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Prospects for Bioenergy with Carbon Capture & Storage (BECCS) in the United States Pulp and Paper Industry

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Abstract

The pulp and paper industry utilizes more biomass for stationary heat and power than any other industry in the United States. In total, pulp and paper mills in the US emit ~150 million metric tons of CO_2 each year, of which 77% is biogenic. Thus, the pulp and paper industry has significant potential to indirectly remove atmospheric CO_2 through bioenergy with CO_2 capture and storage (BECCS). In addition, avenues for CO_2 utilization exist in pulp and paper processing. Here, we analyze the technical and economic potential of integrating carbon capture, utilization, and sequestration (CCUS) technologies at pulp and paper mills in the US through top-down, industry-wide screening and bottom-up, chemical process modeling techniques. We estimate costs of capturing and transporting CO_2 from pulp and paper mills using post-combustion amine chemisorption in the year 2026 with application of the existing federal tax credit for carbon capture and sequestration (Section 45Q). Costs are highly dependent on scenario-specific details, such as waste heat or power generation at the mill, idling or stranded assets, and proximity to suitable geologic storage opportunities. Some CCS implementation scenarios produce significant economic returns for pulp and paper mills, indicating a near-term opportunity to accelerate CCS in the US. Finally, we qualitatively assess alternative techniques for CO_2 capture through process innovation, and opportunities for CO_2 utilization at pulp and paper mills.

Broader Context

Technologies that remove atmospheric CO_2 must be developed and deployed rapidly if we are to avoid the worst effects of climate change. Engineered CO_2 removal technologies, including bioenergy with CO_2 capture and storage (BECCS), require significant advancements to reduce costs and increase scale without environmental or social harm. Currently, there are no operating BECCS plants in the US that utilize biomass combustion for electricity or heat. The lack of BECCS deployment in the US is in large part due to the high cost of electricity from biopower plants, compared to other sources of renewable electricity, such as wind and solar. In addition, a historical lack of policy incentives to capture CO_2 have stymied progress in BECCS deployment. However, the recently revised federal Section 45Q tax credit provides an economic incentive for power generators and industrial manufacturers in the US to capture their CO_2 emissions for permanent storage or utilization. The pulp and paper industry stands to benefit from the 45Q tax credit and is a suitable candidate to lead the deployment of BECCS in the US due to its extensive biomass utilization, existing infrastructure, economies of scale, domain knowledge, and variety of options for CO_2 capture, storage, and/or utilization. This paper identifies multiple CCUS processes that could be profitable at existing mills in the US.

Introduction

To meet the stringent climate targets set forth by the Intergovernmental Panel on Climate Change's (IPCC) Fifth Assessment Report, the United States (US) must deploy bioenergy with CO_2 capture and sequestration (BECCS) to a scale of 1 Gt-CO_2 per year by 2050.[1] Commercial CO_2 capture, utilization, and storage (CCUS) operations must rapidly accelerate to meet such an ambitious target.[2] Near-term opportunities to develop, demonstrate, and deploy CCUS technologies can reduce costs, improve performance, and clarify their sustainable scale. Previous work has highlighted the opportunities to deploy CCUS on existing biogenic CO_2 emissions as a first market for engineered carbon removal. For instance, sequestration of biogenic CO_2 emissions from bioethanol refineries is considered to be a viable opportunity due to its technical and commercial maturity (current emissions are 45 Mt-CO_2 per year) and low-cost of capture, transport, and sequestration.[3] Analyses that identify opportunities to leverage existing infrastructure, technologies, and policies can enhance both near-term and long-term mitigation efforts by deploying existing technologies and developing experience in CCS.[4]

As the largest industrial source of existing biogenic CO_2 emissions in the US, the pulp and paper industry represents a viable opportunity for utilizing and/or sequestering biogenic CO_2 emissions. Many pulp and paper mills involve multiple energy-intensive operations that emit CO_2-containing waste streams, as shown in Figure 1.[5] Some streams are of relatively high purity CO_2 and therefore may be ideal candidates for CCUS. Recent technological innovations in CO_2 capture offer opportunities to cost-effectively integrate CO_2 capture at existing pulp and paper mills.[6–15] However, the complexity associated with the heterogeneity of operations among mills, as well as the sheer quantity of mills, create a challenge when assessing such an opportunity.

The federal government of the US has developed several policies to accelerate innovation and deployment of CCUS. Most notably, Congress has recently revised the Section 45Q tax credit for utilizing or sequestering point-source CO_2.[16] This tax credit will change with time, increasing from \$19 to \$35 per t-CO_2 for utilization and \$31 - \$50 per t-CO_2 for sequestration in geological formations for years 2019 to 2026, after which point the value will remain constant (indexed to inflation). The 45Q tax credit does not distinguish between biogenic and non-biogenic sources of CO_2. Further, the US Department of Energy Loan Guarantee Program can be combined with 45Q federal tax credits to help enable large-scale projects involving advanced technologies. The Department of Energy has \$8.5 billion in unutilized loan guarantee authority for innovative fossil energy technology, including carbon capture and low-carbon power systems.[17] Existing pulp and paper mills that directly utilize CO_2 may be immediately eligible for the 45Q utilization tax credit with minimal upfront costs. In particular, pulp and paper products that contain calcium carbonate fillers and coatings, including multiple grades of paperboard, may be immediately eligible for the tax credit. Additionally, some Kraft pulping mills are currently implementing technologies that utilize CO_2 to precipitate lignin from black liquor, thereby potentially enabling downstream products to be eligible for the 45Q utilization tax credit.[18] Detailed lifecycle assessments (LCAs) are necessary to ensure these products meet the federally-established requirements.

Previous studies have investigated the economic feasibility of capturing CO_2 from pulp and paper mills using traditional amine chemisorbents[9–11], but none have involved an industry-wide, geospatial assessment that quantifies the costs of capturing and transporting biogenic and fossil-derived CO_2 emissions from mills located in the US, with high granularity regarding fuel type and unit operation used for combustion, CO_2 concentrations, and economic impact of the 45Q tax credit. Onarheim et al. conducted a thorough investigation into the techno-economic feasibility of capturing CO_2 emitted from two pulp and paper mills: 1) a generic Kraft market pulp mill and 2) a generic integrated Kraft pulp and board mill.[9,10] Their analysis involved highly detailed chemical process simulations for multiple scenarios, but did not involve an industry-wide analysis. Nabinger et al. quantified the greenhouse gas emissions from a select number of pulp and paper mills in the US, but did not involve a techno-economic analysis for capturing the emissions.[19] Psarras et al. estimated the costs of capturing CO_2 via amine chemisorbents for 21 sectors in the US industrial economy, including the pulp and paper industry.[11] However, in their assessment, emissions are quantified using a dataset from the EPA that does not consistently include biogenic emissions, emissions are not quantified for mill-specific unit operations, emissions are not categorized by fuel type, emissions from different product grades are not assessed, idling assets are not assessed, detailed process simulations are not conducted, and potential revenues from the 45Q tax credit are not taken into account. Regarding CO_2 utilization, to the best of our knowledge, there have been no studies published on integrating CO_2 utilization in the pulp and paper industry though process intensification and innovation for the purpose of removing atmospheric CO_2 and improving process economics through the 45Q tax credit. Thus, there has yet to be an extensive investigation into the economic feasibility of incorporating CCUS technologies into US pulp and paper mills.

Here, we assess the technical and economic feasibility of indirectly removing atmospheric CO_2 through the capture, compression, transportation, and sequestration of CO_2 emissions from the pulp and paper industry in the continental US. This is accomplished by first assessing the entire industry using a top-down approach to identify favorable mills, followed by an assessment of a select number of favorable mills using a bottom-up approach with process engineering methods. All techno-economic analyses use post-combustion amine chemisorption technologies for CO_2 capture. Additionally, alternative techniques for CO_2 capture through process innovation are assessed qualitatively. Finally, pathways for CO_2 utilization at existing pulp and paper mills are assessed qualitatively.

Methodology

Top-Down Analysis, Part I: Industry-Wide Screening

Operable pulp and paper mills in the continental US are assessed for their potential for CO_2 capture and sequestration. All 205 mills included in this analysis meet the eligibility requirements for the Section 45Q utilization tax credit, which places a lower limit on captured mill emissions to 25,000 metric tons per year; 135 mills (66% of total) meet the eligibility requirements for the sequestration tax credit, which places a lower limit of 100,000 metric tons per year.[16] A high level of granularity is taken to assess the CO_2 emissions for each mill, involving the quantification of CO_2 emitted from 1) the entire mill, 2) each fuel consumed, 3) each major operation on-site, and 4) the production of each major product grade, as shown in Figure 2.

Emission stream compositions are quantified using fuel composition and combustion properties typical for each of the major operations.[20] A significant portion of the basis process data used in calculations is derived from the Fisher*Solve* Platform for the year 2018.[21] Visual Basic is used to program an automated model with data inputs from multiple sources and outputs that are presented herein. The major operations include lime kilns, multi-fuel boilers, and recovery boilers; multi-fuel boilers vary considerably in their configuration. As shown in Figure 2, six primary fuels are used at pulp and paper mills to drive the three major operations: black liquor, wastewood, natural gas, coal, petroleum coke, and fuel oil; other minor fuels include diesel, methanol, and tires, to name a few. Emissions are also quantified with respect to eight major product grades: containerboard, market pulp, printing and writing, cartonboard, tissue and towel, specialties, packaging paper, and newsprint.

