

Utilising carbon dioxide for transport fuels: The economic and environmental sustainability of different Fischer-Tropsch process designs

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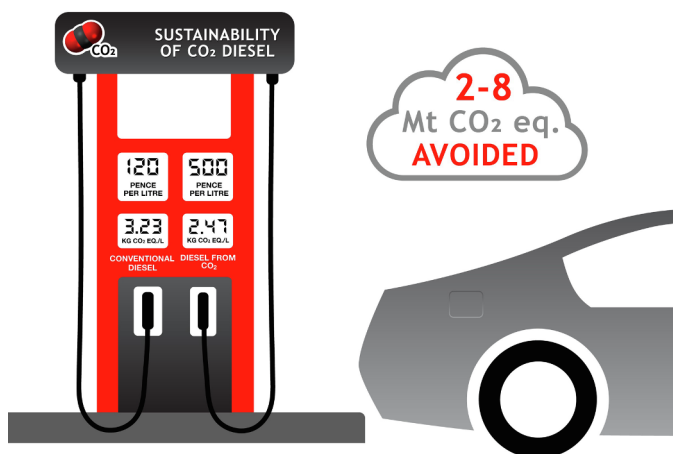
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HIGHLIGHTS

- For large-scale plants, CO₂-derived fuels outperform diesel environmentally.
- The exceptions are global warming and ozone depletion which are lower for diesel.
- Optimising the systems reduces global warming of CO₂ fuels by 70% below diesel.
- CO₂ fuels are not economically viable, costing ~4 times more than fossil diesel.
- Optimising process yields would allow decreasing the subsidies to 8%.

GRAPHICAL ABSTRACT



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ABSTRACT

Producing fuels and chemicals from carbon dioxide (CO₂) could reduce our dependence on fossil resources and help towards climate change mitigation. This study evaluates the sustainability of utilising CO₂ for production of transportation fuels. The CO₂ feedstock is sourced from anaerobic digestion of sewage sludge and the fuels are produced in the Fischer-Tropsch (FT) process. Using life cycle assessment, life cycle costing and profitability analysis, the study considers four different process designs and a range of plant capacities to explore the effect of the economies of scale. For large-scale plants (1,670 t/day), the FT fuels outperform fossil diesel in all environmental impacts across all the designs, with several impacts being net-negative. The only exceptions are ozone depletion, for which fossil diesel is the best option, and global warming potential (GWP), which is lower for fossil diesel for some process designs. Optimising the systems reduces the GWP of FT fuels in the best case by 70% below that of fossil diesel. Assuming a replacement of 9.75–12.4% of fossil diesel consumed in the UK by 2,032, as stipulated by policy, would avoid 2–8 Mt of CO₂ eq./yr, equivalent to 2–8% of annual emissions from transportation. However, these fuels are not economically viable and matching diesel pump price would require subsidies of 35–79% per litre. Optimising production yields would allow decreasing the subsidies to 8%. Future research should be aimed at technology improvements to optimise these systems as well as evaluating different policy mechanisms needed to stimulate markets for CO₂-derived fuels.

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Nomenclature			
$A_{j, \text{ avg}}$	annual cash flow (£/y)	N	evaluation period (years)
A_x	non-manufacturing fixed-capital investment (£)	n	length of the straight-line recovery period
C_1	capacity of a base-case plant (t/day)	NP_{avg}	average annual net profit over the lifetime of the plant (£/yr)
C_2	capacity of a scaled-up plant (t/day)	p	basic price of liquid fuels (pence/l)
$C_{O\&M\text{avg}}$	average annual operating and maintenance costs (£/yr)	PBP	payback period (yr)
d	annual depreciation (£/y)	S_{avg}	average annual revenue from fuel sales over the lifetime of the plant (£/yr)
GP_{avg}	average annual gross profit over the lifetime of the plant (£/yr)	TCI	total capital investment (£)
m	annual production of liquid fuels (l/yr)	TCI_1	total capital investment for a base-case plant (£)
MARR	minimum annual acceptable rate of return on investment (%/yr)	TCI_2	total capital investment for a scaled-up plant (£)
		V	manufacturing fixed-capital investment (£)
		Φ	income tax (%)

1. Introduction

Our dependence on fossil fuels is depleting abiotic resources and contributing to climate change [1]. While substantial reductions in consumption are paramount, research efforts to produce fuels from alternative feedstocks must continue. One such feedstock is carbon dioxide (CO_2). While CO_2 is already being utilised for production of chemicals, such as urea, methanol and polyols [2,3], the overall consumption of CO_2 as a feedstock by the chemicals industry is below 1% of the 49.3 Gt/yr of anthropogenic CO_2 emitted globally [4]. Therefore, CO_2 utilisation strategies should encourage and prioritise the production of products with high demand, such as transportation fuels [5]. This paper focuses on utilisation of CO_2 to produce liquid transport fuels via Fischer-Tropsch (FT) synthesis with the aim of finding out if such fuels would be environmentally and economically sustainable on a life cycle basis relative to fossil and renewable fuels.

Production of liquid fuels via FT synthesis is a well-established practice in industry. However, the feedstocks are largely derived from fossil resources, such as coal or natural gas, particularly in countries with limited reserves of crude oil [6,7]. For example, Sasol in South Africa produces synthetic liquid fuels from coal [8], while ExxonMobil in Qatar [7] and Shell in Malaysia rely on their large natural gas reserves for the production of liquid fuels [5]. In FT synthesis, syngas (carbon monoxide and hydrogen) is converted into a mixture of gaseous and liquid hydrocarbon fuels through a catalyst-aided polymerisation reaction [9]. Although syngas is commonly produced from coal gasification or steam reforming of natural gas, alternative feedstocks, such as biomass or CO_2 , can be used for the production of syngas [5,6,9].

2. Literature review

Over the last decade, several studies evaluated techno-economic feasibility and/or environmental performance of the production of FT liquid fuels from different fossil and renewable feedstocks. For an overview of these studies, see Table 1. Dry [6] and Jaramillo et al. [7] compared the economic viability of different pathways for producing FT fuels from coal and natural gas in South Africa and the United States, respectively. Both studies concluded that the profitability of FT plants depended highly on crude oil prices, carbon taxes and feedstock prices. For example, it was found that FT fuels from natural gas could only compete with conventional fossil fuels at low gas prices (\$0.5 per million cubic feet) and high crude oil prices (\$120 per barrel). Conversely, at low crude oil prices (\$40 per barrel), neither coal-nor gas-derived FT fuels were economically competitive. On the other hand, Jaramillo et al. [7] found that both types of FT fuels could achieve greenhouse gas emissions (GHG) comparable to fossil fuels (87 vs 85 g CO_2 eq./MJ), but only under very optimistic scenarios, involving carbon capture and

storage and low-carbon electricity sources. Most recently, Liu et al. [10] evaluated the economic and GHG emissions of coal and biomass to liquid (CBTL) processes with carbon capture and storage (CCS) for the production of liquid fuels. According to this study, both price and GHG emissions highly depended on regional biomass availability as this determined the optimum biomass-to-coal ratio. It was determined that the ratio of 8/92 provided the best balance between price and GHG reductions estimated at 56 pence per litre and 93.6 g CO_2 eq./MJ, respectively [10].

Although FT fuels from renewable feedstocks were found generally to be more favourable than from fossil resources with respect to life cycle GHG emissions, the latter ranged widely from −20 to 150 g CO_2 eq./MJ of fuel, depending on the unit of analysis, system boundaries, assumptions and allocation methods. For example, some studies excluded transport and pre-treatment of the feedstock [11], while others excluded the use (combustion) of fuels [12].

Some studies considered specifically techno-economic feasibility and/or environmental implications of CO_2 -derived liquid fuels. Those most relevant to this work are summarised in Table 2; for further details on the production pathways and costs, see Graves et al. [13] and Brynolf et al. [14], respectively. For example, Abanades et al. [15] assessed the avoided CO_2 emissions and related mitigation costs of a CO_2 -to-methanol process. The study showed that this particular carbon capture and utilisation (CCU) system was no better than producing methanol from fossil resources in terms of climate change mitigation and was even more costly. On the other hand, Deutz et al. [16] found that blending diesel with CO_2 -derived oxymethylene ethers could reduce the GHG emissions from transport fuels by 22% if hydrogen was produced via water electrolysis using renewable energy. In addition to GHG emissions, this study also estimated the emissions of NO_x and particulates from fuel combustion. Hombach et al. [17] also looked at the production of hydrogen via two different water electrolysis technologies using fossil and renewable energy for the production of FT liquid fuel from CO_2 captured from air considering current (2015) and future projections (2030). The estimated global warming potential (GWP) varied between 64 and 440 g CO_2 eq. per MJ of FT liquid fuels for the current situation, and 7–150 g CO_2 eq./MJ for the future scenario, depending on the electrolysis technology and energy source. The production costs for all cases were found to be 3–5 times higher than the pump price of fossil diesel in Germany. Van der Giesen et al. [18] also estimated the GWP of the production of FT liquid fuels from CO_2 captured from three different sources: combustion of woodchips, combustion of natural gas and direct air capture. Similarly to Hombach et al. [17], the results ranged from 30 to 290 g CO_2 eq. per MJ of fuels, depending on the CO_2 and energy sources. Overall, the direct air capture process powered by solar electricity was the best option with respect to the GWP. Solar energy was also considered in a techno-economic

Table 1
Summary of economic and environmental assessment studies of Fischer-Tropsch (FT) liquid fuels.

Study	Country	Scope	Feedstock	Unit of analysis	System boundaries	Economic assessment	Environmental assessment
Dry [6]	South Africa	Review of FT processes for the production of diesel and the economic viability of existing FT plants	Coal and natural gas	USD per barrel	Cradle to gate	Economic viability of existing FT plants	-
Jaramillo et al. [7]	USA	Environmental and economic assessment of production of FT liquid fuels	Coal and natural gas	Litres of fuel	Cradle to gate	Techno-economic assessment of CTL and GTL plants ^a	Life cycle GHG emissions
Wang et al. [12]	USA	Environmental and economic optimisation of hydrocarbon biorefinery for production of FT liquid fuels	Cellulosic biomass	Gasoline equivalent gallons	Gate to gate	Net present value of a biorefinery	Global warming potential
Pressley et al. [22]	USA	Environmental assessment of production of FT liquid fuels in a refuse-derived fuel facility	Municipal solid waste	One metric tonne of mixed waste	Cradle to gate	-	Global warming potential
Ahmadi Moghaddam et al. [11]	Sweden	Comparison of environmental performance of alternative biogas-derived fuels, including FT diesel	Biogas	Normal cubic meters of raw biogas	Gate to gate (excludes anaerobic digestion for biogas production)	-	Global warming potential
Petersen et al. [23]	South Africa	Comparison of environmental performance of alternative biomass-derived fuels, including FT diesel	Sugarcane bagasse	Fuel required to travel 1 km	Cradle to gate	-	Global warming potential, depletion of abiotic resources, acidification, eutrophication, human toxicity
Liu et al. [10]	USA	Environmental and economic feasibility study of CBTL processes with CCS ^b	Coal and biomass	1,000 MJ energy equivalent	Cradle to gate	Required selling price	Global warming potential, blue water consumption, fossil energy usage

^a CTL: coal to liquid; GTL: gas to liquid.

^b CBTL: Coal and biomass to liquids. CCS: Carbon capture and storage.

analysis of the production of FT liquid fuels in comparison with production of methanol from fossil-derived CO₂ [19]. The results indicated that the capital investment and operating and maintenance costs required for the FT plant were 2.3 times lower than for the methanol plant.

Furthermore, three studies assessed the economic viability of using biogas from sewage sludge to produce FT liquid fuels [5,20,21]. However, they varied in scope. For example, Badgett et al. [20] estimated the potential feedstock prices of various organic waste sources, including sewage sludge, based on the associated costs of their treatment and disposal. The study concluded that sewage sludge could be available at low or negative prices depending on local regulations. Hernandez and Martin [21] went beyond the feedstocks to determine the costs of producing FT fuels from sewage-derived biogas. The authors suggested that the production costs could potentially compete with current selling prices of fossil fuels if 'sustainability tax allowances' were introduced. Dimitriou et al. [5] also considered costs of producing FT fuels from biogas, comparing different process configurations on fuel yields, energy requirements, capital investment and production costs. The results suggested that the production of liquid fuels via FT was not economically feasible, mainly because of the low CO₂ separation and conversion efficiencies as well as the high energy requirements [5]. However, the study did not consider full life cycle costs, focusing only on the production costs.

Similarly, none of the previous studies considered the full life cycle costs or environmental impacts either. Furthermore, no study of FT fuels derived from CO₂ considered any other environmental impacts beyond the GWP (Table 2). This work goes beyond the state-of-the-art to provide a comprehensive evaluation of the life cycle economic and environmental sustainability of the production and consumption of FT liquid fuels derived from CO₂, with the aim of identifying the most promising designs for potential deployment. Life cycle assessment (LCA), life cycle costing (LCC) and profitability analysis have been used for these purposes. The last goes beyond production costs to estimate potential selling prices of fuels. Four representative FT process designs are considered, using CO₂ emitted in anaerobic digestion of sewage sludge during production of biogas, therefore, exploring an alternative and possibly a more profitable application of sludge, which is currently used only as fertiliser. This is described in the next section, alongside the data and assumptions used in the study. The results of the environmental and economic evaluations are presented in Section 4, together with the scale-up of the plants and various sensitivity analyses. Possible policy implications related to these fuels are also considered in this section. The conclusions and recommendations for policy and future work are given in Section 5.

3. Methods

3.1. Process description

This study considers four process designs (PD) for the production of FT fuels from sewage sludge proposed previously by the authors [5]. All designs rely on the best available and proven technologies for CCU and are ready for deployment. They differ in the methods used to separate and capture the CO₂ present in the biogas, as follows:

- PD-MEA: CO₂ capture from raw ('unsweetened') biogas by absorption in monoethanolamine (MEA);
- PD-CHP1: combustion of unsweetened biogas in a combined heat and power (CHP) and utilisation of CO₂ present in flue gases;
- PD-CHP2 (combination of PD-MEA and PD-CHP1): MEA CO₂ capture from unsweetened biogas and utilisation of CO₂ from flue gases generated in combustion of sweetened biogas in a CHP plant; and
- PD-CHP3: MEA CO₂ capture from flue gas generated by combustion of unsweetened biogas in a CHP plant.

Table 2
Summary of economic and environmental assessment studies of CO₂-derived liquid fuels.

