Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information for

A Life Cycle Assessment Case Study of Direct Air Capture and Fischer-Tropsch Fuel Production

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Methods: Conversion factors for chemical reactions

Table S1 lists the conversion ratios by mass for the rWGS reaction (conversion of CO_2 to CO), syngas blending (combining of H_2 and CO), and FTS reaction (conversion of syngas to syncrude). In this case study, only literature data was used for the FTS portion of the process. The base case assumes these ratios, but they are further investigated in the sensitivity analysis. These ratios could change in response to operating conditions or be adjusted for plant upsets.

From van der Giesen et al.,[8] the rWGS scenario is experimental. It is a well-known side reaction in SMR, however it is not performed in isolation. This set-up requires a water sorbent to prevent the reverse reaction (water gas shift) and 2 (or more) reactors to run continuously with the sorbent re-generation cycle. A 100% stoichiometric conversion of CO_2 and H_2 occurs at an experimental scale at lower temperatures and with the removal of water. Water is a by-product of this reaction and could provide some of the water make-up in DAC. Approximately 12% (600 kt/year) of the water make-up requirement (5 Mt/year) in the 1 Mt CO_2/year DAC plant could be supplemented.

Syngas blending is the mixing of H_2 and CO to get an optimal H_2/CO ratio (2:1) before entering the FT reactor. FTS has an 80% extent of conversion to useable fuels. The other 20% are low-quality products and used to sustain the plant in heat, electricity, and steam.

Table S1: Conversion factors for reverse water gas shift (rWGS) reaction, syngas blending, and Fischer-Tropsch synthesis (FTS) reaction

Process	Conversion (kg to kg)	Reference
rWGS (CO_2 to CO)	1.6	van der Giesen et al.[8]
rWGS (H_2 to CO)	0.07	
Syngas blending (CO to syngas)	0.88	
Syngas blending (H_2 to syngas)	0.13	
FTS (syngas to fuel)	2.3	

Methods: Material and energy balance

Table S2 presents a material and energy balance, separated by unit, on the direct air capture (DAC) and Fischer-Tropsch synthesis (FTS) combined process base case scenario. This represents the full-scale mass and energy flows. The DAC process values are from Keith et al.[1] and the FTS process values are from Van der Giesen.[2] Unrounded values may be found from the sources, but they are rounded here to not overstate accuracy. These flows (and their respective emission factors, Table S3) are used to calculate the greenhouse gas (GHG) emissions. Some of these flows are circulating materials (e.g., KOH in the air contactor) and may be a unit output but not a system output.

Construction and decommissioning materials are not included here as they are not “flows” but are discussed with Figure S1.

Table S2: Material and energy flows for each unit in the combined direct air capture (DAC) and Fischer-Tropsch synthesis (FTS) process base case scenario

Unit	Input material or energy	Input flow (Mt/year or MW*)	Output material	Output flow (Mt/year)	Loss material	Loss flow (Mt/year)
Air contactor	Air	2 500	Air with CO_2 removed	2 500	KOH	2.7×10^{-4}

	KOH solution	350	Captured CO_2 (as carbonate)	1.1		
	KOH make-up	2.7×10^{-4}	KOH solution	350		
	Electricity*	10				
Pellet Reactor	Captured CO_2 (as carbonate)	1.1	$CaCO_3$ pellets	2.6	$CaCO_3$ fines	3.0×10^{-2}
	Caustic ($Ca(OH)_2$)	1.9	KOH solution	350		
	Carbonate ($CaCO_3$) seed	5.3×10^{-2}	H_2O on pellets	0.40		
	$CaCO_3$ make-up	3.0×10^{-2}	$Ca(OH)_2$ return	0.22		
	Water (H_2O)	4.9				
	Electricity*	4.0				
Calciner	$CaCO_3$ pellets	2.6	CO_2	1.5		
	Oxygen (O_2)	0.49	Quicklime (CaO)	1.4		
	Natural gas	0.12	$CaCO_3$ seed	5.3×10^{-2}		
	Electricity*	0.8				
Slaker	CaO	1.4	$Ca(OH)_2$	1.6		
	H_2O	0.40	CaO	0.22		
	Electricity*	3.6				
Com-pressor	CO_2	1.5	CO_2	1.5		
	Hydrogen (H_2)	0.20	H_2	0.20		
	Electricity*	30				
Steam Turbine	Steam	0.61	Steam	0.61		
			Electricity*	9.8		
AUX	Electricity*	0.9				
Electro-lyzer	H_2O	1.8	H_2	0.20		
	Electricity*	1 300	O_2	1.6		
rWGS Reactor	CO_2	1.5	CO	0.93		
	H_2	6.5×10^{-2}	H_2O	0.60		
	Copper catalyst	2.0×10^{-6}				
FTS Reactor	CO	0.93	Synthetic fuel	0.46		
	H_2	0.13				
	Cobalt catalyst	2.0×10^{-6}				