Abiding by the laws of thermodynamics, the minimum energy required for separation of CO_2 from each emission stream at each mill is calculated using Equation 1.[22] The incoming and outgoing concentrations of CO_2 and the percent capture rate are the most influential variables with respect to system energy demand for CO_2 capture (Equations 1 – 5). The outgoing concentration of CO_2 is taken to be 97 mol% and the capture rate is taken to be 90% for all cases. The 2nd law efficiency for separation of CO_2 is estimated for each stream at each mill using Equations 2 - 4. The actual energy required for separation of CO_2 is estimated using Equation 5.

$$Min.\ energy\ for\ CO_2\ separation = w_{min} = RT\begin{bmatrix} \left(n_{r,CO_2}\ln\left(X_{r,CO_2}\right) + n_r\ln\left(X_r\right)\right) \\ + \left(n_{p,CO_2}\ln\left(X_{p,CO_2}\right) + n_p\ln\left(X_p\right)\right) \\ - \left(n_{i,CO_2}\ln\left(X_{i,CO_2}\right) + n_i\ln\left(X_i\right)\right) \end{bmatrix} (1)$$

$n_{x,CO_2} = moles\ of\ CO_2\ in\ stream\ x$

$n_x = moles\ of\ nonCO_2\ component\ in\ stream\ x$

$X_{x,CO_2} = mole\ fraction\ of\ CO_2\ in\ stream\ x$

$X_x = mole\ fraction\ of\ nonCO_2\ components\ in\ stream\ x$

$r = residual\ stream$

$p = product\ stream$

$i = inlet\ stream$

$$2nd\ law\ efficiency = \eta = 1 \times 10^{-5} + 0.0014\alpha + 0.0087\ (2)$$

$$\alpha = \frac{capture\ rate}{\log C_f}\ (3)$$

$$C_f = \frac{[CO_2]_{out}}{[CO_2]_{in}}\ (4)$$

$$Actual\ energy\ for\ CO_2\ separation = \frac{w_{min}}{\eta}\ (5)$$

For each mill, existing capital assets are assessed because of their potential to reduce costs. Finally, a geospatial analysis is conducted to determine which mills are co-located with geology suitable for long-term CO_2 sequestration, following methods developed by Sanchez et al.[23]

Bottom-Up Analysis: Case Studies

Chemical process models are developed and assessed using AspenTech process simulation software and process data obtained from the literature to provide engineering metrics necessary for estimating levelized capital and operating expenses for capturing (including compression) CO_2 at four select mills[24,25]. The top-down industry-wide screening is used to select the four mills for detailed analysis, with motivating information provided in Table 1. For Mill 4, we assume on-site waste heat is of sufficient quality to be used in the CO_2 capture system. For each of the four select mills, costs of CO_2 capture ($/t$CO_2$) are estimated for four baseline scenarios, resulting in a total of sixteen baseline cost estimates; see Figure 3 for the logic flow of analysis. All amine chemisorbent systems modeled use biomass fuel for heat and power to ensure a large net removal of CO_2. As shown in Figure 3A, "Steam" scenarios incorporate a biomass boiler subsystem without a turbogenerator, wherein 100% of the steam demand is met from the boiler and 100% of the electrical power is purchased from the grid. "Steam and Power" scenarios incorporate a biomass boiler subsystem with a turbogenerator wherein 100% of the steam and power demands are met by the boiler and turbogenerator. Biomass is the only fuel source used by the boiler subsystem in this analysis. The major unit operations of the biomass boiler subsystem include live-bottom grated fuel bin, dryer, combustor, particle cyclone, electrostatic precipitator, boiler, and turbogenerator (dependent on the scenario).[26] The major unit operations of the amine chemisorption subsystem include pumps for amine circulation, an absorber column with packed bedding, a stripper column with partial reflux, heat exchangers to preheat the stripper column inlet and cool various operations, a water knockout unit to remove water from the CO_2 stream via direct contact condensation, and compressors to pressurize the pure CO_2 stream.[24,27]

A summary of pertinent baseline engineering metrics are presented in Table 2; unless noted, the metrics are consistent across all baseline scenarios. The engineering metrics are determined through a combination of process simulation and literature review.[12,14,27–32] To take reboiler duty variation into account, we conduct a sensitivity analysis across multiple duty values; reboiler duties are defined and used as independent variables in the process models. In addition, we conduct a sensitivity analysis of amine solvent loss rate. The process models developed for each scenario do not undergo intensive engineering optimization and there is some uncertainty; however, the most influential parameters are accounted for, assumptions are conservative, and analyses are conducted consistently to allow for direct comparison between scenarios.

A summary of pertinent baseline economic metrics are presented in Table 3; unless noted, the metrics are consistent across all scenarios. The project year is taken to be 2026, when the 45Q

sequestration tax credit plateaus at \$50 per tonne CO_2. A 2% rate of inflation is used to adjust costs for the year 2026. Capital costs for the biomass boiler subsystem are determined through cost scaling techniques using reference costs from a detailed techno-economic analysis published by the US National Renewable Energy Laboratory.[26] Capital costs for the amine chemisorption subsystems are determined through cost scaling techniques using reference costs from the Aspen Process Economic Analyzer.[24] The major capital costs for the biomass boiler subsystem are broadly broken down into preprocessing, boiler, turbogenerator, gas cleaning, and other accessories. The capital costs for the amine chemisorption subsystem are broadly broken down into compressors, pumps, heat exchangers, absorber column, stripper column, and water knockout. Operating costs are broadly broken down into biomass feedstock, MEA material, cooling water, electricity (for Steam & Power scenarios), other operating and maintenance, and fixed costs (including salaries).[24,25,27,32,33] A value is placed on excess steam in the Steam & Power scenarios with the assumption that such steam will be used on-site for pulp and paper processing. Itemized capital and operating costs for each baseline scenario are provided in the Supplementary Information. To ensure consistency when calculating levelized costs of CO_2 capture, we follow the method of economic analysis used by Keith et al. for CO_2 removal systems.[14] Keith et al.'s methodology for economic analysis, which is adequate for early-stage cost estimations, requires five primary inputs: 1) itemized capital costs, 2) operating costs, 3) plant utilization, 4) capital intensity, and 5) a capital recovery factor (CRF). Levelized costs of CO_2 capture are calculated for each scenario using Equations 6 – 14. A CRF of 8% is used for all scenarios investigated because it corresponds to a 20 year payback period and 5% return on equity. We assume the funding of CO_2 capture systems at existing pulp and paper mills will be through private equity with a guaranteed return of 5%. Therefore, the levelized costs include a 5% return on equity over 20 years. Sensitivity analyses are conducted to account for variation in the cost of biomass, cost of electricity, and return on equity. In addition, scenarios are modified to understand the potential for reducing costs by utilization of existing boilers and turbogenerators on-site.

$$Levelized\ Cost\ of\ CO_2\ Capture\ \&\ Compression = Levelized\ Capital\ Cost + Net\ Operating\ Cost \quad (6)$$

$$Net\ Operating\ Cost = \frac{Operating\ Costs}{CO_2\ Capacity} \quad (7)$$

$$Levelized\ Capital\ Cost = Capital\ Intensity \times \frac{Capital\ Recovery\ Factor}{Utilization} \quad (8)$$

$$Capital\ Intensity = \frac{Total\ Capital\ Cost}{CO_2\ Capacity} \quad (9)$$

$$Capital\ Recovery\ Factor = \frac{i(i+1)^N}{(1+i)^N - 1} \quad (10)$$

$$Weighted\ Average\ Cost\ of\ Capital = i = (Interest\ on\ Debt\ Capital) \times (\%\ Debt\ Financing) + (Return\ on\ Equity\ Capital) \times (\%\ Equity\ Financing) \quad (11)$$

$$N = Project\ life\ (years) \quad (12)$$

$$Total\ Capital\ Cost = Direct\ Capital + Indirect\ Capital \quad (13)$$

$$Direct\ Capital\ Cost = \sum_i Reference\ Cost_i \times \left(\frac{New\ Capacity_i}{Reference\ Capacity_i}\right)^{Scale\ Factor} \quad (14)$$

Top-Down Analysis, Part II: Industry-Wide Cost Assessment

A nonlinear multivariate regression (Equation 15) is constructed using results from the bottom-up analysis combined with published results of cost estimates for a variety of CO_2 capture systems to ultimately estimate costs of capturing CO_2 at all 205 mills across the US.

$$Levelized\ Cost = A1(FlowRate)^{B1} + A2(CapRate)^{B2} + A3(Conc)^{B3} + A \ldots A5(FlowRate * Conc)^{B5} + A6(CapRate * Conc)^{B6} + A7 \ldots * Conc)^{B7} \quad (15)$$

$$A1 = 1001, B1 = 0.0275, A2 = 281.7, B2 = -0.0511, A3 = -8475, B3 = 5.712, A4 = -22.83, B4 = 0.1539, A5 = 95.15, B5 = 0.7852, A6 = -1379, B6 = 0.0115, A7 = 0.0022, B7 = 0.6771, R^2 = 0.8533$$

The nonlinear regression is computed using the Gekko package in Python[34] and a total of 39 data sets, with each set containing four data points: CO_2 flow rate, CO_2 capture rate, CO_2 concentration, and levelized cost of capturing CO_2. Of the 39 data sets used in the regression, 15 are from the bottom-up analysis conducted herein, and the remaining 24 are derived from work published by Psarras et al.[11] wherein techno-economic studies were conducted on CO_2 capture systems for a variety of emission types. Using two separate data sets provides a wide range of operational parameters (CO_2 flow rate, capture rate, and concentration), thereby generating a robust regression. See the Supplementary Information for a detailed explanation of the multivariate regression. The year 2026 is taken to be the year for cost estimation because the 45Q sequestration tax credit levels off at $50 per t-$CO_2$ at this time; a 2% rate of inflation is assumed between the years of 2016, which is the year for cost estimation by the multivariate regression, and 2026, which is the year for cost estimation in this analysis. We assume biomass intake, fuel intake, production capacity, and CO_2 emissions remain constant at all mills between the years 2018 – 2026. Levelized costs are quantified in three ways to provide stakeholders with an understanding of how CO_2 capture will influence their respective operations: 1) cost per tonne CO_2, 2) cost per tonne product, and 3) percent of manufacturing cost. Finally, transportation costs are quantified for each of the 205 mills using results from the geospatial analysis and methods developed by Sanchez et al.[23]

Results

Top-Down Analysis, Part I: Industry-Wide Screening

Approximately 150 million metric tons of CO_2 are emitted annually from the 205 pulp and paper mills selected for this analysis, of which 77% are biogenically derived (Table 4). Compared to other industrial commodities, the production of pulp and paper products is heavily reliant on biogenic fuels, which positions the industry favorably if future carbon emissions policies provide premium incentives for capturing biogenic CO_2. Details regarding the contributions of various fuels across all mills and respective operations are shown in Figure 4 and Table 5.

