En Route to Zero Emissions for Power and Industry with Amine-Based Post-combustion Capture

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ABSTRACT: As more countries commit to a net-zero GHG emission target, we need a whole energy and industrial system approach to decarbonization rather than focus on individual emitters. This paper presents a techno-economic analysis of monoethanolamine-based post-combustion capture to explore opportunities over a diverse range of power and industrial applications. The following ranges were investigated: feed gas flow rate between 1−1000 kg $\cdot$s^{-1}, gas CO_2 concentrations of 2−42%$_{mol}$, capture rates of 70−99%, and interest rates of 2−20%. The economies of scale are evident when the flue gas flow rate is <20 kg $\cdot$s^{-1} and gas concentration is below 20%$_{mol}$ CO_2. In most cases, increasing the capture rate from 90 to 95% has a negligible impact on capture cost, thereby reducing CO_2 emissions at virtually no additional cost. The majority of the investigated space has an operating cost fraction above 50%. In these instances, reducing the cost of capital (*i.e.*, interest rate) has a minor impact on the capture cost. Instead, it would be more beneficial to reduce steam requirements. We also provide a surrogate model which can evaluate capture cost from inputs of the gas flow rate, CO_2 composition, capture rate, interest rate, steam cost, and electricity cost.

KEYWORDS: *CO_2 capture, capture rate, post-combustion capture, industrial CCS, capture cost, techno-economic analysis, decarbonization, BECCS*
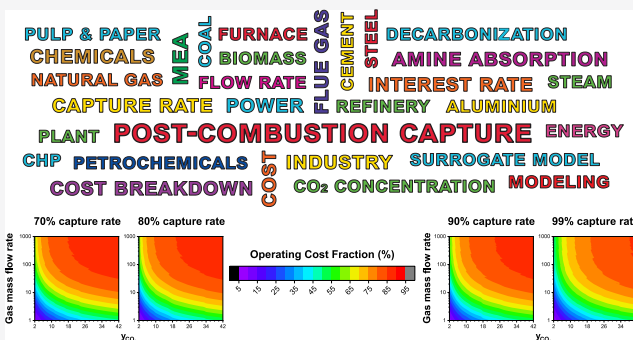
INTRODUCTION

Carbon Capture and Storage. To limit global warming to 1.5 °C, global GHG emissions will need to reach net negative levels by 2050. There needs to be rapid acceleration of decarbonization across all sectors, for example, energy, industry, and transport.[1] The efforts to reduce CO_2 emissions in the power sector have mainly focused on replacing coal with gas and increasing the deployment of renewables.[2] In 2019, the total global coal-fired power capacity was 9800 TWh,[3] with one-third of this being less than 10 years old,[4] and total natural gas capacity was 6300 TWh.[3] As much as 65% of the current global electricity generation continues to be from fossil fuels, predominantly in China, the US, and Asia Pacific regions.[3,5] Carbon capture and storage (CCS) will therefore have a vital role in the deep decarbonization of fossil fuel emissions, also providing dispatchable low carbon capacity and balancing intermittency from renewables.[6−8]

The mitigation of CO_2 emissions from energy-intensive industrial sectors such as cement and iron and steel is especially complex. Due to the international nature of these industries, the challenge will be to maintain cost competitiveness of industrial products.[9] Although electric solutions are technically feasible, these are not currently economically viable.[10] There is a need to identify strategies for industries to lower their CO_2 emissions

without undermining process efficiency or significantly adding to costs. CCS is recognized as the cost effective option for the decarbonization of industrial processes.[11] Compared to power, some industrial flue gases have higher CO_2 concentration (>10%$_{mol}$ CO_2). Such concentration ranges offer opportunities for cost savings with post-combustion capture.[12−14] However, there are also some instances of industrial emissions which can be fairly dilute such as petrochemical or refinery furnaces ($\leq$5%$_{mol}$ CO_2)[12] and aluminum refining ($\leq$10%$_{mol}$ CO_2).[15]

Post-combustion Capture of CO_2 in Power and Industry. Studies on post-combustion capture typically focus on evaluating the technical and economic performance of either power or industrial applications,[14,16,17] with most considering absorption-based systems using monoethanolamine (MEA). Flue gas characteristics can be unique with each application; they can vary greatly in terms of composition, flow rate, process conditions, and the presence of contaminants. Technical

challenges will need to be addressed upon integrating the amine-based absorption process, for example, flue gas pre-treatment. This section provides an overview of the sector-specific developments and advances in absorption-based CO_2 capture.

For power generation, aside from capital costs, the efficiency penalty that is associated with the integration of CCS is an important factor. Therefore, in order to minimize the energy requirements, some studies have focused on identifying the optimum operating conditions of the capture process.[18−21] In other cases, the optimization of process conditions was conducted for an integrated system and considered both the power plant and CCS plant.[22−24] Much of this work is studied under steady-state conditions; however, dynamic modeling has also been carried out to demonstrate the stability of the process with respect to process disturbances (*e.g.*, during flexible operation).[25−29] Flexible operation of amine absorption plants will be an important capability in electricity systems requiring dispatchable low carbon power, for example, to balance intermittency of renewables. The ability to operate amine absorption plants flexibly has also been demonstrated to be technically feasible at various test facilities.[30−36]

The energy requirements of CO_2 separation is primarily a function of the flue gas CO_2 concentration (assuming constant CO_2 capture rate, target product purity, and temperature).[37,38] As combined cycle gas turbine (CCGT) power plants have lower flue gas CO_2 concentrations of $3-4\%_{vol}$, the energy requirements of the capture process are higher compared to the pulverized coal (PC) case ($12-14\%_{vol}$ CO_2 concentration).[39−41] Canepa et al.[22] demonstrated that exhaust gas recirculation (EGR) in CCGT power plants increased the flue gas CO_2 concentration from 4 to $7\%_{mol}$ and reduced flue gas flow rate by 40%, resulting in a capture plant with significantly lower energy penalty and smaller size.[22,42]

For industrial applications, there is significant diversity in flue gas composition and flow rate.[13] Unlike power plants, emissions of CO_2 may come from different locations within the same industrial process and often with dissimilar CO_2 concentration.[16,43−45] Although difficult to achieve in practice, a method of combining flue gas streams (*e.g.*, iron and steel production and petroleum refineries) would likely be required to maximize CO_2 emission reduction.[14] However, a more practical approach is to have local smaller absorbers at each outlet and pump the rich amine to a central stripper. Given the heterogeneous nature of industrial emissions, the integration of CCS should consider industry-specific challenges; consequently, most studies often investigate one industrial sector at a time.