*Units are MW when specified

Methods: Emission factors for material production and energy generation

Table S3 presents the emission factors used in this analysis and the respective sources. It is assumed that the material and energy flows presented by Keith et al.[1] are representative of a full-scale DAC system. These flows must be converted into their respective emissions through emission factors. These emission factors represent the emissions associated with the manufacture of chemicals, generation of electricity, and electricity use of electrolysis. KOH is used in the air contactor, $CaCO_3$ and $Ca(OH)_2$ are used in the pellet reactor, water is used to rehydrate CaO to $Ca(OH)_2$, and natural gas is used in the calciner. Electricity is used to power the equipment and provide energy for electrolysis. Electrolysis is used to produce hydrogen for the rWGS and FTS reactions and oxygen for the calciner.

The electricity CI for baseline scenario is assumed to be that of the British Columbia (BC) grid electricity. CI for existing low-carbon grids, such as BC and Quebec (QC) are indicative of CI for new-build renewables and potentially future grids with increased renewable penetration. The variability of electricity emission intensities due to different generation mixes is investigated in the sensitivity analysis. The CI for Alberta's (AB) grid was used as being indicative of fossil-based electricity supply for sensitivity analysis. The electricity grid intensities of BC and QC, were derived from the National Inventory Report (NIR) 2017.[2] However, there are discrepancies between provincial and federal level reporting, particularly for BC and AB. The electricity grid intensity is investigated in the sensitivity analysis for this reason and the range of electricity grid compositions (and therefore intensities) throughout the world. The emission factors in NIR represent the emissions associated with electricity generation and were developed through a robust reporting protocol. This includes all generation sources (combustion of fossil fuels, hydro, nuclear, etc.). Emissions from ancillary activities, e.g. land flooding for hydro, aren't included. This also excludes energy lost during transmission and distribution, listed as "unallocated energy" in NIR. "Unallocated energy" changes from 2016 to 2018, ranging between unknown (counted as 0) and 1 300 – 22 200 GWh (translates to an additional 0.6 – 9.2 gCO_2e/kWh for BC) over the years. SF_6 emissions are excluded; SF_6 is used as an insulating gas for high voltage equipment. The emissions associated with SF_6 are negligible with an impact of less than 0.1 gCO_2e/kWh and remain similar between 2016 and 2018. The NIR electricity grid emissions intensity for Alberta was not used as it is an over-estimation (by 140 gCO_2e/kWh) based on calculations performed. Alberta's electricity grid intensity is calculated from CANSIM tables 127-0004, 6, and 7[3] instead.

Emission factors for chemicals consumed in the DAC process were retrieved from SimaPro 8.0.2[4] using the "Alloc Def, U" version of the chemical. Only CO_2 emissions are included.

It is important to note that the electricity use of regular and high pressure alkaline water electrolysis include compression. If compression energy use is not included, regular alkaline electrolysis would be in the range of 46 – 50 kWh/kgH_2,[5] which is less intensive than high pressure electrolysis. The emission factor for electrolysis is actually an electricity use factor, which can be converted into GHG emissions with the electricity grid intensity.

Table S3: Emission factors for electricity and various materials used in the DAC process

Material or Process	Emission Factor	Reference
Baseline scenario electricity (gCO_2e/kWh)	13	BC, QC - National Inventory Report 2017[2]
Carbon-free electricity(gCO_2e/kWh)	1.2	
Fossil-based electricity(gCO_2e/kWh)	650	AB - CANSIM[3]
KOH ($tCO_2e/tKOH$)	2.1	SimaPro 8.0.2[4]
$CaCO_3$ ($tCO_2e/tCaCO_3$)	2.1×10^{-3}	
$Ca(OH)_2$ ($tCO_2e/tCa(OH)_2$)	0.79	
Water treatment ($tCO_2e/tH2O$)	9.0×10^{-6}	Racoviceanu et al.[6]
Natural gas, upstream ($KgCO_2e/GJ$ delivered)	6.1	Vaygahn[7]
Alkaline water electrolysis (kWh/kgH_2)	57	Bhandari et al.[5]
High pressure electrolysis (kWh/kgH_2)	50	
PEM electrolysis (kWh/kgH_2)	67	

Methods: Aspen Plus™[9] model

Literature data was used for the FTS process. Thus, a high-level Aspen Plus™[9] model of the FT reactor was created to investigate the effects on the product slate and emissions of change in operating conditions (e.g. temperature, pressure, and space velocity).

The kinetic equation from Mansouri et al.[10] using a power law model is as follows:

$$-r_{CO} = k_0 exp\left(\frac{-E}{RT}\right) P_{CO}^{m} P_{H_2}^{n}$$

Where $-r_{CO}$ = rate of consumption of CO (mol / g_{cat}.h)
k_0 = reaction rate constant (mol / g_{cat}.h.$bar^{-0.4}$), 2.1×10^8
E = activation energy of CO consumption (J/mol), 1×10^5
R = gas constant (J/mol.K), 8.314
T = temperature (K)
P_{CO} = partial pressure of carbon monoxide (bar)
m = reaction order for CO, -0.45
P_{H2} = partial pressure of hydrogen (bar)
n = reaction order for H_2, 0.85
Variables without listed values can change depending on the system.