The lime kilns and multi-fuel boilers use multiple fuels and therefore the emission CO_2 concentrations vary for particular operations across mills. Emissions from lime kilns are derived from fuel combustion and carbonate calcination, and therefore these emission streams have relatively high concentrations of CO_2 (Table 5)[35]; stoichiometric reactions for fuel combustion: $C_xH_yO_z + O_2 \rightarrow xCO_2 + \frac{y}{2}H_2O$ and calcium carbonate calcination: $CaCO_3 \rightarrow CaO + CO_2$. Overall, the total contribution of CO_2 from lime kilns is relatively small (~9%), whereas the contributions from multifuel boilers and recovery boilers are approximately the same (~43, ~48%). The energy demanded by CO_2 separation is most heavily influenced by the concentration of CO_2, which is evident from the relatively low energy demand for capturing CO_2 from lime kiln emissions. Natural gas has a high H/C ratio relative to solid fuels such as biomass, petroleum coke, and coal, and therefore natural gas generates effluent streams with high concentrations of water and low concentrations of CO_2.[20] Thus, natural gas-derived CO_2 emitted from multi-fuel boilers is generally more energy-intensive to capture than that derived from solid fuels. The production rates and corresponding CO_2 emissions of the eight major product grades are shown in Figure 5. As shown in Figures 5 and 6, the production of containerboard constitutes the largest quantity of CO_2 emissions per year, yet has a relatively low emission intensity due to its high production volume and relatively low-energy processing. Newsprint is also produced using a relatively mild, low-energy process.

Tissue and towel and cartonboard are often produced independent from wood pulping and processing, and therefore the on-site fuel consumption for their production is primarily fossil-derived and considerably lower than that for other products that are made at Kraft pulping mills. Thus, newsprint, tissue and towel, and cartonboard have relatively low emission intensities.

Emissions from the production of all grades are primarily biogenically sourced, aside from cartonboard and tissue and towel, as shown in Figure 5. As shown in the Table 6, a substantial quantity of idling kilns and boilers are potentially available to drive new processes, such as CO_2 capture. The costs of capturing CO_2 could decrease if capital expenditures are reduced through the use of idling or stranded assets. In addition, waste heat, if available in sufficient quantity and quality, could reduce costs of capturing CO_2. A geospatial analysis is conducted to determine which mills are co-located with geology suitable for permanent CO_2 sequestration (Figure 7).[23] Of the 205 mills selected for analysis, 88 (43%) are located on geology suitable for sequestration and therefore do not require long-distance transportation of CO_2. Of the 117 mills that are not located on suitable geology, the average pipeline transportation distance to suitable geology is 134km.

Bottom-Up Analysis: Case Studies

Levelized costs of capturing CO_2 (including compression) from the four select mills are estimated over a range of scenarios, as shown in Figure 3A. The initial top-down, industry-wide analysis indicated the energy demands for capturing CO_2 from lime kiln emissions are the lowest of the three major operations, hence the inclusion of a scenario with only lime kiln emissions in this bottom-up analysis. Table 7 shows pertinent process data and cost estimates for the four baseline scenarios outlined in Figure 3A. Costs are estimated in year 2026 and the 45Q

sequestration tax credit of \$50 per tonne CO_2 is applied to all scenarios. Cost estimates below \$0 indicate the tax credit acts a source of income for the particular scenario. Figure 8 provides a breakdown of the levelized capital and operating costs for each baseline scenario investigated. As can be seen in Figure 8, operating costs contribute significantly more than capital costs to the levelized costs of CO_2 capture. As can be seen in Table 7 & Figure 8, the cost estimates for the Combined emission scenarios are near to \$0, ranging from -\$3 to \$5 per tonne CO_2, whereas the cost estimates for the Lime Kiln emission scenarios vary considerably, ranging from -\$19 to \$23 per tonne CO_2. The levelized costs of capturing CO_2 for the Steam & Power + Combined Emissions scenarios are negative for Mills 1-3, and slightly positive for Mill 4, thereby demonstrating the economic feasibility of these scenarios. The lowest cost of CO_2 among the baseline scenarios is the Mill 4, Steam + Lime Kiln Emissions scenario (-\$19/tCO_2) in which on-site waste heat (217 GJ/h) is sufficient for the entire operation and new biomass boiler steam generation is not necessary. Mill 4 is the only mill of the selected four that has waste heat potentially available to use for CO_2 capture.

We assume the waste heat is of high enough quality to be used as steam (4.5 bar, 150°C) in the amine subsystem reboiler. Interestingly, the costs of capturing CO_2 for the other three Mill 4 scenarios are not as low as that for the Steam + Lime Kiln Emissions scenario. This observation can be explained by the fact that the other three scenarios require capital expenses for a biomass boiler subsystem and operating expenses for biomass feedstock, thereby significantly increasing the levelized cost of CO_2 capture. The availability and quality of heat at Mill 4 must be validated before accepting these cost estimates. Nevertheless, this exercise demonstrates the significant potential for cost reduction through the use of on-site waste heat.

According to results from the bottom-up analysis, cost estimates for capturing CO_2 from lime kiln emissions are generally higher than those from combined emissions, which contradicts the energy demand trend determined in the initial top-down analysis; lime kiln emissions require less energy to process than emissions from other operations due to the high concentrations of CO_2. This contradiction can be explained by the relatively small flow rates of CO_2 from lime kilns and thus the lack of economies of scale. The scales of operation for the Combined Emission scenarios are significantly larger in size than those for the Lime Kiln Emission scenarios, evident by the differences in biomass flow and CO_2 capture rates (Table 7). Therefore, the scaling factor (0.7) in the Combined Emissions scenarios outweighs the cost benefits of higher CO_2 concentrations in the Lime Kiln Emissions scenarios. Notably, the biomass input required by the CO_2 capture system for the Mill 1, Steam & Power + Combined Emissions scenario is 3939 tonnes biomass per day, which is 93% of the biomass input to pulp and paper operations at Mill 1. Overall, the Combined emissions scenarios demand significant quantities of biomass, thereby warranting investigation into local availability of biomass resources to determine feasibility[37]; a sensitivity analysis varying biomass costs is conducted to help understand this feasibility.

Mill 4 demands a relatively small quantity of biomass for the Combined Emissions scenarios (831 – 1859 t-biomass/day), because of the on-site waste heat utilization which allows for less biomass consumption. The most effective way to reduce biomass demand is to reduce the reboiler duty and/or utilize existing waste heat. A sensitivity analysis is conducted by varying reboiler duty to help understand how different amine solvents might affect costs. The high costs associated with the Lime Kiln Emissions scenarios are largely due to the small scale of operation. Small biomass boiler systems of capacities in the range of 200 – 300 tbiomass/day have high capital intensities ($/t$CO_2$) relative to large systems due to economies of scale.[38] The Steam & Power scenarios that use biomass to entirely meet steam and power demands have lower levelized costs of CO_2 capture than the Steam scenarios that purchase power. This observation is interesting because scenarios that purchase power do not require large capital expenditures for turbogenerators, and the quantities of biomass required for operation are considerably less than for scenarios that generate steam and power. A sensitivity analysis varying electricity costs is conducted to better understand this observation.

Sensitivity Analyses:

Sensitivity analyses are conducted to understand the effects of important variables on levelized costs of CO_2 capture (Figures 9 - 15). Specifically, reboiler duty, biomass cost, electricity cost, rate of MEA loss, and return on equity are varied. Costs due to transportation are negligible for all four mills since they sit atop suitable geology for long-term sequestration, however, we conduct a sensitivity analysis by varying transportation distances to understand the effects transportation would have if these sites were not located on suitable geology.

Regarding reboiler duty, Figure 9 shows the inflection point at which levelized costs transition from negative to positive is ~3.0 GJ/tCO_2 for the Steam + Combined Emissions and ~3.5 GJ/tCO_2 for the Steam & Power + Combined Emissions scenarios. All mills except Mill 4 have positive costs for the Lime Kiln Emissions scenarios, due to reasons explained in the previous section. Notably, the cost associated with Mill 3, Steam + Lime Kiln Emissions and a reboiler duty of 1.5 GJ/tCO_2 is approximately $0 per tonne CO_2, or break even; Mill 3's lime kiln emissions are of a relatively high concentration of CO_2, hence the lower costs. Scenarios that generate steam only, namely Steam + Combined Emissions and Steam + Lime Kiln Emissions, are more sensitive to reboiler duty values, evident by the larger variation in levelized costs of CO_2 capture when compared to scenarios that generate steam and power. Commercial MEA solvents used at concentrations of 30 wt% in a manner similar to that used in this analysis typically have reboiler duties in the range of 3.0 – 5.0 GJ/tCO_2, and advanced amine solvents currently under development have shown the potential for duties less than 2.0 GJ/tCO_2.[2,12] Therefore, the low reboiler duties analyzed in this sensitivity analysis are proxies for advanced amine solvents.

