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Cost-effective ultra-high CO_2 capture rates for combined cycle gas turbines with monoethanolamine solvent

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ABSTRACT

Natural gas is expected to remain an important power generation fuel for the foreseeable future, but ultra-high CO_2 capture rates near 100% are required to achieve life cycle greenhouse gas (GHG) emissions consistent with net-zero ambitions. Recent studies have found that the marginal cost of abating low concentration CO_2 sources, such as combined cycle gas turbines (CCGTs), with monoethanolamine (MEA) solvent increases dramatically as the capture rate approaches 100%. Yet the assumed process design conditions were suboptimal for minimising CO_2 capture cost for low concentration sources. We perform detailed techno-economic assessments combined with life cycle analyses evaluating the impact of key process design variables for CCGTs with MEA-based CO_2 capture and develop guidelines to determine optimal design parameters to minimise the cost of avoided CO_2 emissions at ultra-high capture rates. We show that, with optimal lean solvent loading and regenerator operating conditions, 99.5% of fossil-CO_2 direct emissions could be captured with marginal cost of avoided CO_2 (MCAC) of 164 USD/tCO_2, while MCAC for 100% fossil-CO_2 capture is 271 USD/tCO_2. MCAC calculated in this study for 100% fossil-CO_2 capture with optimal process conditions is c. 50% lower than in previous studies, establishing a new performance benchmark for ultra-high CO_2 capture. We also show that marginal cost of avoided cradle-to-gate life cycle GHG emissions is only 3–9% higher than MCAC for 96–100% fossil-CO_2 capture. Cost-effective ultra-high capture from CCGTs is particularly significant for achieving climate change mitigation goals given high current costs for permanent atmospheric CO_2 removal, and highly uncertain future costs.

1. Introduction

Combined cycle gas turbines (CCGTs) are expected to remain an important source of dispatchable power generation in many jurisdictions and post-combustion CO_2 capture and storage (CCS) with ultra-high capture rates (>95%) could effectively mitigate direct CO_2 emissions. A key consideration in developing policies and regulations for CCS is determining the required CO_2 capture rate for the direct emission sources. Historically, most studies of CCS assumed a capture rate of 90% or less, but numerous recent studies have shown that higher CO_2 capture rates can be achieved for most industrial emission sources, including CCGTs [1]. Furthermore, regulatory frameworks for CCGT power generation in Canada [2] and the UK [3] are based on CCS with a minimum CO_2 capture rate of 95% for compliance. To achieve net zero life cycle greenhouse gas (GHG) emissions consistent with the Paris Agreement, residual direct and indirect emissions need to be offset with atmospheric CO_2 removal (CDR), and the most economic design CO_2 capture rate will be based on a comparison between the cost of CDR and the marginal cost of reducing direct CO_2 emissions [4].

CDR in climate change mitigation scenarios is most commonly assumed to be afforestation and/or bioenergy with CCS; however, large-scale deployment of either would occupy significant land area and may conflict with sustainable development goals [5]. Furthermore, carbon sequestration through afforestation is not permanent due to potential for carbon losses from disturbances (e.g., fire or disease) [6]. Direct air capture (DAC), removing CO_2 from air using liquid solvents or solid sorbents, and enhanced weathering, accelerating the reaction between atmospheric CO_2 and silicate rocks, could provide permanent carbon storage for geologic timescales and would occupy less land than afforestation or bioenergy, but usage in mitigation scenarios has been limited

Abbreviations: ATWACC, after-tax weighted average cost of capital; CCGT, combined cycle gas turbine; CCS, CO_2 capture and storage; CDR, CO_2 removal; DAC, direct air capture; LCOE, levelised cost of electricity; MCAC, marginal cost of avoided CO_2; MCAGHG, marginal cost of avoided life cycle greenhouse gas emissions; MEA, monoethanolamine; NG, natural gas; PCC, post-combustion CO_2 capture.

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due to uncertain costs and low technology readiness [6].

Regenerative amine-based absorption processes have been used extensively for industrial CO_2 capture over the past century and specifically for post-combustion CO_2 capture (PCC) for merchant CO_2 supply for the past 50 years [7,8]. There are many active areas of research investigating alternative technologies for CO_2 capture such as rotating packed beds, membrane contactors, and solid adsorption to reduce cost and/or energy consumption. Nonetheless, regenerative amine absorption using conventional, vertical mass transfer columns remains the dominant technology of choice for existing and planned industrial-scale CO_2 capture facilities [9]. Amines can be highly effective for CO_2 capture with fast reaction rates, good stability, and low vapour pressure. Aqueous monoethanolamine (MEA) is a common, low-cost generic CO_2 capture solvent; however, there are many other open-source and proprietary solvents and blends available with a range of attributes.

Low-concentration emission sources such as CCGTs (c. 4%mol CO_2) have higher CO_2 capture costs compared to more concentrated sources such as coal power or cement production. A more dilute flue gas stream requires a larger diameter absorber for the same CO_2 emission rate. Furthermore, the reduced interphase mass transfer driving force in the absorber increases the required vessel volume and reduces the achievable rich solvent loading which increases the specific reboiler duty for solvent regeneration. Achieving ultra-high CO_2 capture rates near 100% of added CO_2 also requires consideration of the mass transfer driving force at the top of the absorber as the residual flue gas CO_2 partial pressure approaches the CO_2 fugacity of the lean solvent.

Nonetheless, large-scale pilot testing has demonstrated that ultra-high CO_2 capture from CCGT flue gas is feasible. Up to 99% gross-CO_2 capture from simulated CCGT flue gas (4%mol CO_2) has been demonstrated at Technology Centre Mongstad (3.5 x 2.0 m absorber cross section, 24 m bed height) using 36%wt MEA solvent with solvent regeneration energy of 3.8–4.0 MJ/kgCO_2 (compared to 3.7–3.8 MJ/kgCO_2 at 92% gross-CO_2 capture) [10]. Throughout this paper, gross-CO_2 capture rate refers to the proportion of total CO_2 in the flue gas stream that is removed by the PCC plant, while fossil-CO_2 capture rate refers to net capture of the CO_2 that is added to the flue gas from combustion of the natural gas (NG) fuel (i.e., excluding atmospheric CO_2 that enters the CCGT with the inlet air stream). Ultra-high capture from low concentration CO_2 flue gas has also been demonstrated at Technology Centre Mongstad using the CESAR-1 solvent blend (98% gross-CO_2 capture from 3.5 to 5.0%mol CO_2) [11,12], ION Energy's proprietary ICE-31 solvent (>99% gross-CO_2 capture from 3.9%mol CO_2) [13], and RTI International's proprietary NAS solvent (>99% gross-CO_2 capture from 3.8%mol CO_2) [14]. Similarly, up to 96% gross-CO_2 capture from a simulated CCGT flue gas stream (4%mol CO_2) has been demonstrated at the National Carbon Capture Center (0.6 m diameter absorber) using 4–6 m piperazine solvent and 12 m absorber bed height [15]. A parametric modelling study by Michailos and Gibbins [16] showed that CCGT with MEA-based PCC could achieve high CO_2 capture rates with minimal impact on specific energy consumption (per tCO_2 captured) by increasing the absorber height, using low lean solvent CO_2 loading (0.09–0.15 mol_{CO2}/mol_{MEA}), and increasing the regenerator pressure above 200 kPa; however, their study did not consider process economics.

While technical feasibility has been demonstrated, prior studies have found that the marginal cost of CO_2 capture using MEA solvent for low concentration CO_2 sources becomes prohibitively expensive at high capture rates [4,17,18]. However, these studies used operating conditions (e.g., lean solvent loading and regenerator temperature/pressure) which are suboptimal for achieving ultra-high capture rates with MEA solvent from low concentration sources. Furthermore, solvent intercooling in the absorber column has been identified as an important feature for economically achieving ultra-high CO_2 capture rates [15], but its application in MEA-based ultra-high capture from low concentration CO_2 sources has not been evaluated with low lean solvent CO_2 loading.

Brandl et al. [4] found that the marginal cost of reducing direct CO_2 emissions for a generic capture process with 30%wt MEA solvent is low between 90 and 95% gross-CO_2 capture (c. + 13%), but increases considerably for gross-CO_2 capture rates above 95% from low concentration CO_2 sources (c. + 89% between 95–99% capture). Marginal cost of CO_2 capture in their analysis was defined in a way that is not directly comparable to other studies (change in total cost of capture per p.p. increase in CO_2 capture rate), but their results imply that ultra-high capture rates above c. 97% are not economically feasible for flue gas with 4%mol CO_2. Lean loading of the MEA was identified as a key operating parameter affecting cost of CO_2 capture with different optimal values depending on capture rate and CO_2 concentration in the process stream. Key limitations in their analysis are a low fixed regenerator pressure (159 kPa), the lack of intercooling in the absorption contactor, the use of random packing (Raschig rings) with inferior mass transfer capacity compared to structured packing, and only direct CO_2 emissions were considered rather than overall life cycle emissions.

Du et al. [18] found marginal cost of avoided CO_2 (MCAC) direct emissions of 184 USD/tCO_2 (2018 cost basis) between 95–97.7% CO_2 capture for CCGT, with the marginal cost increasing to very high values for higher capture rates (e.g., 377 USD/tCO_2 at 99% gross-CO_2 capture, 2018 cost basis). MCAC in their calculations incorporated the effect of CCS on power generation efficiency by dividing the change in cost per MWh of electricity generated by the difference in emission intensity (kgCO_2/MWh) for incremental CO_2 capture rate scenarios. Their analysis used 30%wt MEA solvent with structured packing, and optimised solvent flow rate, absorber intercooling, and absorber packing height for each case. However, they used a fixed lean solvent CO_2 loading of 0.20 mol_{CO2}/mol_{MEA} and did not consider indirect GHG emissions. Furthermore, Du et al. used the CCSI model for MEA-based CO_2 capture developed by the US Department of Energy. This model has subsequently been shown to substantially overpredict MEA-CO_2 reaction kinetics at low temperatures in the range employed for the absorber in Du et al. (30-50°C) [19].