Study ^a	Scope	CO ₂ source	Fuel	Unit of analysis	System boundaries	Economic assessment	Environmental assessment
Van der Giesen et al. [18]	Environmental assessment of synthetic hydrocarbon fuels produced from various CO ₂ sources using fossil and renewable energy	Natural gas and biomass combustion and direct air capture	FT ^c liquid fuels	1 MJ of liquid fuels	Cradle to grave	–	Global warming potential
Dimitriou et al. [5]	Identification of most promising process configuration for the conversion of CO ₂ to fuels	Biogas from sewage sludge	FT ^c liquid fuels	Annual production of liquid fuels	Cradle to gate	Techno-economic analysis, production costs	–
Abanades et al. [15]	Assessment of mitigation potential of carbon capture and utilisation processes	Industrially sourced CO ₂	Methanol	kilogrammes of fuel	Cradle to grave	Preliminary assessment of mitigation costs	Global warming potential
Deutz et al. [16]	Environmental assessment of diesel fuel blends with CO ₂ -derived OME ^b produced using renewable energy	Biogas and direct air capture	Dimethoxymethane (OME ₁)	1 km driving a passenger vehicle	Cradle to grave	–	Global warming potential, NOx and particulate emissions
Hernandez and Martin [21]	Optimisation of biogas-derived fuels via tri-reforming	Various sources of biogas incl. sewage sludge	FT ^c liquid fuels	Annual production of fuels	Cradle to gate	Economic evaluation, production costs	–
Badgett et al. [20]	Economic analysis of waste as feedstock for production of transportation fuels, chemicals and power	Sewage sludge, manure and food waste	Biofuels	Wet metric tonne	–	Feedstock prices	–
Hombach et al. [17]	Economic and environmental assessment of synthetic fuels derived from water and CO ₂ via current (2015) and future (2030) technologies	Direct air capture	FT ^c liquid fuels	1 MJ of liquid fuels	Cradle to grave	Production cost	Global warming potential
Tsongidis et al. [19]	Techno-economic evaluation of various pathways for the production of CO ₂ -derived fuels using solar energy	Natural gas and coal combustion	FT ^c liquid fuels and methanol	Annual production of fuels	Cradle to gate	Techno-economic analysis	–
This study	Life cycle environmental and economic sustainability evaluation of FT ^c fuels from CO ₂	Biogas from sewage sludge	FT ^c liquid fuels	1 litre of liquid fuels; 1 MJ of liquid fuels	Cradle to grave	Life cycle costing, profitability analysis, basic and pump price	Global warming potential and ten other env impacts (see Section 3.1)

^a All studies are based in the European Union.

^b OME: Oxymethylene ethers.

^c Fischer-Tropsch.

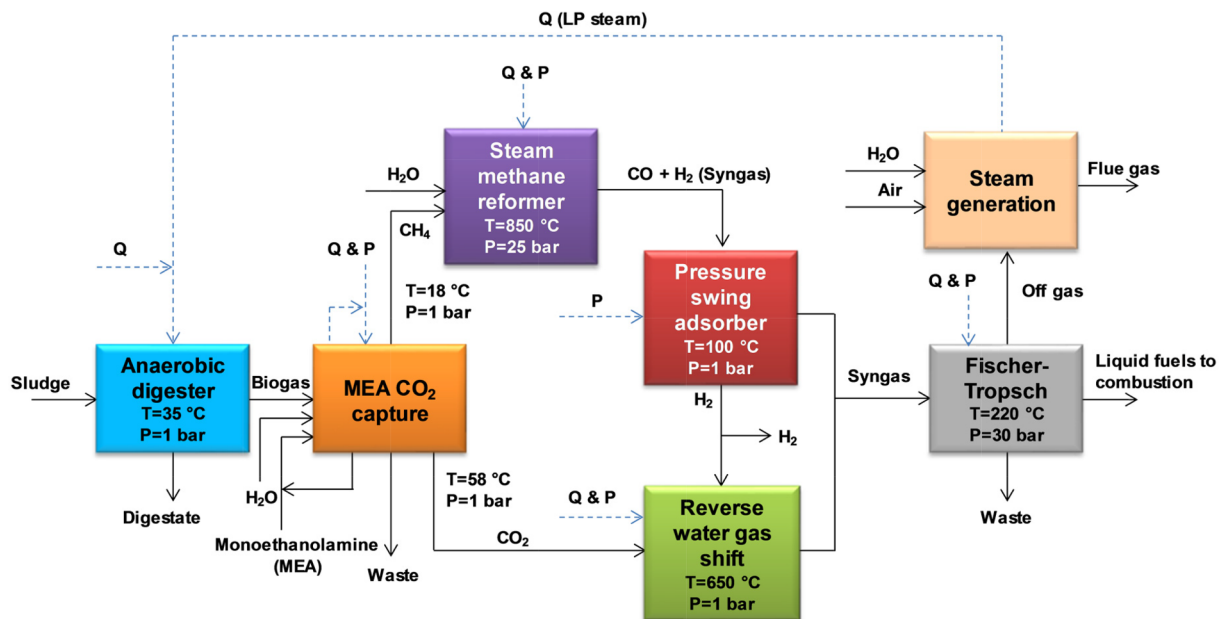


Fig. 1. Process flow diagram for PD-MEA. [Q: heat; P: power; LP: low pressure; — mass flows; — energy flows.]

A schematic overview of each design is given in Figs. 1–4. All four systems comprise an anaerobic digester to produce unsweetened biogas from sewage sludge. The anaerobic digester is operated at 35 °C and 1 bar, with a residence time of 15 days. This is followed by CO₂ capture, syngas production and low-temperature FT synthesis, respectively to produce a mixture of gaseous and liquid fuels. Due to a higher selectivity for heavier hydrocarbons in low-temperature FT synthesis [6], diesel is the predominant liquid fuel produced. The four conceptual designs, simulated through Aspen Plus [24], are described briefly in the following sections; for a more detailed description, see Dimitriou et al. [5].

3.1.1. PD-MEA

As shown in Fig. 1, in this design, CO₂ present in the biogas is captured by absorption in MEA. The CO₂ is then released from the amine solution using a stripping column and the regenerated MEA is recycled to the absorber for re-use, with 99.997% of MEA recovered. The upgraded biogas

and water react in a steam-methane reformer to produce syngas at 850 °C and 25 bar. The excess amount of hydrogen produced in the reformer is recovered using a pressure swing adsorption (PSA) unit similar to the Linde PSA technology. The process operates at 30 °C and 40 bar, which achieves an 85% hydrogen recovery with a purity of 99.999%, whilst the adsorbent is regenerated by lowering the pressure to near atmospheric [5]. The recovered hydrogen is reacted with CO₂ in a reverse-water gas shift (RWGS) reactor to produce syngas at 650 °C and 1 bar. The excess water is removed from the syngas in a condensation unit placed before PSA (in all four designs). However, the nitrogen is not removed from the syngas as the FT process can cope with high nitrogen content [25,26]. All syngas is converted into a mixture of gaseous and unrefined liquid fuels via FT synthesis which takes place at 220 °C and 30 bar. Such relatively low temperatures and high operating pressures, along with a H₂:CO molar ratio of 2:1, favour the production of FT liquid fuels [5]. The gaseous fuels are combusted to produce low-pressure steam, which is used to heat the anaerobic digester.

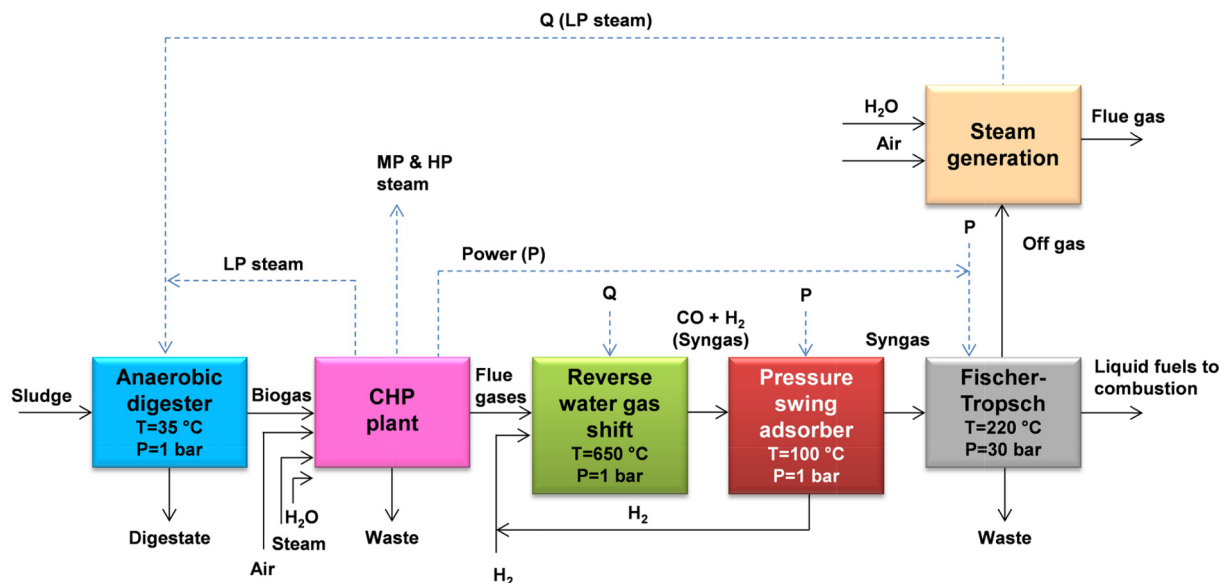


Fig. 2. Process flow diagram for PD-CHP1. [Q: heat; P: power; LP: low pressure; MP: medium pressure; HP: high pressure; — mass flows; — energy flows.]

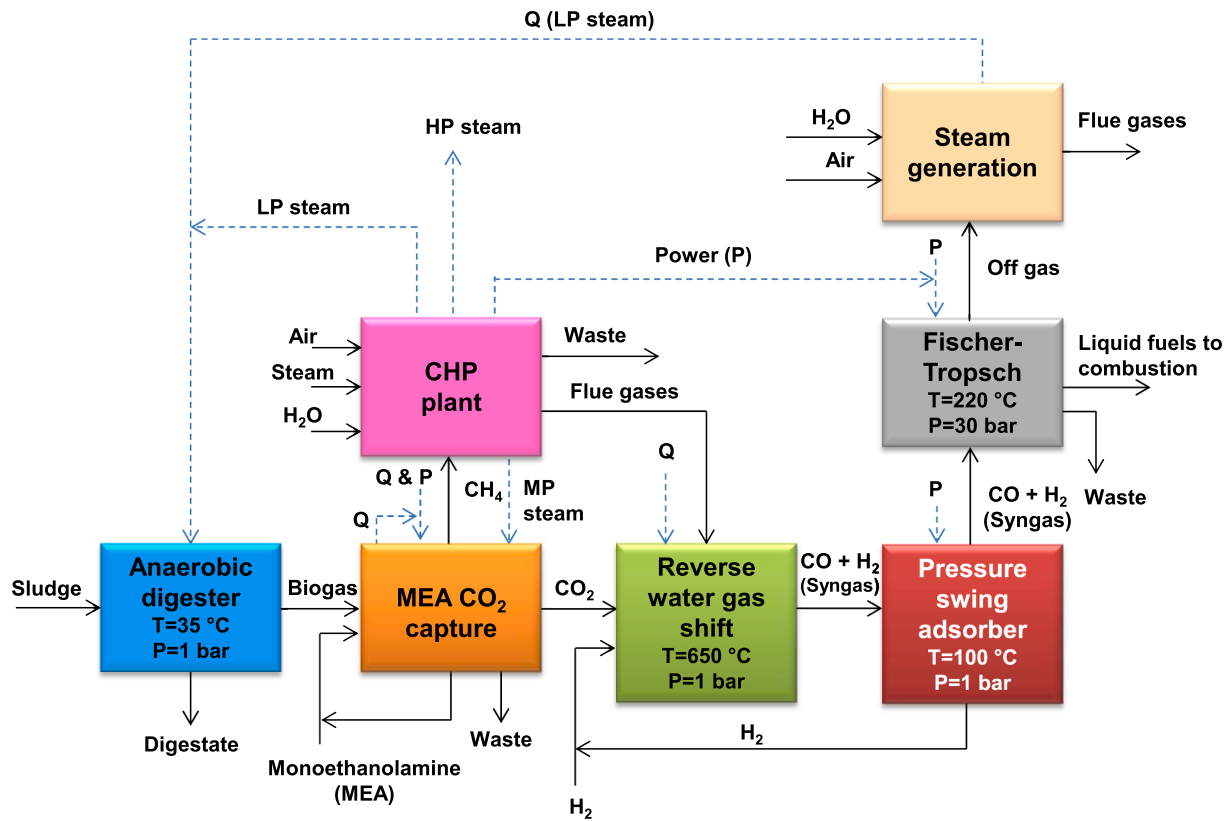


Fig. 3. Process flow diagram for PD-CHP2. [Q: heat; P: power; LP: low pressure; MP: medium pressure; HP: high pressure; — mass flows; — energy flows.]

