are being applied to production of aviation fuel from ethanol today (LanzaJet 2022).

The incumbent pathway to producing FAF is shown in Fig. 2(a). This involves the extraction of conventional crude oil and refining and processing of the crude oil to jet fuel. The bioethanol-based pathway, shown in Fig. 2(b), starts with growing and harvesting of corn in the US Midwest, transport of the corn to an ethanol (dry) mill, production of ethanol and conversion of the ethanol to aviation fuel using the same processes as described in Fig. 1 for CO_2-based ethanol.

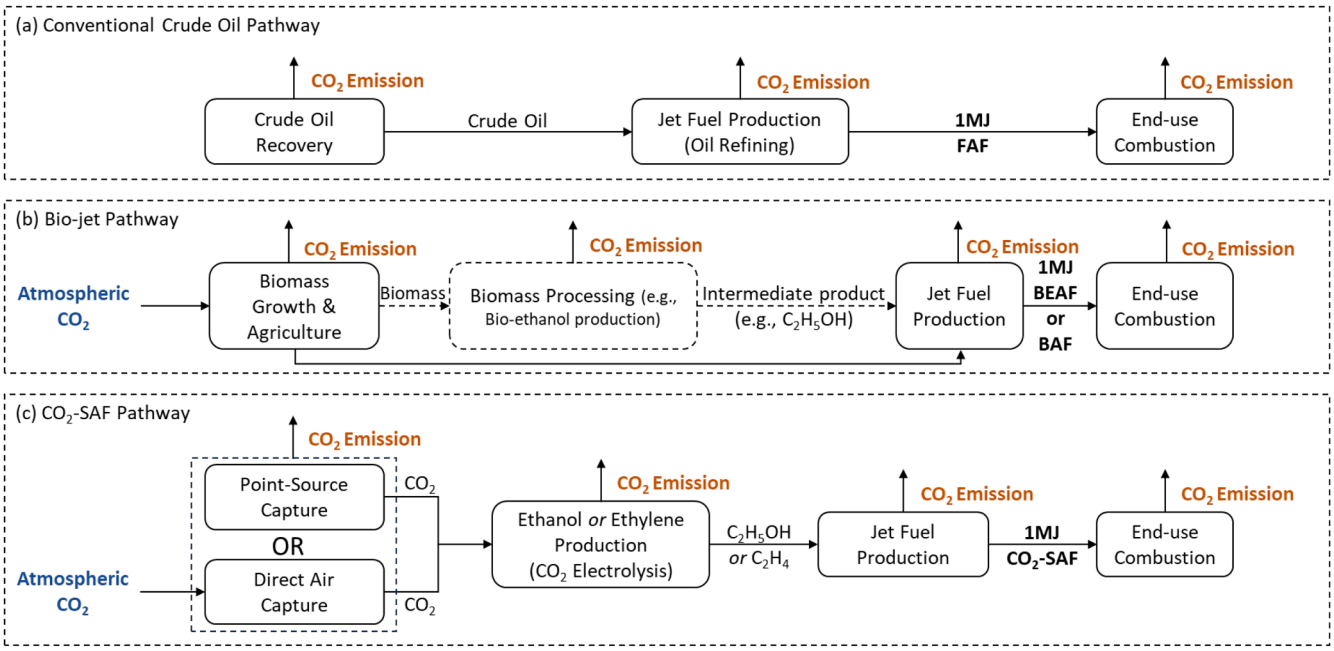

Fig. 2. Pathways (a) FAF and (b) BEAF represent mature pathways to production of aviation fuels and are used as comparisons to the (c) proposed CO_2-SAF pathways.

2.2. Functional unit, impact categories and treatment of multifunctionality

To compare the GHG EI of producing CO_2-SAF with conventional aviation fuel production from crude oil and renewable pathways from biomass, we choose 1 MJ (lower heating value, LHV) producing CO_2-SAF as the functional unit. The function of an aviation fuel is to provide propulsive energy, and this unit allows for fair comparisons between liquid hydrocarbon fuels. Caution should, however, be exercised in comparisons to other fuels, such as hydrogen, as volumetric energy density is a particularly important factor in aviation. We convert GHG emissions to midpoint climate change impacts using 100-year global warming potential (GWP) values provided in the IPCC 6th Assessment Report (IPCC 2022), and report the GHG EI in CO_2-equivalent terms (e.g., gCO_2eq).

Where a unit process generates co-products, and the process cannot be further subdivided, there are two methods that may be employed to isolate the environmental impacts associated with a single product from the system. These are system expansion and substitution (sometimes referred to only as substitution) and allocation (ISO14040 2006, ISO14044 2006, Klöpffer and Grahl, 2014). In the system expansion and substitution approach, we examine the co-products and identify the product for which a change in demand would affect the overall production rate—known as the determining product (e.g., jet fuel). Where the environmental impacts of the determining product are of interest, we can generally subtract the environmental burdens of a relevant production process for the other co-product (known as the dependent product) (Weidema, 2000). In contrast, in allocation, the environmental impacts for the process are shared between the co-products in proportion to a chosen physical attribute (e.g., mass, volume, energy content) or their economic value. We adopted the system expansion and substitution approach for CO_2 supply from PCC (as recommended by Müller et al. (2020)) and the H_2 and O_2 co-products of CO_2 electrolysis (we assumed that O_2 is vented, as it is not an energy product). On the other hand, we adopt energy allocation to address the gasoline and diesel co-products of the final fuel production step, which is typically applied for refinery production.