Danaci et al. [17] reported marginal cost of CO_2 capture of 119 USD/tCO_2 to increase gross-CO_2 capture rate from 90% to 95% for a CCGT, increasing to 280 USD/tCO_2 between 95% and 99% gross-CO_2 capture (c. January 2021 cost basis). Their analysis used 30%wt MEA solvent with a fixed lean amine loading of 0.27 mol_{CO2}/mol_{MEA}, used a simplified single-stage model for absorption and regeneration which had large deviations between model predicted column height and test data, and did not consider indirect GHG emissions.

Mullen and Lucquiaud [20] performed a techno-economic analysis to demonstrate the feasibility of achieving 100% fossil-CO_2 capture from a CCGT with low lean solvent CO_2 loading (0.05–0.20 mol_{CO2}/mol_{MEA}) and elevated regenerator pressure (275 kPa). However, their analysis only evaluated a single marginal cost between 96% and 100% fossil-CO_2 capture and they did not evaluate the effect of varying regenerator pressure or optimise absorber height for given values of lean solvent CO_2 loading. They also used the CCSI process model and did not rigorously evaluate indirect life cycle GHG emissions.

The size of equipment and solvent regeneration energy required for ultra-high CO_2 capture rates from low concentration sources using MEA can be minimised by operating the capture plant with lean solvent CO_2 loading less than 0.15 mol_{CO2}/mol_{MEA} and elevated regenerator pressure (c. > 210 kPa) and reboiler temperature (c. > 125°C) [16], yet these conditions were not employed in the previous studies assessing marginal cost of CO_2 capture. Low lean solvent loading ensures adequate CO_2 mass transfer driving force near the top of the absorber to achieve ultra-high capture as the flue gas CO_2 concentration depletes, avoiding excessive absorber packing height (as in Brandl et al. [4]) or excessive solvent circulation rate (as in Du et al. [18]). Elevated regenerator pressure and reboiler temperature allow low lean solvent loading to be achieved without incurring excessive energy consumption to desorb the CO_2 from the solvent [16]. The lowest lean solvent loading that can

efficiently be achieved at a given regenerator reboiler temperature and packing height results in a regenerator temperature profile with the middle of the packing bed approximately halfway between the reboiler and the top of the regenerator (the regenerator inflection point) [21,22]. Achieving lean solvent loading lower than the inflection point results in excessive water vapour leaving the regenerator with the CO_2 product stream and an associated increase in regenerator heat duty.

Higher regenerator reboiler temperature raises the reaction rate of MEA thermal degradation (carbamate polymerization), but this can be partially mitigated by operating with lower lean solvent loading which reduces the carbamate concentration near the bottom of the regenerator. The net increase in thermal degradation rate up to a reboiler temperature of 136°C is expected to be manageable (c. + 70 g_{MEA}/tCO_2) within the context of overall MEA oxidation and degradation rates throughout the process without materially affecting the process economics [23].

As the capture rate from CCGT direct process emissions approaches 100% of the added CO_2, life cycle GHG emissions become dominated by indirect sources such as fuel and material supply chains [24]. Life cycle GHG emissions associated with NG supply chains vary widely because of different regulations and production practices (e.g., 4.2 − 14 gCO_2e/MJ in ref. [25]). Fugitive methane is a particular concern for NG life cycle emissions, but government regulations and leak detection/repair programmes can successfully abate fugitive methane [26,27]. Life cycle GHG emission intensity of CCGTs can be within the range of wind and photovoltaic power generation when installed with ultra-high CO_2 capture and fueled with NG from existing best practices for reducing supply chain GHG emissions [24]. To the knowledge of the authors, no prior studies have assessed the marginal cost of CO_2 capture from CCGTs (or other low CO_2 concentration sources) on a life cycle GHG emissions basis.

2. Methods

We perform techno-economic and life cycle assessments (LCAs) of a 1x1 H-Class CCGT power generation facility (649 MW net power output excluding CCS) with PCC using 35%wt MEA solvent.

Detailed steady-state process models for the PCC plant design cases were developed in Aspen Plus [28] based on methodologies and thermodynamic property models for MEA-CO_2 capture which have been extensively validated against 4 datasets from 3 different pilot plants, including a wide range of operating conditions (e.g., 75–99% gross-CO_2 capture, 0.08–0.27 mol_{CO2}/mol_{MEA} lean solvent CO_2 loading) and absorber/regenerator heights representative of industrial-scale vessels [19,29]. This model has been shown to more accurately predict CO_2 capture from CCGT flue gas and MEA-CO_2 reaction kinetics at low absorber temperature than the CCSI model [19,29] used in Du et al. [18]. The Aspen Plus MEA-CO_2 capture process model used in this study is available in ref. [19]. The CCGT and CO_2 compressor were modelled in Aspen Hysys [30].

2.1. Process description

The CCGT process is based on the design for a 1x1 H-class CCGT in the US NETL baseline study of power generation technologies [31]. CCGT operating parameters were selected to match the overall full load performance in the NETL study at sea level with 15°C ambient temperature and 60% relative humidity (detailed description in Supplementary Material). Low pressure (LP) steam from the CCGT steam cycle is diverted from the inlet to the LP steam turbine to supply heat for solvent regeneration. The inlet pressure to the LP steam turbine is adjusted so that the steam condenses 5°C above the PCC regenerator reboiler solvent outlet temperature.

The PCC plant design is based on a conventional regenerative MEA absorption process (Fig. 1). Two parallel PCC plants were assumed for all cases to maintain vessel diameters similar to the process design in the study used for the baseline cost data (c. 12 m diameter for the absorber). The outlet flue gas from the CCGT heat recovery steam generator is cooled by evaporating water in spray coolers upstream and downstream of the flue gas blower. The cooled flue gas enters the absorber and flows up through 2–3 sections of Mellapak 252Y structured packing in the absorber to promote mass transfer with the MEA solvent flowing down from the top. Absorber bed height is a variable in this study, with an assumed maximum height of 10 m for each packing section. Intercoolers remove heat from the solvent between the packing sections. A water wash section is included to remove MEA from the flue gas exiting the absorber. Rich MEA solvent loaded with CO_2 leaves the bottom of the absorber and is heated in the lean-rich heat exchanger before entering the regenerator (also referred to as stripper/desorber) above 2 x 10 m sections of Mellapak 252Y structured packing. The vapour stream

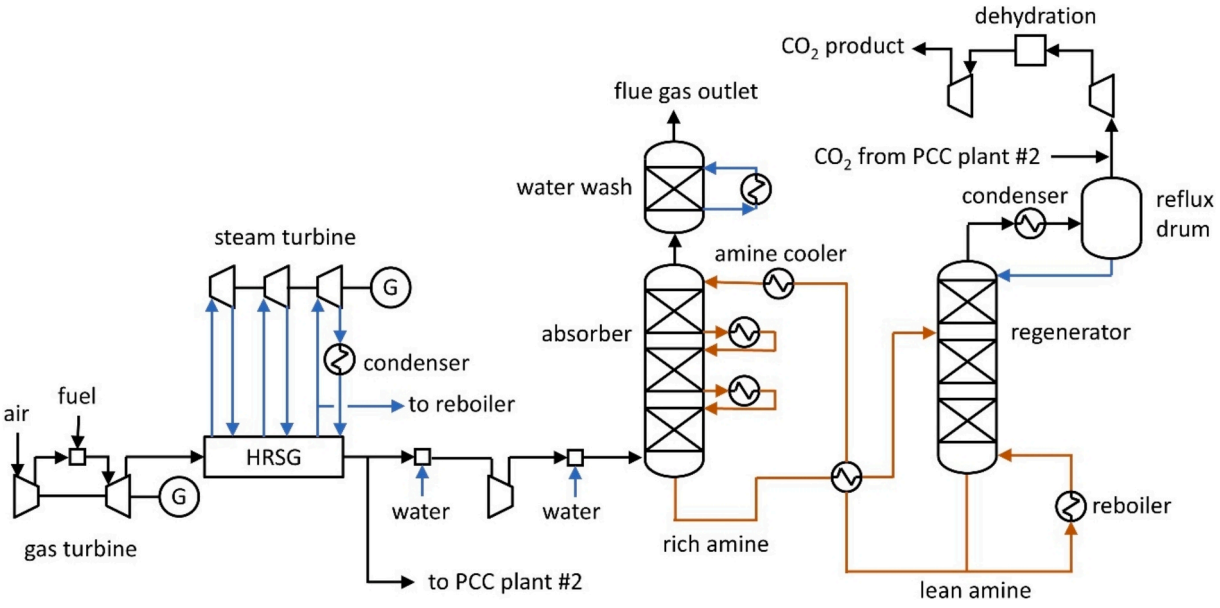


Fig. 1. Simplified schematic of CCGT process with PCC assumed in this study. 2 PCC plants operate in parallel to process the CCGT flue gas stream. CO_2 compressor has 4 stages with intercooling (2 upstream of dehydration and 2 downstream), detail not shown in schematic.

leaving the top of regenerator is cooled to condense water which is returned above a 4 m wash section of Mellapak 252Y structured packing in the top of the regenerator. The CO_2 product stream flows to a compressor with 2 stages of compression and intercooling, followed by desiccant dehydration, and 2 more stages of compression with intercooling to 15.2 MPa (final stage is supercritical). A reboiler supplies the heat input at the bottom of the regenerator column using LP steam extracted from the CCGT. The lean solvent from the bottom of the regenerator is cooled in the lean-rich heat exchanger and lean amine cooler before returning to the top of the absorber. Details on the process design assumptions are provided in the Supplementary Material.