Aspen Plus V10[9] was used with the PENG-ROB thermodynamic package. The reactor chosen was "RPlug" in Aspen Plus™ set at 2 m long with 400 tubes of 1 in diameter to model a multitubular fixed-bed reactor. The operating conditions are: pressure of 30 bar, temperature of 220°C, and flowrates of 5.0 kg/s CO and 1.45 kg/s H_2. Catalyst properties and packing are: density of 8900 kg/m^3 (pure cobalt), 0.2 cm particle diameter, 0.4 void fraction, and a shape factor of 1. Note that this reactor is a high-level model to look at the fuel product slate and the energy content. Further optimization would be required for actual operation and to be pairable with the CO_2 feed from a commercial-sized DAC plant.

Methods: Construction and decommissioning emissions breakdown (2 methods)

Construction and decommissioning emissions are associated with the construction of the plant (e.g. material production, equipment manufacture, and their transport) and the retirement of the plant (e.g. demolition). This could be a significant source of emissions, as plants require lots of material to construct, and should be investigated. Two methods to estimate construction and decommissioning emissions were performed using EIO-LCA and a similar plant comparison, the results are shown in Figure S1.

EIO-LCA was performed using eiolca.net[11], a tool developed at Carnegie Mellon University. It estimates the materials and energy and resulting emissions from different industry sectors. This method looks at the entire supply chain and estimates economy-wide emissions. It provides a very comprehensive look at the emissions generated by any sector. Some uncertainties inherent to EIO-LCA are due to sector aggregation, cost estimates, and old or uncertain data. Sectors available for EIO-LCA analysis are very limited and many of them are combined. For example, power generation and supply includes all methods of power generation, whether fossil or renewable. The emissions are also based on economic activity. If the cost itself is an estimate, the GHG results will reflect that estimate. The US 2002 Producer model which is used for this LCA uses public data from 2002. Industries and their emissions have changed since then, as well as reporting guidelines.

The tool is free for public use. It can be accessed online at www.eiolca.net and selecting the "Use the Tool" tab. The default model was used for this LCA. The industry and sector can be selected from the drop-down list or by searching key words (sectors used listed in Table S4). Input the cost of the equipment in M$ and select GHG results to display, then run the model. The results will be presented in a table with the emissions from different sectors (e.g. power generation, transportation, etc.) and the total. The emissions are also further divided into different categories (e.g. CO_2, CH_4, etc.).

Table S4 lists the costs and GHG emissions for the DAC and FTS plants. The emissions are normalized to a per year amount assuming a 20-year plant lifetime. The DAC equipment costs were obtained from Keith et al.[1] and the FTS equipment costs were estimated with the "Equipment Costs" tool by Kobe[12]. based on rough sizing calculations. These 2016 costs were also adjusted to the 2002 dollar with CEPCI. Emissions from power generation were specifically looked at, as they typically contributed the most to overall emissions and electricity grid in the USA has changed significantly from 2002. Schivley et al.[13] found that the average emissions intensity from 2001 to 2017 in the USA decrease by 30%. This was accounted for in the construction and decommissioning estimate for DAC and FTS.

Table S4: Cost and emission details using the EIO-LCA US 2002 Producer model for the DAC and FTS plants

Equipment		**Industry sector assumed in the EIO-LCA**	**Equipment cost (M$, 2016)**	**Equipment cost (M$, 2002)**	**Total CO_2e (t)**	**CO_2e from power generation (t)**
Air contactor		Air purification and ventilation equipment	114	83.4	54500	17500
Pellet reactor		Metal tank, heavy gauge	76.9	56.2	53100	13200
Calciner-slaker		Metal tank, heavy gauge	43.8	32.0	30200	7520
CO_2 Compressor		Air and gas compressor	17.2	12.6	7100	2560
Steam Turbine		Turbine and turbine generator set units	6.70	4.89	1950	630
Fines Filter		Other fabricated metal	17.6	12.9	10800	3390
Other DAC equipment*	Pumps	Pump and pumping equipment	40	29.2	16400	6060
	Heat exchangers	Power boiler and heat exchanger	30	21.9	17200	5010
	Piping	Plastics pipe and pipe fitting	26.9	19.6	18400	4760
Buildings		Prefabricated wood building	2.50	1.83	962	307
Electrolyzer		Storage battery	4.39	3.21	3340	1220
rWGS reactor		Metal tank, heavy gauge**	N/A***	0.10	1130	282
FT reactor				0.10		
Distillation tower				1.0		
H_2O knockout drums (2)				0.4		
Other FTS equipment*	Pumps	Pump and pumping equipment		0.50	225	83
	Compressor	Air and gas compressor		0.50	225	81
	Piping	Plastics pipe and pipe fitting		0.50	375	97

*Cost split between other equipment is estimated.