To help identify opportunities for CCS in the U.S. industrial sector, Bains et al.[12] developed a simple capture cost correlation as a function of flue gas CO_2 concentration. For initial deployment of CCS in the U.S., it was concluded that the sectors which are able to cost effectively achieve the greatest reduction in CO_2 emissions would be ethanol, cement, and ammonia production.[12] To date, the majority of the research on industrial applications of post-combustion capture has been to evaluate the technical and economic feasibility in relation to the cement, oil refining and iron and steel sectors,[14,16,43−45] with a few studies on various other industries.[14]

Initial assessments identified technologies that could be applied to capture CO_2 from cement plants, specifically studying post-combustion capture, pre-combustion capture, and oxy-combustion.[43] Pre-combustion would only mitigate the fuel-derived CO_2, not the larger emissions from the decomposition of carbonate minerals. Compared to oxy-combustion technol-

ogy, post-combustion capture using MEA has a higher CO_2 abatement cost—two times the cost per ton of CO_2 captured. However, post-combustion capture significantly reduces the net CO_2 emissions of the cement plant, avoiding 77% of the CO_2 emissions with a 85% capture rate, whereas oxy-combustion only avoided 52% of the CO_2 emissions. However, as high as 93% of the CO_2 emissions could be avoided if the post-combustion capture rate was increased to 95%.[43] Recent studies demonstrate that using advanced solvents (such as KS-1, CANSOLV, or AMP/PZ) for post-combustion capture would reduce energy requirements and thus significantly decrease the cost of capture and cost of CO_2 avoided compared to MEA-based systems.[16,46−50] The cost of advanced chemical absorption in cement plants was shown to be equivalent or even lower than other CCS technologies such as membranes, oxyfuel, and hybrid systems.[16] In the context of new-build cement plants, calcium looping technologies can provide improved techno-economic performance in comparison with other capture technologies, with lower costs and better energy efficiency.[48,49,51] However, the key advantage of post-combustion capture is the ability to retrofit the technology—a cost effective approach to reduce the emissions of existing cement plants.

In the case of the iron and steel industry, a range of measures exist for CO_2 emission reduction; these include improving energy efficiency (*e.g.*, sinter plant heat recovery), minimizing energy consumption, and using alternative fuels or reducing agents (*e.g.*, wood charcoal).[52] In addition to these measures, CCS could further decrease CO_2 emissions of integrated steel mills.[53] Post-combustion capture can avoid 50−60% of the CO_2 emissions, assuming a 90% capture rate using MEA regardless of the varying gas CO_2 content.[44,51] Some of the gas streams (*e.g.*, blast furnace gas) contain significant amounts of carbon monoxide; therefore, including a shift reactor could further increase CO_2 capture up to 85−99.5%,[52] thereby potentially further avoiding more CO_2 emissions. However, vacuum pressure swing adsorption (VPSA)[16] and calcium looping[51] have been shown to be slightly more cost effective than post-combustion capture. These technologies have not been tested at large scale, and challenges could be experienced in the scale-up.[16] In the case of VPSA, operating under vacuum may also present challenges in terms of sealing connections, and mechanical vacuum generation to the pressures suggested (5 kPa_a) at reasonable cost.

For oil- and gas-related applications, different process configurations for the retrofit of amine-based absorption in refineries of varying complexities have been proposed.[45,54] However, to integrate MEA-based CO_2 capture, flue gas desulfurization of some gas streams would be required. In addition, heterogeneity of the flue gases around the facility presents some technical challenges.[45] Andersson et al.[54] evaluated opportunities to use excess heat from the refinery process for amine regeneration in the capture plant. Specific capture cost increases with greater amounts of captured CO_2 due to increased supplementary heating requirements (provided by heat pumps).[54] In a study of CO_2 captured from waste gas combustion units, applying optimum operational conditions was shown to reduce energy consumption by 16%, where a response surface methodology was being used for optimization.[55] This highlights the value of optimization in determining the ideal operating conditions.

The pulp and paper industry is an energy-intensive sector with most of the CO_2 emissions arising from the combustion of

Table 1. Summary of Investigations into Capture Costs (unless Otherwise Indicated) in US\$·$t_{CO_2}^{-1}$ at Higher Capture Rates in the Published Literature[a]

	NGCC cases			Coal cases			
	Capture rate [%]						
Study	90	95	99	90	95	99	Solvent
Feron et al.,[62b]	93.9	93.1	101.3	65.1	65.4	69.0	30%$_{wt}$ MEA
Brandl et al.,[68c]	77.6	87.1	167				
Abu-Zahra et al.,[63b,c]				46.5	46.9	49.7	
Jiang et al.,[65b,c]				63.8	64.0	69.8[d]	
Hirata et al.,[66c,e]				1.00	1.01	1.04	KS-1

[a]An exchange rate of 1€:1.18US\$ was used where required. [b]Costs are on a CO_2 avoided basis. [c]Values are digitized from a figure. [d]Cost is for 99.7% capture rate. [e]Costs are scaled relative to 90% capture rate due to commercial confidentiality.

biomass onsite.[56] Coupling the pulp and paper production process with CCS could have the potential to provide net removal of atmospheric CO_2, that is, negative emissions as a bioenergy with CCS (BECCS) technology.[57,58] Studies indicate that post-combustion capture of CO_2 from the recovery boiler, and bark boiler flue gases offer the greatest CO_2 reduction potential.[57−59] The recovery boiler accounts for as much as 70−80% of the emissions for a pulp mill.[53] Absorption-based CO_2 capture is often considered due to the low CO_2 concentration of 12−13%$_{mol}$ in the flue gases from the recovery boilers,[56,58,59] which is similar to coal flue gas (12−14 %$_{mol}$ CO_2). The cost of CCS for pulp and paper mills depends on the process configuration and the CO_2 transport distance.[58] There are opportunities to reduce cost and achieve greater CO_2 removal through using alternative process configuration and heat integration.[56]

There is some interest in exploring new applications for CO_2 capture technologies in other industries. For instance, CO_2 capture from silicon refining processes[53] could potentially lower the carbon intensity of silicon supplied for photovoltaic cell manufacture. There have also been some alternative pathways for BECCS proposed. The integration of CO_2 capture in biomass-fired combined heat and power plants could be promising in countries with established biomass supply chains such as Sweden.[60] A comparative techno-economic study of CO_2 capture technologies for waste-to-energy plants[61] demonstrated that post-combustion capture using advanced amines has significantly lower CO_2 avoidance cost compared to a membrane-based system and MEA-based absorption. Unlike the advanced amine and MEA options, the membrane-based system was unable to achieve net-negative emissions as only partial capture was being considered to reduce energy penalty.[61]

The vast majority of literature on CO_2 capture uses an assumed capture rate of 90%. The capture rate has major implications on the residual CO_2 emissions in both power[62] and industrial[43] applications. The techno-economic evaluation by Abu-Zahra et al.[19,63] was one of the first studies to demonstrate that higher capture rates of 95% and 99% would only lead to marginal increases in energy consumption and cost. More recently, the use of higher capture rates with CCS (i.e., 99%) is recognized as a valuable and cost-effective strategy to meet global climate change targets as it enables power generation with zero emissions.[64,65] A comprehensive techno-economic analysis of natural gas and coal-fired power plants with capture rates of 99% confirmed that there were no technical limitations and a low marginal cost increase relative to 90% capture.[62,64] A follow up to this work showed that capture rates as high as 99.7% would only result in a small increase to marginal cost.[65] In the case of

coal-fired power plants, co-firing with 5−10% biomass could enable zero and even net negative CO_2 emissions at comparable costs to the 99.7% capture case.[64,65] These findings concur with an industry-led study by Hirata et al.,[66] which also demonstrated that 99.5% capture rate with the Advanced KM CDR process using KS-1 solvent applied to a coal-fired power plant would incur a small increase in marginal cost relative to 90% capture. A similar study was also carried out by Gao et al.[67] using 5 m Piperazine, which showed that increasing capture rate from 90 to 99% increased heat duty requirements by 5% with no increase in absorber height. Of those investigating the impact of higher capture rates on cost, their findings are summarized in Table 1.