3.1.2. PD-CHP1

In PD-CHP1, untreated biogas is used in a CHP plant to produce electricity and heat which are used within the CCU process (Fig. 2). The

flue gases, containing CO_2 , air and water vapour, react with hydrogen in a RWGS reactor to produce syngas at 650 °C and 1 bar. As in PD-MEA, the excess hydrogen is recovered via PSA and reused in the RWGS

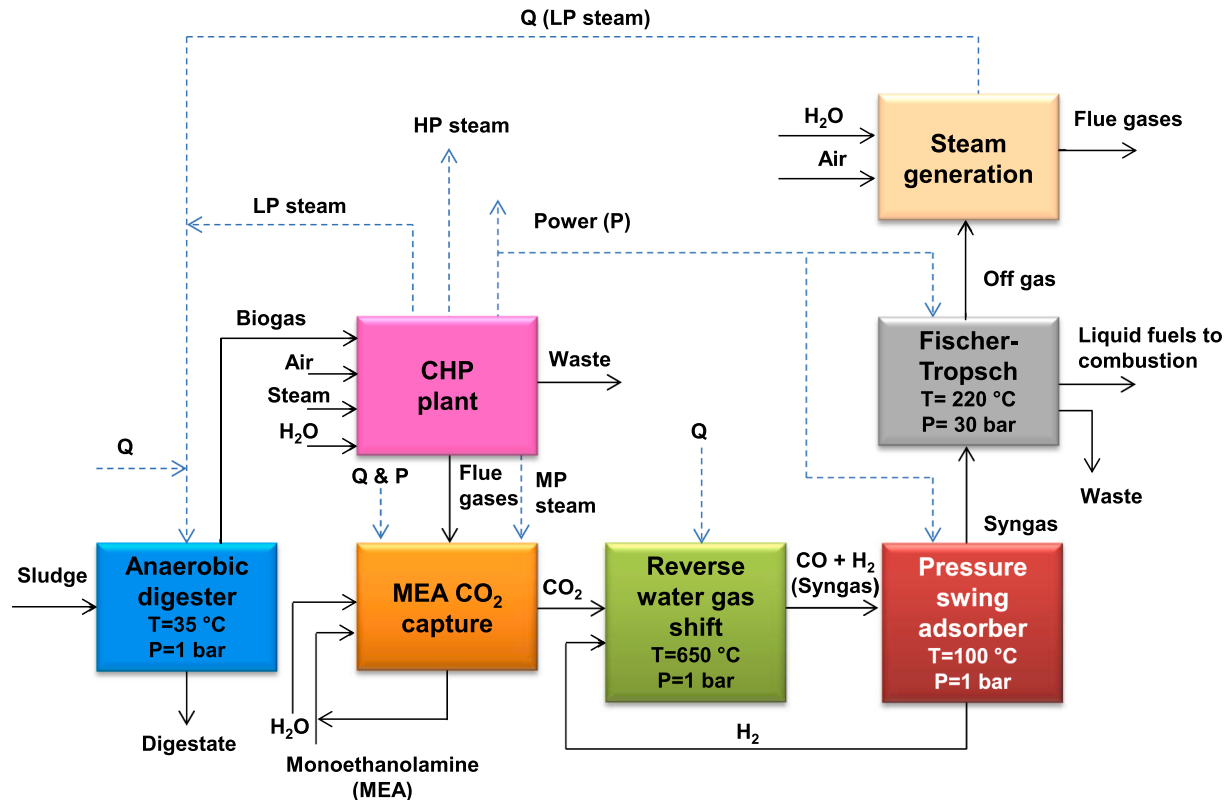


Fig. 4. Process flow diagram for PD-CHP3. [Q: heat; P: power; LP: low pressure; MP: medium pressure; HP: high pressure; — mass flows; — energy flows.]

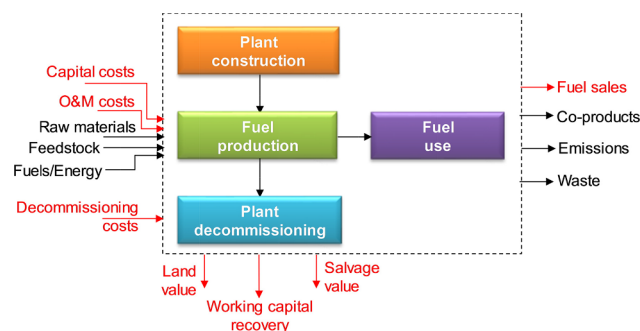


Fig. 5. System boundaries for the estimation of life cycle environmental impacts and costs of Fischer-Tropsch liquid fuels produced from CO₂. [For the details of fuel production, see Figs. 1–4. Transport of raw materials not shown for simplicity. The black font and arrows denote the flows considered in the estimation of environmental impacts and the red refer to the costs and revenues included in the economic evaluation. O&M costs: operating and maintenance costs.]

reactor. The syngas is converted into gaseous and liquid fuels in the FT unit and the gaseous fuels are used to heat the anaerobic digester.

3.1.3. PD-CHP2

PD-CHP2 combines the previous two designs, utilising CO₂ from two sources: unsweetened biogas and flue gas generated in combustion of sweetened biogas (Fig. 3). The CO₂ in the raw biogas is first captured in MEA and then released using the same process as in PD-MEA. The upgraded biogas is then combusted in a CHP plant and the CO₂ in the resulting flue gas reacted with hydrogen to produce syngas following the method in PD-CHP1.

3.1.4. PD-CHP3

This design assumes that the untreated biogas is combusted in a CHP plant and the CO₂ in the flue gas is then captured in the MEA unit. After recovery in a stripping column, CO₂ is used in the RWGS reactor to produce syngas (Fig. 4). The rest of the process is identical to PD-CHP1.

3.2. Study scope and inventory data

The following life cycle stages are considered in both the environmental and cost analyses (Fig. 5): transportation of sewage sludge and other raw materials to the FT plant, capture and utilisation of CO₂ from biogas to produce liquid fuels in the four designs described previously, use of fuels for road transport and plant construction and decommissioning at the end of its lifetime. The latter is assumed at 20 years in the base case but the effect of different plant lifetimes is explored through a sensitivity analysis. The process for refining the produced liquid fuels is not considered as it is the same across the different process designs considered. This is not expected to influence the outcomes of the study as previous studies [11,22,23] found that the contribution of energy consumption in the refining process was below 1%.

The unit of analysis (functional unit) is defined as ‘production and consumption of 1 litre of FT liquid fuels derived from CO₂’. All four process designs are assumed to use the same amount of sewage sludge to obtain the CO₂, but they produce different amounts of liquid fuels due to different configurations and efficiencies (Table 3).

The inventory data are summarised in Tables 4 and 5, expressed per litre of liquid fuels. These have been estimated from the material and energy balances obtained through Aspen simulation, based on the annual operation of the plant [5]. The transport distances for the sludge have been assumed at 100 km and for hydrogen and MEA at 50 km. The life cycle inventory data for the materials, energy, transport and other background systems used for LCA modelling have been sourced from Ecoinvent 2.2 [27]. The production process is assumed to be based in the UK.

Table 3

The amounts of liquid fuel produced in different designs [5].

Production	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
Hourly production (kg/h)	43.23	33.95	33.79	29
Hourly production (l/h) ^a	50.9	41.6	41.3	34.9
Annual production (l/yr) ^b	407,350	332,436	330,061	278,846
Lifetime production (million litres) ^c	8.147	6.649	6.601	5.577

^a Estimated using fuel densities obtained from Aspen simulations, as follows: PD-MEA: 0.849 kg/l; PD-CHP1: 0.817 kg/l; PD-CHP2: 0.819 kg/l; PD-CHP3: 0.832 kg/l. The densities differ across the designs due to slight differences in the composition of the produced liquid fuels.

^b Based on 8000 h of operation.

^c Based on the plant lifetime of 20 years.

As can be observed in Table 4, PD-MEA and PD-CHP3 produce enough hydrogen to satisfy own demand, while the other two designs require additional hydrogen. This has been assumed to be produced elsewhere by steam reforming of natural gas [28]. The heat demand is met via the CHP plant, combustion of off-gases and steam from natural gas. Electricity is sourced from the CHP plant and the grid, except for PD-MEA, where the electricity demand is fully met by the grid as this option does not include a CHP plant. The UK electricity mix in 2017 [29] has been used to estimate the life cycle impacts of grid electricity; for details, see Figs. S1 and S2 in the SI.

As indicated in Table 5, all four systems produce different co-products for which they have been credited for avoiding environmental impacts from products made in other production systems. It has been assumed that the nutrients in the digestate displace an equivalent amount of industrially-produced fertilisers (see Section S2 in the SI). For the three CHP-based systems, the excess steam and electricity produced in the CHP plant is assumed to replace the equivalent amounts of steam from natural gas and grid electricity. The systems have also been credited for the recycling of construction materials remaining after the decommissioning of the plant. The quantities of the end-of-life materials for different plant capacities are specified in

Table 4

Materials and energy used in the production of Fischer-Tropsch liquid fuels.

Inputs	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
Raw materials (kg/litre)				
Sewage sludge (wet) ^a	218.2	267.4	269.3	318.8
Biogas ^b	4.21	5.16	5.20	6.15
CO ₂ ^b	2.5	3.1	3.1	3.7
Monoethanolamine ^c	2.92x10 ⁻⁴	0	3.68x10 ⁻⁴	3.41x10 ⁻³
Hydrogen (total)	0.236	1.20	1.21	0.344
Produced internally ^d	0.236	0.41	0.42	0.344
Produced externally ^e	0	0.79	0.79	0
Water	22	28	28	26
Steam ^f	0	23	23	27
Process energy (MJ/litre)				
Heat (total)	78	95	104	108
Steam produced internally ^g	29	56	54	40
Natural gas supplied externally	49	40	50	68
Electricity (total)	15	50	51	13.7
Produced internally ^h	0	15	18	13.6
Grid electricity	15	35	33	0.1

^a Based on the amount of sludge of 11,111 kg/h.

^b Based on 214.44 kg of biogas in 11,111 kg of sludge and the content of CO₂ in biogas of 60%.

^c Fresh and make-up.

^d Excess hydrogen recovered by pressure swing adsorption.

^e Produced by steam reforming of natural gas.

^f Steam used as a reactant in the process, produced from natural gas and heavy fuel oil.

^g Steam produced in CHP plant and off-gas burner.

^h Power produced in the CHP plant.

Table 5
Co-products and waste generated in the production of CO₂ Fischer-Tropsch liquid fuels.

Outputs	Displaced product	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
<i>Co-products</i>					
Digestate ^a (kg/litre)	Nitrogen fertiliser (as N)	0.35	0.43	0.44	0.52
	Phosphate fertiliser (as P ₂ O ₅)	0.63	0.77	0.78	0.92
	Sulphur fertiliser (as SO ₄)	0.37	0.46	0.46	0.55
Hydrogen (kg/litre)	Hydrogen ^b	0.016	–	–	–
Medium-pressure steam (MJ/litre)	Steam ^c	–	1.44	–	–
High-pressure steam (MJ/litre)	Grid electricity	–	1.97	1.80	2.35
Electricity (MJ/litre)	Grid electricity	–	–	–	1.23
<i>Waste</i>					
Liquid waste (kg/litre) ^d	–	3.7	31	32	36

^a For calculations, see Tables S1–S4 and the calculations in Section S2 in the Supplementary Information.

^b From steam reforming of natural gas.

^c From natural gas.

^d Treated as industrial wastewater.

Table 6
Recyclable construction materials available after decommissioning of the plant.

Material	Displaced product	PD-MEA	PD-CHP plants	Medium-scale plant	Large-scale plant
Steel (t)	Virgin steel	373	299	22,300	33,600
Concrete (t)	Sand	115	923	69,000	104,000
	Gravel	115	923	69,000	104,000

Table 6, estimated using data in Ecoinvent as explained in Section 3.3.5.

Given that diesel is the predominant liquid fuel produced, all the impacts and costs have been allocated to it. In reality, other liquid fuels would also be produced in the refining process, including gasoline; however, due to a lack of data, it is not possible to speculate on their respective amounts.

The CO₂ emitted during the combustion of the fuel has not been considered as it is biogenic in nature (derived from sewage sludge). This is congruent with the ISO 14067 standard for estimating greenhouse gas emissions [30]. The impacts and costs of sewage sludge are also excluded as it is considered a waste material. However, the impacts of its transport are considered, together with the related economic costs, as explained in the next section.

3.3. Cost data and assumptions

The life cycle costs considered in this work are outlined in Fig. 5 and described in more detail in the following sections. All the data represent the ‘overnight’ costs with no discounting applied, following the method in Peters et al. [31].

3.3.1. Sewage sludge costs

The sludge is assumed to be free of charge but its transportation costs have been taken into account. These comprise fuel and labour costs given in Table 7. The fuel costs have been calculated based on the total amount of diesel used by a 27 t truck over 100 km, the distance assumed between the wastewater treatment and the FT plants. It has been assumed that 1.58 litre of diesel is needed to transport 1 t of sludge over 100 km [27] and the average price of diesel in 2017 was 120 pence per litre [32].

The costs of labour for transporting the sludge comprise wages and non-wage costs, such as sick leave, employer’s pension contributions, holidays and other employment overheads. The wages have been estimated based on the hourly rate for a large goods vehicle (LGV) driver [33] and the total hours worked over 20 years. An LGV driver earns on average £22,750/yr, equivalent to an hourly rate of £10.42 [33]. Non-wage costs represent 27% of the total labour costs [34] and are shown

Table 7
Costs of sludge transport over the plant lifetime of 20 years.^a

Item	All process designs ^b
Sludge consumption (t)	1,777,760
Diesel consumption (t)	2,384
Labour hours (hr)	68,586
Diesel costs (£)	3,377,823
Labour costs (£)	976,522
Wages (£)	714,442
Non-wage costs (£)	262,080
Total sludge costs (£)	4,354,345

^a 65,843 trips in a 27 t truck over 20 years @ 100 km each.

^b All process designs are assumed to consume the same amount of sludge but produce different amounts of liquid fuels (see Table 3).

in Table 7, together with the total sludge costs.

3.3.2. Fuel production costs

The production costs comprise capital, operating and maintenance costs as summarised in Table 8 for the four process designs [5]. The unit production costs, also shown in the table, are based on the total production of liquid fuels over the lifetime of the plant (for the latter, see Table 3).

3.3.3. Basic price of fuels and the payback period

To ensure the profitability of the plant and a minimum expected return on investment, it is necessary to determine the minimum price at which fuels need to be sold. This excludes the excise duty and VAT applied to road fuels [35] and is normally referred to as the ‘basic price’. The basic price has been estimated using the modified payback period (PBP) method that incorporates the minimum annual rate of return (MARR). The PBP method is widely used to determine the

Table 8
Production costs for the four process designs over the lifetime of the plant.^a

Costs	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
Total capital investment (M€)	30	32	34	36
Operating & maintenance costs (M€) ^b	102.4	126.4	132.4	132.4
Total production costs (M€)	132.35	158.35	166.35	168.35
Unit fuel production costs (£/l) ^c	16.2	23.8	25.2	30.2

^a Based on data from Dimitriou et al. [5], adjusted for costs of sludge transport not considered there.

^b Includes costs for sludge transport (see Table 7).

^c Based on the amount of liquid fuels produced over 20 years given in Table 3.

economic feasibility and compare different investment opportunities [31,36], which is the aim of this part of the study. The application of the PBP method is described below.

An initial payback period (PBP) is determined from a chosen minimum acceptable rate of return (MARR):

$$PBP = \frac{0.85}{MARR + \frac{0.85}{N}} \quad (\text{years}) \quad (1)$$

where:

PBP payback period (years).
MARR minimum acceptable rate of return (%/yr).
N evaluation period (yr).

Four values of MARR have been considered here (8%, 16%, 20% and 24%) to cover a range of risk levels, from low to high.