Regarding MEA loss, Figure 10 shows the inflection point at which levelized costs transition from negative to positive is ~1.7 kg/tCO_2 for the Steam + Combined Emissions and ~2.6 kg/tCO_2 for the Steam & Power + Combined Emissions scenarios. All mills except for Mill 4 have positive costs for the Lime Kiln Emissions scenarios, due to reasons explained in the previous section. Costs associated with Mill 4, Steam + Lime Kiln Emissions are highly sensitive to MEA rate loss, with a minimum cost of -\$26/tCO_2; the contribution of MEA cost to overall operating cost is significant for this particular scenario since no biomass is consumed. Notably, the cost associated with Mill 3, Steam & Power + Lime Kiln Emissions and a MEA loss rate of 1.1 kg/tCO_2 is approximately \$0/tCO_2, or break even; Mill 3's lime kiln emissions are of a relatively high concentration of CO_2, hence the lower costs. Relative to Steam only scenarios, the Steam & Power scenarios are overall more sensitive to MEA loss rates because of the increased biomass flow rate rates and thus larger quantities of CO_2 to process.

Regarding biomass cost, Figure 11 shows the inflection point at which levelized costs transition from negative to positive is ~\$50/tbiomass for the Steam + Combined Emissions and

~\$60/tbiomass for the Steam & Power Combined Emissions scenarios. Costs associated with Mill 3, Steam & Power + Lime Kiln Emissions are negative with a biomass cost of \$20/tbiomass, but all other costs for Mill 3, Lime Kiln Emissions are positive. The costs associated with Mill 4, Steam + Lime Kiln Emissions do not change with biomass cost because this particular scenario relies entirely on waste heat and does not require biomass energy for operation. Relative to Steam only scenarios, the Steam & Power scenarios are more sensitive to biomass cost because of the increased biomass flow rates. The use of natural gas-fueled boiler and turbogenerator systems would likely reduce costs of CO_2 capture due to the very low cost of natural gas energy in the US, relative to biomass energy. However, the net removal of CO_2 from the atmosphere would be reduced if natural gas was used in place of biomass fuel.

Regarding electricity cost, Figure 12 shows the inflection point at which levelized costs transition from negative to positive is ~\$0.045/kWh for the Steam + Combined Emissions scenarios. The costs for all Steam & Power scenarios do not change from baseline with variation in electricity price since all power necessary for operation is derived from biomass. Costs associated with Mill 4, Steam + Lime Kiln Emissions are highly sensitive to electricity cost, with a minimum cost of -\$27/tCO_2; the contribution of electricity cost to overall operating cost is significant for this particular scenario since no biomass is consumed.

Regarding return on equity, Figure 13 shows the inflection point at which levelized costs transition from negative to positive is ~2.0% for Steam + Combined Emissions and ~5.0% for Steam & Power + Combined Emissions. All mills except for Mill 4 have positive costs for the Lime Kiln Emissions scenarios, due to reasons explained in the previous section. Relative to Steam only scenarios, the Steam & Power scenarios are more sensitive to return on equity due to the larger capital expenditure required. For a return on equity of 10%, which is common for industrial investments, all mills except Mill 4 have positive costs of CO_2 capture. Therefore, near-term investments into CO_2 capture at pulp and paper mills in the US will likely not be driven solely by economics, but rather a combination of economics and environmental stewardship.

Regarding transportation distance, Figure 14 shows the effects of increasing transportation distance on levelized costs of capturing and transporting CO_2. The four mills selected for the bottom-up analysis are co-located with suitable geology and thus do not require transportation of CO_2 for long-term sequestration, however, understanding how transportation could affect cost is important. The average transportation distance among the 117 mills that do not sit atop suitable geology for long-term sequestration is ~130km, and thus 130km is selected as the median point in the sensitivity analysis. As can be seen in Figure 14, the lime kiln scenarios are affected by transportation distance more than the combined scenarios, which is due to economies of scale; constructing pipeline to transport CO_2 is highly expensive for relatively small CO_2 flow rates. There are only two scenarios with negative costs for transporting CO_2 a distance of 130km (the average distance for mills not co-located with suitable geology), namely Mill 1, Steam & Power + Combined Emissions and Mill 4, Steam + Lime Kiln Emissions. Thus, the majority of scenarios are not economical for the average transportation distance of 130km, and stakeholders should prioritize mills that are co-located with suitable geology.

Figure 15 shows the levelized costs of CO_2 capture for best and worst case scenarios in which the highest and lowest performing values from the sensitivity analyses are applied, respectively. These scenarios are not highly likely, but they show the range of potential costs. The best case scenario might be plausible with an optimized system using advanced amine solvents located in an area with an abundance of low cost biomass or curtailed solar/wind energy. The worst case scenario shows costs typically considered very high for post-combustion CO_2 capture, and would likely not be economical even with stringent carbon emission policies.

Figure 16 shows the levelized costs for scenarios in which idling boilers and turbines are brought online to provide the heat and power for CO_2 capture. To be conservative, 10% of the installed capital cost for a new boiler + turbogenerator system is assumed necessary to bring an idling boiler + turbogenerator system online. The reductions in costs compared to the baseline scenarios are significant. The largest reductions in costs are experienced by the Steam & Power Emissions scenarios, due primarily to the avoidance of steep capital costs for boilers and turbogenerators.

Top-Down Analysis, Part II: Industry-Wide Cost Assessment

Results from the bottom-up analysis are used to construct a multivariate regression that is applied in a top-down fashion to estimate levelized costs of capturing CO_2 for all 205 mills in the year 2026, the results of which are shown in Table 8. To account for the 45Q tax credit, a \$50 per tonne CO_2 reduction in cost is incorporated in each cost estimate shown. Costs range from -\$0.6 to \$12.7 per tonne CO_2, with recovery boiler- and lime kiln-derived CO_2 being the lowest cost options of the three main operations. Given that levelized costs are inversely proportional to CO_2 concentration and flowrate, the aforementioned trend in costs can largely be explained by the following: CO_2 concentrations of lime kiln emissions are significantly high, CO_2 concentrations and flow rates of recovery boiler emissions are moderately high, and CO_2

concentrations of multifuel boilers are moderately low, corroborated by Table 5. Compared with costs estimated in the bottom-up analysis, the top-down analysis provides similar cost estimates for capturing combined emissions, however, cost estimates for lime kiln emissions are relatively low. Thus, the regression is better suited for large flow rates of CO_2 wherein economies of scale are more readily achieved, including flue streams from multi-fuel boilers, recovery boilers, and combined emissions. If existing capital assets, such as boilers and turbines, can be utilized, the small scales of operation for lime kiln scenarios become more cost competitive since reduced capital costs make economies of scale less important. Overall, economic results from the top-down approach demonstrate the robustness of the regression and its ability to accurately estimate costs of capturing CO_2 emissions from a multitude of fuel types and large-scale industrial operations.

The costs of capturing CO_2 from combined emissions are analyzed on the basis of product mass (\$/tproduct) and CO_2 mass (\$/tCO_2), as shown in Figure 17A. Costs on the basis of product mass trend differently than those on the basis of CO_2 mass, which is due to the variation in emission intensities. For example, the cost of capturing CO_2 emitted from the production of containerboard is relatively moderate on the basis of CO_2 mass, but relatively low on the basis of product mass, which is because costs on the basis of product mass are proportional to emission intensities. Incremental costs (Figure 17B) are also proportional to emission intensities, but in addition, they are inversely proportional to manufacturing costs, and thus the dynamics are more complex. For example, tissue and towel products have relatively low emission intensities and high manufacturing costs, and thus incremental costs of capturing CO_2 are relatively low. Incremental cost data shown in Figure 17B provide stakeholders with an understanding of how the capture of combined CO_2 emissions would affect their margins. Overall, costs on the basis of product mass are relatively small and therefore incorporating the capture of combined CO_2 emissions would not add significant cost to pulping operations. Using results from the geospatial analysis (Figure 7), costs are estimated for transporting CO_2 to suitable geology for long-term sequestration. The average cost of transportation among mills that do not sit atop suitable geology is \$24.8 per tonne CO_2 in the year 2026 (assuming 2% rate of inflation and not accounting for the 45Q tax credit), and thus mills co-located with suitable geology should be prioritized for initial deployment of CO_2 capture and sequestration in the US pulp and paper industry.

Process Innovation

Two opportunities to reduce costs of CO_2 capture exist through process innovation at existing Kraft pulping mills: 1) partial oxy-fuel combustion of lime kilns with post-combustion CO_2 capture and 2) integrated alkaline solvent CO_2 capture.