Previous work by the authors[68] employs a different shortcut model to investigate scenarios where direct air capture of CO_2 and sequestration (DACCS) would be economically preferable to increased capture rates, and the impact of a range of US financial incentives on capture cost. They found for gas concentrations ≤8%$_{mol}$ that it was more cost effective to employ DACCS beyond certain capture rates. However, this conclusion is directly related to the chosen cost of DACCS (140 US\$· $t_{CO_2}^{-1}$). Their work also employs a constant molar feed flow rate, that is, the analysis is a function of CO_2 concentration and capture rate. In terms of process design, the columns utilize random packing, and a conventional process layout without restriction on equipment size is used, for example, only one absorber and one stripper. In this work, we investigate the cost of capture as a function of feed mass flow rate, CO_2 concentration, and capture rate. Structured column packing is used, and equipment is parallelized (i.e., consider multiple items of the same unit) which is required to be within reasonable size limits. Unlike our previous work, in this study, we consider the implications of higher CO_2 capture rates in both power and industrial applications. Furthermore, we provide insights into total annualized cost and operating cost fraction (OCF), breakdowns of capital and operating costs, impact of advanced solvents with reduced reboiler duty (compared against MEA), and the effect of the cost of steam, across the three operating variables. A data-driven surrogate model is also developed and provided for others to integrate into their work.

Study Objectives. Most of the aforementioned works employ rigorous, discretized process models developed in commercial software, either using existing model libraries (e.g., in Aspen Plus, ProTreat, gCCS) or user-developed models (e.g., gPROMS or MATLAB). This approach offers an accurate representation of the process, providing axial profiles for the absorber and stripper, while accounting for the changes in the vapor−liquid equilibrium (VLE) and mass transfer. The disadvantage of discretized models is that they are not
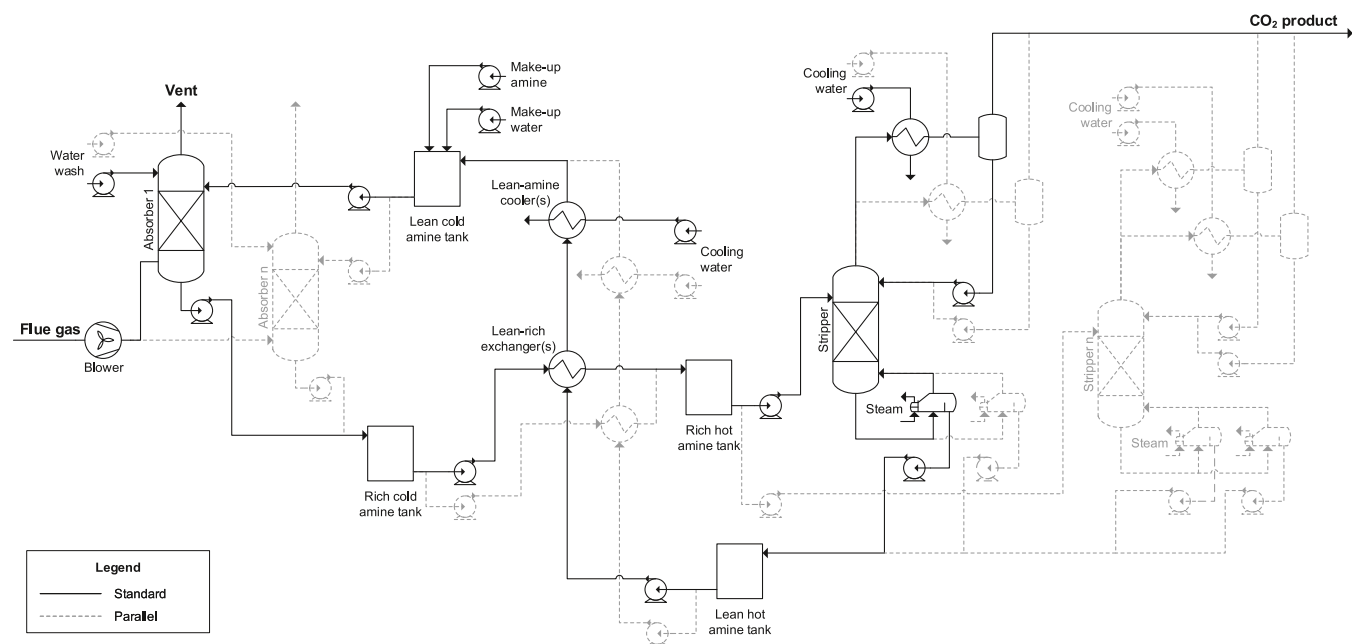
Figure 1. Process flow diagram of the model. Unit operations indicated with dashed gray lines are enumerated as required.

particularly suited for investigating a wide range of operating conditions efficiently. Consequently, studies typically focus on assessing a single application, for example, only power, or comparing similar applications, for example, cement and iron and steel. A more efficient modeling approach could facilitate techno-economic screening of post-combustion capture performance across a wider range of flue gas CO_2 concentration and flow rates, thereby enabling evaluation of cross-sector applications, that is, both power and industry. Furthermore, the evaluation of a wide range of applications using the same methodology and techno-economic assumptions provides capture costs that are comparable, which can be valuable for system-level assessments, for example, energy system optimization or integrated assessment modeling.

For this work, we have opted for a single-stage rate-based model to achieve a balance between swift and representative solutions. We explore opportunities for amine-based CO_2 capture across a comprehensive range of post-combustion capture applications. We consider a range large enough to cover most industrial and power plant applications, including BECCS, up to capture rates of 99%. In addition to the cost of capture over the range of operating conditions, we also provide breakdowns of the capital and operating costs and investigate the impact of the cost of capital.

This paper also provides additional value by presenting a surrogate model to determine CO_2 capture cost from the input parameters of the gas flow rate, CO_2 composition, capture rate, interest rate, steam cost, and electricity cost. This simple surrogate model along with a goodness-of-fit analysis are available as Supporting Information.

■ METHODS

The model of the MEA-based absorption process that is used in this study was published previously[69] and is illustrated in Figure 1. A feature of this model is that process equipment items are enumerated as necessary in order to keep sizing within reason (*i.e.*, column fabrication limitations and commercially available pumps). We present these size limits in Table S4. A range of

updates were made to the published version to expand the range of applicability, and these changes are summarized in Appendix A and described in Supporting Information (Section S5). The model assumes a single stage for both the absorber and stripper to allow swift solutions, but is rate-based in order to capture the effects of mass transfer over the wide CO_2 concentration range investigated.

The key process and economic parameters varied across a range in order to assess the techno-economic performance of the process include

- Feed gas CO_2 concentrations (y_{CO_2}) between 2 and $42\%_{mol}$,
- Feed gas flow rates between 1 and 1000 kg·s^{-1},
- CO_2 capture rates between 70 and 99%,
- Interest rate between 2 and 20%.

The range of conditions considered in this study account for the diversity in industrial post-combustion capture applications, both in size/scale and gas CO_2 concentration.