Using the PBP equation based on fixed capital investment and annual cash flow, the basic price can be estimated as follows:

$$PBP = \frac{V + A_x}{A_{j, \text{avg}}} \quad (\text{years}) \quad (2)$$

where:

V manufacturing fixed capital investment (£).
 A_x non-manufacturing fixed capital investment (£).
 $A_{j, \text{avg}}$ average annual cash flow (£/y).

The sum of the manufacturing fixed-capital investment (V) and non-manufacturing fixed-capital investment (A_x) represents the total fixed capital investment, assumed to be 95% of the TCI.

The average annual cash flow is equal to:

$$A_{j, \text{avg}} = (S_{\text{avg}} - C_{O \& M} + d\Phi) \times (1 - \Phi) + d\Phi \quad (\text{£/yr}) \quad (3)$$

where:

S_{avg} average annual revenue from fuel sales over the lifetime of the plant (£/yr).
 $C_{O \& M}$ average annual operating and maintenance costs (£/yr).
 Φ income tax (40%).
d annual depreciation (£/y).

The average annual revenue (S_{avg}) is equal to:

Table 9

Data for the estimation of the basic price of liquid fuels [5].

Parameter	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
TCI (M£)	30	32	34	36
$C_{O \& M}$ avg (M£/yr)	5.1	6.3	6.6	6.6
Φ (%)	40	40	40	40
m^a (l/yr)	407,350	332,436	330,061	278,846
V (M£)	20.349	21.706	23.062	24.419
A_x (M£)	8.151	8.694	9.238	9.781
N (yr)	20	20	20	20

^a Annual production based on the amount of liquid fuels produced over 20 years given in Table 3.

Table 10

End-of-life costs as a function of tangible fixed assets.

Assets and costs	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3	Source
Tangible fixed assets (M£) ^a	16.257	17.341	18.425	19.509	Dimitriou et al. [5]
Decommissioning and closure costs (M£) ^b	0.699	0.746	0.792	0.839	Hicks et al. [37]

^a 57% of the fixed capital investment shown in Table 9 [31].

^b 4.3% of the tangible fixed assets [37].

$$S_{\text{avg}} = p \times m \quad (\text{£/y}) \quad (4)$$

where:

p basic price of liquid fuels (£/l).
m annual production of liquid fuels (l/yr).

Therefore, by substituting Eqs. (3) and (4) into Eq. (2), and rearranging to solve for price (p), the basic price of fuels can be estimated as:

$$p = \frac{\frac{V + A_x}{PBP} - \frac{d\Phi}{(1 - \Phi)} + C_{O \& M} + M_{\text{avg}}}{m} \times 100 \quad (\text{pence/l}) \quad (5)$$

where:

100 factor to convert £ to pence.

The annual depreciation has been estimated using the straight-line method, as this is often used in profitability analyses that do not consider the time value of money [31]:

$$d = \frac{V}{n} \quad (\text{£/yr}) \quad (6)$$

where:

n length of the straight-line recovery period (9.5 years for petrochemical plants).

The variables in Eq. (5) are summarised in Table 9, based on the data in Dimitriou et al. [5].

3.3.4. Decommissioning costs

Decommissioning and closure costs include plant dismantling, site decontamination and remediation. It has been assumed that these costs correspond to 4.3% of the plant's tangible fixed assets, such as machinery, buildings and land [37]. The tangible fixed assets costs are equivalent to 57% of the fixed capital investment shown in Table 9 [31]. Both the tangible fixed assets and the related decommissioning costs are presented in Table 10.

A percentage of the initial investment can also be recouped from the recovery of the working capital [38] and from the sales of land and used equipment [31]. It has been assumed that the salvage value related to the latter amounts to 20% of the fixed-capital investment [24]. It has also been assumed that all the working capital is recovered and that the price of land corresponds to 2% of the total capital investment. The end-of-life revenue can be found in Table 11.

Table 11

End-of-life revenue [24,31,38]

Revenue	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
Salvage value (M£)	5.70	6.08	6.46	6.84
Working capital recovery (M£)	1.50	1.60	1.70	1.80
Land value (M£)	0.60	0.64	0.68	0.72
Total (M£)	7.80	8.32	8.84	9.36

Table 12
Scaled-up capacities and cost estimates for the four process designs.^a

	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
<i>Plant capacity (t liquid fuels/day)</i>				
Base case	1.04	0.81	0.81	0.70
Medium capacity	850	850	850	850
Large capacity	1,670	1,670	1,670	1,670
<i>Total capital investment costs (M£)</i>				
Base case	30	32	34	36
Medium capacity	1,685	2,073	2,210	2,572
Large capacity	2,527	3,108	3,314	3,856
<i>Operating & maintenance costs (M£/yr)^b</i>				
Base case	5.1	6.3	6.6	6.6
Medium capacity	449	559	583	680
Large capacity	758	945	980	1145

^a The medium and large capacities considered here are based on existing Fischer-Tropsch plants as reported in Dimitriou et al. [5]. They refer to the production of liquid fuels. Annual operation: 8,000 h.

^b Includes costs of sludge transport.

3.3.5. Plant scale-up

The production rates considered in this study correspond to a daily production of < 1 t of liquid fuels (see Table 12), which is significantly lower than the production rates of actual FT plants. For example, the Bintulu GTL plant in Malaysia produces 1670 t/day of liquid fuels [39]. Therefore, to provide more realistic estimates of costs and profitability, the production capacity of the process designs considered here have been scaled up to a medium (850 t/day) and large (1670 t/day) capacity plants using the six-tenths factor rule as follows [5]:

$$TCI_2 = TCI_1 \times \left(\frac{C_2}{C_1} \right)^{0.6} \quad (£) \quad (9)$$

where:

TCI_1 total capital investment for a base-case plant (£).

TCI_2 total capital investment for a scaled-up plant (£).

C_1 capacity of a base-case plant (t/day).

C_2 capacity of a scaled-up plant (t/day).

The scaled-up TCI costs of the four process designs are shown in Table 12, along with the operating and maintenance costs. As can be seen, all the scaled-up designs are assumed to have the same capacity but they retain the respective ratios between the parameters, including the feedstock requirements and fuels produced as in the base case.

The six-tenths factor rule has also been used to estimate the environmental impacts of the scaled-up designs, replacing the TCI in Eq. (9) with the life cycle environmental impacts associated with the construction of the production plants. As the impacts of constructing the FT plants are not available, data for constructing an organic chemical plant have been used instead, with an annual production capacity of 50,000 t. These data have been sourced from Ecoinvent [27] and the impacts of construction scaled to the small, medium and large capacities of the FT plants considered here. The estimated data for the amounts of construction materials needed for different capacities can be found in Table 6.

4. Results and discussion

4.1. Life cycle environmental sustainability

The LCA modelling has been carried out in GaBi V6.4 [40]. The CML 2001 method [41] has been used to estimate the following 11 environmental impacts included in this method: global warming potential (GWP), abiotic depletion of elements (ADP elements) and fossil resources (ADP fossil) potentials, acidification potential (AP), eutrophication potential (EP), human toxicity potential (HTP), ozone layer depletion potential (ODP), photochemical oxidants creation potential (POCP), freshwater aquatic ecotoxicity potential (FAETP), marine aquatic ecotoxicity potential (MAETP) and terrestrial ecotoxicity potential (TETP).

The results for the four process designs are shown in Figs. 6 and 7. The GWP is discussed first as mitigating climate change is the main driver for CCU. This is followed by a discussion of the other environmental impacts. The contributions of different life cycle stages

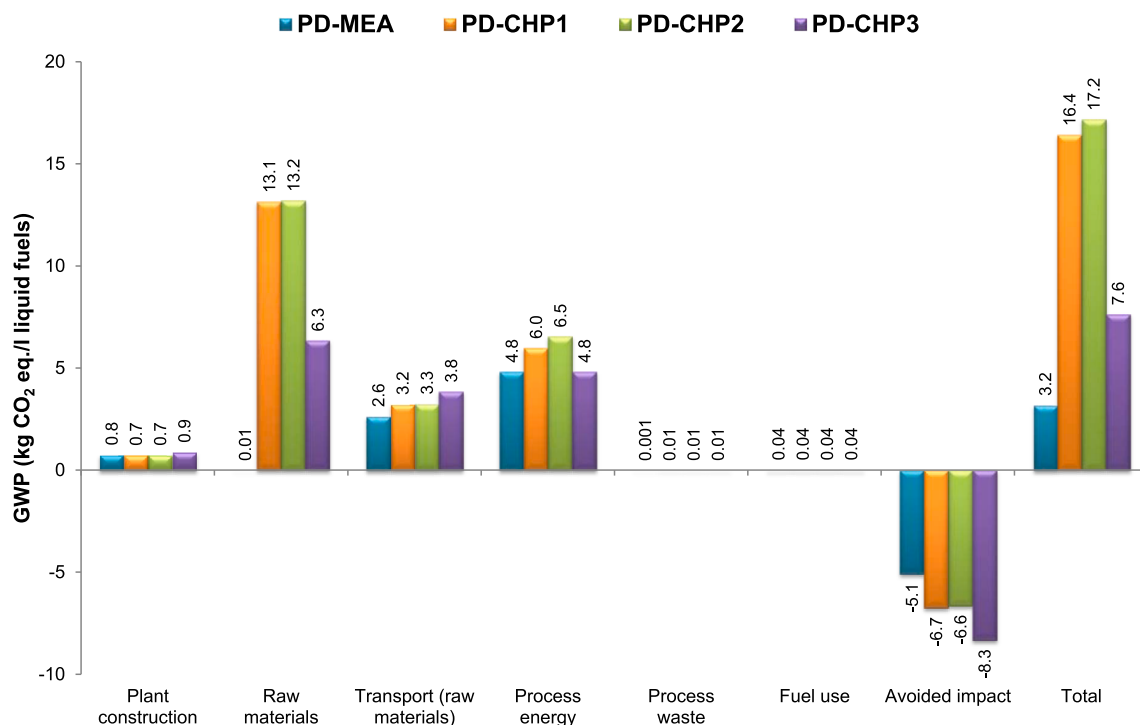


Fig. 6. Global warming potential of the four process design with the contribution of different life cycle stages.

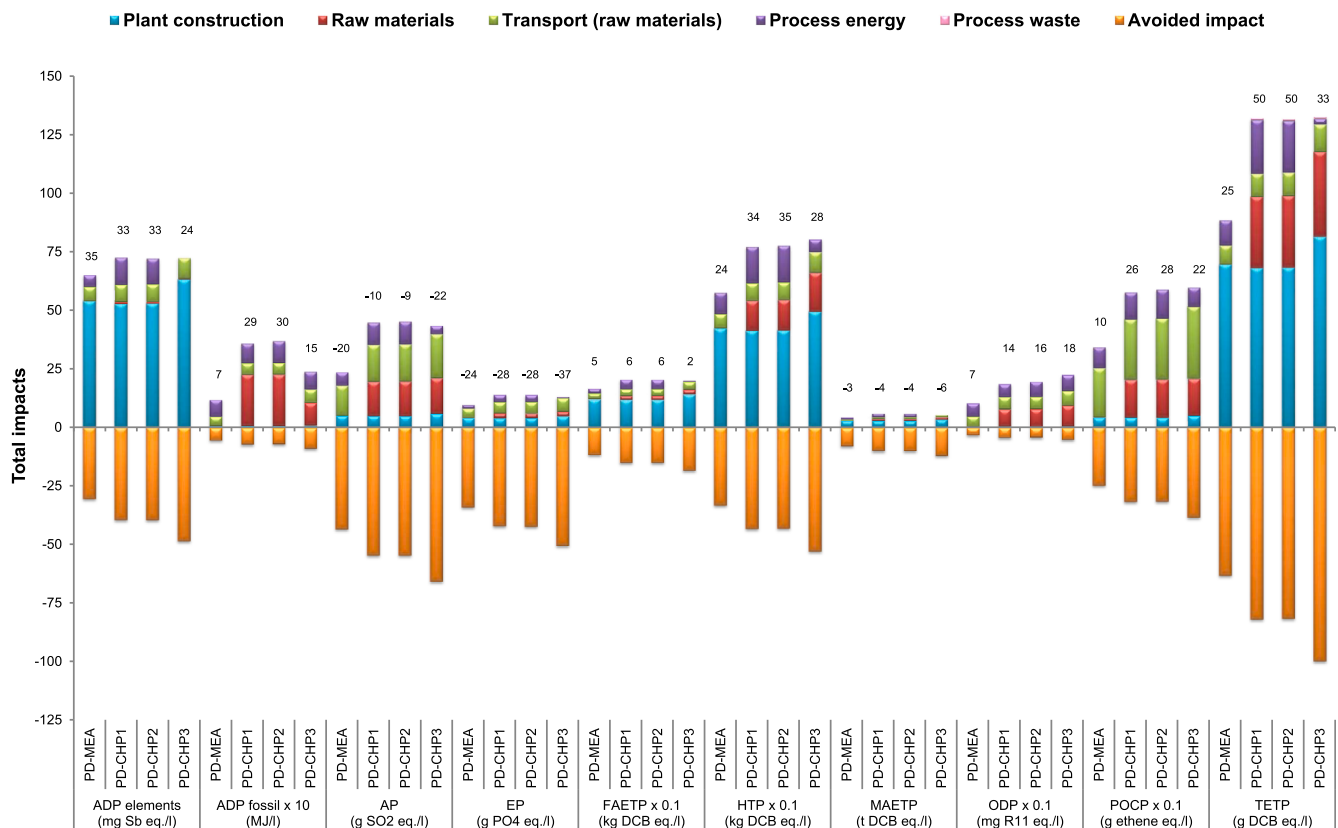


Fig. 7. Life cycle environmental impacts of the four process designs. [The values on top of the bars represent the total impact after the system has been credited for the co-products. The values of some impacts have been scaled to fit. The original values can be obtained by multiplying the value shown on top of the bars by the scaling factor given on the x-axis. Impact categories: ADP elements: abiotic depletion potential of elements; ADP fossil: abiotic depletion potential of fossil resources; AP: acidification potential; EP: eutrophication potential; FAETP: fresh water aquatic ecotoxicity potential; HTP: human toxicity potential; MAETP: marine aquatic ecotoxicity potential; ODP: ozone depletion potential; POCP: photochemical oxidants creation potential; TETP: terrestrial ecotoxicity potential. DCB: dichlorobenzene.]

mentioned below refer to the total impacts before the system credits.

4.1.1. Global warming potential

As can be seen in Fig. 6, the lowest GWP is estimated for PD-MEA (3.2 kg CO₂ eq./l) and the highest for PD-CHP2 and PD-CHP1 (17.2 and 16.4 kg CO₂ eq./l, respectively). The impact from PD-CHP3 is around two times lower than from the other two CHP-based designs (7.6 kg CO₂ eq./l) as all its hydrogen and electricity are produced internally (see Table 4).