We assess the impact of lean/rich solvent CO_2 loading, absorber packing height, and CO_2 capture rate on the cost of electricity generation and marginal cost of avoided emissions based on both direct CO_2 emission intensity and cradle-to-gate life cycle global warming (GW) intensity. We consider three assumptions for regenerator reboiler temperature with variable pressure to achieve a range of lean solvent CO_2 loading: 126, 131, and 136°C. 126°C is the maximum reboiler temperature in the pilot plant dataset used to validate the process model [29], but higher regenerator reboiler temperatures have been proposed in other studies to improve the economics of ultra-high capture rates [16,20].

For the CCGT and NG supply assumed in this study, 99.14% gross-CO_2 capture is equivalent to 100% fossil-CO_2 capture.

2.2. Economic model

Levelised cost of electricity (LCOE) was calculated based on estimates of the capital and operating expenses and assumed financial parameters (Table 1). The methodology and assumptions follow Sampath et al. [32]. This study uses real US dollars (USD) with a cost base of January 2025 and assumes no real price escalation. Costs were adjusted to a January 2025 base using the US producer price index for mining, utilities, and manufacturing industries (US PPI) [33]. The project costs are based on construction in the USA. The baseline after-tax weighted average cost of capital (ATWACC) is based on a market capitalisation weighted average of companies in the power generation industry [32]. We assess the sensitivity of LCOE and MCAC to ATWACC, NG price, and capacity factor.

The LCOE (USD/MWh) for each design case is calculated based on the estimated capital cost, annual operating and maintenance costs (AOM), fuel costs (FC), and annual electricity production (AEP, MWh) using eqn. (1). Annual electricity production is based on the net electricity output after deducting ancillary loads associated with the PCC plant.

$$LCOE = \frac{TASC^{*}FCR + AOM + FC}{AEP} \tag{1}$$

Total as-spent capital (TASC) for each case is calculated from the total overnight cost (TOC) using eqn. (2) and the fixed charge rate (FCR) is determined using eqns. 3–6.

Table 1
Baseline financial assumptions used in this analysis. Based on ref. [32].

item	assumed value
plant life	30 years
capacity factor	90%
asset life for depreciation	28 years, straight line
construction duration	3 years
construction spend profile	10%/60%/30% per year
effective tax rate	25%
ATWACC	5.19%
NG price	$3.25/GJ (HHV)

$$TASC = TOC^{*}\left(1 + \sum_{m=1}^{3}\sum_{n=1}^{m}(ATWACC^{*}\%Capital_n)\right) \tag{2}$$

Where TOC is the total plant cost for all equipment required for the process (equipment/materials, engineering, procurement, construction, and contingency) plus pre-production expenses, inventory, financing, and owner's expenses, and $\%Capital_n$ is the share of capital expenses in year n.

$$FCR = CRF^{*}PFF \tag{3}$$

$$CRF = \frac{ATWACC}{\left(1 - \frac{1}{(1+ATWACC)^{t}}\right)} \tag{4}$$

$$PFF = \frac{1 - TR^{*}PVD}{1 - TR} \tag{5}$$

$$PVD = \frac{1}{CL}\sum_{n=1}^{CL}\frac{1}{(1+ATWACC)^{n}} \tag{6}$$

Where CRF is the capital recovery factor, t is the plant life, PFF is the project financing factor, TR is the effective tax rate, PVD is the present value of depreciation, and CL is the asset class life for depreciation.

Capital and operating expenses for the CCGT are based on data published for a 1x1 H-class CCGT in the US NETL baseline study of power generation technologies [31]. The capital cost estimates for the PCC plant for each design case were calculated using a multi-level factorial cost model of the absorber, regenerator, and CO_2 compressor/conditioning subsystems which were further broken down into major components. Reference capital and operating cost estimates are based on a detailed front end engineering design (FEED) study for a MEA-based post-combustion CO_2 capture plant for the Panda Sherman 719 MW F-class CCGT in Texas, USA [34]. A detailed description of the capital and operating cost estimate assumptions is provided in the Supplementary Material.

MCAC (USD/tCO_2) between two capture rate cases with different direct CO_2 emission intensities (CDI, tCO_2/MWh) is determined using eqn. (7).

$$MCAC = \frac{LCOE_2 - LCOE_1}{CDI_1 - CDI_2} \tag{7}$$

We also present results for average cost of avoided CO_2 (CAC) (USD/tCO_2) for cases with CCS compared to an unabated CCGT (with no CCS) using eqn. (8).

$$CAC = \frac{LCOE_{CCS} - LCOE_{unabated}}{CDI_{unabated} - CDI_{CCS}} \tag{8}$$

The marginal cost of avoided life cycle greenhouse gas emissions (MCAGHG, USD/tCO_2e) between two capture rate cases with different cradle-to-gate life cycle global warming impact intensities (GWI, tCO_2e/MWh) is determined using eqn. (9).

$$MCAGHG = \frac{LCOE_2 - LCOE_1}{GWI_1 - GWI_2} \tag{9}$$

2.3. LCA model

Life cycle GHG emissions were assessed using the Ecoinvent database (Version 3.10) [35] as implemented in SimaPro [36] for background inventory and IPCC AR6 GHG characterisation factors (100-year global warming potential, aligned with UNFCCC GHG reporting and most prior studies) [37]. The LCA in the current study is focused on quantifying the effect of CO_2 capture rate on life cycle global warming impact. CO_2 capture rate has previously been shown to have little effect on other life

cycle impacts (e.g., eutrophication, toxicity) [24]. CCGT power generation must be fueled from a low GHG emission intensity NG supply chain to be consistent with net-zero GHG emission ambitions. Therefore, baseline life cycle inventory for the NG supply chain in this study is based on a detailed LCA of existing low-emission production practices and technologies (1.46 gCO_2e/MJ_{LHV}), including electrification, low pressure vapour recovery, and reducing flaring, venting, and fugitive emissions [24]. Global average NG supply (14.9 gCO_2e/MJ_{LHV}) was also considered to assess the sensitivity of the results to NG supply chain emissions. LCA process data for the CCGT and PCC plant were based on data published in the US NETL baseline study of power generation technologies [31] and a FEED study for a MEA-based PCC plant [34] respectively. The CCGT/PCC process data include materials and energy consumption during construction, operating consumables, waste disposal, maintenance, and trace pollutants. The process data do not include decommissioning or CO_2 sequestration infrastructure as these have previously been shown to make insignificant contributions to overall life cycle GHG emission intensity for CCGTs with PCC [24]. Details of the life cycle process data assumptions and Ecoinvent background processes selected are provided in the Supplementary Material.

3. Results and discussion

3.1. Optimal process design conditions for ultra-high capture

The cost of abating CO_2 emissions from CCGTs with PCC is minimised by selecting the highest permissible temperature for the regenerator reboiler (Fig. 2) considering thermal degradation of the MEA and available thermodynamic data for process design. Regenerator operating pressure can be adjusted to achieve different lean solvent CO_2 loadings at that temperature. For each reboiler temperature there is an optimal lean solvent CO_2 loading which minimises LCOE and corresponds to the regenerator inflection point where the temperature profile is relatively linear between the reboiler and top of the columns (e.g., 0.13 mol_{CO2}/mol_{MEA} in Fig. 3). This is the lowest solvent CO_2 loading that can be achieved efficiently at the reboiler temperature and assumed regenerator height without rapidly increasing specific reboiler duty due to increasing vapour outlet temperature at the top of the regenerator which leads to excess water in the vapour outlet stream (e.g., 75.6 t/h water vapour flow at 0.10 mol_{CO2}/mol_{MEA} v. 47.7/46.8 t/h at 0.13/0.15 mol_{CO2}/mol_{MEA} in Fig. 3). The excess water vapour is condensed from the CO_2 product in the overhead condenser and returned to the regenerator as liquid water with a resulting loss of energy from the regenerator. Lean solvent CO_2 loadings higher than optimal slightly increase LCOE primarily due to increased capital cost to provide a higher solvent circulation rate to achieve the same CO_2 capture rate.

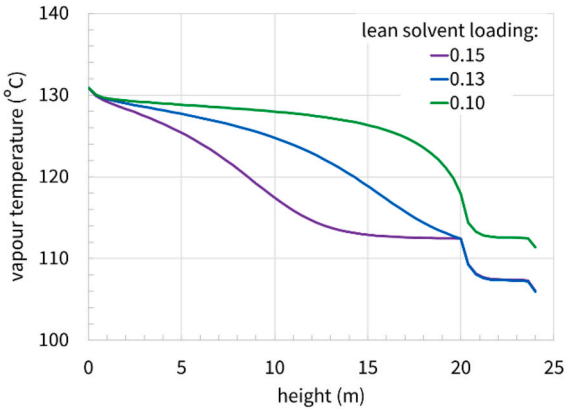


Fig. 3. Typical regenerator temperature profiles. Vapour temperature versus regenerator height for 0.15 mol_{CO2}/mol_{MEA} (purple line), 0.13 mol_{CO2}/mol_{MEA} (blue line), and 0.10 mol_{CO2}/mol_{MEA} (green line) with a regenerator reboiler temperature of 131°C, regenerator pressure of 244/241/238 kPa respectively, and gross-CO_2 capture rate of 99.14% (including CO_2 in the CCGT inlet air stream). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Higher regenerator reboiler temperature increases the regenerator operating pressure for a constant lean solvent CO_2 loading. The associated reductions in the CO_2 compressor power and specific reboiler duty, due to increased ratio of CO_2 to water in the vapour within the regenerator, approximately offset the reduction in steam turbine power output from increasing the LP steam turbine inlet pressure to maintain the temperature differential in the reboiler. Meanwhile, the increased regenerator pressure reduces capital costs associated with the regenerator (smaller vessel) and CO_2 compressor (less shaft power due to lower overall compression ratio) resulting in a net reduction in LCOE with increasing regenerator reboiler temperature.