For the economic analysis, we calculate the capture cost, the relative contributions of capital and operating costs, the marginal cost of capture, and assess the effect of cost of capital on the capture cost. Itemized breakdowns of the capital and operating costs are also provided to investigate the change in distribution of cost with the variation of key parameters. For the breakdown of operating cost, cooling water refers to the make-up water required to compensate for evaporation from the system; town water refers to potable water used to maintain the water balance of the amine solution; and electricity accounts for the energy required to operate the feed blower and pumps. The capture cost is defined by eq 1, where TAC is the total annualized cost (eq 2). The TAC is the sum of the annual operating cost, and the annuity of the loan obtained for the capital costs *via* the capital recovery factor (eq 3).

$$\text{capture cost} = \frac{\text{TAC}}{CO_2 \text{ captured}} \tag{1}$$

$$\text{TAC} = \text{capital cost·CRF} + \text{operating cost} \tag{2}$$

$$CRF = \frac{i(1 + i)^n}{(1 + i)^n - 1} \qquad (3)$$

where i represents the interest rate and n is the economic life of the plant. In cases where the marginal cost of capture is presented, it was calculated using a central finite difference to approximate the derivative of the conventional function (eq 4).

$$Marginal\ cost = \frac{\partial(capture\ cost)}{\partial(variable)} \qquad (4)$$

where 'variable' represents the parameter of interest, that is, flow rate, capture rate, composition, and so forth. A key difference of this work is the approach used to determine the capital cost; in the original implementation,[69] the IChemE Factorial Cost Estimation method was used.[70,71] This method is fairly comprehensive, however, it is not particularly amenable to undertaking comparisons over a large range as the installed cost factor is a function of purchase cost (and hence size). This results in outputs with artefacts that are not related to the operation of the process. To decouple these effects and simplify data interpretation, a constant Lang factor of 3.6 was used. The implications of this are that the capital costs presented here are underestimated, as the IChemE method can easily return installed cost factors above 5. Furthermore, other fixed costs such as land, insurance, labor, and maintenance are also not accounted for in the capture costs. These factors are not only a function of scale but also plant location and emission source/ industry and will vary widely. Rubin et al. present a thorough discussion of these factors.[72] Therefore, we cannot generalize how these factors will impact the capture cost, as each is typically taken as a percentage of the capital or operating cost. As such, the effect on the capture cost will vary significantly depending on the specific scenario, that is, whether it is capital intensive or not. Furthermore, using the same fixed factor over such a wide range of feed gas conditions is debatable, as a small plant with 5 kg·s^{-1} feed flow will require a different proportion of capital and operating costs relative to a 500 kg·s^{-1} plant. Nevertheless, we present a comparison in Figure S3 for the 90% capture rate case using a fixed set of parameters taken from Jiang et al.[65] These results are indicative only. Cost increases from 1.5 to over 5 times are seen, depending on the scenario.

A summary of relevant economic parameters are presented in Table 2. It should be kept in mind that the costing approach used

Table 2. Summary of Economic Parameters Used in This Work

parameter	value
lang factor	3.6
CEPCI	616.3
economic life	25 years
interest rate	10%
MEA cost[21,63]	1100 US\$ ·t^{-1}
steam cost[70]	19.4 US\$ ·t^{-1}
electricity cost[70]	113 US\$ ·MWh^{-1}

in this work is sufficient to establish trends and corresponding insights. However, for the purposes of decision making, more rigorous costing should be undertaken for the applications of interest by an EPC firm.

Throughout this work, a lean loading of 0.27 mol$_{CO_2}$·mol$_{MEA}^{-1}$ was used. It was found to result in the lowest capture cost for the range of feed concentrations investigated (Figure S1). This

minimum occurs as a result of the trade-off between increased steam consumption at lower lean loadings, and increased capital cost due to greater amine circulation rate at higher lean loadings. In order to form a comparison between MEA and advanced solvents with reduced reboiler duty, it is assumed that the absorbent has the same characteristics as MEA (cost, physical properties, VLE); however, the reboiler duty is scaled by a factor of 2.7/4.0.[47]

We compared our model to other published works employing a detailed discretized model. This data is presented in the Supporting Information (Section S2.3). First, we performed a cost comparison for a range of applications. We did not observe a distinct trend: the cost values returned by our model are either above, below, or in line with those of the other publications. Agreement is obtained within 0.22% in one case, and up to 188% deviation in the worst case, with the remainder being between 1 and 36%. We attribute this observation to varying modeling approaches and techno-economic assumptions. In fact, this variation was one of the main motivations for undertaking this study and produce a comparison over a range of flue gas conditions on the same techno-economic basis.

Second, where possible, we matched the operating conditions of this model to another set of published works to compare process design output. Overall, we observe good agreement between metrics such as the liquid-to-gas ratio, specific reboiler duty, and column diameters. However, no general trend appears with respect to packed height. We do expect that some variation will exist for the absorber at conditions where the driving force at the top are poor, that is, low feed CO_2 concentration combined with a higher capture rate and/or high solvent lean loading. In this model, the column diameter is first determined based on the operating flooding percentage; then, the volume of packing required is determined based on the mass transfer and amount of CO_2 to be absorbed. The packing height is then obtained as the quotient of those two values. As the columns are treated as a single stage and mass transfer properties are calculated based on the log-mean of conditions at the inlet and outlet of the columns, low driving force at the top of the absorber can yield excessive packing volume. For instance, when comparing to pilot plant data for 4%$_{mol}$ CO_2,[36] variations as much as twice the packing volume can be returned. Conversely, when compared to detailed modeling for a 12%$_{mol}$ CO_2 case,[20] agreement within 11% can be achieved. For the stripper design, operating temperature and pressure has a great impact on the design. In this model, the stripper bottom temperature and pressure are set based on the lean loading desired to be achieved. Therefore, it was not straightforward to match the stripper operating conditions to the reference works. Some combinations result in flashing of the feed which impacts the diameter, and operating conditions alter the driving force, and hence height. Section S2.3 should be referred to for further numerical comparisons, as well as the work by Fosbøl et al., where they conducted a "multi-laboratory study", whereby the same specifications were provided to a number of groups to undertake detailed process modeling and the variability was analyzed. They found that up to 10% variation in outputs was common, and the outlet gas flow rate from the columns could disagree up to 20%, and the condenser duty by up to 15%.

A range of example power and industrial applications are used for comparison, presented in Table 3. The data for the applications were obtained from a range of sources,[43,44,73,74] and characteristics of the coal-fired applications were calculated based on a described method in Danaci et al.[69] For the biomass