The reason for the MEA-based design being the best option for this impact is a much lower requirement for the raw materials relative to the other three designs (see Table 4). These consumables, particularly steam and hydrogen, are the main hotspots for the CHP-based designs, contributing 60% to the GWP of PD-CHP1 and PD-CHP2 and 40% for PD-CHP3. Process energy adds a further 25–30% to the total for these three designs. It can be noted in Fig. 6 that the energy consumption is slightly greater in the PD-CHP1 and PD-CHP2 systems because of the need to heat and compress the large volume of flue gases produced in the CHP plant that are fed downstream (see Figs. 2 and 3). Even though PD-MEA has the lowest energy requirements (see Table 4), its impact related to the energy consumption is similar to PD-CHP3 because all the electricity is supplied from the grid. For that reason, and due to a low input of raw materials, the process energy contributes 58% to the total GWP of PD-MEA.

The contribution of transport is also significant, ranging from 14% for PD-CHP1 and PD-CHP2 to 31% for PD-MEA. On the other hand, the shares of process waste management and use of fuels are negligible across the designs (< 1%). It should be noted that the impact from the use of fuels shown in Fig. 6 is related to the emissions of nitrous oxide

from the car exhaust catalyst and volatile organic compounds due to incomplete combustion [42]. As mentioned earlier, the emissions of CO₂ from combustion are not considered, since they are of biogenic origin.

As can also be observed in Fig. 6, the system credits for the various co-products and recycling of end-of-life materials have a significant effect on the total GWP. PD-CHP3 has the greatest avoided GHG emissions of 8.3 kg CO₂ eq./l, largely due to the amount of fertiliser and grid electricity it displaces. The avoided impact is the lowest for the MEA-based design (5.1 kg CO₂ eq./l) due to a lack of electricity or heat generation in this system.

4.1.2. Other environmental impacts

PD-MEA has the lowest ADP fossil, HTP, ODP, POCP and TETP while CHP-3 is the best option for the remaining five categories (Fig. 7). The main hotspots are the plant construction, raw materials and their transport. Plant construction is particularly significant for ADP elements, HTP and TETP, contributing 50–90% to the total across the designs due to the use of metals for the plant infrastructure and emissions of heavy metals during their production. Raw materials are dominant for the ADP fossil of the CHP-based designs, causing 40–60% of the impact, largely due to the use of hydrogen and steam. They also contribute 17–35% to the AP, HTP and TETP of these designs. Finally, sludge transport causes 30–60% of AP, EP, ODP and POCP across the designs.

The credits for the co-products are significant for most categories and process designs. For AP, EP and MAETP, the credits are greater than the impacts caused, resulting in net-negative overall values. This is mainly due to the avoided production of mineral fertilisers and grid

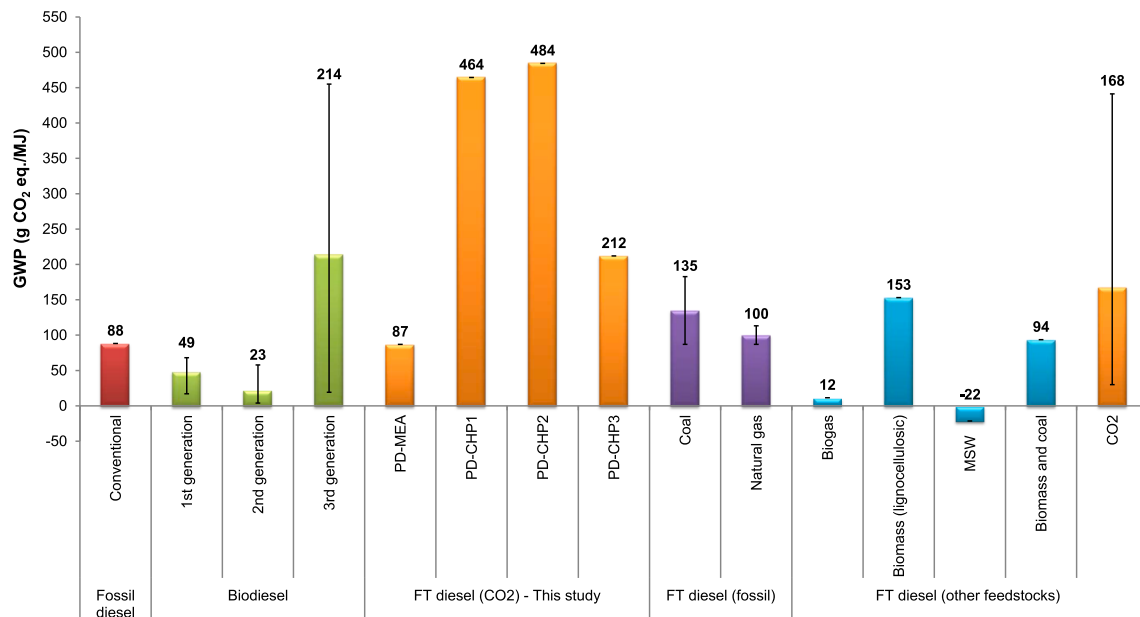


Fig. 8. Global warming potential of Fischer-Tropsch fuels produced from CO₂ in comparison with diesel from fossil and renewable resources. [Data for the other fuels sourced from: Jaramillo et al. [7], Azapagic and Stichnothe [43], Defra [44], Jeswani and Azapagic [45], Ecoinvent Centre [27], Wang et al. [12], Pressley et al. [22], Ahmadi Moghaddam et al. [11], Liu et al. [10], van der Giesen et al. [18], Hombach et al. [17]. Error bars indicate the range of results reported in the literature. MSW: municipal solid waste.]

electricity. As for the GWP, the share of process waste management is negligible across the designs and impacts.

4.1.3. Comparison with other fuels

With the exception of fossil diesel, comparison of CO₂-derived FT fuels with other fuels is only possible for GWP as data for the other impacts are not available in the literature. Since different fuels have differing energy content, for the purposes of comparison, the GWP values are expressed per MJ instead of per litre.

As indicated in Fig. 8, of the four designs, only PD-MEA can compete

with fossil diesel as their GWP is almost identical (87 and 88 g CO₂ eq./MJ, respectively). Compared to the 1st and 2nd generation biodiesel, the CO₂-derived fuel has at best 30% higher (PD-MEA) and at worst seven times greater impact (PD-CHP2). However, in comparison to the 3rd generation (algal) biofuels, PD-MEA has on average a 2.5 times lower GWP.

PD-MEA also has a lower impact than the other FT fuels shown in Fig. 8, except for diesel from biogas and MSW. The latter is the best option overall, with a net negative value of -22 g CO₂ eq./MJ due to the credits for co-products [22].

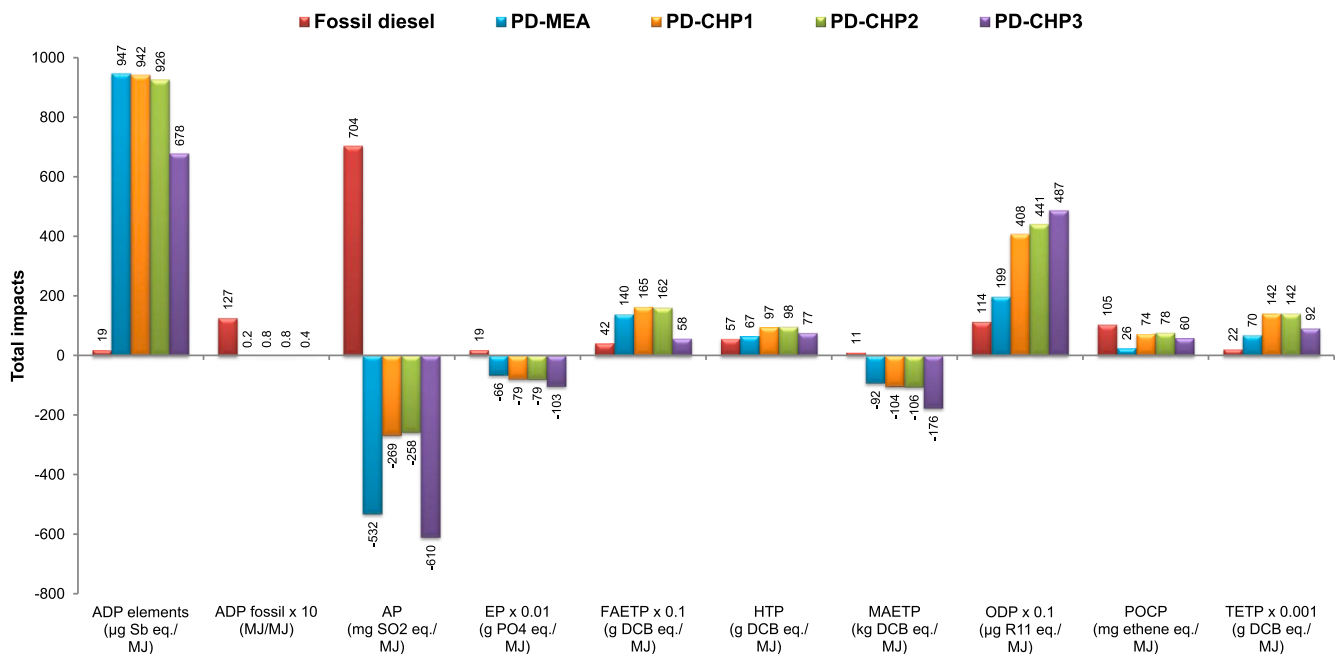


Fig. 9. Life cycle environmental impacts of Fischer-Tropsch fuels produced from CO₂ in comparison with fossil diesel. [The values for some impacts have been scaled to fit. The original values can be obtained by multiplying the value shown on top/bottom of the bars by the scaling factor given on the x-axis. For the impacts nomenclature, see Fig. 7. Data for the impacts from fossil diesel sourced from Ecoinvent [27]. For GWP, see Fig. 8.]

It can also be seen in Fig. 8 that all CHP-based designs have a much higher impact than any other fuels, with the exception of biodiesel from algae and FT diesel derived from direct air capture of CO₂, with which these designs are comparable. Therefore, from a climate change perspective, these designs are not viable for the production of FT liquid fuels.

The picture is less clear for the other environmental impacts, with the FT fuels from CO₂ having lower ADP fossil, AP, EP, MAETP and POCP and fossil diesel being a better option for ADP elements, FAETP, HTP, ODP and TETP (Fig. 9). For example, fossil diesel has 35–50 times lower ADP elements and 2–7 times lower FAETP, HTP, ODP and TETP. However, its ADP fossil is 220 times higher. FT fuels are also significantly better for AP, EP and MAETP, which are net-negative due to the system credits.

The next section considers the economic viability of FT liquid fuels in comparison with conventional diesel.

4.2. Life cycle economic sustainability

The estimated life cycle costs, revenue and profits for different MARR values are shown in Fig. 10. As can be observed, over the 20-year lifetime, operation and maintenance (O&M) are the main contributors (78%) to the total costs, with the latter ranging from £133 million for PD-MEA to £169 million for PD-CHP3. The revenues over the period vary from £211–£390 million (PD-MEA) to £262–£477 million (PD-CHP3) across the MARR values. This includes the revenue from the recovery of land, salvage and working capital of £7.8–9.4 million

(Table 11). The resulting total profits are estimated in the range of £66 million for PD-MEA to £79 million for PD-CHP3 for the lowest MARR of 8%, increasing to £244 million and £293 million, respectively, for the MARR of 24%. Assuming the latter, the payback period across the designs is 3 years, while for the lowest MARR considered (8%), this goes up to seven years (Table 13).

Therefore, the results suggest that PD-MEA is the least costly but PD-CHP3 is the most profitable design, despite the latter having high O&M costs. This is due to the highest estimated basic price of liquid fuels for this design, which ranges from 4,736 to 8,609 pence per litre (£47.36–£86.09/l), depending on the MARR (Table 13). The fuel prices for the other designs range from 2,601–4,808 pence/l (PD-MEA) to 3,875–6,952 pence/l (PD-CHP2). These prices are 60–190 times higher than the average basic price of fossil diesel in 2017 of 44.4 pence/l (120 pence at pump [32]). These prices are so much higher than for diesel because of the small production scale considered. For that reason, the effect of scaling-up the plants to capacities comparable to existing fuel production facilities is explored further on, in Section 4.4.

Prior to that, the next section considers through a sensitivity analysis if the economic as well as the environmental sustainability of the four designs could be improved by targeting the respective hotspots.

4.3. Sensitivity analysis

The key parameters that could affect the environmental sustainability of FT fuels are displacement of mineral fertilisers, energy consumption, sludge transport distance, production yield and plant

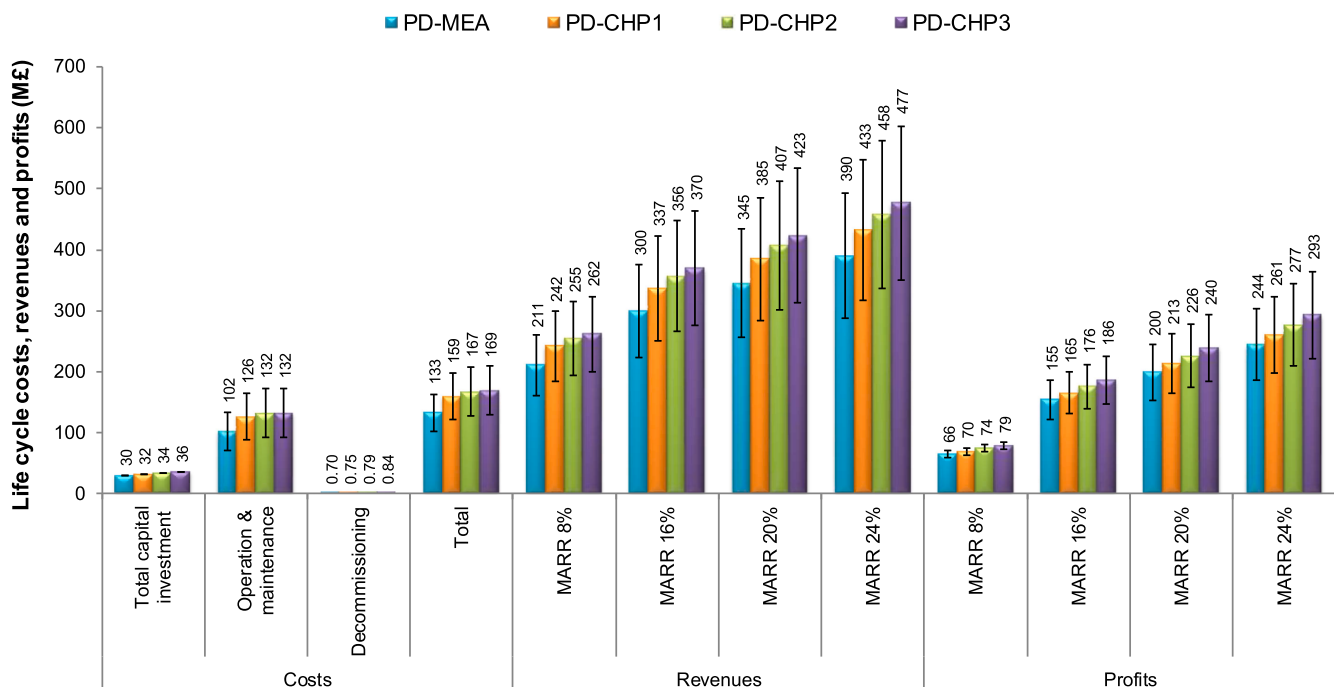


Fig. 10. Life cycle costs, revenues and profits for the four process designs. [The values shown on top of the graph bars refer to the plant lifetime of 20 years. The lower and upper error bars correspond to the lifetimes of 14 and 26 years, respectively, considered in the sensitivity analysis. MARR: minimum acceptable rate of return.]