The increase in MEA degradation with higher regenerator temperature is expected to be manageable up to at least 136°C, but long term testing under actual PCC plant operating conditions with solvent reclaiming is required to confirm model predictions and demonstrate continuous solvent reclaiming to maintain low levels of contaminants which may catalyse degradation [23]. Nonetheless, increased thermal degradation of MEA in the regenerator is not expected to materially affect the overall PCC plant economic calculations. The economic model in this study assumes a total MEA consumption rate of 2 kg/tCO_2, based on the Panda Sherman FEED study [34]. The regenerator reboiler temperature in the Panda Sherman process design is 131°C. MEA consumption accounts for 2.1% of the LCOE (including reclaimer waste

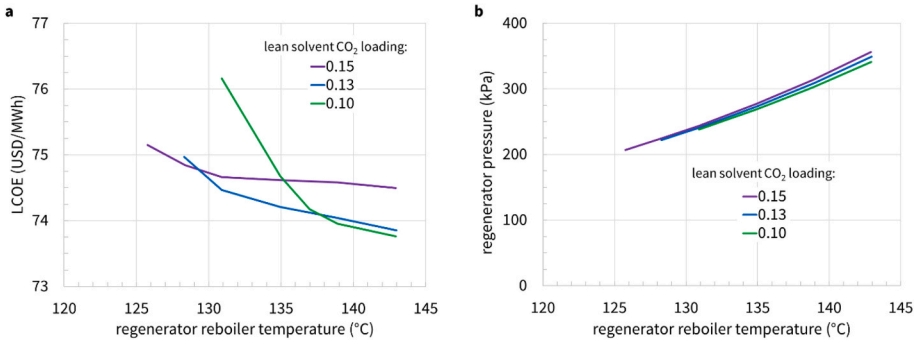


Fig. 2. Impact of regenerator reboiler temperature and lean solvent CO_2 loading on LCOE and regenerator pressure. (a) LCOE versus regenerator reboiler temperature for 1x1 H-class CCGT with 100% fossil-CO_2 capture (99.14% gross-CO_2 capture, including CO_2 in the CCGT inlet air stream) and lean/rich solvent CO_2 loadings of 0.15/0.45 (purple line), 0.13/0.46 (blue line), and 0.10/0.46 (green line) mol_{CO2}/mol_{MEA}. 3 x 10 m absorber beds with solvent intercooling between each bed. (b) Regenerator pressure versus regenerator reboiler temperature to achieve lean solvent CO_2 loadings of 0.15 (purple line), 0.13 (blue line), and 0.10 (green line) mol_{CO2}/mol_{MEA} for the cases in panel a. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

disposal). The predicted increase in MEA thermal degradation for operation at 136°C v. 126°C is 70 g/tCO_2, equivalent to 3.5% of assumed total MEA losses and 0.07% of LCOE.

Further testing is also required to validate the thermodynamic models for MEA-water-CO_2 at elevated temperatures. The process thermodynamic model used in this study was regressed using vapour-liquid equilibrium data between 40 and 120°C and has been shown to provide accurate predictions for pilot plant regenerator temperatures up to at least 126°C [19]. Publicly available vapour-liquid equilibrium data at temperatures greater than 120°C is not available. Nonetheless, the extrapolation of the thermodynamic model over the range of regenerator reboiler temperatures in this study is expected to provide sufficiently accurate results for the study objectives. The regenerator process model result which has the greatest impact on overall PCC plant process economics is specific reboiler duty, the amount of energy input required to the reboiler per unit mass of CO_2 captured. Process model predictions of regenerator vapour-liquid equilibrium and reboiler duty are heavily constrained by the large water vapour fraction within the tower (>95% mol at the bottom) and different thermodynamic models typically provide similar predictions. The regenerator pressure and specific reboiler duty predictions in this study for 131 and 136°C are very similar (within c. 1%) to values predicted in Michailos and Gibbins [16] and Mullen and Lucquiaud [20] respectively using a different thermodynamic model (CCSI).

The optimal lean solvent CO_2 loading at the chosen regenerator reboiler design temperature results in the lowest LCOE for any CO_2 capture rate between 95% and 100% fossil-CO_2 capture with constant absorber bed length (Fig. 4). Below 98% gross-CO_2 capture, lean solvent CO_2 loadings slightly above optimal provide similar LCOE (<0.1% higher due to higher capital cost associated with increased solvent circulation rate). Above 98% gross-CO_2 capture, lean solvent CO_2 loadings higher than optimal increase LCOE due to reduced CO_2 transfer near the top of the tower as the flue gas CO_2 partial pressure approaches the lean solvent CO_2 fugacity. Lean solvent CO_2 loadings lower than optimal increase LCOE considerably due to increased heat input to the regenerator reboiler when operating beyond the inflection point.

When variable absorber bed length is considered, the optimal bed length changes considerably with design capture rate, while the corresponding optimal rich solvent CO_2 loading remains relatively constant (Fig. 5). Attempting to achieve higher rich loading than optimal while maintaining capture rate requires large increases in the absorber bed height due to development of a rich mass transfer pinch at bottom of the absorber column as the solvent CO_2 fugacity approaches the gas phase inlet CO_2 partial pressure. Designing the absorber with lower rich solvent CO_2 loading than optimal increases the specific reboiler duty (at constant capture rate) which increases LCOE by more than the diminishing marginal cost benefit from reducing absorber bed length, which is dominated by packing volume required for mass transfer in the top portion of the absorber.

Based on the foregoing results, we used the following method to optimise the MEA-based PCC process design for variable CO_2 capture rates for the given flue gas flow/composition (Fig. 6):

1. Select the highest permissible regenerator reboiler temperature based on consideration of solvent degradation and available thermodynamic property data.
2. Determine the optimal lean solvent CO_2 loading. Calculate LCOE for a range of lean solvent loadings (by adjusting the regenerator pressure) while maintaining constant CO_2 capture rate by adjusting solvent circulation rate with fixed absorber bed height of 30 m. The optimal lean loading will have the minimum LCOE; it is the lowest lean solvent CO_2 loading that can be efficiently achieved at the given reboiler temperature and is characterised by a sharp increase in specific reboiler duty with further reduction in lean loading (c. + 0.1 GJ/tCO_2 for 0.01 mol_{CO2}/mol_{MEA} decrease in lean solvent CO_2 loading).
3. Determine the optimal rich solvent CO_2 loading. Calculate LCOE for a range of solvent circulation rates while maintaining a constant CO_2 capture rate (within range of 95–99%) by adjusting absorber bed height with lean solvent CO_2 loading fixed at the optimum determine in step 2. The case with the lowest LCOE will provide the optimal rich loading, which is the highest loading that can be achieved with the given flue gas composition without causing a large increase to absorber bed length which outweighs the cost benefit from the reduction in specific reboiler duty from higher CO_2 loading. In this study that corresponded to in-situ solvent CO_2 fugacity at the bottom of the absorber of approximately 90% of the flue gas CO_2 partial pressure.
4. The optimal lean and rich solvent CO_2 loadings can be used to determine the required solvent circulation rates for a range of CO_2 capture rates and the given inlet flue gas flow/composition. Absorber bed length is adjusted to achieve the target CO_2 capture rate with the calculated solvent flow rate and optimal lean solvent CO_2 loading.

This process will provide a near-optima combination of lean and rich solvent CO_2 loading and corresponding absorber height for a target CO_2 capture rate and selected regenerator reboiler temperature because LCOE is relatively insensitive to small changes in lean/rich loading (c. ± 0.01) near the optima (Fig. 4 and Fig. 5).