Table 3. Capture Costs in US\$·$t_{CO_2}^{-1}$ at Different Capture Rates for the Reference Post-combustion Capture Applications

application	y_{CO_2} [%mol]	flow rate [kg·s⁻¹]	symbol	capture cost [US\$·$t_{CO_2}^{-1}$] capture rate [%]				
				70	80	90	95	99
500 MW NGCC	3.98	791	○	58.7	59.3	61.7	64.7	73.4
500 MW NGCC 50% EGR	8.19	401	△	49.0	49.2	50.3	51.8	55.9
500 MW high-rank coal	11.7	503	◇	45.6	45.8	46.7	47.8	50.6
500 MW low-rank coal	12.5	1077	□	44.8	45.1	45.9	46.8	49.4
500 MW biomass	14.4	503	▷	44.1	44.3	44.9	45.8	48.0
1 MMtpa cement	18.6	162	▽	43.4	43.4	43.9	44.5	46.3
1 MMtpa steel	23.2	164	☆	42.3	42.3	42.6	43.2	44.8
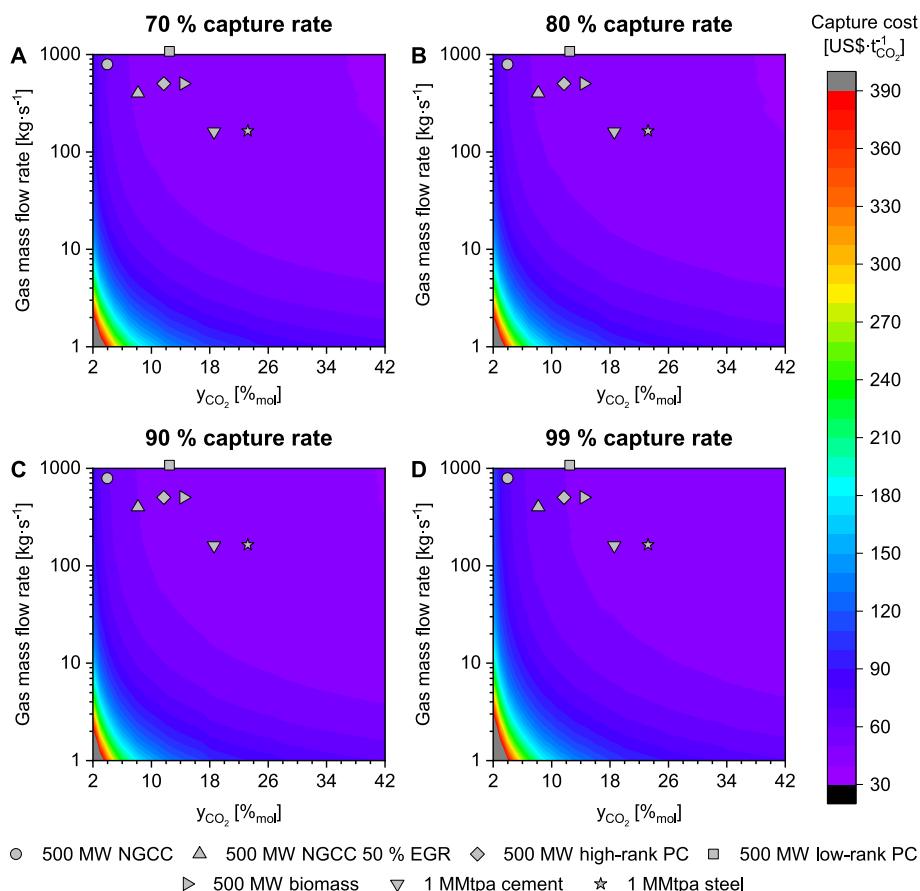
Figure 2. Cost of capture as a function of gas composition and flow rate at four different capture rates: 70 (A), 80 (B), 90 (C), and 99% (D).

application in particular, Ali et al. have provided data for a U.S. forestry residue with an LHV of 18.1 MJ·kg⁻¹, $48\%_{wt}^{A.R.}$ carbon, and excess combustion air of 15%.[74]

RESULTS AND DISCUSSION

Power and Industrial Applications of CO_2 Capture. A summary of the capture costs for the reference applications considered is provided in Table 3, which includes gas-, coal-, and biomass-fired power plants, as well as cement and steel production. The capture costs for the coal-fired power applications are comparable with those for biomass power due to similarities in flue gas composition. As shown in Table 3, capture costs increase with greater capture rate and lower gas CO_2 composition.

Figure 2 presents the capture costs as a function of the gas mass flow rate and CO_2 concentration at different CO_2 capture rates. In the context of the power and industrial applications considered, capture cost at 99% capture rate are not disproportionately higher compared to 90% capture. In the majority of cases, 10% additional CO_2 can be captured for ≤10% additional cost per ton of CO_2. For flue gases with $\leq 8\%_{mol}$ CO_2, a decision would need to be made whether the additional cost remains worthwhile. In a net-zero emission context, we expect the additional costs are still beneficial when compared to the cost of negative emission offsets (currently 1100 US\$·$t_{CO_2}^{-1}$[175]).

The disadvantage of higher capture rates is the increased up-front capital, the extent of which can be seen in Figure 6 for selected cases. A net-zero future will require more investment compared to a more general CO_2 emission reduction relative to a base year, such as those ratified in the Kyoto Protocol. However, in the long term, the marginal cost of maximizing CO_2 capture from point sources is small on per ton of CO_2 basis.
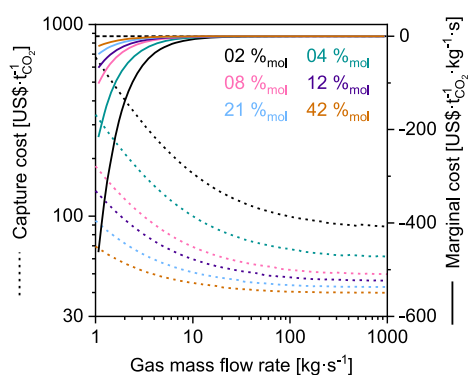
Figure 3. Capture cost (dashed lines) and marginal cost of capture (solid lines) as a function of the gas flow rate at a capture rate of 90% for selected gas CO_2 compositions ($2\%_{mol}$ to $42\%_{mol}$). Figure S10 shows a zoomed region of the higher flow rate area.

Figure 5. Capture cost and marginal cost of capture as a function of capture rate for selected gas compositions, at a gas flow rate of 500 kg · s^{-1}.

These findings are in agreement with previous work by other authors.[62,65,66] However, the exception is the NGCC case, which shows a greater increase in capture cost (19%) compared to Feron et al. (8%).[62] This is attributed to the difference in the modeling approach. The method employed here uses the log-mean of the inlet and outlet conditions to determine reaction and mass transfer parameters over the whole column. A discretized model takes advantage of the higher gas concentrations at the bottom of the absorber to a greater effect, resulting in a smaller packing volume compared to the single stage method—the impact of this would be more pronounced at lower feed gas concentrations.

At a given gas concentration (y_{CO_2}) in Figure 2, the most striking observation across the capture rates is that the impact from economies of scale diminish once gas flow rates increase beyond 20 kg·s^{-1}. In the context of plant size and technology readiness levels (TRLs), these results indicate that on a US\$ per t_{CO_2} basis, most of the cost reduction occurs as plant size increases from pilot scale (TRL 5) to demonstration scale (TRL 7) and up to commercial scale (TRL 8/9). The Petra Nova Carbon Capture plant is an example of a commercial-scale system, which is designed to capture 1.4 million t_{CO_2} per year, that is, a feed flue gas flow rate of ∼50 kg·s^{-1}.[76] Once the plant size reaches commercial scale, any further reduction in cost will need to come from other areas, such as more efficient manufacturing practices.