2.3. Inventory and assumptions

We have derived the mass and energy balance for each of the eight CO_2-SAF pathways shown in Fig. 1 from previous studies (see SI for more details) (Geleynse et al., 2018; Nabil et al., 2021; Müller et al., 2020) and use them as the basis for the life cycle inventory for the study. The key assumptions for this study are presented in Table 1. The GHG footprint of equipment installation such as CO_2 electrolyser, the capture, separation units, is not included in this analysis. In addition, the GHG EI of

Table 1
Overview of the assumptions utilized in this study. Avoided burdens refers to the values used in substitution.

Parameters	Range	Base Value
System Technological Parameters		
Faradaic efficiency of CO_2 electrolysis (Nabil et al., 2021)	0.5 to 0.9	0.7
Dehydration process yield (Geleynse et al., 2018; Han et al., 2017)	0.7 to 0.95	0.86
Oligomerization process yield (Geleynse et al., 2018; Han et al., 2017)	0.7 to 0.95	0.86
Hydrogenation process yield (Geleynse et al., 2018; Han et al., 2017)	0.5 to 0.8	0.67
Pressure Swing Adsorption (PSA) Energy [kWh/m^3] (Nabil et al., 2021)	0.25	0.25
Background Parameters		
Average GHG EI of electricity [gCO_2eq/kWh] (IEA 2019, EIA 2021)	10 to 600	100
Average electricity consumption of water supply [kWh/ton] (Griffiths-Sattenspiel and Wilson, 2009)	0.08 to 0.15	0.11
Average carbon intensity of thermal energy [gCO_2eq/MJ] (USEIA 2021)	15 to 90	50
Energy Intensity, LHV		
Jet fuel [MJ/kg] (IEA 2020, Song, 2020)		42.8
Hydrogen [MJ/kg] (IEA 2020, Song, 2020)		120
Diesel [MJ/kg] (IEA 2020; Song, 2020)		45.5
Gasoline [MJ/kg] (IEA 2020; Song, 2020)		46
Avoided Burdens		
Feedstock CO_2 [$gCO_2eq/kg\ CO_2$] (Müller et al., 2020)	950 to 590	800
H_2 Production (water electrolysis) [kWh/kg] (Bertuccioli et al., 2014)	50 to 55	50

FAF and BEAF were calculated using the Greenhouse Gases, Regulated Emissions, and Energy use in Technologies (GREET) model, an LCA tool developed by Argonne National Laboratory (Argonne National Laboratory, 2023).

Because CO_2 is a feedstock to the process, we treat it as a product flow. As such, we include the elementary flows (and associated impacts) associated with its production in the life cycle (as shown in Fig. 1). We assume the flue gas capture system removes 90 % of the CO_2 in flue gas, a level that has been commercially achieved (Sleep et al., 2021; Wang and Song, 2020). We conservatively assume that CO_2 capture results in emission of 200 gCO_2eq/kg CO_2 supplied (the average of CO_2 capture from flue gas) and, following the system expansion and substitution approach described by Müller et al. (2020), results in a GHG EI of -800 gCO_2eq/kg CO_2 supplied. The meaning of this negative value varies between sources of CO_2. Where the CO_2 is removed directly from the atmosphere (e.g., using DAC) or indirectly (e.g., from biomass growth), the negative sign represents a removal of CO_2 from the atmosphere. In contrast, where CO_2 is captured during fossil fuel combustion (e.g., power generation, cement manufacture), the negative value represents a *reduction* in emissions from the same facility where combustion occurs. Thus, supplying 1 kg of CO_2 will either result in an emissions reduction of 800 g of CO_2 when the CO_2 source is PCC or remove 800 g of CO_2 from the atmosphere by DAC. Because the GHG EI of 1 kg CO_2 supply varies based on the source of CO_2 and the capture process, we vary GHG emissions for capture from as low as 50 (for PCC) to a high of 410 gCO_2eq (for DAC), which results in a net GHG EI of CO_2 supply for all CO_2 sources, the cradle-to-gate footprint of captured CO_2 is negative ranging from -950 to -590 gCO_2eq/kg (Müller et al., 2020). This indicates that supplying 1 kg of CO_2 can reduce (or remove) 590 to 950 gCO_2eq, depending on the carbon capture techniques (e.g., remove CO_2 by DAC, reduce emissions by point source capture) in 2020, with the potential to increase to 990 gCO_2eq in future (Müller et al., 2020).

The GHG EI of electricity grids varies widely from location to location based on the electricity generation options employed. We assume the electricity is supplied from a power system with a GHG EI of 100 gCO_2eq/kWh as illustrated in Table 1. Such an intensity is higher than that required in net-zero scenarios, but less than a quarter of the global average GHG EI of electricity supply in 2021 (IEA 2022). We vary the GHG EI of electricity from 10 to 1000 gCO_2eq/kWh to reflect a wide range of potential electricity sources, where the lower end represents renewable electricity from wind power (Hertwich et al., 2015) and the higher represents electricity generated from lignite (brown) coal (EIA 2021). Thermal energy for electrochemical ethylene and ethanol production (Fig. 1), is assumed to be supplied by electricity. For downstream conversion of these products to jet fuel, we assume thermal energy is supplied by natural gas—as in commercial processes today—with a GHG EI of 50 gCO_2eq/MJ for all (USEIA 2021). Consistent with use of the substitution approach adopted, excess co-product hydrogen from CO_2 electrolysis—after accounting for consumption in the downstream hydrogenation step shown in Fig. 1—substitutes for production of H_2 from water electrolysis (requiring, ~50 kWh/kg H_2 production) (Bertuccioli et al., 2014). We assume that O_2 is vented.