3.2. *Marginal cost of abating direct CO_2 emissions*

When optimal PCC process design conditions and absorber bed length are selected to minimise LCOE, gross-CO_2 capture rates up to 98%

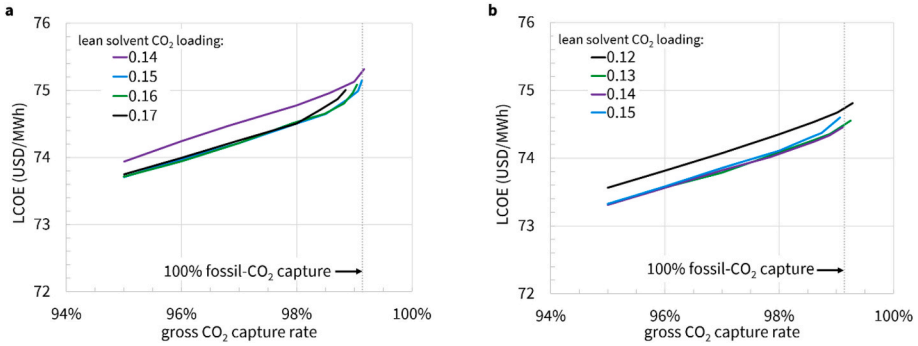


Fig. 4. Impact of CO_2 capture rate and lean solvent CO_2 loading on LCOE. LCOE versus gross-CO_2 capture rate (including CO_2 in the CCGT inlet air stream) for 1x1 H-class CCGT with variable lean solvent CO_2 loading. 3 x 10 m absorber beds with solvent intercooling between each bed. (a) 126°C regenerator reboiler temperature with lean solvent CO_2 loadings of 0.14 (purple), 0.15 (blue), 0.16 (green), and 0.17 (black) mol_{CO2}/mol_{MEA}. (b) 131°C regenerator reboiler temperature with lean solvent CO_2 loadings of 0.12 (black), 0.13 (green), 0.14 (purple), and 0.15 (blue) mol_{CO2}/mol_{MEA}. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

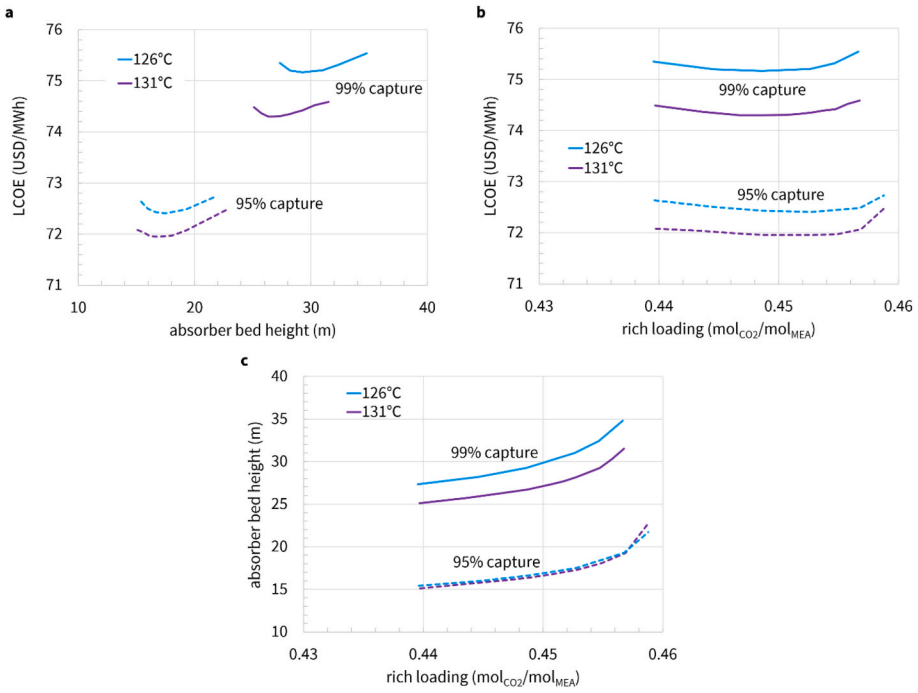


Fig. 5. Impact of rich solvent CO_2 loading on LCOE and absorber bed height. Constant gross CO_2 capture rates of 99.14% (solid lines) and 95% (dashed lines) for 1x1 H-class CCGT with regenerator reboiler temperature of 126°C and lean solvent CO_2 loading 0.15 mol_{CO2}/mol_{MEA} (blue lines) and 131°C and 0.13 mol_{CO2}/mol_{MEA} (purple lines). Gross-CO_2 capture rates include CO_2 in the CCGT inlet air stream. (a) LCOE versus absorber bed height. (b) LCOE versus rich solvent CO_2 loading. (c) Absorber bed height versus rich solvent CO_2 loading. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

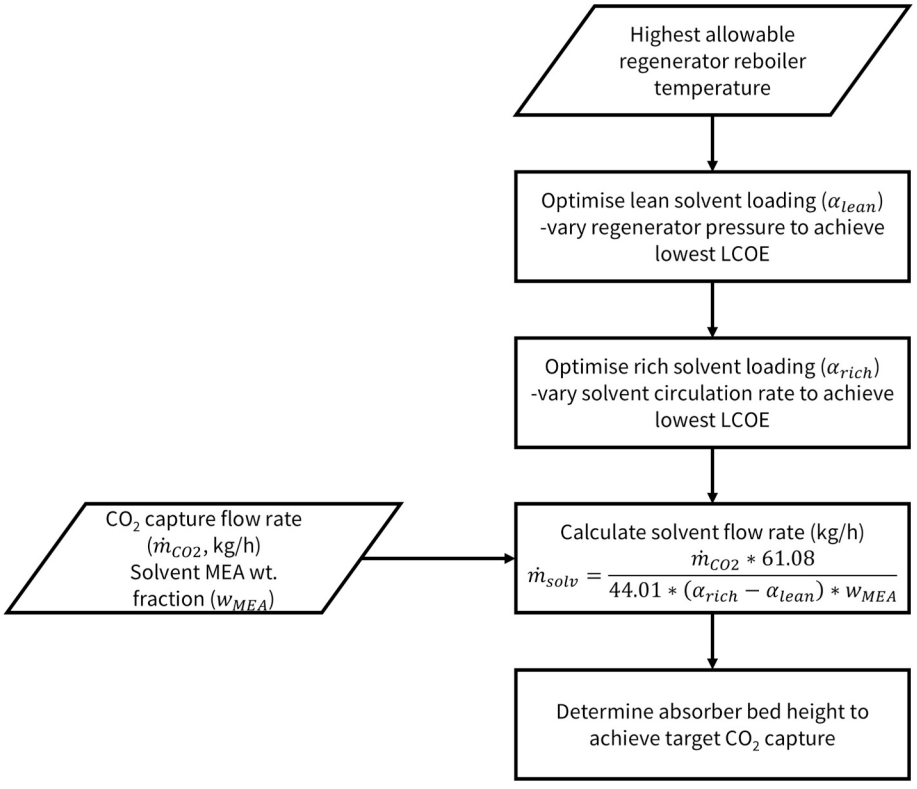


Fig. 6. Flowchart of process to optimise MEA-based PCC plant design conditions for variable CO_2 capture rates for a given flue gas flow/composition.

(99% fossil-CO_2 capture) can be achieved with relatively low MCAC in all three regenerator reboiler temperature scenarios (97–146 USD/tCO_2, Fig. 7). The MCAC for gross-CO_2 capture rates greater than 98% is considerably lower when the regenerator reboiler operates at higher temperatures to efficiently achieve lower lean solvent CO_2 loading which increases the CO_2 mass transfer driving force at top of absorber.

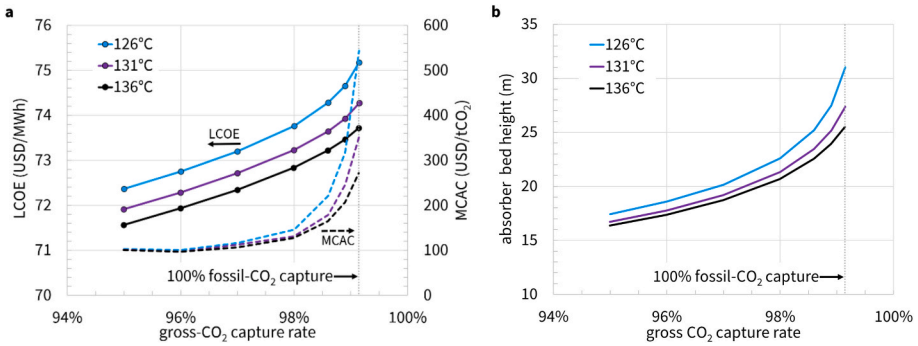


Fig. 7. Impact of CO_2 capture rate on LCOE and MCAC with optimal absorber bed height. (a) LCOE (solid lines, LHS) and MCAC (dashed lines, RHS) versus gross-CO_2 capture rate for 1x1 H-class CCGT with regenerator reboiler temperature/pressure of 126°C/207 kPa and lean solvent CO_2 loading 0.15 mol_{CO2}/mol_{MEA} (blue lines), 131°C/241 kPa and 0.13 mol_{CO2}/mol_{MEA} (purple lines), and 136°C/278 kPa and 0.11 mol_{CO2}/mol_{MEA} (black lines). Gross-CO_2 capture rates include CO_2 in the CCGT inlet air stream. Absorber bed length adjusted to achieve design CO_2 capture rate with optimal rich solvent CO_2 loading. 3 absorber beds with solvent intercooling between each bed. MCAC shown at 95% gross-CO_2 capture is relative to unabated CCGT. (b) Optimal absorber bed length versus gross-CO_2 capture rate for the cases in panel (a). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

While all three regenerator reboiler temperature scenarios in this study exhibit non-linear increases in absorber bed height and LCOE with CO_2 capture rate, a regenerator reboiler temperature of 136°C limits the MCAC to 164 USD/tCO_2 at 98.6% gross-CO_2 capture (99.5% fossil-CO_2 capture) and 271 USD/tCO_2 at 99.14% gross-CO_2 capture (100% fossil-CO_2 capture). For all three regenerator reboiler temperature scenarios, the MCAC between 95–96% gross-CO_2 capture is slightly lower than MCAC between no CCS and 95% gross-CO_2 capture. Therefore, average cost of avoided CO_2 compared to unabated CCGT operation is slightly lower for 96% gross-CO_2 than 95%. While MCAC increases considerably at capture rates near 100%, the effect on average CAC is minimal because the marginal cost applies to very small proportion of total emissions; average CAC at 100% fossil-CO_2 capture is only 1.7–3.4% higher than 95% gross-CO_2 capture (Supplementary Fig. 1).