The results for the advanced solvent case are presented in Figure S7 and Table S1. In the NGCC and cement applications
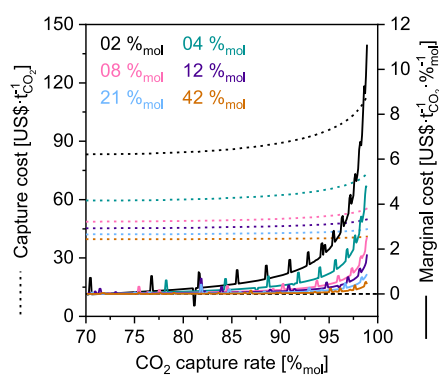
at 90% capture rate, the costs are 50.1 and 32.7 US\$·$t_{CO_2}^{-1}$, respectively. For reference, an NETL study determined that the cost of 90% capture from a 600 MWe NGCC plant using CANSOLV technology is 71.1 US\$·$t_{CO_2}^{-1}$.[77] However, the costing approach employed in their work includes additional indirect capital and operating costs, which are excluded in the costs in this work. Aside from reducing steam expenses, a reduced reboiler duty affects capital costs. A smaller heat exchange area is required, and the stripper diameter can also be reduced as less boilup is sent to the column. The percentage difference in cost between the cases is presented in Figure S8. There is a general trend of increasing reduction toward the upper right, which is in-line with the greater contributions of operating costs in this region (Figure 7).

The greatest variation and highest capture costs occur in the lower left region of Figure 2A−D, where y_{CO_2} <20$\%_{mol}$ and gas flow <20 kg·s^{-1}. Deployment at scales below 10 kg·s^{-1} would be unlikely as the total cost becomes dominated by capital costs. Advancements in process intensification, for example, rotating packed beds,[78,79] could potentially provide significant reductions in the capital cost and improve the economic viability of smaller scale absorption systems. In applications where capture costs of absorption may be considered expensive, there are potential opportunities for other technologies such as adsorption and membranes.

Effect of Scale and Gas CO_2 Composition. The impact of the flue gas flow rate on capture cost is shown in Figure 3 for a fixed capture rate of 90%. The cost to capture one ton of CO_2 reduces with the increasing flow rate due to economies of scale;
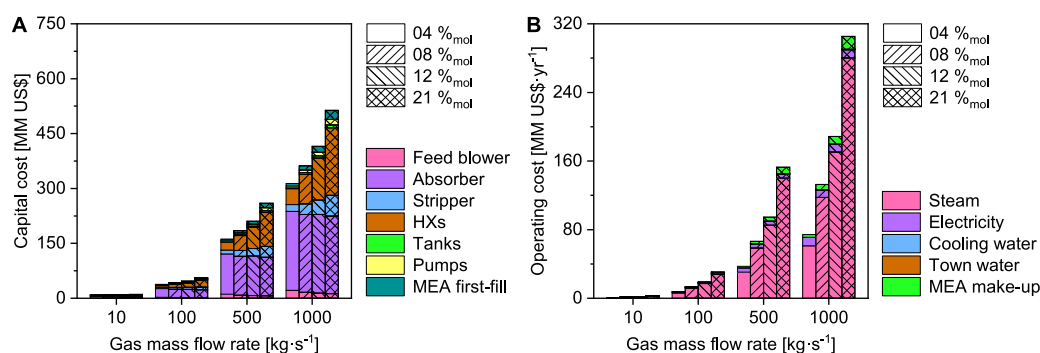

Figure 4. Breakdown of (A) capital cost and (B) operating cost at selected gas flow rates and compositions for a capture rate of 90%, where "HX" represents heat exchangers.
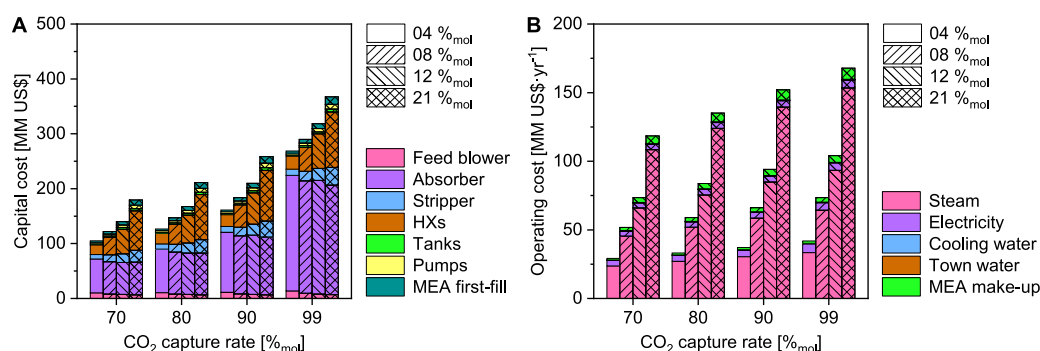
Figure 6. Breakdown of (A) capital cost and (B) operating cost at selected capture rates and gas compositions, for a gas flow rate of 500 kg·s^{-1}.

thus, the marginal cost of capture is negative. The effect of scale has the greatest impact on cost reduction at lower gas flow rates <20 kg·s^{-1} and low CO_2 composition ($\leq$4%$_{mol}$, *e.g.*, NGCC flue gas), which corresponds to the highest absolute values of marginal cost. However, cost reductions due to scale become insignificant once gas flow rates increase beyond 20 kg·s^{-1}, that is, where marginal cost is zero, which concurs with Figure 2 observations. In this region of negligible marginal cost, a potential optimization opportunity exists. Two plants of half size could be deployed and allow only half of the capture capacity to be lost during plant turnarounds. However, more detailed *NPV* analysis are required to determine whether this advantage is offset by increased maintenance requirements. Operating twice the number of equipment increases the incidents of regular and emergency maintenance. Increased land requirements may also be a potential disadvantage.

The capture cost consists of contributions from both capital and operating costs (eqs 1 and 2). The breakdown of capital and operating costs at selected flow rates is illustrated in Figure 4. Both capital and operating costs increase with higher mass flow rates. The cost of the absorber represents the largest share of the total capital cost. There is a marginal reduction in absorber cost as the feed gas CO_2 concentration increases. Although the increased gas CO_2 concentration results in a shorter absorber, the column diameter also increases to meet flooding requirements and accommodate the greater liquid flow rates required to achieve 90% CO_2 capture.

At a given gas flow rate, the ratio between the absorber and stripper costs does not remain constant as gas CO_2 composition increases. This suggests that general design assumptions, or rules of thumb,[80,81] are inadequate when modeling a broad range of separation applications (*e.g.*, large range of gas CO_2 composition). Consequently, rate-based sizing methods to determine the absorber and stripper dimensions are needed, instead of using an equilibrium approach, which assumes a number of theoretical stages and HETP.

Prima facie, the cost contribution of the stripper appears small relative to the absorber. Two main factors cause this outcome. The first is the operating temperature: the increased temperature results in significantly faster kinetics which reduces the packing height required. Second, the gas flow rate is significantly smaller (boilup from the reboiler) than the absorber, which reduces the column diameter. Therefore, the packing volume required in the stripper is substantially smaller than the absorber. We have compared our sizing results to a range of other published works in Supporting Information (Section S2.3.2).

The cost of the heat exchangers is the second largest portion of the total capital cost. Heat exchanger costs increase with higher feed gas CO_2 concentration due to the increased area requirement. As gas CO_2 concentration increases, greater heat exchanger area arises from the need for higher absolute reboiler duty and a greater amine circulation rate. Consequently, this increases the size of the reboiler, condensers, lean-rich exchangers, and lean amine coolers. As a result, at high gas CO_2 concentration of 21%$_{mol}$, the proportion of cost for heat exchangers becomes comparable to the absorber.