Partial Oxy-Fuel Combustion of Lime Kiln

A substantial number of mills that utilize oxygen for delignification purchase oxygen from external suppliers because the quantities of oxygen demanded are not large enough to justify the acquisition and operation of large air separation units. A possible justification for the use of air separation units could be the reduction of CO_2 capture costs through partial oxy-fuel combustion in lime kilns. Therefore, Kraft pulping mills that utilize large quantities of pure oxygen for delignification might be suitable for CO_2 capture via partial oxy-fuel combustion in the lime kiln with post-combustion CO_2 capture. Partially substituting air intake with pure oxygen generates emissions with relatively high concentrations of CO_2 and thereby reduces the energy required for CO_2 capture, as shown in Figure 18; the increase in CO_2 concentration is due to the reduction in nitrogen introduced to the system via air. In addition, preliminary studies have demonstrated an increase in lime kiln capacity with oxy-fuel substitution.[8]

Integrated Calcium Looping CO_2 Capture

Calcium looping CO_2 capture is an established method of separating CO_2 from a gaseous stream through four chemical reactions, as shown in Figure 19B.[2,39] The company Carbon Engineering is commercializing a technology to remove CO_2 from the air using chemistry similar to that shown in Figure 19B, with the main difference being the use of potassium hydroxide in place of sodium hydroxide.[14] An advantage to using alkaline chemistry for CO_2 capture is the ability for solvent regeneration; as shown in Figure 19, all chemicals are regenerated. Interestingly, Carbon Engineering's inspiration for capturing CO_2 using alkaline chemistry came from studying the Kraft pulping process, in which sodium and calcium hydroxides are used in a closed system for biomass pulping, as shown in Figure 19A.[40] Carbon Engineering chose potassium hydroxide over sodium hydroxide because the former is better at reacting with dilute concentrations of CO_2 in the air. Kraft chemistry for biomass pulping is very similar to that used for calcium looping CO_2 capture, as shown in Figure 19, except the substrate is biomass, instead of carbon dioxide. Figure 20A shows a simplified process flow diagram of Kraft pulping, and Figure 20B shows an altered process flow diagram wherein biomass is replaced with flue gas. Thus, Kraft pulping mills have the potential for calcium looping CO_2 capture through process intensification. The theoretical CO_2 absorption capacity of NaOH is higher than that of monoethanolamine (MEA), with 0.9 tonnes of NaOH and 1.39 tonnes of MEA required to capture 1 tonne of CO_2.[39] Many of the critical components necessary for large scale calcium looping CO_2 capture currently exist at Kraft pulping mills: alkali reactors, liquid/solid separators, alkali regenerators, lime kilns, and causticizing plants. Residual alkali (NaOH) that does not react with biomass during pulping in alkali reactors is available for reaction with CO_2 to form additional sodium carbonate. Flue gas from various on-site sources, such as recovery boilers and multi-fuel boilers, could be passed through pulping liquor to capture a percentage of the CO_2. The biomass- and CO_2-derived sodium carbonate would then move through the existing process with little to no modifications required. The operations in which alkali CO_2 capture could occur include the pulping digestor, recovery boiler, or a new operation dedicated to CO_2 capture. The new operation could involve a mineralization reactor in which flue gas reacts with sodium hydroxide to form sodium carbonate, which could then combine with existing sodium carbonate prior to entering the causticizer. The CO_2-derived sodium carbonate transfers CO_2 to calcium carbonate which is then calcined to liberate CO_2. Figure 20B incorporates complete oxy-fuel combustion in the lime kiln to provide a pure stream of CO_2 post-calcination (and postcondensation of water vapor). Research shows the conversion of a lime kiln from air- to oxy-fuel increases kiln capacity, which would be necessary if additional $CaCO_3$ is being processed due to the incorporation of CO_2 capture in the upstream pulping process.[8] Therefore, the aforementioned synergies with Kraft pulping and calcium looping CO_2 capture create an opportunity for pulp and paper mills to cost-effectively integrate CO_2 capture at scale with minimal capital expenditure.

CO_2 Utilization

The Section 45Q tax credit provides the potential for pulp and paper mills to improve cash flow through on-site CO_2 utilization.[16] To be eligible for the utilization tax credits, taxpayers must prove through life cycle assessment that the CO_2 is captured and permanently isolated from the atmosphere or displaced from being emitted into the atmosphere. Two potential pathways for CO_2 utilization in pulp and paper manufacturing are lignin precipitation and calcium carbonate filling.

Lignin Precipitation

Lignin precipitation involves a process similar to that described in the previous section on calcium looping CO_2 capture wherein flue gas is bubbled through black liquor to lower the pH and precipitate lignin.[15,41] To achieve a high purity lignin product, sulfuric acid is used to remove inorganics. Valmet has patented a technology, Lignoboost, that achieves a high rate of lignin precipitation through the use of CO_2 for pH adjustment. Domtar is using the lignoboost process at their pulping mill located in Plymouth, NC.[18] Typically, high purity CO_2 is purchased for lignin precipitation, and thus CO_2 captured onsite could substitute purchased CO_2 and thereby potentially qualify for the 45Q utilization tax credit.

Calcium Carbonate Filling

Pulp and paper mills that use calcium carbonate fillers typically employ a pathway similar to the one shown in Figure 21, wherein mined calcium carbonate is calcined to produce lime (CaO).[42] Lime (CaO), which is much lighter than calcium carbonate, is transported to a satellite site nearby to a pulp and paper mill where various grades of calcium carbonate filler are made using purchased CO_2. The calcium carbonate filler may be eligible for the 45Q utilization tax credit if CO_2 from a nearby pulp and paper mill is used instead of the purchased CO_2, because in this way CO_2 is displaced from being emitted into the atmosphere, as shown by the dashed stream in Figure 21. Lignin precipitation and calcium carbonate filling both require full life cycle assessments to guarantee eligibility with the Section 45Q utilization tax credit.

Conclusions

The pulp & paper industry in the United States emits approximately 150 million metric tons of CO_2 per year, of which 77% is biogenically derived. Lime kilns, multi-fuel boilers, and recovery boilers are responsible for approximately 9%, 43%, and 48% of total CO_2 emissions. Approximately 90% of total CO_2 emissions are derived from the combustion of black liquor (~50%), wastewood (~25%), and natural gas (~15%). Lime kiln emissions are more concentrated in CO_2 than those from multi-fuel boilers and recovery boilers due to fuel- and calcium carbonate-derived CO_2. The top-down, industry-wide analysis indicates CO_2 concentration is the most influential variable when estimating energy demands of CO_2 capture. Fuels with high H/C ratios, such as natural gas, generate low concentrations of CO_2 via combustion, whereas fuels with low H/C ratios, such as coal and biomass, generate relatively high concentrations of CO_2. Therefore, within the scope of the top-down analysis, capturing CO_2 from coal and biomass combustion is generally less expensive than capturing CO_2 from natural gas combustion. Idling capital assets available at pulp and paper mills warrant further investigation into the feasibility of using such for CO_2 capture. 88 of the 205 mills assessed in the top-down, industry-wide analysis are located on geology suitable for sequestration and therefore do not require long-distance transportation of CO_2. The average distance to suitable geology for mills that are not co-located is ~130km. The bottom-up analysis estimates baseline costs of CO_2 capture for the four select mills to range from -\$19 to \$23 per tonne CO_2 (in year 2026 with \$50 per tonne CO_2 tax credit applied). The bottom-up analysis shows operating costs dominating capital costs for CO_2 capture at all four mills. For all four mills in the bottom-up analysis, levelized costs of CO_2 capture are near to zero for the baseline combined emissions scenarios (in year 2026 with \$50 per tonne CO_2 tax credit applied). Overall, the bottom-up analysis estimates the capture of CO_2 from combined emissions to be less expensive than that of CO_2 from lime kiln emissions only, due primarily to differences in economies of scale; this observation contradicts energy demand trends from the top-down analysis. According to the bottom-up analysis, waste heat at Mill 4 enables low cost CO_2 capture, particularly for the small scale scenario: Steam + Lime Kiln Emissions, wherein the levelized cost is -\$19 per tonne CO_2. Thus, waste heat, if of sufficient quality and availability, can substantially reduce costs of CO_2 capture. The Combined Emissions scenarios demand substantial quantities of biomass and thereby require investigation into local biomass resource availability. Sensitivity analyses indicate reboiler duty, biomass cost, electricity cost, rate of MEA loss, return on equity, and transportation distance affect levelized costs to varying degrees. Reboiler duty, biomass cost, and transportation distance are the most influential parameters analyzed. Lime Kiln Emissions scenarios are proven to be most sensitive to transportation distance due to their low flow rates of CO_2 and thus high pipeline cost intensities. Retrofitting idling biomass boilers and turbogenerators for CO_2 capture significantly lowers levelized costs, particularly for Steam & Power Emissions scenarios. The use of natural gas-fueled boiler and turbogenerator systems would likely reduce costs of CO_2 capture due to the very low cost of natural gas energy in the US, relative to biomass energy. However, the net removal of CO_2 from the atmosphere would be reduced if natural gas was used in place of biomass fuel. A multivariate regression is generated using data from the bottom-up analysis and data from the literature[11] to estimate costs of capturing CO_2 at all 205 mills selected for this study. Cost estimates of CO_2 capture from the top-down, industry-wide analysis range from -\$0.6 to \$12.7 per tonne CO_2 (in year 2026 with \$50 per tonne CO_2 tax credit applied). The top-down economic analysis finds that, overall, capturing lime kiln- and recovery boiler-derived CO_2 is less costly than multi-fuel boiler-derived CO_2. For mills that are not co-located with suitable geology, the average cost of transporting CO_2 to suitable geology is \$24.8 per tonne CO_2 (in the year 2026, not accounting for the 45Q tax credit). The average incremental cost of capturing combined CO_2 emissions, expressed as percent of manufacturing cost, is ~1% for the different product grades assessed. Process intensification and innovation offer an opportunity to reduce the costs of CO_2 capture through 1) partial oxy-fuel combustion of lime kilns with post-combustion CO_2 capture and 2) integrated calcium looping CO_2 capture. Utilizing Kraft chemistry for large scale calcium looping CO_2 capture at existing pulp and paper mills is of particular interest due to the potential for innovation and thus high impact. Pathways for CO_2 utilization, such as lignin precipitation and calcium carbonate filling, hold the potential for cash flow improvement through the Section 45Q utilization tax credit. Detailed lifecycle assessments in compliance with the US Internal Revenue Service are required to validate eligibility for the utilization tax credit.

Conflicts of Interest: Authors declare no conflicts of interest.