Table 13

Estimated basic price of fuels and the payback period for the four process designs for the plant lifetime of 20 years and different MARR values.

Price	MARR (%)	Payback time (yr)	PD-MEA	PD-CHP1	PD-CHP2	PD-CHP3
Basic price (pence per litre)	8	6.9	2,601	3,645	3,875	4,736
	16	4.2	3,705	5,081	5,413	6,672
	20	3.5	4,256	5,800	6,183	7,640
	24	3.0	4,808	6,518	6,952	8,609

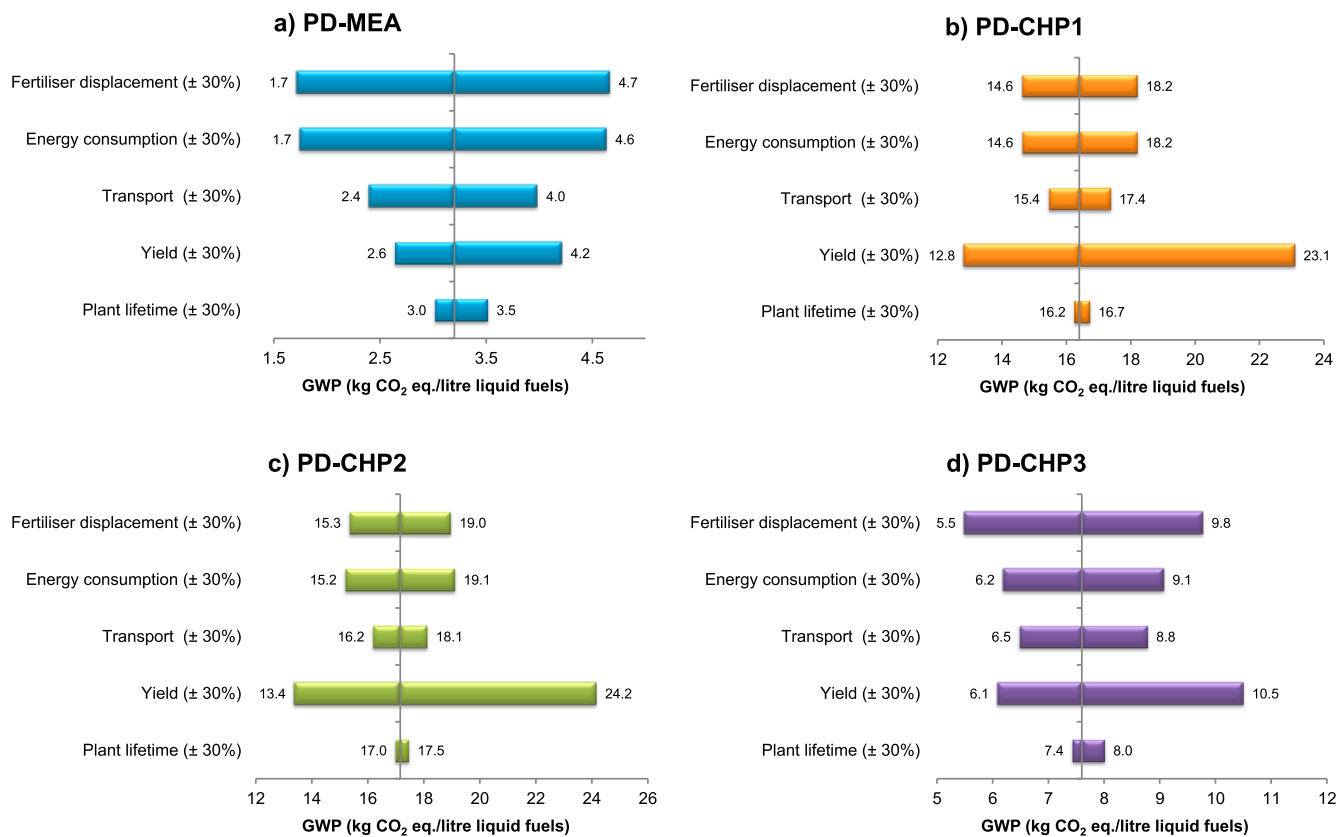


Fig. 11. Changes in the global warming potential (GWP) with variations in different parameters. [The vertical lines in the graphs denote the GWP values in the base case. For PD-MEA, GWP of 3.23 kg CO₂ eq./l is equal to the GWP of fossil diesel. For the other three designs, the equivalent values equal to the GWP of fossil diesel are: 3.11 kg CO₂ eq./l (PD-CHP1), 3.12 kg CO₂ eq./l (PD-CHP2) and 3.17 kg CO₂ eq./l (PD-CHP3) kg CO₂ eq./l.]

lifetime. The last two, as well as the total capital investment, could also influence the economic sustainability. Therefore, these parameters are considered within the sensitivity analysis, with each being varied in turn by $\pm 30\%$ around their original values; for the latter, see Sections 3.2 and 3.3.

4.3.1. Effect on the environmental sustainability

The effect of different parameters on the GWP of the four designs is shown in Fig. 11; the results for the other impacts can be found in Figs. S3–S6 in the SI. As indicated in Fig. 11, the GWP of PD-MEA is more sensitive to the changes in the displacement of mineral fertilisers and energy consumption than to the other three parameters. For example, if the system can displace an additional 30% of mineral fertilisers resulting from variations in the NPK content in the digestate, its GWP would be nearly two times lower than in the base case and, as a consequence, almost half the GWP of fossil diesel. The same reductions would be achieved if the energy consumption was reduced by 30%. Furthermore, shortening the sludge transport distance by 30%, would reduce the impact by a further 25%, while increasing the yield and the plant lifetime by the same percentage would lead to the GWP reductions of 18% and 6%, respectively. Therefore, optimising these parameters simultaneously could lead to significant reductions in GHG emissions from the PD-MEA system, making it much more competitive with fossil diesel. It would also mean that FT liquid fuels from CO₂ would be able to meet the requirement of the EU Renewable Energy Directive (RED) that alternative diesel fuels must have at least 60% lower life cycle GHG emissions than conventional diesel [46].

For the other three designs, a very different pattern can be observed in Fig. 11. For instance, the GWP of PD-CHP1 and PD-CHP2 is most sensitive to the changes in process yields and least affected by the variation in the plant lifetime. This is due to the raw materials being the

main hotspot in both designs. The impact of PD-CHP3 is also most affected by the yield but also by the fertiliser displacement, energy consumption and sludge transportation distances. However, from a climate change perspective, none of these variations achieves significant reductions to make liquid fuels from these designs competitive with fossil diesel. For example, the minimum GWP achieved for PD-CHP1 and PD-CHP2 is 12.8 kg CO₂ eq./l (362 g CO₂ eq./MJ) and 13.38 kg CO₂ eq./l (378 g CO₂ eq./MJ), respectively, which is still four times higher than the GWP of fossil diesel. The GWP of PD-CHP3 drops to 5.48 kg CO₂ eq./l (152 g CO₂ eq./MJ) when the displacement of mineral fertilisers is increased by 30%, but this is still nearly two times higher than the impact of diesel.

All process designs show significant variations of $\pm (40\text{--}120\%)$ in most other impact categories with the changes in the displacement of mineral fertilisers (Figs. S3–S6). ADP fossil and ODP are the only exceptions, which are mostly affected by changes in the yield, as well as energy consumption and sludge transport distances in the case of PD-MEA. The second most influential parameter is the process yield, with the impacts changing by $\pm (20\text{--}75\%)$ across most categories and designs. The only exceptions are POCP of PD-MEA and HTP of PD-CHP1 and PD-CHP2 which are not affected by the yield as they are largely related to the plant infrastructure and sludge transport, respectively. It can also be observed that the lifetime of the plant affects ADP elements, FAETP, HTP, MAETP and TETP, which vary by $\pm (20\text{--}145\%)$. Changes in the sludge transportation distances lead to a $\pm (20\text{--}40\%)$ variation in AP, FAETP and POCP. Finally, ADP fossil, ODP and POPC of PD-MEA, as well as AP and FAETP of PD-CHP1 and PD-CHP2, are sensitive to the changes in energy consumption ($\pm 20\text{--}30\%$).

Compared to fossil diesel, at the most favourable conditions, the FT liquid fuels from some process designs become a better option for HTP, FAETP and TETP than fossil diesel (Tables S4 and S5); this is in addition

to the other five impacts for which they were better in the base case (Fig. 9). For example, increasing the displacement of mineral fertilisers by 30% reduces the HTP of PD-MEA and PD-CHP3 by 31% and 36% relative to diesel and TETP by 18% and 32%, respectively. In addition, FAETP of all process designs is also reduced significantly on the base case, with PD-CHP3 becoming net-negative for this impact and a better option than fossil diesel. However, ADP elements and ODP still remain higher across the designs than the equivalent impacts of diesel. Even by extending the plant lifetime by 30%, the ADP elements is still 16–36 times higher than for diesel. Finally, ODP remains 25–30% higher than that of diesel for the best values of any of the parameters considered.

4.3.2. Effect on the economic sustainability

The effect of the plant lifetime on the LCC and profitability of the different designs is shown in Fig. 10. Increasing the lifetime from 20 to 26 years, increases the LCC by 24%, while reducing it to 14 years lowers the costs by 24% across the designs. It can also be observed that the LCC of PD-MEA over 26 years are comparable to the CHP-based plants if their lifetime is kept at 20 years. This would also lead to a competitive profitability of this process compared to the rest whilst maintaining lower fuel prices. For example, for a MARR of 8%, PD-CHP1 would earn a profit of £70 million over 20 years at a basic price of fuel of 3,645 pence per litre. By comparison, PD-MEA would gain £71 million over 26 years for a price of fuel of 2,466 pence per litre.

The basic fuel price would also be reduced with an increase in the yield and a reduction in TCI. For a MARR of 8%, increasing the yield by 30% would decrease the fuel price across the designs by 23% (Fig. 12). However, if the yield was 30% lower than in the base case, the fuel price would increase by 43%. Increasing the TCI by 30% would increase the basic prices by 20–30% while reducing the TCI by 30% would decrease them by 33% across the designs. Similar trends are found for the other MARR values (Figs. S7–S9 in the SI). However, under the best conditions, these prices would still be much higher than the price of fossil diesel. As mentioned earlier, this is largely due to the small-scale production considered so far. The issue of scale is explored further in the next section.

4.4. Plant scale-up

This section examines the effect of the economies of scale on the environmental and economic sustainability of the four process designs considered in the previous sections. Two production capacities are considered: medium and large, producing 850 and 1,670 tonnes of liquid fuels per day, respectively. The scaling-up has been carried out following the methodology explained in Section 3.3.5.

4.4.1. Life cycle environmental sustainability

Only the impacts of the large-scale plants are discussed here as the trends are similar to the medium scale. The results for the latter can be found in Figs. S10 and S11 in the SI.

As shown in Fig. 13, the FT fuels from the scaled-up plants outperform fossil diesel in all impact categories across all the designs. The only exceptions are the ODP, for which fossil diesel is the best option, and the GWP, which is lower for fossil diesel than for the CHP-based designs. However, PD-MEA now has a 24% lower GWP than fossil diesel and is the best option from a climate change perspective. Nevertheless, it still has a higher impact than the 1st or 2nd generation biofuels. For the other impacts, PD-CHP3 now emerges as the most sustainable alternative among the four designs, except for ADP fossil and POCP, where the MEA design is better and, as previously mentioned, ODP for which fossil diesel is preferred.

It is notable that, with the scaling-up, ADP elements, FAETP, HTP and TETP improve substantially and become net-negative. This is due to a much lower requirements for the plant infrastructure per unit of FT fuels produced. For that reason, the reduction in ADP elements is particularly significant – from being 35–50 times higher than that of fossil diesel in the base case (Fig. 9), it is now around 25–50 times lower (Fig. 13). The change in the remaining impacts (ADP fossil, AP, EP, MAETP, ODP and POCP) with the scaling-up is negligible. The reason for this is that the contribution of the plant infrastructure to these impacts is minimal, both in the base case (Fig. 7) and scaled-up plants (Fig. S12). However, as larger plants are typically more resource efficient, it is likely that all these and the remaining impacts would improve further. In the absence of real data, it is not possible to consider fully the effect of the economies of scale on the resource consumption and the related reductions in impacts. Instead, this is explored in a sensitivity analysis, focusing on PD-MEA, which outperforms fossil diesel for all impacts but ODP.

Varying the same parameters as in the base-case sensitivity analysis (Section 4.3.1) by $\pm 30\%$ would change the GWP of the scaled-up PD-MEA as shown in Fig. 14; the effect on the other impacts can be seen in Fig. S13 in the SI. As in the base case, the amount of mineral fertilisers displaced and the process energy consumption have the greatest effect on the GWP. In the best case, the GWP reduces to 1 kg CO₂ eq./l or 27 g CO₂ eq./MJ, which is around 70% lower than the impact of fossil diesel. However, in the worst case, the GWP of FT liquid fuels increases to 3.95 kg CO₂ eq./l (107 g CO₂ eq./MJ), 18% higher than for fossil diesel. Decreasing sewage transport distances and increasing the yield by 30% each reduces the impact to 1.68 and 1.92 kg CO₂ eq./l, which is 50% and 40% lower than for fossil diesel, respectively. The equivalent changes in the opposite direction lead to a 1–8% higher impact than that of diesel. The GWP is unaffected by the changes in the plant lifetime as the infrastructure plays a much smaller role in this impact for large plants.