The sum of the avoided GHG emissions from CO_2 supply and H_2 production is the total reduction in GHG EI. In cases where CO_2 is removed from the atmosphere in DAC, the result reflects both removals and avoided GHG emissions from H_2 production and substitution. In principle, while a reduction and a removal are physically distinct, if large quantities of CO_2 continue to be emitted from human activities, there is no practical difference between them.

3. Results and discussion

3.1. Mass and energy balance for CO_2-SAF synthesis

In this section, the mass and energy balance for synthesis of SAF from captured CO_2 for each of the four pathways (i.e., one-step ethanol, two-step ethanol, one-step ethylene, and two-step ethylene) is discussed. As shown in Fig. 1, both ethanol and ethylene can be produced from captured CO_2 via an electrochemical process and then CO_2-SAF can be synthesised. In a CO_2 to ethanol to SAF process, dehydration is needed to convert ethanol to ethylene. Table 2 summarizes the overall mass and energy balance to produce 1 kg of SAF starting from captured CO_2 (sample calculations are detailed in SI). The whole system requires CO_2, electricity, water, and thermal energy as inputs. Hydrogen, oxygen, diesel, and gasoline are the co-products of the multifunctional system along with the SAF. According to Table 2, the one-step CO_2 electrolysis required approximately 4 times more CO_2 because maximum achievable single pass CO_2 conversion in the one-step route is 25 % while the rest is lost to carbonate/bicarbonate formation as highlighted in our recent study (Nabil et al., 2021). Likewise, the electricity requirement for one-step synthesis is ~12–19 % higher than two-step for both intermediates and attributed to higher overpotential (total 1.4 V) required for one-step reactions (detail description available in Table S1). Thermal energy is required only for the dehydration, oligomerization and hydrogenation processes as the electrochemical process needs electricity only. Hydrogen is produced from CO_2 electrolysis and is used in the hydrogenation process to produce SAF, and the excess H_2 (reported in Table 2) is treated as a co-product. The detailed mass and energy balances for each unit process can be found in Fig. S1. The results also demonstrated that the largest amount of excess hydrogen was produced from the ethanol one-step pathway. The co-products also include O_2 due to CO_2 electrolysis under the assumptions of 70 % Faradaic efficiency (FE) for both ethanol and ethylene along with 1.4 V overpotential for one-step route and 0.6 V overpotential for two-step route (CO to products) as reported previously (Nabil et al., 2021). For product separation, a RadFrac distillation column was designed in Aspen Plus (V10) for liquid products while pressure swing adsorption was considered for gaseous products (Nabil et al., 2021).

3.2. WtP global warming potential of ethanol and ethylene pathways

The GHG EI to produce SAF are calculated based on the assumptions presented in Table 1 and inventory in Table 2. The results are presented in Fig. 3, in which we use the functional unit of 1 MJ aviation fuel.

The WtP GHG EI for the base case (stars) and range that encompass high and low single-parameter uncertainties for technological parameters (red bars) and all parameters (blue bars) are shown in Fig. 3(a), while the contribution to the total GHG EI for the ethylene two-step

Table 2
Mass and energy balance for reaction feedstocks and utility requirement for producing 1 kg CO_2-SAF from captured CO_2 using base case parameters (refer to Table 1). These results do not include the CO_2 capture process (e.g., PCC or DAC).

Mass & Energy Balance	Ethanol	Ethanol	Ethylene	Ethylene
Input	One-Step	Two-Step	One-Step	Two-Step
CO_2 Consumed [kg]	4.26	4.26	3.67	3.67
Electricity [kWh]	87.06	72.58	45.38	32.90
Water [kg]	2.91	1.94	2.50	1.67
NG Thermal [m^3]	0.15	0.15	0.11	0.11
CO_2 conversion rate*	25.8 %	100 %	25.0 %	100 %
Mass Output				
SAF [kg]	1.00	1.00	1.00	1.00
Diesel [kg]	0.29	0.29	0.29	0.29
Gasoline [kg]	0.15	0.15	0.15	0.15
H_2 [kg]	0.05	0.03	0.04	0.02
O_2 [kg]	5.17	3.45	4.45	2.96
Energy Output – Energy density adopted from (Song, 2020)				
SAF [MJ]	43.5	43.5	43.5	43.5
Diesel [MJ]	13.20	13.20	13.20	13.20
Gasoline [MJ]	6.9	6.9	6.9	6.9
H_2 [MJ]	6	3.6	4.8	2.4

Note: * conversion rate in a single pass; unconverted CO_2 is recycled in the system.