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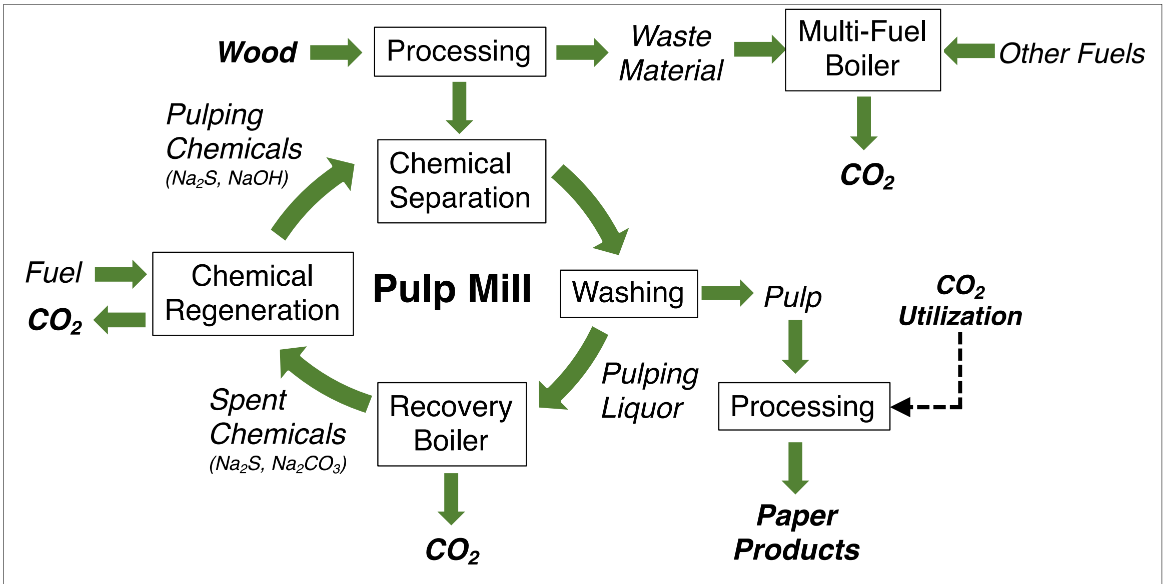

Figure 1: A simplified process flow diagram of a Kraft pulp mill.

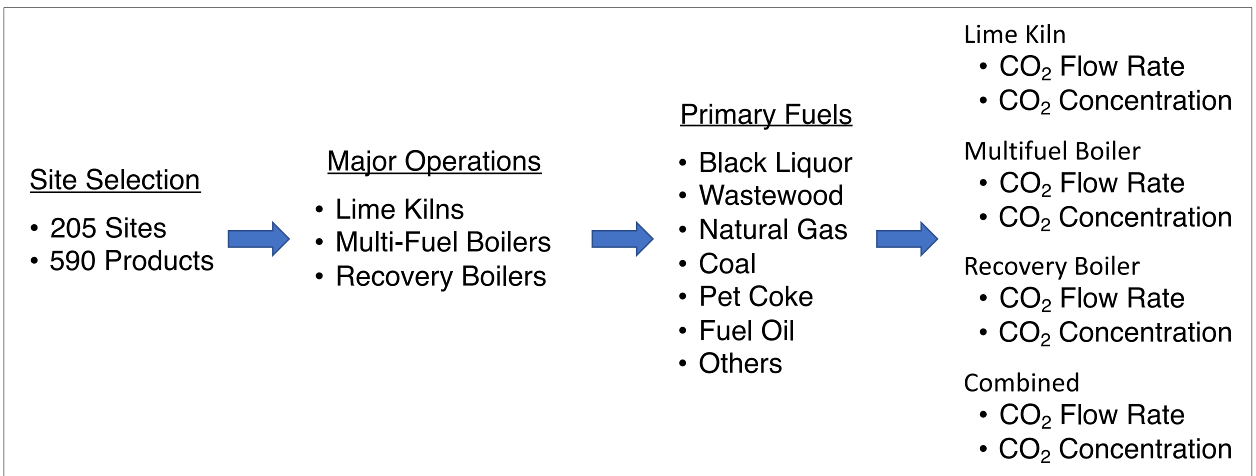

Figure 2. Logic flow to quantify CO_2 emissions in multiple ways for each site. (Year: 2018)

Table 1. Descriptions of the four mills selected for detailed analysis

Site Name	Site Type	Products	CO_2 Capacity	Motivation from Top-Down, Industry-Wide Analysis
Mill 1	Virgin & Recycled Integrated	Containerboard	Lime Kiln: 0.204 $MtCO_2/y$ Combined: 2.59 $MtCO_2/y$	• High CO_2 concentration of combined emissions (14.83 mol%) • Integrated site that produces pulp and board products • Co-located with suitable geological storage • Eligible for 45Q sequestration tax credits
Mill 2	Virgin Integrated	Cartonboard	Lime Kiln: 0.223 $MtCO_2/y$ Combined: 2.89 $MtCO_2/y$	• Largest quantity of CO_2 emissions of all sites (2.89 $MtCO_2/y$) • High percentage biogenic emissions (> 85%) • Integrated site that produces pulp and board products • Co-located with suitable geological storage • Eligible for 45Q sequestration tax credits
Mill 3	Virgin Integrated	Market Pulp	Lime Kiln: 0.417 $MtCO_2/y$ Combined: 2.00 $MtCO_2/y$	• High CO_2 concentration of lime kiln emissions (27.15 mol%) • High percentage biogenic CO_2 (> 85%) • Co-located with suitable geological storage • Eligible for 45Q sequestration tax credits
Mill 4	Virgin & Recycled Integrated	Containerboard, Cartonboard	Lime Kiln: 0.183 $MtCO_2/y$ Combined: 1.23 $MtCO_2/y$	• Large quantity of waste heat available on-site • Integrated site that produces pulp and board products • Co-located with suitable geological storage • Eligible for 45Q sequestration tax credits

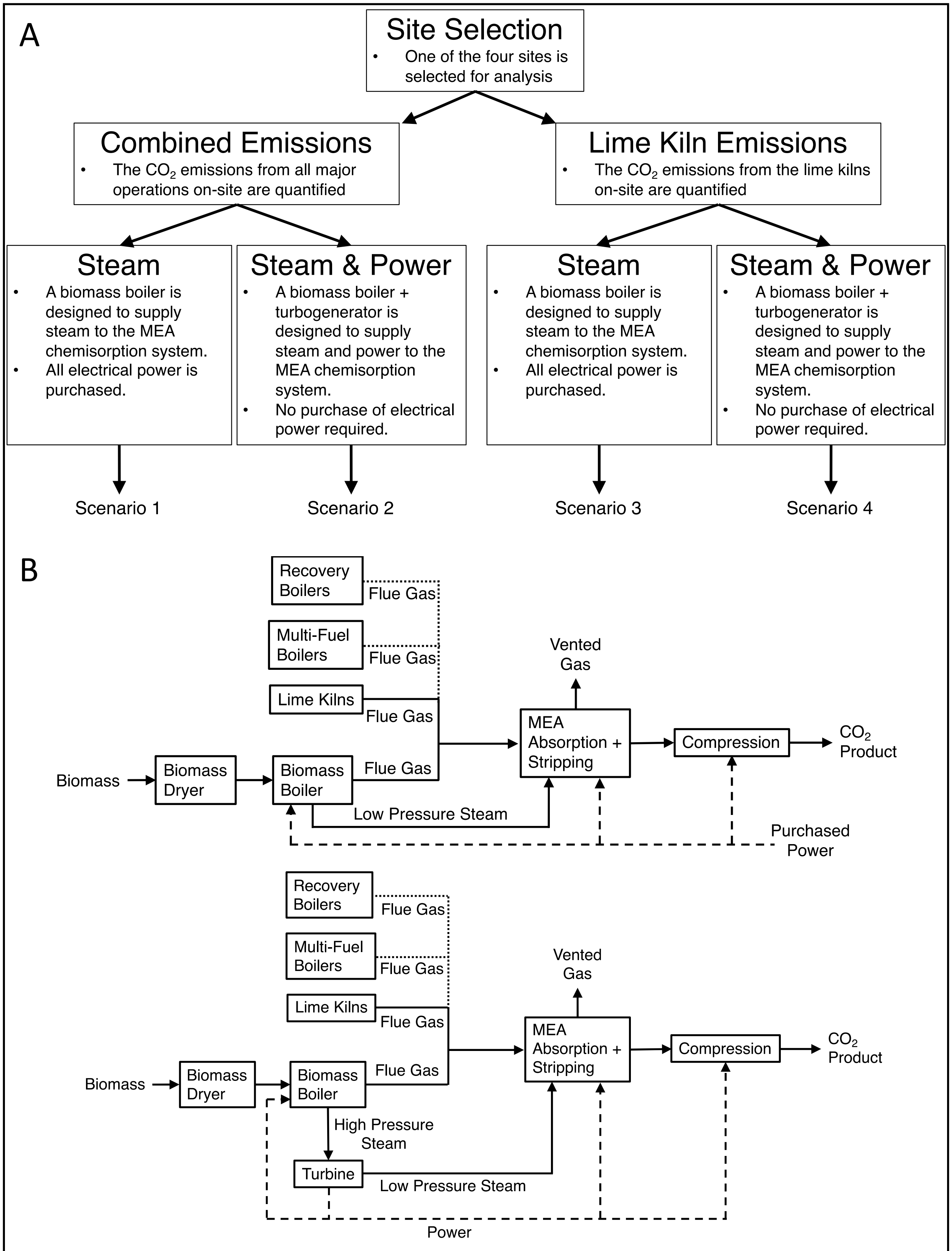

Figure 3. A) Descriptions of the four scenarios investigated for each of the four select sites. B) Simplified process flow diagrams of the "steam" (top) and "steam & power" (bottom) processes.

Table 2. Baseline technical details consistent with all scenarios, unless noted otherwise.