**Equipment was combined as it was in the same category and cost was under recommended threshold to analyze with EIO-LCA.

***Cost estimation performed for the FTS plant was in 2002 dollars.

The construction and decommissioning emissions of a similar plant, steam methane reforming (SMR), were also looked at for comparison. For the purpose of this comparison, the similarity between SMR, DAC and FTS was established based on that the three processes use the same number of major units, e.g. the steam reformer, high temperature shift, low temperature shift, and pressure swing adsorption in SMR, compared to the air contactor, pellet reactor, calciner, and slacker in DAC. Spath and Mann[14] looked at the materials required for construction of an SMR facility and based the emissions estimate on that. Concrete, steel, aluminum, and iron were included in materials. SMR is a common process and well understood. The size estimate of the plant and materials required should be accurate. To be comparable to the combined DAC and FTS process, the emissions from SMR were doubled. However, this emissions estimate is not as comprehensive as EIO-LCA as it only includes construction materials.

The comparison of emissions is shown in Figure S1. EIO-LCA gives a construction and decommissioning CI at 0.49 gCO_2e/MJ fuel, whereas the similar plant comparison would put it at 0.23 gCO_2e/MJ fuel. This difference is likely due to EIO-LCA accounting for economy-wide impacts. It includes many more sectors than construction materials. However, keep in mind this data may be under-reported or aggregated, leading to a higher estimate of actual emissions.

The pellet reactor, air contactor, and other DAC equipment each make up approximately 25% of the total EIO-LCA construction and decommissioning emissions. 17% is from the calciner-slaker and the remaining 8% (miscellaneous) includes all other equipment. FTS is part of the miscellaneous equipment. The FTS cost estimates are much lower than those for DAC. DAC cost estimates are also more in-depth and specific, whereas FTS was based on rough equipment sizes and costing methods.

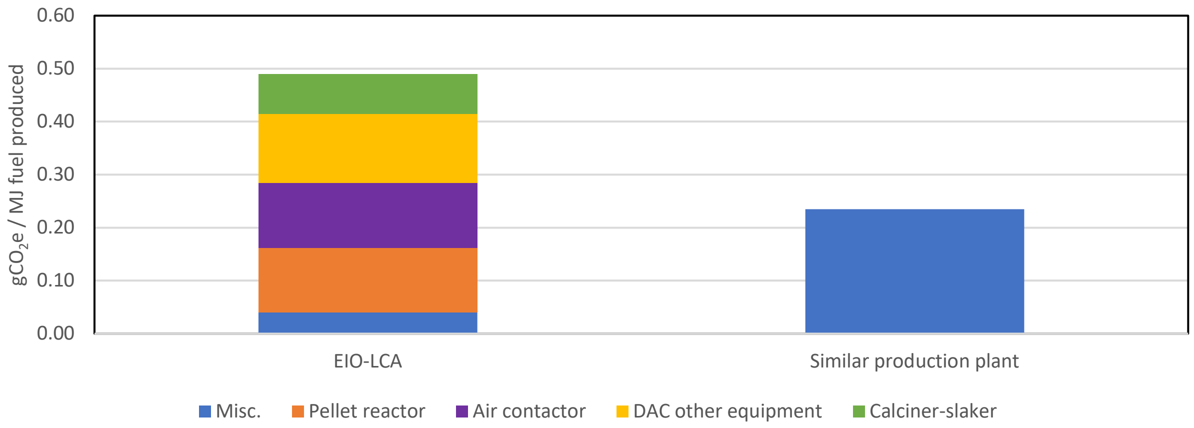

Figure S1: Construction and decommissioning emissions estimates from two methods: EIO-LCA (left) and comparison to a similar production plant (right) for the DAC and FTS plant

Results: Combined DAC and FTS sensitivity analysis (functional unit: per gCO_2 captured from air)

Two metrics can be used to investigate the CI of the DAC and FTS process, a per gCO_2 captured from air and a per MJ fuel produced basis. The sensitivity analysis on a per MJ fuel basis can be found in the main paper and is discussed there. The sensitivity on a per gCO_2 captured from air basis for the combined DAC and FTS process, Figure S2, is very similar to that. The majority of the discussion holds true from the main paper. The only discrepancy, the effect of FTS conversion, is discussed here.

Depending on operating conditions, catalyst condition, etc., the extent of conversion of FTS may change. The base case assumes 80% of the energy from syngas is converted into useable fuels, with the other 20% being light ends (i.e. gases) that are used to generate heat or electricity. If a lower extent of conversion was achieved, 80% of the base case assumption, then the CI increases to 0.52 gCO_2e/gCO_2 captured, a

1% increase. This isn't as impactful as with the per MJ fuel basis, as the major change from this is less fuel being produced, i.e. less MJ fuel produced. This means the denominator by which normalization occurs decreases, leading to larger changes on a per MJ fuel basis. However, as mentioned in the paper, this relationship may not be linear due to gas recycle or changes in the product slate.