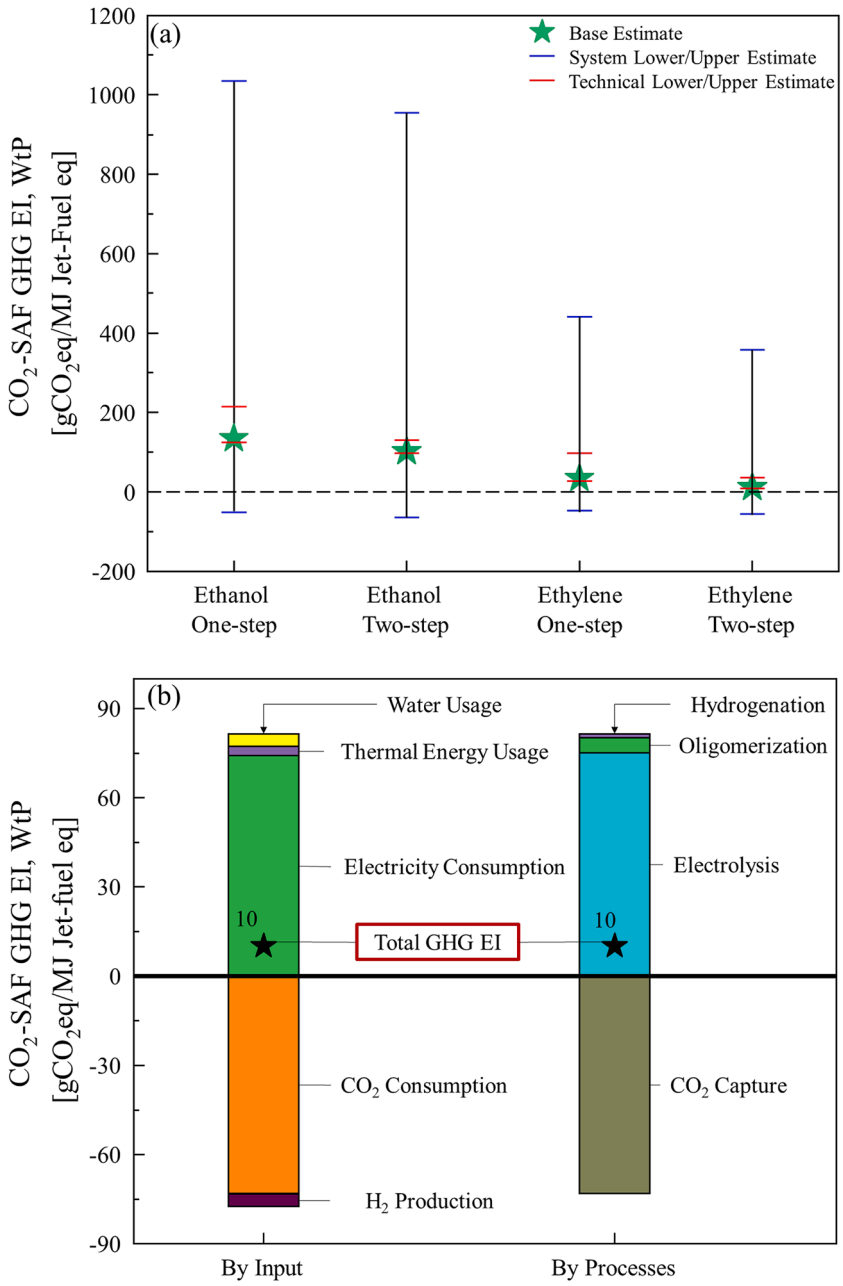

Fig. 3. WtP GHG EI (gCO_2eq/MJ Jet-Fuel equivalent) of (a) the four combinations of different intermediates and one- and two-step pathways, where the base case result is indicated by a star, the lower and upper limits (red bars) resulting from varying technological parameters in Table 1, and lower and upper limits (blue bars) resulting from varying all parameters indicated in Table 1; and, (b) contribution of each input (left) and process (right) to the base case result for the two-step process through ethylene. The relative contributions by input and process for the other three pathway's base case results are similar to those shown in (b) and can be found in Fig. S2.

pathway is shown in Fig. 3(b). Variations in the technical parameters between their high and low values (as shown in Table 1), which are intended to capture uncertainties in the future evolution of the technology, affect the resulting GHG EI much less than variations in the background system parameters. These results show that, all else equal, the ethylene pathways tend to have a lesser GHG EI than the ethanol pathways and there is a negligible difference between the one- and two-step pathways. The two-step ethylene process has the lowest GHG EI in the base case, which is 10 gCO_2eq/MJ, while the one-step ethanol process has a base case GHG EI of 137 gCO_2eq/MJ.

The largest contributor to the resulting GHG EI across all pathways is the electricity consumption of CO_2 electrolysis. Fig. 3(b) shows this results for the two-step ethylene pathways, and the relative contribution is similar across all processes (Fig. S2) (Nabil et al., 2021). Fig. 3(b) also clearly shows that positive GHG emissions resulting from energy generation and feedstock production are largely—although not entirely—offset by capturing CO_2 from the air or an existing point source, avoiding H_2 production from water electrolysis, and avoiding product of gasoline and diesel (both of which are co-products, as shown in Table 1). As a reference, production of gasoline and diesel as a co-product is assumed to avoid upstream emissions from fossil fuel production and refining of 19.7 and 18.5 gCO_2eq/MJ, respectively, based on averages for the United States (Bauer et al., 2022; Cooney et al., 2017; Howarth and Jacobson, 2021).

The lower GHG EI of ethylene as intermediate for both CO_2 electrolysis pathways compared to ethanol results from two factors. First,

the electricity required to produce ethylene from CO_2 is almost half compared to ethanol for both one- and two-step routes. This is because of the molar mass of ethanol is larger and requires more energy to synthesize from CO_2. Additionally separation energy for ethylene is only accompanied by pressure swing adsorption (PSA) as opposed to ethanol that typically requires energy intensive distillation (Nabil et al., 2021). Secondly, when producing aviation fuel from ethylene, the dehydration process is not needed, which limits overall energy requirement.