Energy content of biomass fuel (LHV) (GJ/dry tonne)	16.8
1st law conversion efficiency of fuel energy (LHV) to steam enthalpy (Steam scenario: S, Steam & Power scenario: S&P)	S: 80% S&P: 69%
1st law conversion efficiency of fuel energy (LHV) to electrical power (Steam & Power scenario)	S&P: 11%
Biogenic emissions from boiler (tCO_2/t-biomass)	1.51
CO_2 concentration of boiler flue gas (mol%)	15.5%
Amine chemisorbent (MEA) concentration (wt%)	30%
Temperature of absorber column	~ 50°C
Temperature of stripper column	~ 120°C
Rate of amine loss (kg MEA/tCO_2 captured)	2.3
Reboiler Duty (GJ/tCO_2)	3.5
Total CO_2 capture rate (tCO_2 captured/tCO_2 generated)	90%
Concentration of CO_2 output (mol%)	97%
Pressure of CO_2 output (bar)	150
CO_2 compression power demand (kWh/tCO_2)	132

Table 3. Baseline economic metrics consistent with all scenarios, unless noted otherwise

Plant utilization	90%
Project year	2026
Capital scaling factor	0.7
Indirect capital costs (% of total direct capital)	35%
Capital cost contingency (% of total direct & indirect capital)	10%
Debt financing	0%
Debt interest rate	n/a
Equity financing	100%
Return on equity	5%
Payback period (years)	20
Capital recovery factor (CRF)	8%
Cost of biomass ($/dry short ton)	$60
Cost of excess steam (4.5 bar) ($/tonne)	$8.6
Cost of MEA chemisorbent ($/kg)	$5.9
Cost of electricity ($/kWh)	$0.07

Table 4. Cumulative CO_2 emissions differentiated by origin of fuel

Total CO_2 Emissions ($MtCO_2/y$)		
Fossil	34	23%
Biogenic	115	77%
Total	149	100%

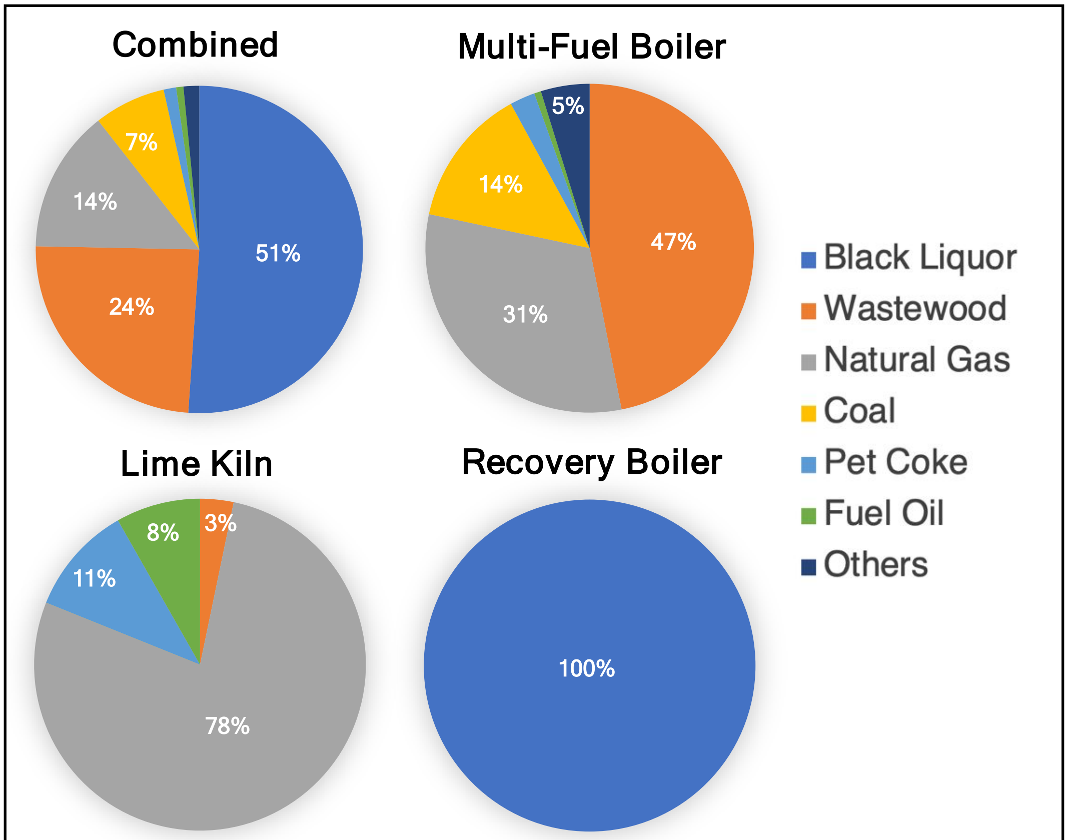

Figure 4. Contributions to CO_2 emissions by various fuels for the major operations across all pulp and paper mills in the US

Table 5. Cumulative CO_2 emissions by operation and their contribution to total emissions. Average CO_2 concentration in emission streams from the respective operations and the corresponding energy required for CO_2 separation; energy demand for CO_2 separation not including compression

CO_2 Emissions by Operation				
Operation	**CO_2 Emissions ($MtCO_2$/y)**	**Contribution to Total Emissions**	**Avg. CO_2 Concentration (mol%)**	**Energy Demand (kJ/mol-CO_2)**
Lime Kiln	13.7	9%	21%	13.1
Multi-Fuel Boiler	64.1	43%	9%	40.7
Recovery Boiler	71.4	48%	13%	24.9
Combined	149.2	100%	10%	37.4

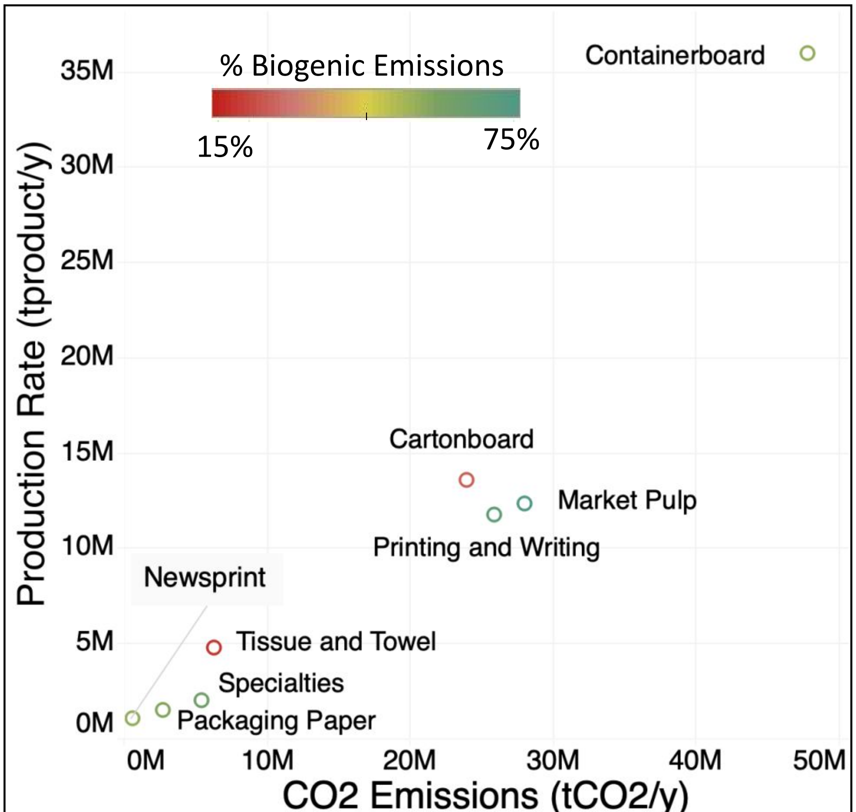

Figure 5. Annual production rates of the eight major product grades and their corresponding CO_2 emissions. The percentage of biogenic emissions for each major product grade shown via color.

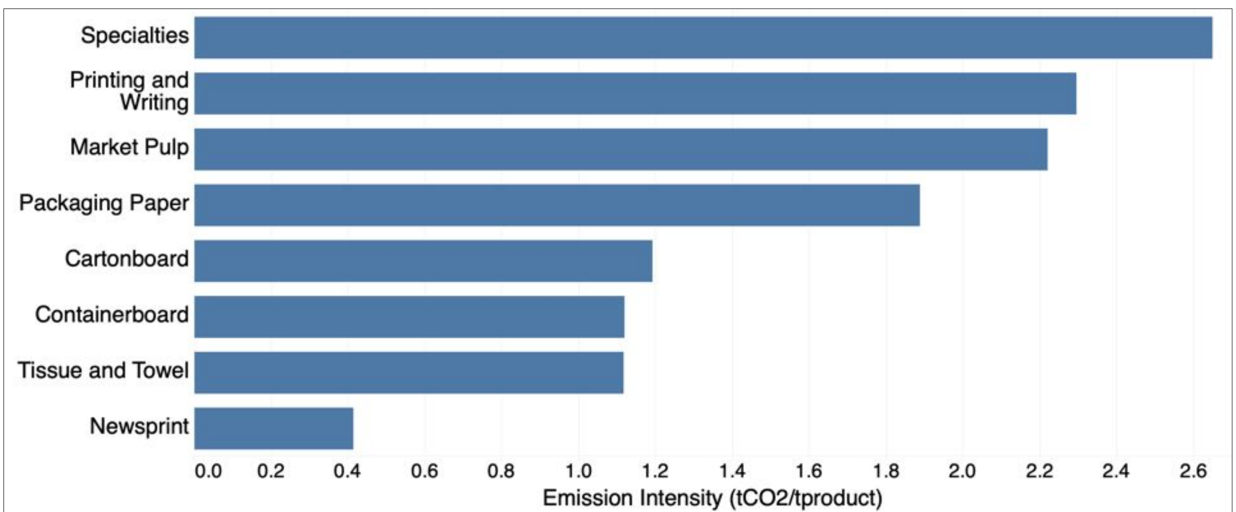

Figure 6. Emission intensities from on-site manufacturing for eight grades of pulp & paper products

Table 6. Quantities of operating and idling assets across all sites

Site Assets		
Operation	**# Operating**	**# Idling**
Lime Kiln	124	14
Multi-Fuel Boiler	406	103
Recovery Boiler	150	16