The cost of the feed blower decreases (*i.e.*, reduced blower size), with increased gas CO_2 concentration due to two main factors. At higher gas CO_2 concentration, the volumetric flow is lower owing to the greater gas density; also, the absorber pressure drop is lower due to the smaller column height.

As shown in Figure 4B, operating costs are dominated by the cost of steam. However, the absolute cost of steam will depend on the scenario assumptions (*e.g.*, steam extracted from a power plant steam cycle or generated by a dedicated boiler). To capture the effect of this variation, a data set was generated for selected capture rates at different steam costs (5, 10, and 40 US$·t^{-1}). These results are presented in Figures S4−S6. In situations where steam consumption is substantial, that is, high feed concentrations and flow rates, noticeable savings are possible if a lower cost steam supply can be sourced. The amount of steam required is proportional to the amount of CO_2 captured and commensurate with reboiler duty. Thus, steam requirements and cost increases with higher gas flow and CO_2 concentration. Similarly, electricity consumption increases with higher gas CO_2 concentration; this is due to the higher pumping duty associated with increased amine circulation rates. The MEA make-up increases proportionally with CO_2 composition and gas flow rate, owing to the assumption that MEA make-up is a function of the captured CO_2; data sourced from Chapel et al.[82]

Effect of Capture Rate. Figure 5 shows the marginal cost of capture as a function of the capture rate for a flow rate of 500 kg·s^{-1}. The peaks in the marginal cost curves represent the addition of equipment items; negative peaks correspond with either a minimum in capture cost or equipment items being enumerated and resized in such a fashion that results in a lower capture cost.

A comparison of Figures 3 and 5 indicates that the capture cost is significantly more sensitive to feed gas flow rate (specifically when flow is <20 kg·s^{-1}) compared to CO_2 capture rate. The capture cost curves and corresponding marginal cost curves in Figure 5 remain relatively constant between 70 and 90%, indicating that capture costs are primarily dictated by the gas flow rate (*i.e.*, constant 500 kg·s^{-1} gas flow, resulting in similar capture costs).

For most of the equipment items in Figure 6A, the capture rate has a negligible impact on the capital cost, with the
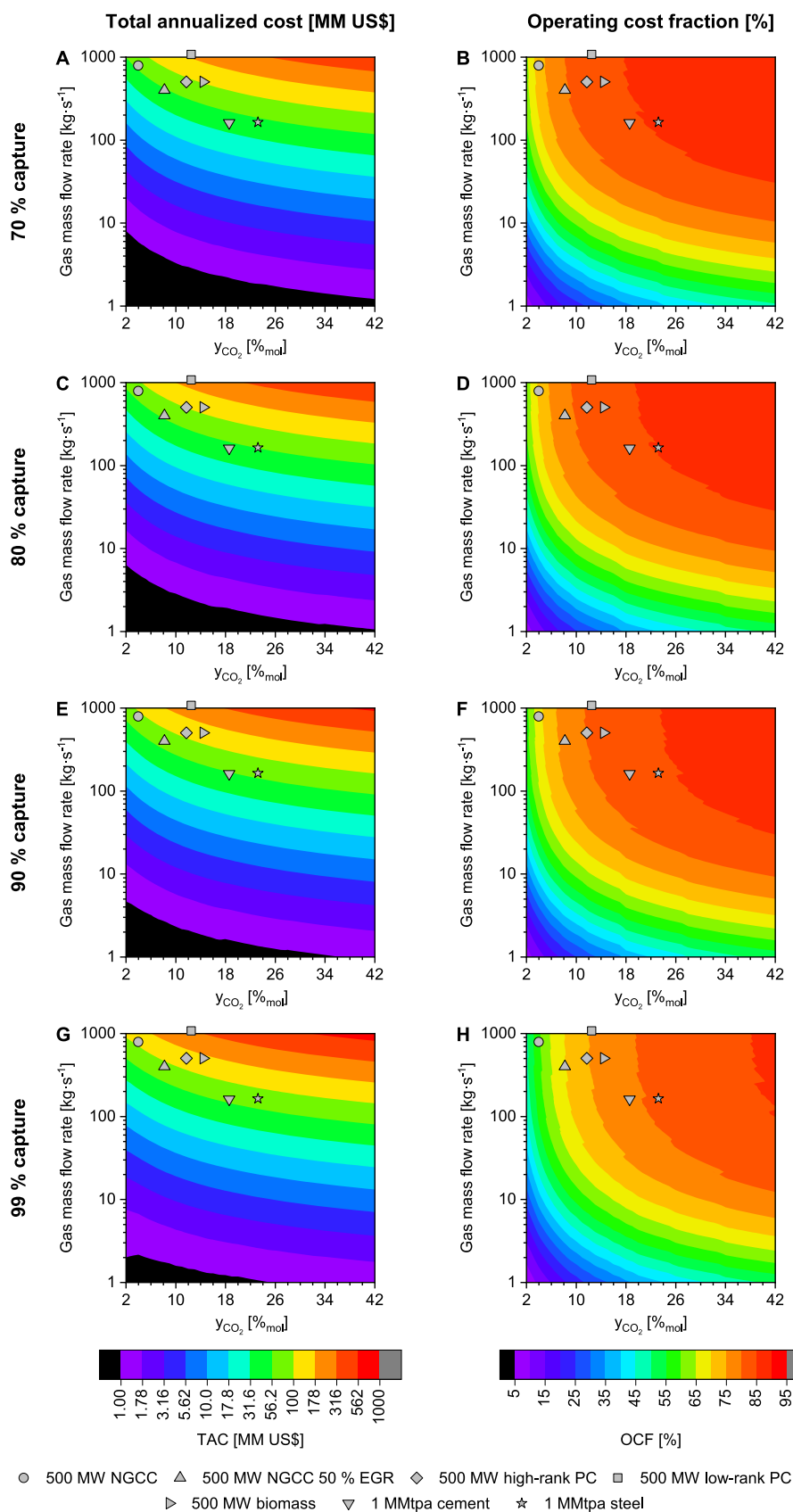
Figure 7. TAC (A,C,E,G) and the OCF (B,D,F,H) as a function of gas CO_2 composition and flow rate for selected capture rates. The colormap for the TAC plots (left column) uses a logarithmic scale.

exception being the absorber which increases in cost between 90 and 99% capture rate. The outlet gas CO_2 concentration has an