3.3. Single parameter sensitivity analysis of WTP GHG EI

The model parameters for each unit process were systematically varied to assess their influence on the resulting WtP GHG EI. A shown in Table 1, a conservative base case scenario for FE of CO_2 electrolysis has been considered (70 %), which is consistent with typical current densities of 300 mA/cm^2 in 2023 (Nabil et al., 2021). For all other cases, although most of the preeminent studies report a FE of ~50 % for liquid products, a stretch-target FE of 90 % has been assumed for both the intermediates (De Luna et al., 2019; Nabil et al., 2021). The dehydration, oligomerization and hydrogenation are mature commercial scale technologies with stable efficiencies around 85 %, 85 % and 70 % respectively (Geleynse et al., 2018).

As evident from Fig. 4, the GHG EI of all pathways is most sensitive to the GHG EI of electricity supply due to the considerable electricity consumption. In an electricity system that is largely based on renewables, the GHG EI of electricity could be reduced to 10′s of gCO_2eq/kWh and the WtP GHG EI of CO_2-SAF could, thus, be below zero. However, renewable-based systems also require storage solutions to manage variability and ensure stable power supply to the process, which is not considered at this stage. Of course, this ignores the final use of the fuel, which will be examined in the following section. The GHG EI of CO_2 supply is the second most influential parameter on the resulting GHG EI of CO_2-SAF. As seen from the figure, the improvement of efficiency will not considerably reduce GHG EI. However, improving hydrogenation efficiency will result in less hydrogen consumption and lead to a slightly larger H_2 credit to avoid CO_2 emissions. The impact of emissions from thermal energy and water supply is negligible.

Because we adopt published values for the GHG EI of CO_2 supply (Table 2), GHG emissions from DAC and PCC do not vary with the GHG EI of electricity or thermal energy in the model used here. While DAC and PCC both require electricity and thermal energy, electrochemical reduction of CO_2 requires one- to two-orders of magnitude more energy than separation of CO_2 from air and flue gas, respectively. If the relationship between GHG EI of CO_2 supply and GHG EI its energy inputs were to be modeled explicitly, GHG EI of electricity would become even more influential.

3.4. Well-to-Wake (WtW) GHG EI

Today, aviation fuel is almost exclusively produced from crude oil (FAF), and it has been commercially produced (in relatively small quantities) from bioethanol (BEAF) using the ethanol-to-jet processes described in Section 2.1. In this section, the system boundaries were expanded to include end-use combustion emissions (as shown in Fig. 2), and the four proposed CO_2-SAF pathways were compared with the BEAF and FAF as demonstrated in Fig. 5. As presented in Table 1, we use a GHG EI for electricity supply of 100 gCO_2eq/kWh to represent a system that could be renewable dominated but still use some small amount of fossil generation (e.g., for reliability). While 100 gCO_2eq/kWh is less than a quarter of the global average in 2021 (IEA 2023), there are some locations with even lower GHG EI values today (e.g., the Canadian Province of Quebec, at 1.7 gCO_2eq/kWh (ECCC 2023)). Thus, these results should be seen as representing a future application of the technology along the path towards net-zero.

Fig. 5(a) shows that the total WtW GHG EI of the base cases for each of the four pathways ranges from nearly 200 gCO_2eq/MJ for the one-