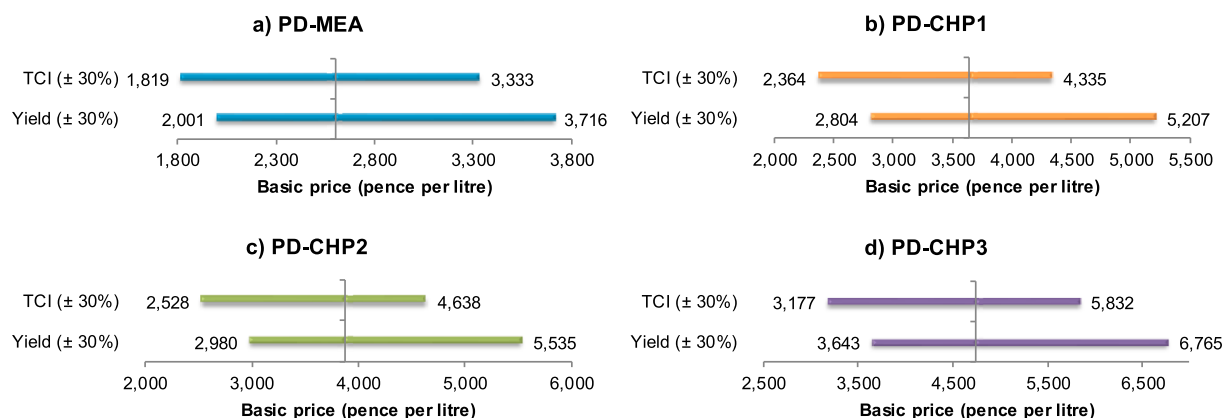


Fig. 12. Changes in the basic fuel price with the total capital investment (TCI) and production yield for a minimum acceptable rate of return of 8%. [The vertical lines in the graphs represent the basic price in the base case.]

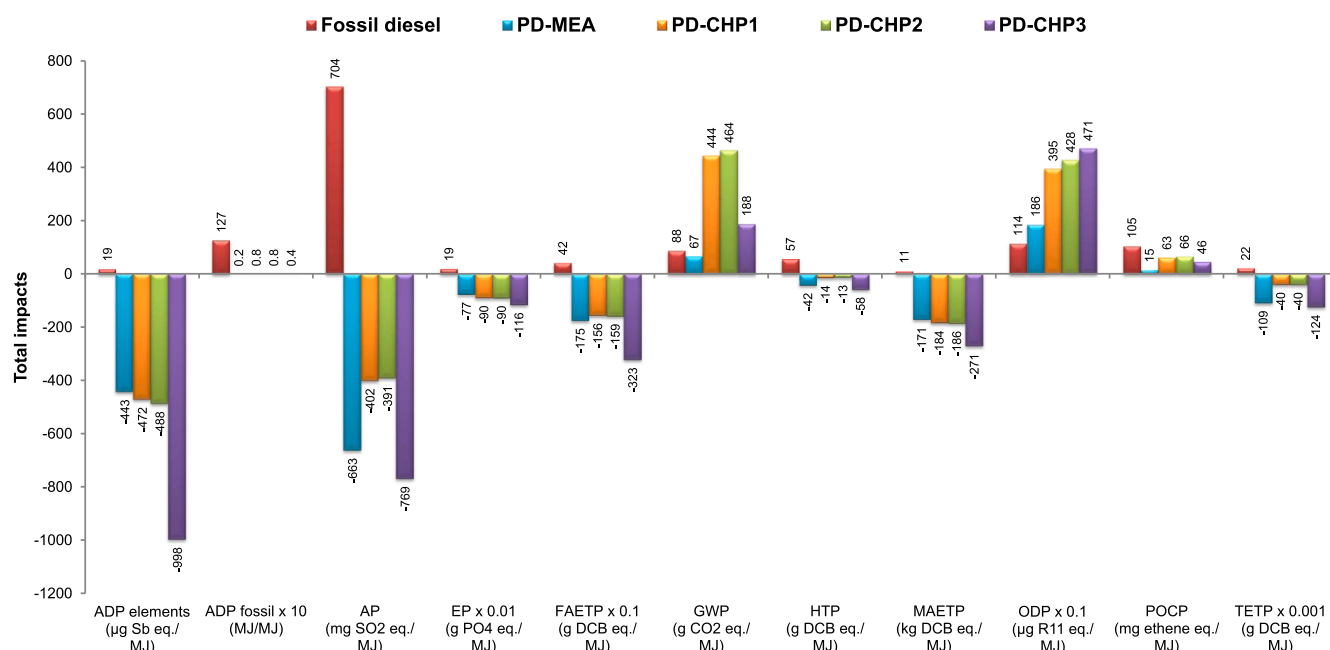


Fig. 13. Life cycle environmental impacts of Fischer-Tropsch fuels produced from CO₂ in a large-scale plant (1,670 t/day) in comparison with fossil diesel. [The values for some impacts have been scaled to fit. The original values can be obtained by multiplying the value shown on top/bottom of the bars by the scaling factor given on the x-axis. GWP data for fossil diesel sourced from: Azapagic and Stichnothe [43], Defra [44], Jeswani and Azapagic [45], and Ecoinvent Centre [27]; data for other environmental impacts sourced from Ecoinvent [27]. See Fig. 7 for impacts acronyms.]

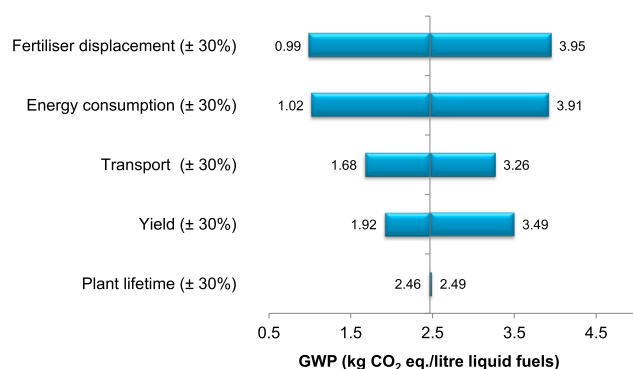


Fig. 14. Changes in the global warming potential (GWP) for PD-MEA with variations in different parameters for a large-scale plant (1,670 t/day). [The vertical lines in the graphs denote the original GWP value (2.47 kg CO₂ eq./l). GWP of 3.23 kg CO₂ eq./l is equal to the GWP of fossil diesel.]

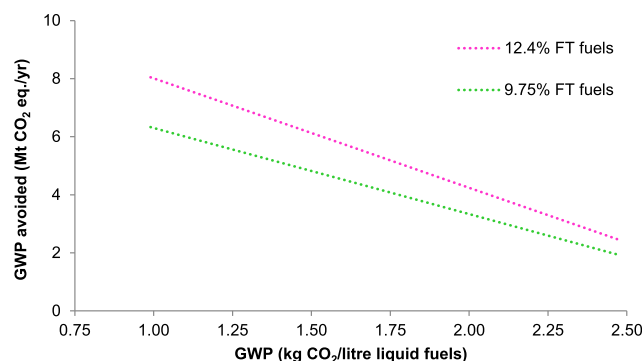


Fig. 15. Global warming potential (GWP) potentially avoided by displacing fossil diesel based on the Renewable Transport Fuels Obligation (RTFO). [Values shown for PD-MEA. Based on consumption of fossil diesel in the UK in 2017, sourced from BEIS [48] and RAC [47]. RTFO: Renewable Transport Fuels Obligation [49].]

The above results can be used to estimate the potential for mitigating the climate change impact from the transport sector by displacing a certain share of fossil diesel with FT liquid fuels. In 2017, 30.4 billion tonnes of diesel were consumed in the UK [47], emitting 95 Mt CO₂ eq. of GHG emissions [48]. The revised Renewable Transport Fuels Obligation (RTFO) stipulates through the fuel blending obligation that 9.75% of fossil fuels should be replaced by alternative fuels by 2020 and 12.4% by 2032 [49]. Assuming hypothetically that all of this replacement is met through CO₂-derived fuels from large-scale PD-MEA plants, gives the maximum GHG mitigation potentials in Fig. 15. This has been calculated considering the base-case and the improved GWP values shown in Fig. 14, assuming that the actual plants will be optimised and hence their impacts reduced rather than increased. In that case, FT liquid fuels could avoid around 2–8 Mt CO₂ eq. annually by displacing fossil diesel, with 2 and 2.5 Mt CO₂ eq. avoided in the base case for the displacements of 9.75% and 12.4% of fossil diesel, respectively. Therefore, there is a clear potential for FT fuels produced in large-scale PD-MEA plants to contribute towards mitigation of the climate change impact from transport. This would also lead to reductions in all other life cycle impacts considered here, ranging from 8% and 13% for POCP and ADP fossil to 170–480% for MAETP and ADP elements for the displacement of 9.75% and 12.4% of fossil diesel, respectively. The only exception is ODP which would increase by 7% and 8% (Table S6).

Their economic potential at large-scale production is explored next.

4.4.2. Life cycle economic sustainability

The life cycle costs and profits of the scaled-up process designs are presented in Fig. 16 for a MARR of 8%; the profit values for the other MARRs can be found in Fig. S13 in the SI. As can be seen in Fig. 16, the LCC for the medium-size plants are 80–100 times higher than in the base case, while the larger plants have 130–160 times greater costs, depending on the design. However, the larger plants are more profitable due to the economies of scale. In the worst case, a medium-scale PD-MEA would earn around £3.7 billion in profits over 20 years, while in the best case, large-scale PD-CHP3 would have a profit of around £8.4 billion.

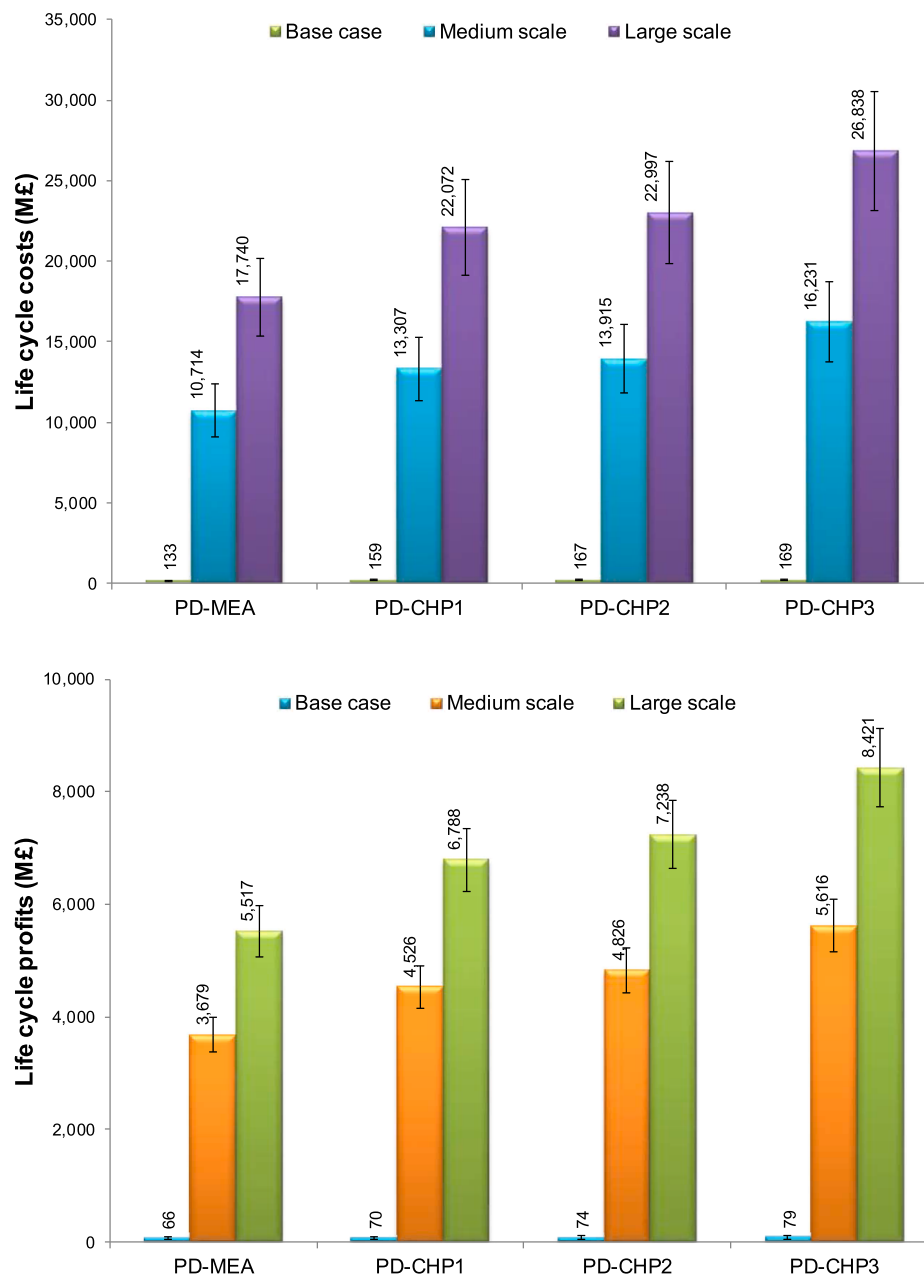


Fig. 16. Life cycle costs (top) and profits @ 8% MARR (bottom) for different designs and production capacities. [Base case: > 1 t/day; Medium scale: 850 t/day; Large scale: 1,670 t/day. The values shown on top of graph bar refer to the plant lifetime of 20 years while the lower and upper error bars correspond to the costs for the lifetime of 14 and 26 years, respectively.]

Both the costs and profits are affected by the plant lifetime. For example, extending the lifetime of the PD-MEA from 20 to 26 years would increase its profits by 8% while decreasing it to 14 years would lead to 8% lower net earnings (Fig. 16).