exponential effect on the absorber column sizing. At the higher capture rates, there is a reduction in concentration driving force,
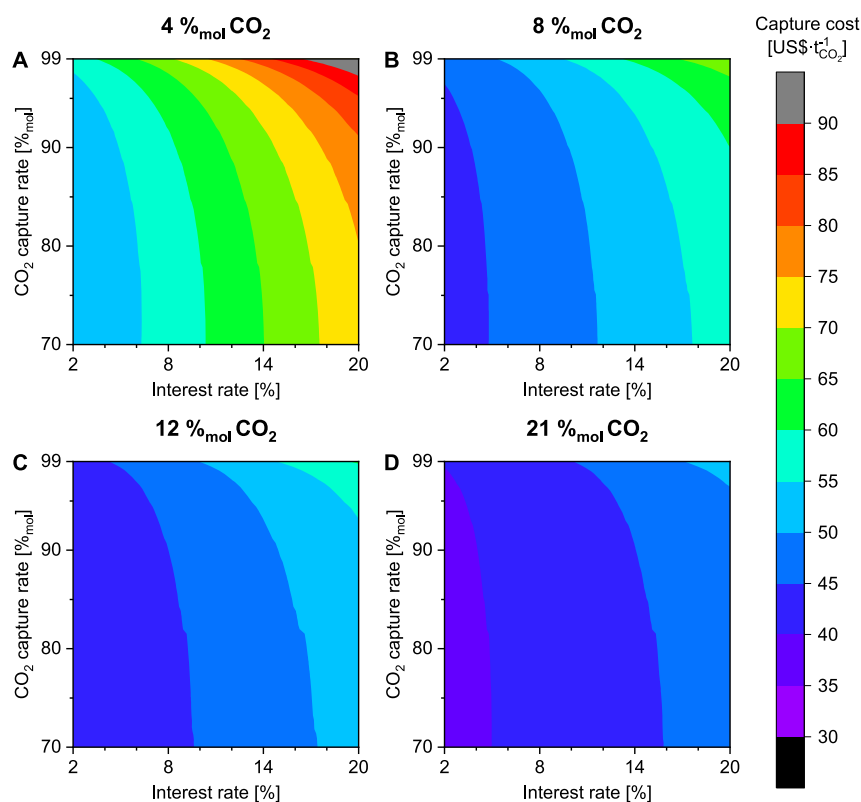
Figure 8. Capture cost as a function of the interest rate (*i.e.*, cost of capital) and capture rate for selected gas CO_2 compositions, at a gas flow rate of 500 kg·s^{-1}.

Table 4. Summary of Capture Costs (US$·$t_{CO_2}^{-1}$) at 2% (Minimum) and 20% (Maximum) Interest Rates, IR, Calculated for Different Capture Rates and a Flue Gas Flow Rate of 500 kg·s^{-1}

y_{CO_2}	IR	capture rate [%]				
[%$_{mol}$]	[%]	70	80	90	95	99
4	2	50.5	50.6	51.5	53.0	57.5
	20	73.6	75.0	79.0	84.4	99.3
8	2	43.3	43.4	43.8	44.6	46.6
	20	57.1	57.9	59.9	62.5	69.7
12	2	41.0	41.1	41.5	42.0	43.3
	20	51.8	52.3	53.9	55.8	60.6
21	2	38.9	39.0	39.2	39.4	40.2
	20	47.2	47.5	48.4	49.4	52.2

Table 5. Summary of Updates Applied to the Published Model

parameter	refs
pressure drop	85
VLE	86–89
rich amine physical properties	90
CO_2 liquid-phase diffusivity	91
CO_2 gas-phase diffusivity	92,93
mass transfer coefficients	85,94
enhancement factors	95–99

requiring additional packing height to absorb the CO_2. The additional height increases pressure drop across the absorber, thereby requiring more blower work and increasing blower costs. The change in absorber cost between 70 and 80% capture rate is relatively small.

As previously mentioned, operating costs are a function of the amount of CO_2 captured. Thus, operating cost increases proportionally with the capture rate. As the lean and rich CO_2 loading are fixed for a given flue gas concentration, increasing the capture rate results in increased amine circulation rate, requiring slightly more electricity for the pumps. Although capital and operating costs increase with capture rate, these increases are not substantial. Consequently, the capture cost increase for 99% capture is marginal compared to 90% capture for CO_2 concentrations above 4%$_{mol}$ (Figure 5).

Ratio of Operating-to-Capital Costs. The OCF, given in eq 5, represents the operating cost as a percentage of the TAC.

$$OCF = \frac{operating\,cost}{TAC} \tag{5}$$

where the OCF provides insights into the project economics, identifying areas of potential cost saving. In cases where operating cost dominates, the total project cost may be many multiples of the upfront capital cost.

Figure 7 shows how the TAC and OCF vary as a function of gas flow and CO_2 composition at different capture rates. For a given capture rate and gas flow rate, the TAC increases with feed gas CO_2 concentration (*i.e.*, horizontal cross-section on a TAC plot in Figure 7), which is attributable to the increase in capital and operating costs (Figure 4). The corresponding OCF also increases with gas CO_2 concentration, indicating that operating costs increase at a greater rate than the TAC. For a given gas CO_2 composition, the TAC and OCF increase with greater feed gas flow rates. Figure 7B,D,F,H shows a general decrease in OCF with the capture rate.

For the reference applications considered (symbols plotted on Figure 7), the OCF is at least 50% in all cases, with most cases being around 65% and some as high as 85%. With the majority of the cases having high OCF above 65%, any cost reduction benefit will mainly come from operating costs. As steam is the predominant operating cost, measures to reduce steam consumption would be beneficial. This is reflected in the advanced solvent capture cost data presented in the Supporting Information (Section S2.6), as discussed earlier. The equivalent TAC and OCF data for the advanced solvent case is shown in Figure S9. There is a general reduction in both; however, OCF remains above 50% for the reference cases.

Cost reduction may also be achieved with waste heat recovery or heat integration to reduce steam requirements. However, it should not be assumed that waste heat or excess steam will be available at all process facilities. In a case where OCF is 70%, the reduction in capture cost offered by a 25% decrease in operating cost is equivalent to the reduction provided by a 58% decrease in capital cost. On the other hand, when the OCF is 40%, the same 25% decrease in operating costs is equivalent to a 17% reduction in capital cost. Thus, efforts toward process intensification would be better directed toward the latter cases.

Effect of Cost of Capital. To demonstrate the effect of cost of capital, capture cost was evaluated for a range of interest rates between 2 and 20%, illustrated in Figure 8 and Table 4. This was evaluated as a function of the capture rate at different feed CO_2 compositions and a gas flow rate of 500 kg·s^{-1}. The value of 2% interest rate represents a government-funded project, whereas 20% is representative of the expected return received by a private company if instead they invested the capital cost of the project.[83,84]

The 4%$_{mol}$ CO_2 case shows the greatest variation in the capture cost, with diminishing variability as gas concentration increases. As shown by eqs 1−3, the interest rate has the greatest impact on capture cost in scenarios that are capital cost-intensive, that is, those with low OCF, such as the scenarios with low feed gas CO_2 composition and/or low flow rate in Figure 7. For NGCC power plant applications, sourcing suitable project financing may have an appreciable impact on project cost. However, variations of two to three percentage points in interest rate have little impact on the capture cost for industrial applications with higher flue gas CO_2 concentration, that is, scenarios with high OCF.

We have evaluated the capture cost for MEA-based post-combustion CO_2 absorption for a range of flue gas conditions between capture rates of 70 and 99%. We have shown that post-combustion amine absorption has equivalent viability in both industrial and power applications, and the cost of 99% CO_2 capture is not significantly higher than 90%. Although the capture costs increase with a greater capture rate and lower gas CO_2 composition, the additional few percent of CO_2 that can be captured by increasing capture rate from 90 to 95%, for essentially no additional cost, will be significant in terms of climate change mitigation efforts.

APPENDIX A

Modifications were made to the published model[69] in order to widen the range of applicability. The updated aspects are tabulated in Table 5 and are described in further detail in Supporting Information (Section S5).

ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c07261.

> Capture cost surrogate model (XLSX)
>
> Capture cost surrogate model (ZIP)
>
> Additional results, surrogate model goodness-of-fit, and description of updates to process the model (PDF)

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Notes

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