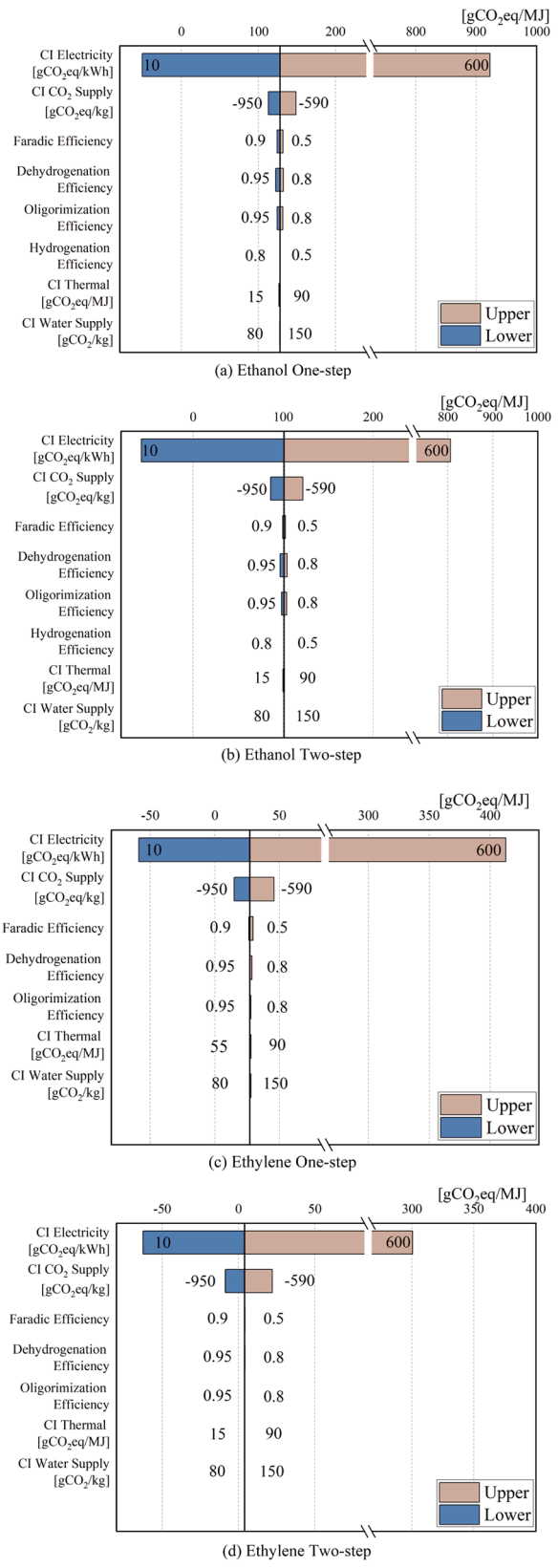

Fig. 4. Sensitivity analysis showing the impact of most sensitive parameters of WtP GHG EI for (a) one-step ethanol, (b) two-step ethanol, (c) one-step ethylene and (d) two-step ethylene routes. The ranges for carbon intensities of electricity, CO_2 supply as well as the Faradaic efficiency for CO_2 electrolysis and efficiencies of unit processes of aviation fuel production are in accordance with the values provided under Table 1.

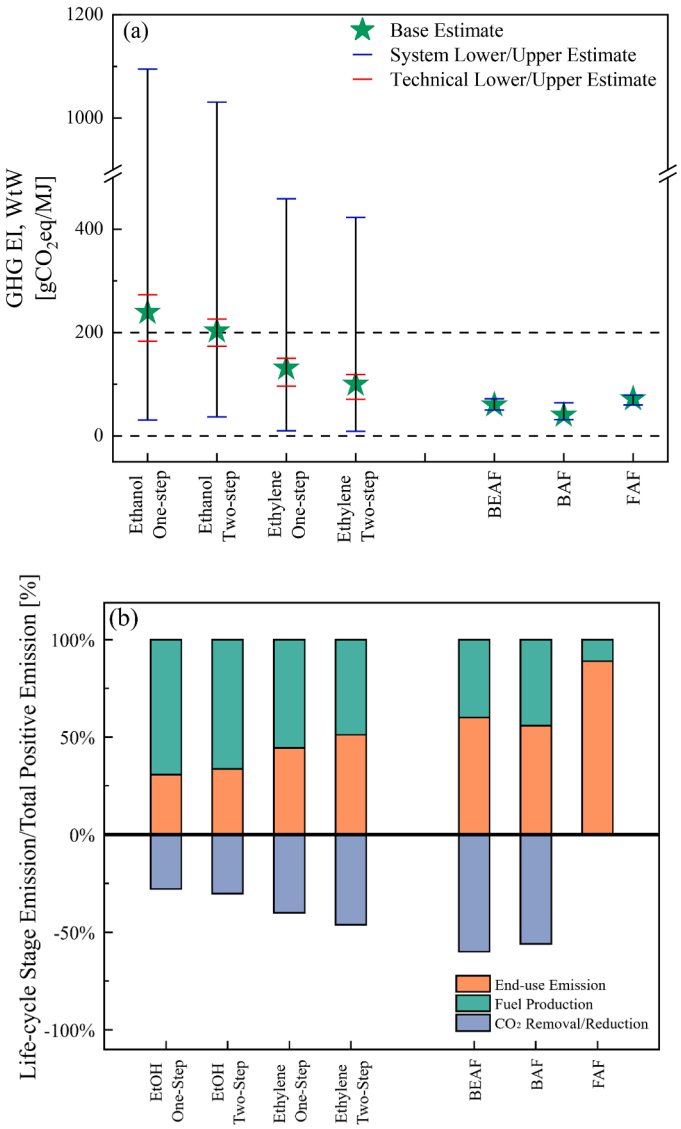

Fig. 5. Prospective WtW GHG EI comparison for CO_2-SAF pathways with BEAF and FAF of (a) total estimates with uncertainties shared the same definition with Fig. 3 and (b) percentage breakdowns including end-use emissions and CO_2 supply. GHG EI of electricity in low-carbon economy is assumed at 100 gCO_2eq/kWh (direct), and that of CO_2 supply from DAC and PCC is assumed to be -800 gCO_2eq/kg with the PCC capture rate at 90 % (Sleep et al., 2021). DAC and PCC CO_2 are represented by the CO_2 Removal/Reduction bars but are not differentiated because they have the same quantitative result of reducing the WtW GHG EI, albeit by different means.

step pathways via either intermediate to 70 gCO_2eq/MJ for either two-step pathways. Comparing the results for the CO_2-SAF pathways with those for BEAF, BAF, and FAF, we see that all of the CO_2-SAF pathways are generally higher but could be competitive with advantageous choices of system parameters (e.g., very low GHG EI of electricity and CO_2 supply). However, only the pathways using ethylene as an intermediate are competitive in the base case. Previous studies show that the WtP GHG EI of aviation fuels from both conventional crude oil and biotechnologies vary with GHG EI of grid electricity. Using the GHG EI of the U.S. grid in 2020 of 410 gCO_2/kWh (Kelly and Elgowainy, 2018), the WtW GHG EI of FAF was estimated from GREET (using energy allocation) to vary between 90 and 110 gCO_2eq/MJ (de Jong et al., 2017, Wu et al., 2006, Wong, 2008), and 45.8 to 75.4 gCO_2eq/MJ for BEAF (Han et al., 2017, Wong, 2008, European Commission, 2015, Stratton et al., 2010). The range is wider for BEAF because of the number of feedstock variations (e.g., corn, sugarcane, cellulosic biomass, etc.) possible. The case where CO_2 is supplied from PCC have the same GHG EI as those where CO_2 is supplied from the air as long as the GHG EI of CO_2 supply are same. That said, the GHG EI of PCC will tend to be lower than DAC given the latter's higher energy intensity. The conditions under which the GWP of CO_2-SAF could lower than the incumbent fossil and biofuel options are explored in the subsequent section.