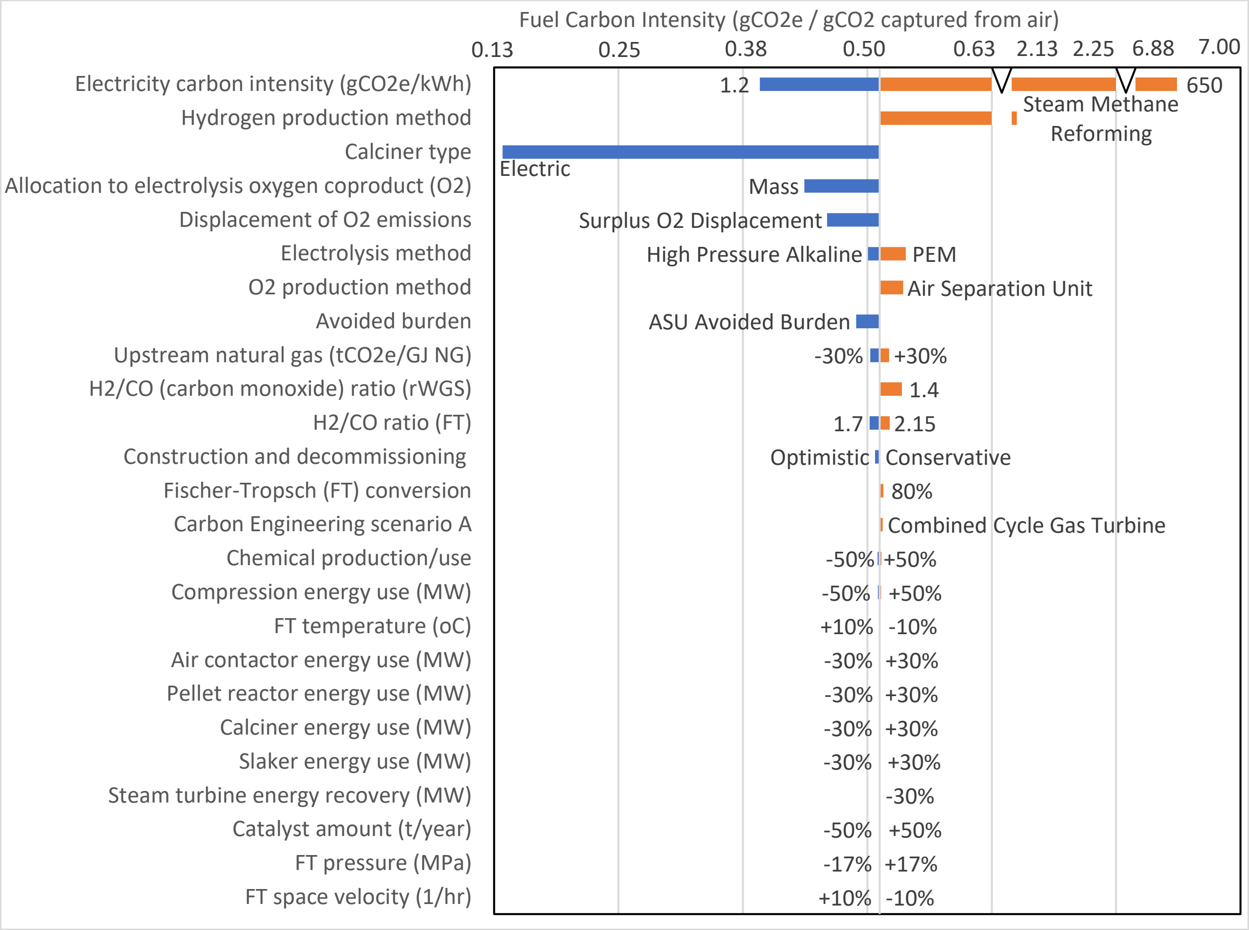

Figure S2: Sensitivity analysis of various parameters on the carbon intensity of the combined DAC and FTS process. The functional unit of per g CO_2 captured from air is shown. Base case assumptions can be found in Table 1 (main paper).

Results: DAC-only sensitivity analysis (functional units: per gCO_2 captured from air and per MJ fuel)

DAC can also be paired with CO_2 utilization methods other than synthetic fuel production, such as, EOR. It is important to investigate the sensitivity and uncertainty in DAC process, shown in Figure S3. Only the per gCO_2 captured from air basis is used as it is a better metric for DAC system performance.