Based on the profitability analysis of the scaled-up plants, the estimated basic prices of FT fuels decrease considerably with the increase in capacity (Fig. 17). For the large-scale plant (1,670 t/day), PD-MEA has the lowest basic price of 185 pence/l at 8% MARR, increasing to 300 pence/l for the MARR of 24%. Thus, even in the best case, this is still four times higher than the basic price of fossil diesel considered here (44.4 pence/l). However, it should be noted that a biodiesel was also significantly more expensive than fossil diesel a few years ago. For example, in 2013, their respective wholesale prices in the EU were €0.85 and €0.47/l [50] but the gap has closed since due to a wider deployment of biodiesel, as mandated by the RED, and the increasing prices of diesel.

The basic prices for the other three designs range from 221 to 275 pence/l for 8% MARR and 357–447 pence/l for the MARR of 24%. Thus, in the worst case for the large plant (PD-CHP3 @ 24% MARR), FT fuels are ten times more expensive than fossil diesel. For the medium-scale capacity, the basic prices range from 226 to 561 pence/l across the designs and MARRs; this is 5–13 times greater than the price of diesel. These prices are also unfavourable in comparison to other FT liquid fuels reported in the literature. For example, the basic price of FT diesel produced from natural gas ranges from 9 to 47 pence/l [7]. For coal-derived diesel, it is 30 pence/l [7] and around 56 pence for fuels produced in a CBTL plant [10]. However, CO₂-derived FT fuels compare more favourably with the basic price of algae biodiesel, which can be as high as 2,300 pence/l depending on estimates [51].

Assuming the same taxation for CO₂-derived FT fuels as for the conventional road fuels, with 63% of the pump price being the excise duty and VAT, the pump prices of these fuels would range from 500

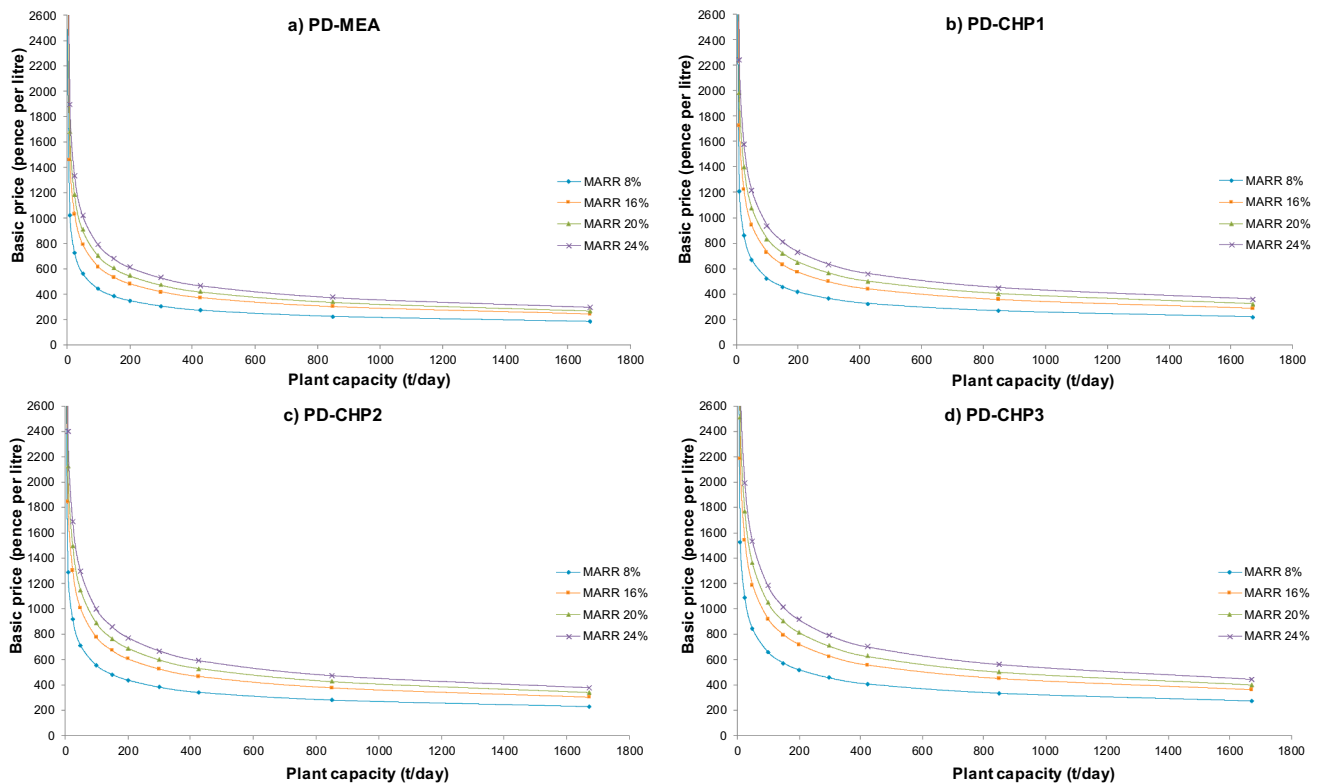


Fig. 17. Estimated basic price (excluding excise duty and VAT) of Fischer-Tropsch liquid fuels for different plant capacities. [The y-axis has been capped at 2,600 pence/l to enable a better comparison between process designs. The basic prices for small (base-case) capacity can be found in Table 13.]

pence (large-scale PD-MEA @ 8% MARR) to 1,516 pence (medium-scale PD-CHP3 @ 24% MARR), compared to the assumed 120 pence for a litre of fossil diesel (Table 14). Therefore, these fuels would not be viable without either reducing the taxation, providing subsidies and/or mandating their use through policy. Similar applies to biofuels, which are still not economically viable and would not be produced without strong policy incentives [52]. In addition to the fuel blending obligation mandated through the RTFO, this also included a 20 pence/l excise duty exemption until 2012.

The issue of taxation and incentives is considered in Fig. 18, which shows that subsidies would be required in all cases, ranging from 35% to 79% of the pump price of 120 pence/l, depending on the process design, production scale and the MARR.

The tax margins and subsidies are explored further through a sensitivity analysis by varying the TCI and production yields by $\pm 30\%$. The results in Fig. 19 suggest that in the best case, subsidies could be reduced to 8% but in the worst case, they would need to be at the level of 70–85%.

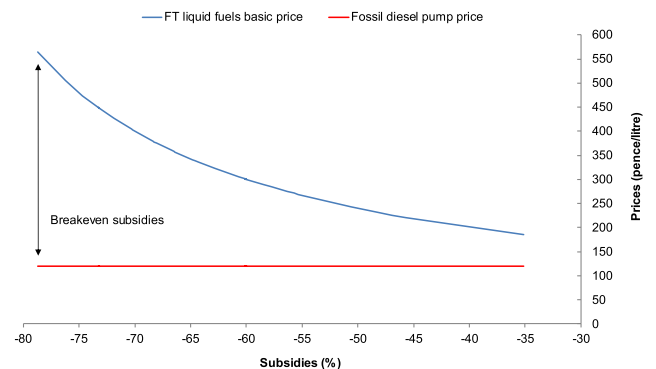


Fig. 18. Subsidies required for Fischer-Tropsch liquid fuels to match fossil diesel pump prices. [Based on the range of basic prices for the medium (850 t/day) and large (1,670 t/day) plant capacities for different process designs and MARRs as shown in Fig. 17.]

Table 14

Hypothetical price at pump of Fischer-Tropsch liquid fuels for different plant capacities considering the same road taxation as for fossil diesel (pence per litre).

Price/tax	PD-MEA				PD-CHP1				PD-CHP2				PD-CHP3			
	8%	16%	20%	24%	8%	16%	20%	24%	8%	16%	20%	24%	8%	16%	20%	24%
<i>Medium-scale plant (850 t/day)</i>																
Basic price	226	301	339	376	269	358	403	447	284	379	427	474	336	449	505	561
Excise duty and VAT ^a	385	513	577	641	458	610	686	762	483	645	726	807	572	764	860	955
Price at pump	610	814	915	1,017	728	968	1,089	1,209	767	1,024	1,153	1,282	908	1,212	1,364	1,516
<i>Large-scale plant (1670 t/day)</i>																
Basic price	185	243	271	300	221	289	323	357	232	305	341	378	275	361	404	447
Excise duty and VAT ^a	315	413	462	511	376	492	550	608	396	519	581	643	469	615	688	761
Price at pump	500	656	733	811	597	781	873	965	628	824	922	1,021	744	976	1,092	1,208

^a 63% of the total pump price.

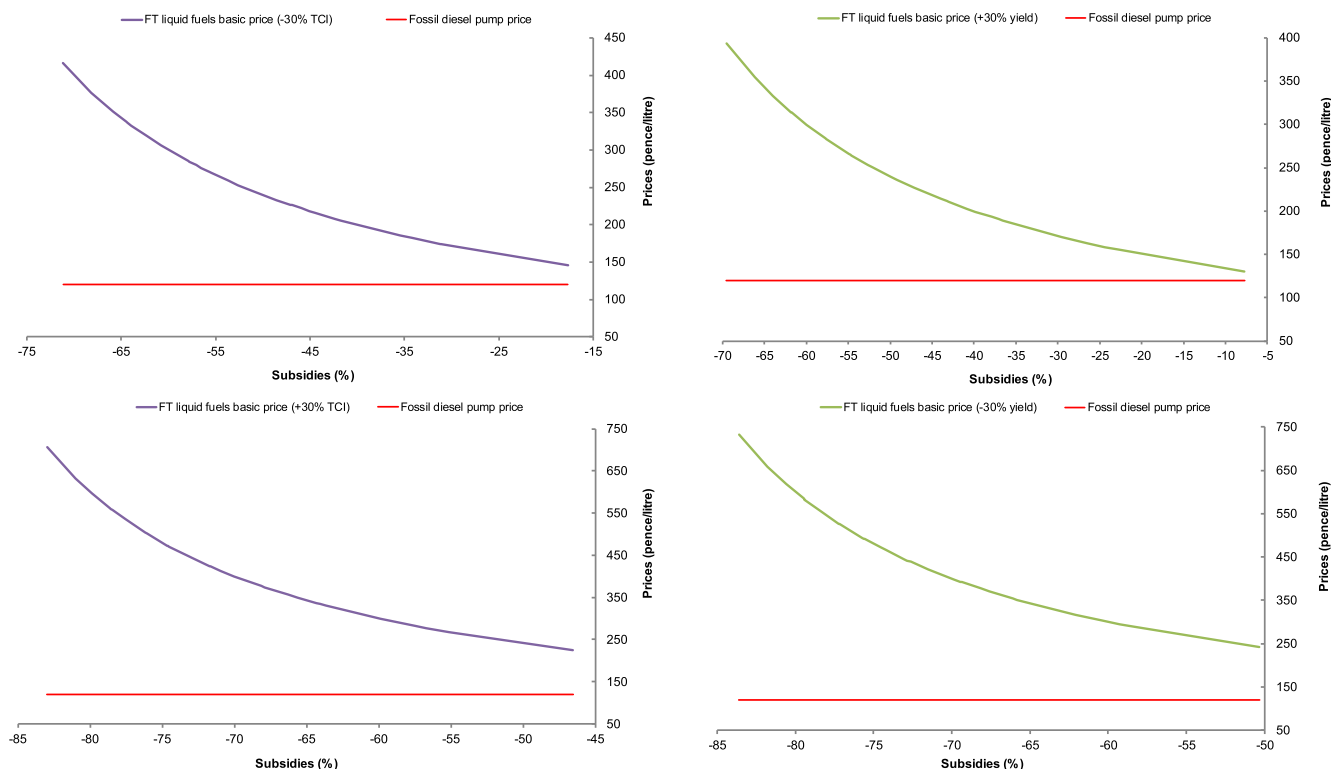


Fig. 19. Effect of the total capital investment (TCI) and production yield on subsidies and taxation. [Based on the range of basic prices for the medium (850 t/d) and large (1,670 t/d) plant capacities for different process designs and MARRs as shown in Fig. 17.]

5. Conclusions

This paper has evaluated the life cycle environmental and economic sustainability of FT liquid fuels produced from CO₂ generated during anaerobic digestion of sewage sludge. Four process configurations have been considered and the sustainability evaluated for a range of plant capacities to explore the effect of the economies of scale. For a small scale production (< 1 t fuels/day), the lowest global warming potential (GWP) of 87 g CO₂ eq./MJ estimated for PD-MEA matches the GWP of fossil diesel. However, the highest impact of 484 g CO₂ eq./MJ (PD-CHP2) is 5.5 times higher than that of diesel. Liquid fuels from PD-MEA also have the lowest other environmental impacts compared to the fuels from the other three designs. In comparison with diesel, PD-MEA fuels are better for five impact categories while diesel is environmentally more sustainable for the remaining five impacts.

The results suggest that PD-MEA is the least costly but PD-CHP3 is the most profitable design. However, the latter has the highest estimated basic price, ranging from 4,736 to 8,609 pence per litre, depending on the MARR. The lowest fuel prices of 2,601–4,808 pence/l are found for PD-MEA. These are 60–190 times higher than the average basic price of fossil diesel in 2017 of 44.4 pence per litre.

Both the environmental and economic sustainability improve significantly with the economies of scale. For the large-scale plants (1,670 t/day), the FT fuels outperform fossil diesel in all impacts across all the designs, with several impacts being net-negative. The only exceptions are the ODP, for which fossil diesel is still the best option, and the GWP, which is lower for fossil diesel than for the CHP-based designs. Optimising the key parameters for the scaled-up plants leads to the GWP of FT fuels from PD-MEA being up to 70% lower than that of diesel. Therefore, FT fuels from CO₂ have a potential to reduce GHG emissions from the transport sector. Following the RTFO fuel blending obligation, and assuming a hypothetical replacement of 9.75–12.4% of diesel by FT fuels from CO₂, would mitigate 2–8 Mt of CO₂ eq./yr, equivalent to around 2–8% of emissions from the transport sector.

The fuel prices are also significantly lower for the large plants,

reducing to 185 pence per litre in the best case (PD-MEA at 8% MARR), but this is still four times higher than the price of diesel. Thus, these fuels would not be economically viable without either reducing the taxation, providing subsidies and/or mandating their use through policy, in a similar manner the biofuels markets have been stimulated. The results obtained in this work suggest that matching the diesel pump price of 120 pence/litre would require subsidies between 35% and 79%, depending on the MARR and the scale of production. If the production yields can be improved by 30%, subsidies could be reduced to 8%.

While the economies of scale and policy will help to improve the competitiveness of these fuels, further technological developments are also essential to optimise key process parameters. These include more efficient CO₂ capture and conversion processes as well as novel sorbents and catalysts aimed at reducing energy consumption and increasing fuel production yields. Future research should also explore different policy mechanisms for stimulating development of the markets for CO₂-derived fuels.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apenergy.2019.113560>.

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