It is also clear from Fig. 5(a) that none of the CO_2-SAF fuels are "carbon neutral" (i.e., having a GHG EI of zero). This can be explained by the results in Fig. 5(b), which show that the CO_2 consumption (whether from PCC or removal) is equal to that emitted from CO_2-SAF combustion across all pathways. The combustion of CO_2-SAF (or BEAF) releases the CO_2 consumed from the capture unit (or photosynthesis) back to the atmosphere. In addition, for BEAF, CO_2 is also released during ethanol fermentation. For example, growth of the biomass feedstock removes the equivalent of 60 gCO_2/MJ (biogenic, on average) and the same amount of CO_2 is released (i.e., 60 gCO_2eq/MJ) back to atmosphere as a result of fermentation and combustion of the resulting fuel (Han et al., 2017). Thus, producing net-zero emission CO_2-SAF is impossible (absent geological sequestration of some removed CO_2), as this would require the fuel production system to emit zero GHG emissions. The estimations of WtW GHG EI for all pathways with respect to variations of either GHG EI of electricity or CO_2 supply can be found in Fig. S3, respectively.

3.5. *Impact of sensitive parameters: GHG EI of electricity and CO_2 supply source*

In this section, we examined variation in WtW GHG EI as a result of changing the GHG EI of electricity and CO_2 supply as shown in Fig. 6, As discussed in Section 3.2, the realistic WtP GHG EI of the proposed four electrochemical pathways are much larger than the existing conventional and biomass technologies because of the high GHG EI of the U.S. electricity grid, which ranged from 240 to 740 gCO_2eq/kWh in 2023 (USEPA 2023).

Technical advancements in CO_2 capture and increasing use of non-emitting generation technologies (e.g., renewables, nuclear) can be expected to reduce the GHG EI of CO_2 and electricity supply in the future. Fig. 6 shows that using CO_2 from a DAC unit (a GHG EI of CO_2 supply of -900 gCO_2eq/kg) in the pathways with ethylene as the intermediate product requires a GHG EI of electricity from 120 to 180, and 78 to 136 gCO_2eq/kWh to compete with FAF, and BEAF, respectively. The ethanol pathways require 57 to 77, and 27 to 43 gCO_2eq/kWh to compete with FAF and BEAF, respectively for similar scenarios.

In addition to the general trend towards lower GHG EI of CO_2 supply and grid electricity, the location of a CO_2-SAF facility could be chosen to minimize the resulting GHG EI of CO_2-SAF. For example, in 2021, while the average grid electricity of Canada was approximately 110 gCO_2eq/kWh (ECCC 2023), this number hides variation between provinces (each of which have an independently operated electricity grid) ranging from 700 gCO_2eq/kWh (Saskatchewan) to near zero (Quebec). Deploying these CO_2-SAF pathways in Canadian provinces or states in the US with grid GHG EI <200 gCO_2eq/kWh, such as BC, MB, ON, QC in Canada, WA, ID NH, and SD in U.S. could reduce GHG emissions (see Fig. S4) (USEPA 2023). Deploying these pathways in other regions may not be environmentally beneficial today due to the GHG intensive electricity supply.

With the CO_2 capture technology development and rapid development of renewable electricity, the CO_2-SAF pathways in this study have he potential to be competitive technologies—at least based on GHG EI—in the coming decades. Replacing FAF by CO_2-SAF could, therefore, deliver a GHG emissions benefit. That said, while the theoretical lowest GHG EI of proposed CO_2-SAF with CO_2 supplied from DAC could approach zero, the process can not generate "negative emissions" as removed CO_2 is not being stored permanently (Tanzer and Ramírez, 2019).