The electricity carbon intensity is the most impactful parameter. The baseline case assumes a low CI renewable electricity supply(13 gCO_2e/kWh). Process emissions for the baseline DAC case emissions more than triple when assuming a fossil-based electricity supply, from 0.043 to 0.14 gCO_2e/gCO_2 captured from air. Note this change in CI is relative to the change in electricity CI (an increase of 50 times). When going to the nearly carbon-free electricity supply, there is only a 6% decrease in the CI from 0.043 to 0.040 gCO_2e/gCO_2 captured.

Emissions from oxygen production have the next greatest impact. In the base case, oxygen is assumed to have zero emissions as it is a co-product from electrolysis. If pairing DAC with other processes, oxygen may not necessarily be produced as a co-product. Thus, oxygen would have emissions associated with it. If produced by electrolysis and using mass allocation, which would allocate 94% of emissions to oxygen, DAC CI would increase by 81% to 0.078 gCO_2e/gCO_2 captured from air. Volume allocation, on the other hand, would have a negligible effect on the DAC CI, as nearly 100% of the emissions are still attributed to hydrogen. Market value (0.064 gCO_2e/gCO_2 captured) and mole allocation (0.056 gCO_2e/gCO_2 captured) lie in between these two extremes. Also, with the case of the DAC and FTS system, if electrolysis is used to produce hydrogen and consequentially oxygen, there is an avoided burden from producing oxygen with an ASU. This would give credit for a lower CI of 0.019 gCO_2e/gCO_2 captured. However, this would more likely be credited to the FTS side of the process, as the electrolyzer's main use is to produce hydrogen for FTS. The most common method of producing oxygen is with an ASU. This would result in a DAC CI of 0.066 gCO_2e/gCO_2 captured, an increase of 53%. This would be a lower-CI option than mass-allocated electrolysis.

Natural gas upstream emissions, i.e. the emissions associated with producing and delivering NG, are the next biggest factor to affect the DAC CI. An increase or decrease of 30% in NG production emissions results in a 23% increase or decrease in the DAC CI to 0.052 or 0.033 gCO_2e/gCO_2 captured from air respectively. This of course only affects the case with an oxy-fired calciner.

On replacing the oxy-fired calciner in the baseline case, with an electric calciner, the DAC CI would decrease by 26% to 0.032 gCO_2e/gCO_2 captured from air. If we assume an 80% electric heating efficiency for the electric calciner, then there would only be a 15% decrease from the base case DAC CI to 0.036 gCO_2e/gCO_2 captured. Note that the impact of electric heating efficiency is only seen on a per gCO_2 captured basis.

Chemical (KOH, $CaCO_3$, and $Ca(OH)_2$) production emissions have a slight impact. With an increase or decrease of 50% in the production emissions, the DAC CI will increase or decrease by 4% to 0.044 gCO_2e/gCO_2 captured or 0.041 gCO_2e/gCO_2 captured, respectively. Note that this sensitivity also applies to the use of these chemicals within the DAC process.

An optimistic estimate of construction and decommissioning emissions would decrease the DAC CI by 7% to 0.040 gCO_2e/gCO_2 captured from air. However, this would have a limited scope of emissions compared to the base case estimate. The base case includes emissions throughout the supply chain, the optimistic estimate may miss transportation emissions, raw material processing, etc.

Other parameters, e.g. energy use and recovery and CE's scenario A (using a CCGT instead of electricity), have negligible impacts on the CI of the DAC process.