The size of discretisation steps affects the calculated MCAC, particularly for capture rates > 98%. MCAC calculated in this study using optimal PCC process design conditions is considerably less than previous research when comparing similar discretisation steps. Du et al. [18] reported MCAC of 391 USD/tCO_2 between 97.7% and 99.0% gross-CO_2 capture from CCGT flue gas (adjusted to 2025 cost base using US PPI and same calculation methodology as the current study) v. 198 USD/tCO_2 in this study between 98.0% and 99.14% gross-CO_2 capture. Du et al. used a higher lean solvent CO_2 loading of 0.20 mol_{CO2}/mol_{MEA} and achieved 99.0% gross-CO_2 capture by increasing solvent circulation rate to reduce rich solvent CO_2 loading which caused a considerable increase in specific reboiler duty (4.10 to 4.32 GJ/tCO_2). The process design approach used in this study maintained constant specific reboiler duty (3.66 GJ/tCO_2). Danaci et al. [17] reported marginal cost of CO_2 capture of 280 USD/tCO_2 between 95.0% and 99.0% gross-CO_2 capture for CCGT flue gas v. MCAC of 135 USD/tCO_2 in this study between 95.0% and 99.14% gross-CO_2 capture. Insufficient details were reported in Danaci et al. [17] to enable adjusting their result to a 2025 cost basis (US PPI −13% in 2021 v. 2025) or a detailed comparison of results with this study. However, the high lean solvent CO_2 loading used in Danaci et al. (0.27 mol_{CO2}/mol_{MEA}) likely increased costs for ultra-high capture from low concentration sources.

There may be discontinuities in the relationship between LCOE/MCAC and capture rate due to limitations on allowable individual bed height between liquid redistribution. While there is no technical hard limit on individual bed height, limitations may be imposed based on commercial considerations (e.g., packing supplier performance guarantee). The results in Fig. 7 are based on the total absorber bed height split into 3 sections; however, total bed height required for gross-CO_2 capture rates less than c. 97% could be accommodated in 2 sections (assuming allowable individual packed bed height of 10 m). 3 beds are required to achieve greater than 97% gross-CO_2 capture for the process conditions and packing material assumed in this study. For a regenerator reboiler temperature of 131°C, the transition from 2 beds to 3 would cause MCAC to be 184 USD/tCO_2 between 97% and 98% gross-CO_2 capture v. 135 USD/tCO_2 assuming constant 3 beds and 178 USD/tCO_2 for 3 beds between 98% and 98.6% (Supplementary Fig. 2). Detailed analyses of specific design cases could consider using packing with higher surface area to intensify mass transfer and achieve higher capture rate without increasing the number of beds. Higher packing surface area could also be employed as part of a design strategy to future proof CCS installations as a means of increasing capture rates to adapt to changes in regulatory requirements or economic conditions.

3.3. Marginal cost of mitigating life cycle GHG emissions

Life cycle GW intensity is primarily determined by the CO_2 capture rate with only a 3% increase in life cycle indirect GHG emission intensity between 95% and 99.14% gross-CO_2 capture (Fig. 8). Regenerator reboiler temperature does not materially impact life cycle GW intensity over the range assessed in this study. With low emission NG supply, MCAGHG is 3% higher than MCAC at 96% gross-CO_2 capture, increasing to 5–9% higher at 99.14% gross-CO_2 capture. MCAGHG increases slowly from 100 to 103 USD/tCO_2e at 96% to 131–151 USD/tCO_2e at 98%. At 98.9% gross-CO_2 capture, MCAGHG exceeds 200 USD/tCO_2e for all regenerator reboiler temperatures and increases rapidly for higher capture rates. MCAGHG at 100% fossil-CO_2 capture is 285 USD/tCO_2e for a regenerator reboiler temperature of 136°C (0.11 mol_{CO2}/mol_{MEA} lean solvent CO_2 loading) and increases to 594 USD/tCO_2e for a regenerator reboiler temperature of 126°C (0.15 mol_{CO2}/mol_{MEA}). Similar to MCAC, the MCAGHG between 95–96% gross-CO_2 capture is slightly lower than MCAGHG between no CCS and 95% gross-CO_2 capture.

At 98.6% gross-CO_2 capture, residual direct CO_2 emissions account for 9% of life cycle GW intensity which is dominated by indirect emissions associated with the NG supply chain (43%) and operations and maintenance (28%, primarily supply of make-up MEA and disposal of solvent reclaimer waste, Supplementary Fig. 3). Construction and direct non-CO_2 emissions account for 8%/11% of life cycle GW intensity respectively. There are opportunities to reduce indirect emissions throughout the supply chain which may be employed in the future as industries and jurisdictions progress toward achieving GHG abatement commitments. The life cycle GW impact from the low emission NG supply scenario assumed for the baseline in this study could be further reduced (e.g., fugitive emission abatement, low-carbon electricity supply, electrification of drilling/completions equipment) [24]. The MEA consumption rate assumed in this study is conservative compared to results demonstrated in pilot plant testing, and could be minimised by

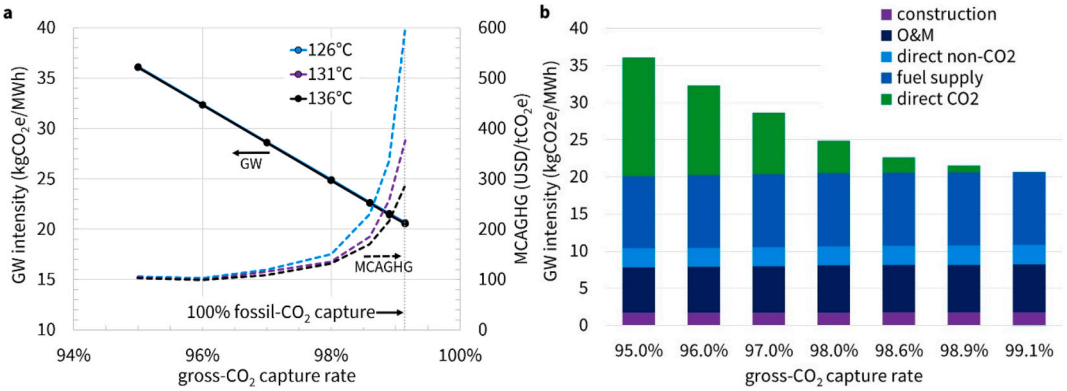


Fig. 8. Impact of CO_2 capture rate on life cycle GW intensity and MCAGHG. (a) Life cycle cradle-to-gate GW intensity (solid lines, LHS) and MCAGHG (dashed lines, RHS) versus gross-CO_2 capture rate for 1x1 H-class CCGT with regenerator reboiler temperature/pressure of 126°C/207 kPa and lean solvent CO_2 loading 0.15 mol_{CO2}/mol_{MEA} (blue lines), 131°C/241 kPa and 0.13 mol_{CO2}/mol_{MEA} (purple lines), and 136°C/278 kPa and 0.11 mol_{CO2}/mol_{MEA} (black lines). Gross-CO_2 capture rates include CO_2 in the CCGT inlet air stream. Absorber bed length adjusted to achieve CO_2 capture rate with optimal rich solvent CO_2 loading. 3 absorber beds with solvent intercooling between each bed. NG supply chain based on existing low-emission production practices (1.46 gCO_2e/MJ_{LHV}) described in Cownden and Lucquiaud [24]. MCAGHG shown at 95% capture is compared to unabated CCGT. (b) Breakdown of contributions to cradle-to-gate life cycle GW intensity for the cases in panel (a) with regenerator reboiler temperature of 126°C and lean solvent CO_2 loading 0.15 mol_{CO2}/mol_{MEA}. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

adding degradation inhibitors and/or effective reclaiming [38]. Life cycle emissions from MEA supply (3.1 $kgCO_2e/MWh$) could be mitigated through decarbonisation of the chemical manufacturing processes (e.g., low emission electricity/NG supply or deploying CCS to mitigate emissions from ethylene plants [39]). Alternative disposal strategies for the solvent reclaimer waste could reduce GHG emissions compared to unabated incineration assumed in this study (2.2 $kgCO_2e/MWh$).

With global average NG supply, life cycle GW intensity (112–126 $kgCO_2e/MWh$ for 95–99.14% gross-CO_2 capture) is dominated by NG supply chain emissions: 79% at 95% gross-CO_2 capture, increasing to