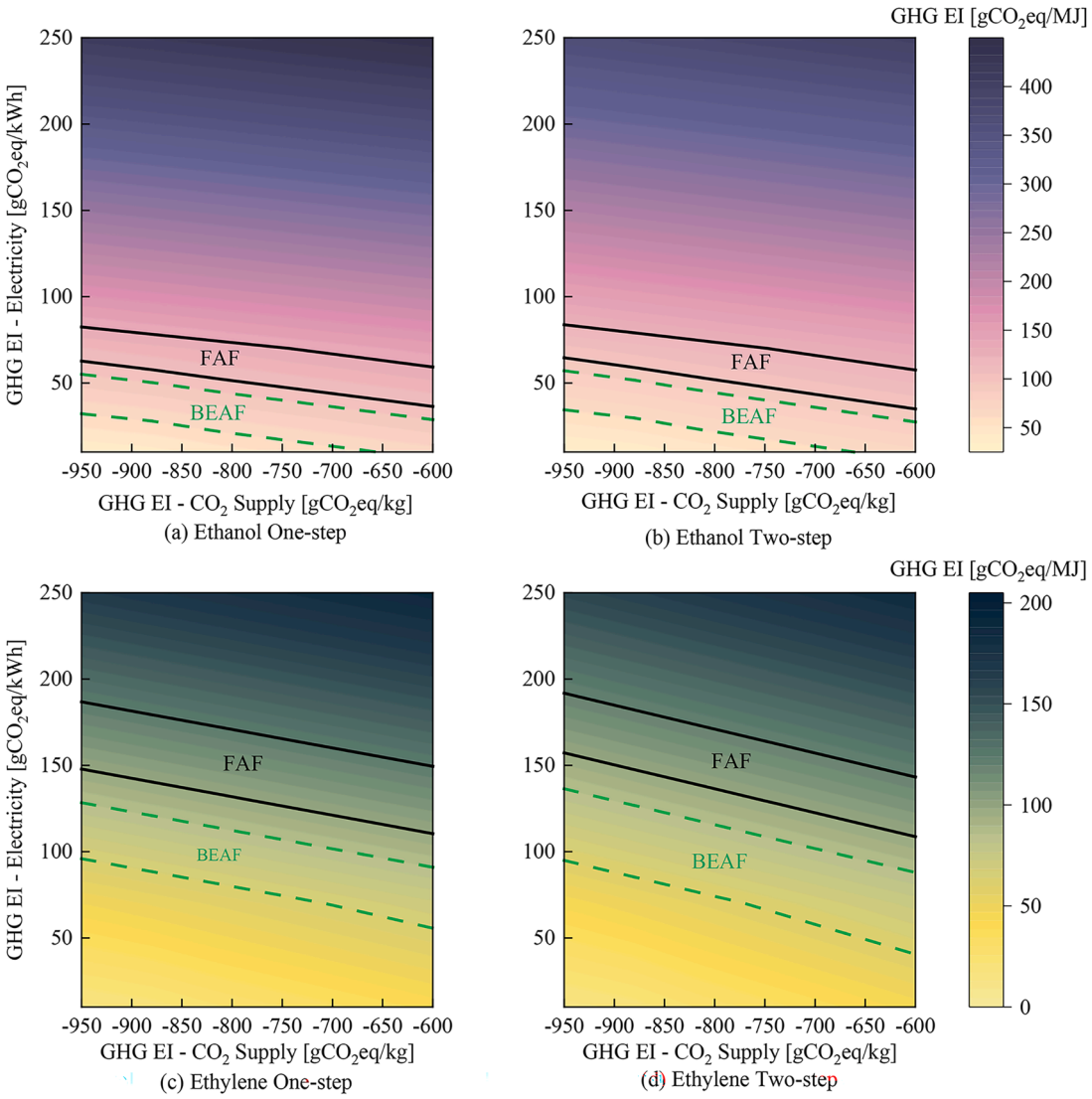

Fig. 6. Effect of GHG EIs of electricity and CO_2 supply from DAC on WtW GHG EI by the method of substitution for (a) one-step ethanol, (b) two-step ethanol, (c) one-step ethylene and (d) two-step ethylene routes for CO_2-SAF production. The x-axis and y-axis representing the combination of GHG EI of electricity and CO_2 supply, and the color gradient stand for the estimated GHG emission intensities for each pathway in the unit of gCO_2eq/MJ. GHG EI of FAF and BEAF range are collected from GREET based on energy allocation method (Argonne National Laboratory, 2023).

4. Conclusions

In this research, we evaluated the GHG EI of producing SAF from CO_2 through four pathways that couple CO_2 captured from DAC and PCC units with conversion of CO_2 to intermediate products via electrolysis, and subsequent production of SAF using mature technologies. Our results show that, from a global warming perspective, ethylene is better than ethanol as an intermediate as pathway for conversion to SAF via indirect electrochemical routes (Nabil et al., 2021), and that there is not a meaningful difference in GHG EI between one- and two-step pathways. Results of a single parameter sensitivity analysis indicate that the WtP GHG EI is most sensitive to the GHG EI of electricity due to large electricity demand. The GHG EI of CO_2 supply also has a direct effect on GHG EI as it is the feedstock to the system. Given how small the differences are between the various pathways considered, factors that are outside the direct control of technology developers (e.g., GHG EI of the electricity grid and the CO_2 supply) may determine which technology is a success. By expanding the system boundary to a WtW includes end-use combustion, we demonstrate that all the investigated pathways can outperform FAF and BEAF when low-GHG EI electricity inputs (below 180 gCO_2e/kWh) and low-GHG EI CO_2 supply (less than -900 gCO_2e/kWh) are utilized. This highlights the importance of strategically selecting geographic areas for deploying the proposed CO_2-SAF pathways, as such choices could enable the production of fuels that achieve meaningful GHG EI reductions compared to FAF and BEAF. We estimate the minimum WtP GHG EI of CO_2-SAF to be 10 gCO_2eq/MJ for the two-step ethylene pathway, however, this does not include emissions from infrastructure used in the system. Finally, it is important to note that the proposed pathways cannot be classified as negative emission technologies, as CO_2 sourced from DAC is not permanently removed from the atmosphere due to its end-use combustion. With additional data on CO_2 electrolysis performance, future work could enhance the study by integrating process-level simulations of the entire system (including CO_2 capture) with LCA to optimize process configurations and link electro-reduction Faradaic efficiency to catalyst performance. The field would also benefit from investigating the cost implications of these pathways compared to aviation biofuels.

CRediT authorship contribution statement

Haoming Ma: Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Resources, Methodology, Formal

analysis, Data curation, Conceptualization. **Shariful Kibria Nabil:** Writing – review & editing, Writing – original draft, Visualization, Resources, Methodology, Formal analysis, Data curation, Conceptualization. **Keju An:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Emily Nishikawa:** Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Md Golam Kibria:** Writing – review & editing, Validation, Supervision. **Joule A. Bergerson:** Writing – review & editing, Validation, Formal analysis, Conceptualization. **Zhangxin Chen:** Writing – review & editing, Investigation, Funding acquisition, Conceptualization. **Sean T. McCoy:** Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ccst.2025.100477.

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