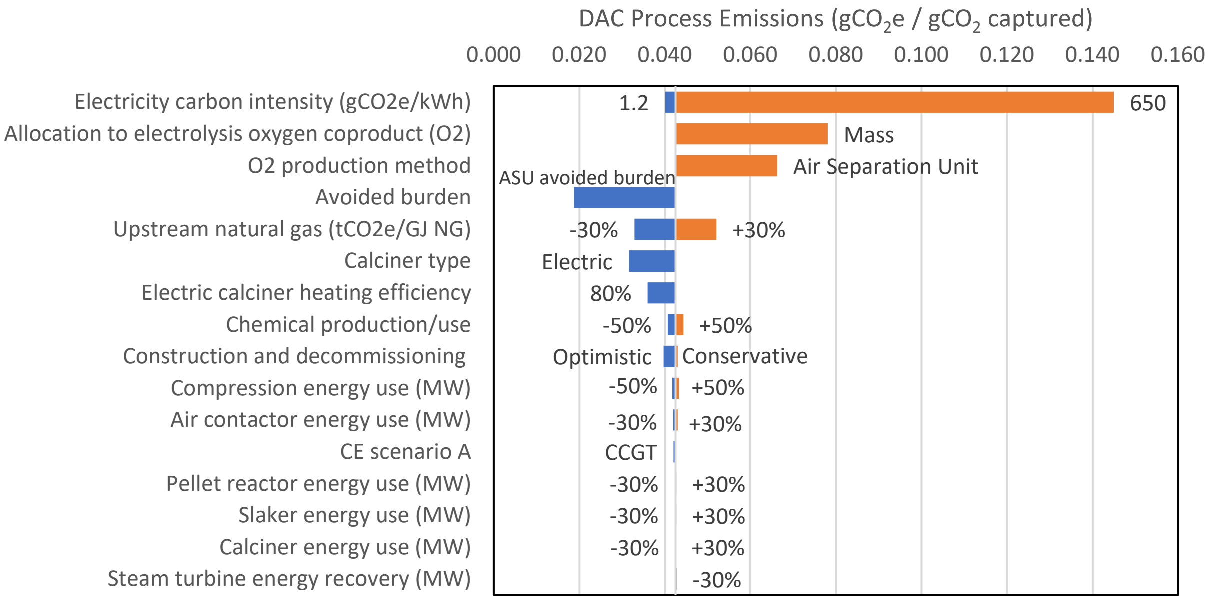

Figure S3: Sensitivity analysis of various parameters on the DAC process emissions on a per gCO_2 captured from air basis. Base case assumptions can be found in Table 1 (main paper).

Results: FTS-only sensitivity analysis (functional units: per gCO_2 captured and per MJ fuel)

It is also important to see how much of the combined impact is from the FTS process, displayed in Figure S4. For FTS, the CI values will be stated on a per MJ fuel produced basis as the product of FTS is synthetic fuels.

Again, the electricity carbon intensity is the most impactful parameter. If assuming a nearly carbon-free electricity supply (such as, the Quebec grid), the FTS process emissions will decrease from 7.6 to 0.92 gCO_2e/MJ fuel, an 88% decrease. With a fossil-based electricity supply, the CI increases to 366 gCO_2e/MJ fuel, over 48 times greater than the base case CI. The change in CI is almost 1:1 with the change in electricity supply carbon intensity, e.g. the change from baseline case (13 gCO_2e/kWh) to a fossil-based electricity supply is an increase of 50 times in electricity CI. FTS is more sensitive to the choice of electricity supply than DAC. This is due to the requirement for hydrogen, produced through electrolysis, which is very electrically intensive.

Using the most common method of hydrogen production, SMR, instead of electrolysis would result in an increase in the CI from 7.6 to 118 gCO_2e/MJ fuel, over 15 times greater than the base case. Further discussion on using SMR for synthetic fuel production from atmospheric CO_2 can be found in the paper.

Emissions from oxygen production also have a significant impact. Mass allocation of emissions from electrolysis to oxygen and hydrogen will result in a decrease of 86% in the CI to 1.1 gCO_2e/MJ fuel. This method of allocation credits oxygen with the majority (94%) of the emissions from electrolysis. Volume allocation, on the other hand, will only result in a 5% decrease in CI to 7.2 gCO_2e/MJ fuel. Market value and mole allocation CI values reside between the two aforementioned methods. Credit could also be given for “carbon-free” surplus oxygen (not including any used for the oxy-fired calciner) produced through electrolysis instead of ASU in industry. This would decrease the FTS CI by 39% to 4.6 gCO_2e/MJ fuel. Another credit is the avoided burden by using “carbon-free” oxygen from electrolysis in the oxy-fired calciner instead of oxygen from an ASU. A 17% decrease in CI to 6.3 gCO$_2$e/MJ fuel would result from this credit. Note that these are all accounting changes in the CI and don't affect the absolute amount of carbon entering the atmosphere.

A factor that has a significant impact is the extent of conversion of FTS. A decrease in extent of 20% from the base case will result in a 24% increase in the FTS CI to 9.4 gCO$_2$e/MJ fuel. This is discussed in the combined sensitivity analysis (Figure S2) and is due to the decreased amount of fuel produced.

Electrolysis type is important to FTS. Using high pressure alkaline electrolysis will result in a decrease in CI of 12% to 6.6 gCO$_2$e/MJ fuel. PEM electrolysis will result in an increase in the CI of 16% to 8.8 gCO$_2$e/MJ fuel.

There are two reactions in the FTS process that involve H_2 and CO: rWGS and FTS. H_2 is a feed to the rWGS reaction (along with CO_2), and CO and H_2O are produced. H_2 and CO are blended to make syngas, which is converted to synthetic fuels through the FT reaction. The H_2/CO ratio can be changed depending on the desired product slate or other operating conditions. For rWGS, changing the ratio to 1.4 from the base case 1, an increase in the FTS CI of 12% from 7.6 to 8.5 gCO$_2$e/MJ fuel will result. The H_2/CO ratio should not drop below 1 for optimal conversion of CO_2. For Fischer Tropsch, the H_2/CO ratio could be increased or decreased most commonly to 1.7 or 2.15 respectively. A ratio of 1.7 will result in a decrease of 9% in the CI to 6.9 gCO$_2$e/MJ fuel, and a ratio of 2.15 will result in an increase of 4% in the CI to 7.9 gCO$_2$e/MJ fuel.

Other parameters (construction and decommissioning, compression, catalyst production or use, CE's scenario A, and FTS operating conditions) have negligible impacts on the CI.