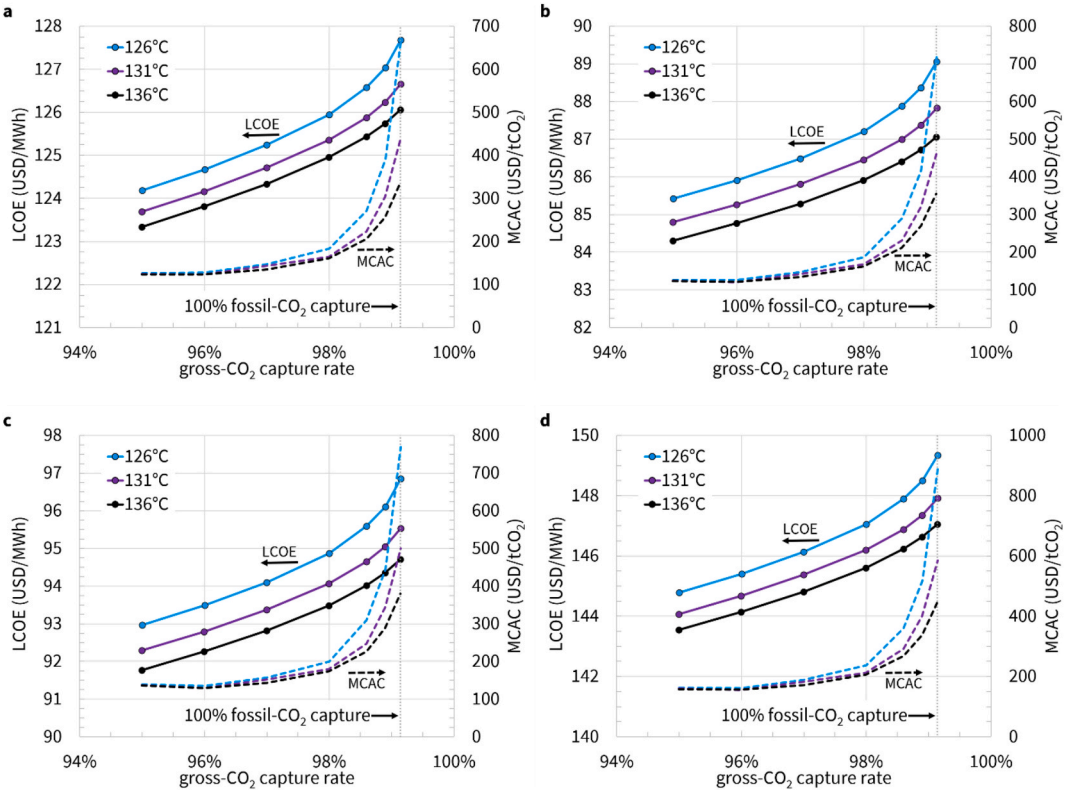


Fig. 9. Impact of financial assumptions on LCOE and MCAC for variable CO_2 capture rate with optimal absorber bed height. LCOE (solid lines, LHS) and MCAC (dashed lines, RHS) versus gross-CO_2 capture rate for 1x1 H-class CCGT with regenerator reboiler temperature/pressure of 126°C/207 kPa and lean solvent CO_2 loading 0.15 mol_{CO2}/mol_{MEA} (blue lines), 131°C/241 kPa and 0.13 mol_{CO2}/mol_{MEA} (purple lines), and 136°C/278 kPa and 0.11 mol_{CO2}/mol_{MEA} (black lines). Gross-CO_2 capture rates include CO_2 in the CCGT inlet air stream. Absorber bed length adjusted to achieve design CO_2 capture rate with optimal rich solvent CO_2 loading. 3 absorber beds with solvent intercooling between each bed. (a) NG price of 10.25 USD/GJ with baseline ATWACC (5.19%) and capacity factor (90%). (b) ATWACC of 8% with baseline NG price (3.25 USD/GJ) and capacity factor (90%). (c) Capacity factor of 60% with baseline ATWACC (5.19%) and NG price (3.25 USD/GJ). (d) Capacity factor of 60% and NG price of 10.25 USD/GJ with baseline ATWACC (5.19%). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

90% at 100% fossil-CO_2 capture (Supplementary Fig. 4). The higher dependence on NG supply chain emissions increases MCAGHG compared with the low emission NG supply scenario, considerably so for capture rates > 98%. MCAGHG increases slowly from 105 to 109 USD/tCO_2e at 96% to 140–162 USD/tCO_2e at 98%. MCAGHG at 100% fossil-CO_2 capture is 324 USD/tCO_2e for a regenerator reboiler temperature of 136°C (0.11 mol_{CO2}/mol_{MEA} lean solvent CO_2 loading) and increases to 769 USD/tCO_2e for a regenerator reboiler temperature of 126°C (0.15 mol_{CO2}/mol_{MEA}). MCAGHG with global average NG supply is 5–6 USD/tCO_2e higher than the low-emission NG supply scenario at 96% gross-CO_2 capture, increasing to 39–175 USD/tCO_2e higher at 100% fossil-CO_2 capture.

3.4. Sensitivity of MCAC to financial assumptions

NG price, ATWACC, and capacity factor do not materially affect the optimal PCC plant process design conditions, but all 3 financial assumptions have a substantial effect on MCAC (Fig. 9). Typical UK NG supply cost (10.25 USD/GJ) [40] causes a 22–28% increase in MCAC compared with the baseline assumption (typical US NG supply cost, 3.25 USD/GJ) over the range of capture rates and regenerator reboiler temperatures considered in this study. The intent of these two NG price scenarios (low-cost domestic production and high-cost imported liquefied NG) is to consider the effect of expected long-term average NG prices on policy decisions related to target CO_2 capture rate. Future work could consider the effect of short-term market pricing for NG on operational decisions for built PCC plants. Increasing the ATWACC to 8%, from the baseline assumption of 5.19%, results in a similar increase in MCAC (25–32%). Reducing capacity factor to 60%, from the baseline assumption of 90%, causes an increase of 33–34% in MCAC at 95% gross-CO_2 capture increasing to 40–41% at 100% fossil-CO_2 capture. The combination of higher NG price (10.25 USD/GJ) and reduced capacity factor (60%) increases MCAC 61–65% compared to the baseline assumptions. Capacity factor, ATWACC, and NG price have a greater impact on MCAC than other financial assumptions (Fig. 10). However, MCAC is less than 226 USD/tCO_2 for 98.6% gross-CO_2 capture and 136°C regenerator reboiler temperature (0.11 mol_{CO2}/mol_{MEA} lean solvent loading) for the variability in financial assumptions considered in this analysis. The most significant opportunities to reduce MCAC are lower PCC plant capital cost and lower ATWACC. Variability in LCOE and MCAC between regenerator reboiler temperature scenarios up to a gross-CO_2 capture rate of 98% is small relative to the impact of key economic assumptions.

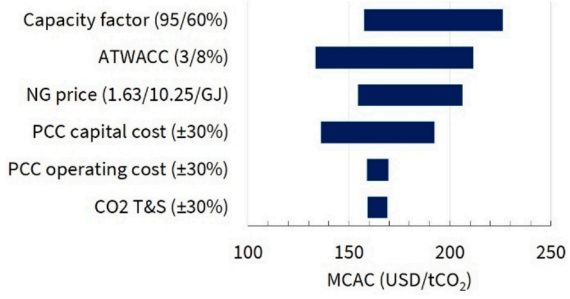


Fig. 10. Sensitivity of MCAC to key economic assumptions. Range of MCAC with variation in model economic assumptions for 1x1 H-class CCGT with gross-CO_2 capture rate of 98.6%, regenerator reboiler temperature/pressure of 136°C/278 kPa and lean solvent CO_2 loading of 0.11 mol_{CO2}/mol_{MEA}. Gross-CO_2 capture rates include CO_2 in the CCGT inlet air stream. Absorber bed length adjusted to achieve design CO_2 capture rate with optimal rich solvent CO_2 loading. 3 absorber beds with solvent intercooling between each bed.

3.5. Discussion and limitations

The presence of absorber intercooling does not materially affect LCOE at 95% gross-CO_2 capture for the CCGT process assumed in this study; however, it becomes increasingly important as CO_2 capture rate increases and the residual CO_2 partial pressure in the flue gas approaches the CO_2 fugacity of the lean solvent. For a lean solvent loading of 0.15 mol_{CO2}/mol_{MEA}, it is not economically feasible to achieve 100% fossil-CO_2 capture using the process configuration assumed in this study without absorber intercooling. To achieve 100% fossil-CO_2 capture without intercooling using a lean solvent loading of 0.11 mol_{CO2}/mol_{MEA}, the required absorber bed length is 6% longer resulting in a 0.5% increase in LCOE. Absorber intercooling has also been shown to reduce solvent degradation rates (c. −45 g_{MEA}/tCO_2 for CCGT flue gas in ref. [23]). Cooling the solvent to a lower temperature at the lean inlet and top intercooler than assumed in this study would allow for a small reduction in absorber bed height; however, it would require a different flue gas cooling strategy to ensure solvent water balance (e.g., packed bed direct contact water cooler) which would increase the cost of the flue gas conditioning system and was not considered in this study.

Other solvents would have different relationships between LCOE/MCAC and CO_2 capture rate. Solvents with lower CO_2 fugacity than MEA at low solvent CO_2 loading may have lower MCAC for ultra-high capture rates; while, it may be more difficult for solvents with higher CO_2 fugacity at low solvent CO_2 loading (e.g., piperazine [41] and CESAR-1 [42]) to achieve ultra-high capture due to reduced CO_2 mass transfer driving force at the top of the absorber. A detailed comparison of LCOE/MCAC between solvents and/or alternative CO_2 capture technologies with common economic assumptions is outside of the scope of the present study and would be difficult for proprietary solvents due to lack of publicly available information for performance and cost.

Capital cost estimates derived in this study for the PCC plant align closely with independent estimates for PCC plants based on a proprietary solvent (Cansolv) in the US NETL baseline study of power generation technologies for the same CCGT [31]. Ranges of total plant cost for the optimal PCC plant designs with 95% gross-CO_2 capture in this study are 587-617MM USD compared to an increase in total plant cost of 591MM USD for 95% gross-CO_2 capture from the baseline case of no CCS in the NETL study. The corresponding comparison for 97% gross-CO_2 capture is 602-633MM USD in this study versus 603MM USD in the NETL study. The cost estimates in this study and the NETL baseline assume construction in the US. There will be regional variability in capital cost (e.g., cost of labour) which could affect the LCOE and MCAC, but a detailed examination of geographic variability is beyond the scope of the present study.

Exhaust gas recycle from the outlet of the CCGT heat recovery steam generator to the inlet of the compressor could also reduce the LCOE and MCAC for CCGT with CCS by increasing the flue gas CO_2 partial pressure [43]; however, techno-economic analysis of the effect of exhaust gas recycle was outside of the scope of the present study.

A detailed assessment of constructability is not included in the present study; however, absorber diameter in the optimal design cases (11.9–12.2 m) is similar to the absorber in the Panda Sherman FEED study (11.8 m) [34] while absorber overall straight section height is taller – 48.6–49.6 m at 95% gross-CO_2 capture, increasing to 57.7–63.2 m at 99.1% gross-CO_2 capture versus 44.3 m in Panda Sherman FEED. The regenerator vessel diameter is smaller in the present study compared to the Panda Sherman FEED (5.2–5.7 m v. 6.7 m) while the regenerator overall straight section height is substantially the same (c. 43.5 m).

GHG mitigation policies which include carbon pricing in excess of the MCAC based on anticipated local economic conditions (e.g., NG price, cost of capital) would directly incentivise ultra-high CO_2 capture. The economically optimal CO_2 capture rate for CCGT with CCS in the context of achieving systemic net-zero GHG emissions could be determined by comparing the MCAC with the cost of high permanence CDR to

offset residual direct (and indirect) GHG emissions. However, there is wide variance in cost estimates of high permanence CDR reflecting different assumptions (e.g., timing, technology scale-up, installed capacity, energy supply, location) [6]. Young et al. [44] estimate net GHG removal costs of c. 380 USD/tCO_2e for DAC with potassium hydroxide solvent and 450 USD/tCO_2e for enhanced weathering for first-of-a-kind plants in the US, with cost decreasing to c. 180 and 200 USD/tCO_2e respectively for $GtCO_2$/year scale deployment (2019 cost basis, US PPI −28% v. 2025). Similar "author judgement ranges" of long-term future costs of 100–300 and 50–200 USD/tCO_2 for DAC and enhanced weathering were provided in Fuss et al. [45] and Babiker et al. [6]. Estimated current cost for solid sorbent DAC in Europe using grid-supplied electricity is c. 580–2500 €/tCO_2 net removal with high temperature heat pumps for sorbent regeneration and c. 540–1000 €/tCO_2 with industrial waste heat (2019 cost basis) [46].

Current cost estimates for high permanence CDR are considerably higher than the current cost of achieving ultra-high CO_2 capture from CCGT flue gas with MEA and optimal process design conditions. Further, the pathway to achieving forecast future CDR cost reductions is uncertain. Therefore, it is important to develop techniques to minimise the cost of achieving near 100% fossil-CO_2 capture from flue gas. 100% fossil-CO_2 capture during transient variation of flue gas flow rate (5200–8000 Nm^3/h) and composition (9–14%mol CO_2) has been demonstrated during 75 h of pilot plant testing [47] and greater than 97% gross-CO_2 capture has been demonstrated during a simulated CCGT startup using solvent storage [48]. Long-term pilot plant testing will be important to identify and develop solutions for operational issues that may arise during ultra-high CO_2 capture such as optimising process control, dynamic process response, solvent management, impact of flue gas variability, and impact of flow maldistribution within the absorber packing on capture rate. Developing real-time monitoring and control strategies to enable consistent attainment of ultra-high CO_2 capture with the regenerator operating near optimal conditions to minimise energy consumption is an area of active research [22]. Dynamic process modelling and predictive control may provide additional insight into transient behaviour and opportunities for optimisation (e.g., refs. [49,50]) but is beyond the scope of the present study. For MEA, future work to improve thermodynamic models at temperatures $> 120°C$ and assess the impact of higher regeneration temperatures on solvent degradation under realistic operating conditions is required. It is also crucial to implement opportunities to reduce NG supply chain emissions through global deployment of low-emission production practices.

The sensitivity of LCOE and MCAC to cost of capital highlights a role for government and policymakers in minimising the perceived financial and/or regulatory risk associated with construction of PCC facilities to reduce the cost of capital for PCC project proponents. The effect of reduced capacity factor on MCAC needs to be considered as CCGTs are expected to increasingly provide dispatchable generation to support higher penetration of intermittent wind and photovoltaic power generation in many jurisdictions. While reduced plant capacity factor increases the absolute value of LCOE and MCAC, it does not materially affect the optimal PCC plant design conditions and MCAC with plant capacity factor of 60% remains below current estimates for high integrity, high permanence CDR. Nonetheless, rigorous multi-variable optimisation could be performed with case-specific economic assumptions and material/labour cost data to further refine optimal PCC plant design conditions. In jurisdictions where low capacity factor is combined with higher NG prices, the economically optimal CO_2 capture rate may be below 98% gross-CO_2 if CDR achieves anticipated future cost reductions.

4. Conclusion

This study combines for the first time rigorous techno-economic analysis and LCA to determine the effects of optimising key process design parameters for ultra-high CO_2 capture from CCGT flue gas using a regenerative MEA solvent absorption process. MCAC to achieve 100% fossil-CO_2 capture in this study with optimal process design parameters is c. 50% lower than previous studies. Considerably lower MCAC shown in this study may help establish guidelines or regulatory requirements for residual CO_2 emissions from capture plant installations. This study also establishes a new standard for economically viable CO_2 capture rate for MEA-based absorption, a common benchmark used for comparison against other solvents and capture technologies.

Lower lean solvent CO_2 loading in this study (0.11–0.15 mol_{CO2}/mol_{MEA} v. 0.20 in Du et al. [18] and 0.27 in Danaci et al. [17]) enables ultra-high capture with reasonable absorber column height while maintaining constant rich solvent CO_2 loading and, therefore, constant regenerator specific reboiler duty. Elevated regenerator reboiler temperature and pressure in this study (131-136°C and 207–278 kPa) enables low lean solvent CO_2 loading to be achieved with low regenerator specific reboiler duty (3.7–3.8 GJ/tCO_2).

Other novel contributions of this research include quantifying the difference between marginal cost of mitigating direct CO_2 emissions and life cycle GHG emissions for CCGTs, and developing guidelines to minimise the cost of ultra-high CO_2 capture from a given CCGT flue gas stream based on operating strategies that have been successfully demonstrated to minimise regenerator specific reboiler duty [21]:

1. Select the highest permissible regenerator reboiler temperature.
2. Determine the minimum lean solvent CO_2 loading that can be efficiently achieved at that reboiler temperature.
3. Determine the cost optimal rich loading based on trade-off between absorber height and regenerator duty.
4. The solvent circulation rate for a range of capture rate scenarios can be determined from the economically optimal lean/rich solvent loadings.

The baseload CCGT with MEA-based PCC in this study achieved 99.5% fossil-CO_2 capture with MCAC of 164 USD/tCO_2 with optimal process design conditions and a regenerator reboiler temperature of 136°C, while 100% fossil-CO_2 capture could be achieved with MCAC of 271 USD/tCO_2. While the rate of MEA thermal degradation is expected to be positively correlated with reboiler temperature, the absolute contribution to overall MEA degradation/losses is not expected to materially affect process economics up to at least 136°C [23]. Future studies could perform similar optimisations for other CO_2 capture solvents.

Higher NG prices or cost of capital than the baseline typical US-based values in this study (3.25 USD/GJ NG price, 5.19% ATWACC) increased MCAC, but 98.8% fossil-CO_2 capture could be achieved with 10.25 USD/GJ NG or 8% ATWACC for a marginal cost of 161/162 USD/tCO_2 respectively. The results show that anticipated load factor for CCGTs will be an important consideration for design CO_2 capture rates as intermittent renewable energy provides an increasing share of generation in many jurisdictions, particularly where this is combined with higher NG prices.

Marginal cost of reducing life cycle GW impact is 3–9% higher than abating direct emissions over the range of process design conditions in this study with a low-emission NG supply chain. For 100% fossil-CO_2 capture, MCAGHG varies from 285 to 594 USD/tCO_2e for regenerator temperatures between 126 and 131°C – within the wide range of current and future cost forecasts for high permanence CDR. This highlights the need for flexibility in policies, regulations, and PCC plant design to be able to adapt as CDR technology evolves while also encouraging point-source PCC deployment by reducing financial risk for project proponents. Detailed case-specific comparisons between CDR and MCAGHG for PCC should carefully consider permanence, system boundaries, policy incentives, and carbon accounting rules. Nonetheless, the results show that ultra-high CO_2 capture (>99% of fossil-CO_2) from CCGTs with MEA is expected to be economic compared to current and foreseeable CDR technology to achieve net zero life cycle GHG emissions. Understanding process design options to minimise the cost of ultra-high CO_2

capture from CCGTs is particularly important to support net-zero ambitions given the current cost of high permanence CDR and uncertain path to reduce future costs.

Life cycle GW impact of CCGT power generation is considerably higher with global average NG supply chain emissions (14.9 gCO_2e/MJ_{LHV}) than the baseline low-emission scenario in this study (1.46 gCO_2e/MJ_{LHV}) – 112 v. 21 $kgCO_2e/MWh$ at 100% fossil-CO_2 capture – and MCAGHG is also higher, reinforcing the critical importance of low emission NG production practices. Further research to quantify the effect of higher reboiler temperature on thermal degradation of MEA under actual operating conditions with suitable solvent reclaiming to remove impurities and validate thermodynamic models for MEA-CO_2-water at temperatures greater than 120°C and solvent CO_2 loadings lower than 0.20 mol_{CO2}/mol_{MEA} is recommended.

CRediT authorship contribution statement

Ryan Cownden: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Mathieu Lucquiaud:** Writing – review & editing, Supervision, Resources, Methodology, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Ryan Cownden reports financial support was provided by Natural Sciences and Engineering Research Council of Canada (NSERC). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecmx.2026.102313.

Data availability

The Supplementary Material provides the data that was used in the analysis.

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