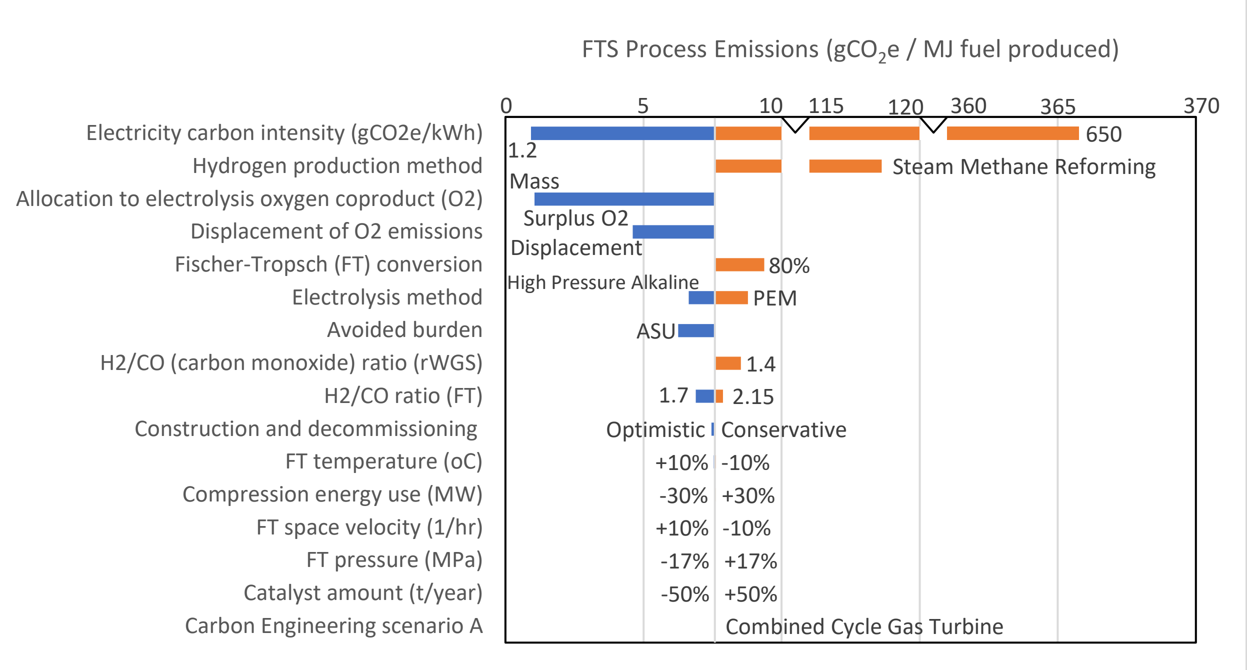

Figure S4: Sensitivity analysis of various parameters on the FTS process emissions on a per MJ fuel produced basis. Base case assumptions can be found in Table 1 (main paper).

References

1 D. W. Keith, G. Holmes, D. St. Angelo and K. Heidel, *Joule*, 2018, **2**, 1573–1594.

2 E. and C. C. Canada, Canada’s greenhouse gas inventory, https://www.canada.ca/en/environment-climate-change/services/climate-change/greenhouse-gas-emissions/inventory.html, (accessed March 8, 2019).

3 S. C. Government of Canada, Data, https://www150.statcan.gc.ca/n1/en/type/data, (accessed March 8, 2019).

4 G. Wernet, C. Bauer, B. Steubing, J. Reinhard, E. Moreno-Ruiz and B. Weidema, *Int. J. Life Cycle Assess.*, 2016, **21**, 1218–1230.

5 R. Bhandari, C. A. Trudewind and P. Zapp, *J. Clean. Prod.*, 2014, **85**, 151–163.

6 A. I. Racoviceanu, B. W. Karney, C. A. Kennedy and A. F. Colombo, *J. Infrastruct. Syst.*, 2007, **13**, 261–270.

7 E. Senobari Vayghan, Thesis, University of Calgary, 2016.

8 C. van der Giesen, R. Kleijn and G. J. Kramer, *Environ. Sci. Technol.*, 2014, **48**, 7111–7121.

9 Aspen Plus, https://www.aspentech.com/en/products/engineering/aspen-plus, (accessed March 14, 2019).

10 M. Mansouri, H. Atashi and A. A. Mirzaei, *J. Thermodyn. Catal.*, , DOI:10.4172/2157-7544.1000113.

11 C. M. University, CMU - Economic Input-Output Life Cycle Assessment - Carnegie Mellon University, http://www.eiolca.net/index.html, (accessed March 8, 2019).

12 K. A. Kobe, *J. Chem. Educ.*, 1958, **35**, A506.

13 G. Schivley, I. Azevedo and C. Samaras, *Environ. Res. Lett.*, 2018, **13**, 064018.

14 P. L. Spath and M. K. Mann, *Life Cycle Assessment of Hydrogen Production via Natural Gas Steam Reforming*, 2000.