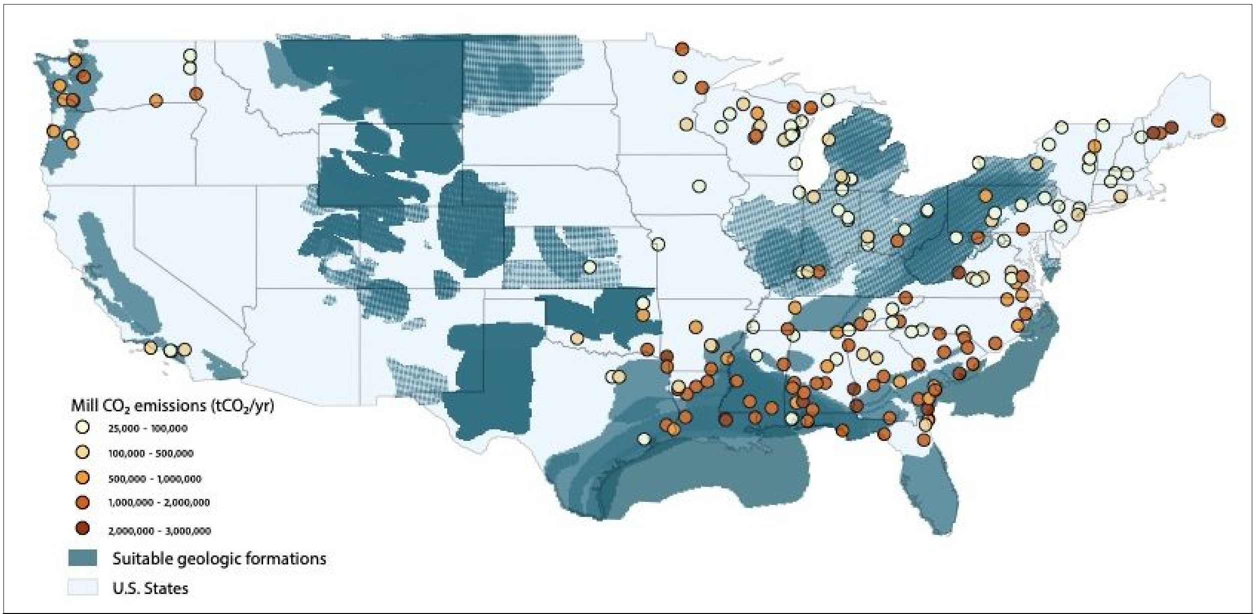

Figure 7. Geospatial plot showing the location and relative CO_2 capacity of all 205 pulp and paper mills. Suitable geology for sequestration overlaid in green.

Table 7. Pertinent process data and cost estimates of CO_2 capture ($/t$CO_2$) for the four baseline scenarios outlined in Figure 3A. Waste heat at Mill 4 is assumed to be of high enough quality for use as steam in the amine subsystem reboiler. Year of cost estimation: 2026. 45Q tax credit: $50/tCO2.

Baseline Scenario 1: Steam + Combined Emissions

	Mill 1	Mill 2	Mill 3	Mill 4
P&P Mill Biomass Flow Rate (dry short ton/day)	4225	5263	7320	3444
CO_2 System Biomass Flow Rate (dry short ton/day)	3140	3162	2438	831
CO_2 Concentration (mol%)	15.1%	13.0%	14.9%	13.1%
CO_2 Captured (tCO_2/y)	4.01E+06	4.03E+06	3.11E+06	1.60E+06
Power Demand (MW)	61	63	47	24
Steam Demand (GJ/h)	1600	1611	1242	640
Cost of Capture & Compression ($/t$CO_2$)	$2	$5	$4	$2

Baseline Scenario 2: Steam & Power + Combined Emissions

	Mill 1	Mill 2	Mill 3	Mill 4
P&P Mill Biomass Flow Rate (dry short ton/day)	4225	5263	7320	3444
CO_2 System Biomass Flow Rate (dry short ton/day)	3939	4088	3026	1859
CO_2 Concentration (mol%)	15.1%	13.2%	14.9%	13.6%
CO_2 Captured (tCO_2/y)	4.37E+06	4.45E+06	3.38E+06	2.07E+06
Power Demand (MW)	77	80	59	36
Steam Demand (GJ/h)	1744	1778	1349	827
Cost of Capture & Compression ($/t$CO_2$)	-$3	$0	-$2	$1

Baseline Scenario 3: Steam + Lime Kiln Emissions

	Mill 1	Mill 2	Mill 3	Mill 4
P&P Mill Biomass Flow Rate (dry short ton/day)	4225	5263	7320	3444
CO_2 System Biomass Flow Rate (dry short ton/day)	247	243	506	0
CO_2 Concentration (mol%)	18.8%	18.6%	21.4%	20.9%
CO_2 Captured (tCO_2/y)	3.16E+05	3.10E+05	6.45E+05	1.83E+05
Power Demand (MW)	4.9	4.8	9.9	2.9
Steam Demand (GJ/h)	126	124	258	73
Cost of Capture & Compression ($/t$CO_2$)	$23	$23	$12	-$19

Baseline Scenario 4: Steam & Power + Lime Kiln Emissions				
	Mill 1	Mill 2	Mill 3	Mill 4
P&P Mill Biomass Flow Rate (dry short ton/day)	4225	5263	7320	3444
CO_2 System Biomass Flow Rate (dry short ton/day)	320	314	640	296
CO_2 Concentration (mol%)	18.4%	18.2%	20.8%	18.2%
CO_2 Captured (tCO_2/y)	3.48E+05	3.42E+05	7.06E+05	3.16E+05
Power Demand (MW)	6.2	6.1	12.5	5.8
Steam Demand (GJ/h)	139	137	282	126
Cost of Capture & Compression ($/$tCO_2$)	$18	$18	$8	$3

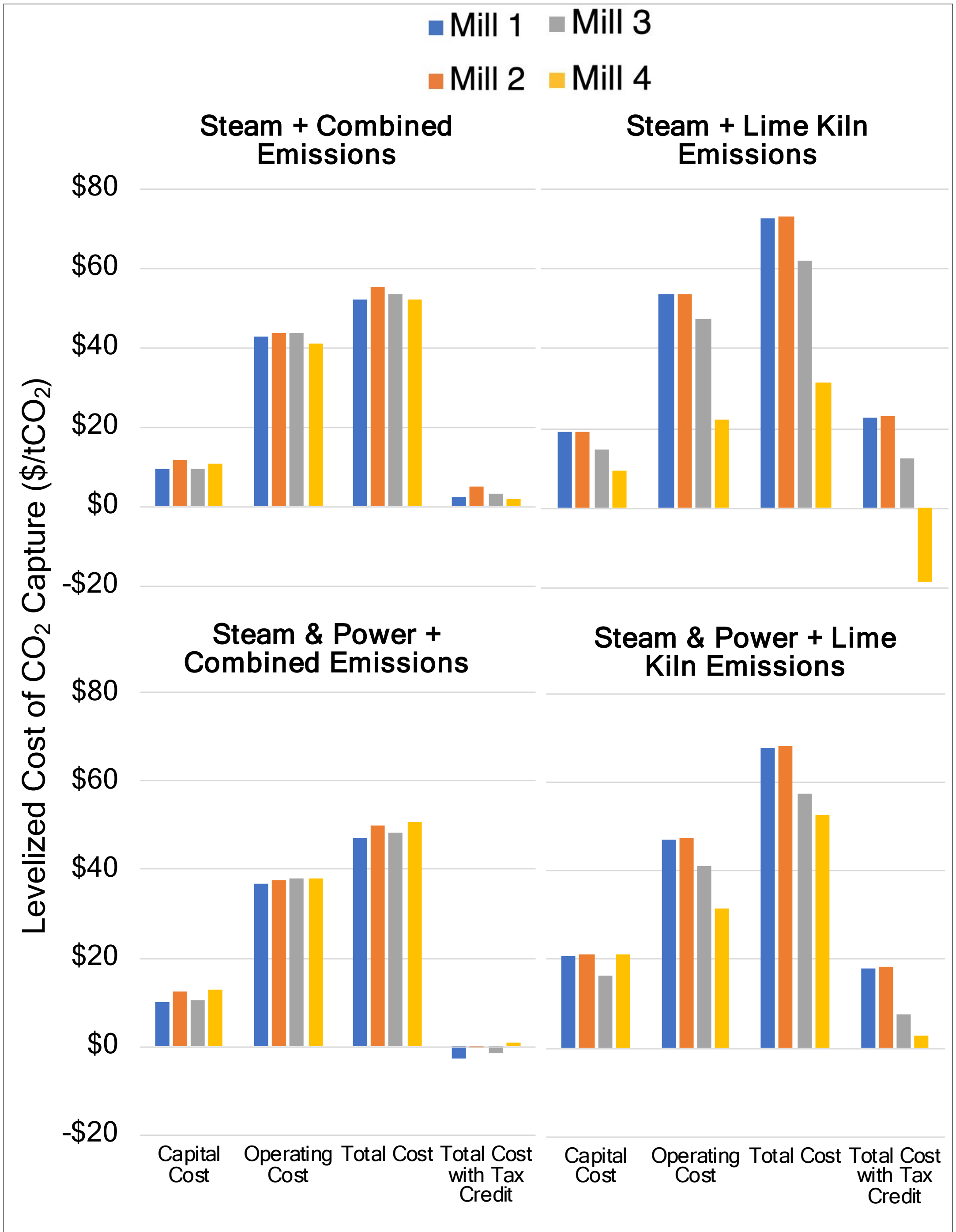

Figure 8. Levelized capital, operating, and total costs of CO_2 capture for baseline scenarios. Year of cost estimation: 2026. 45Q tax credit: $50/tCO2.

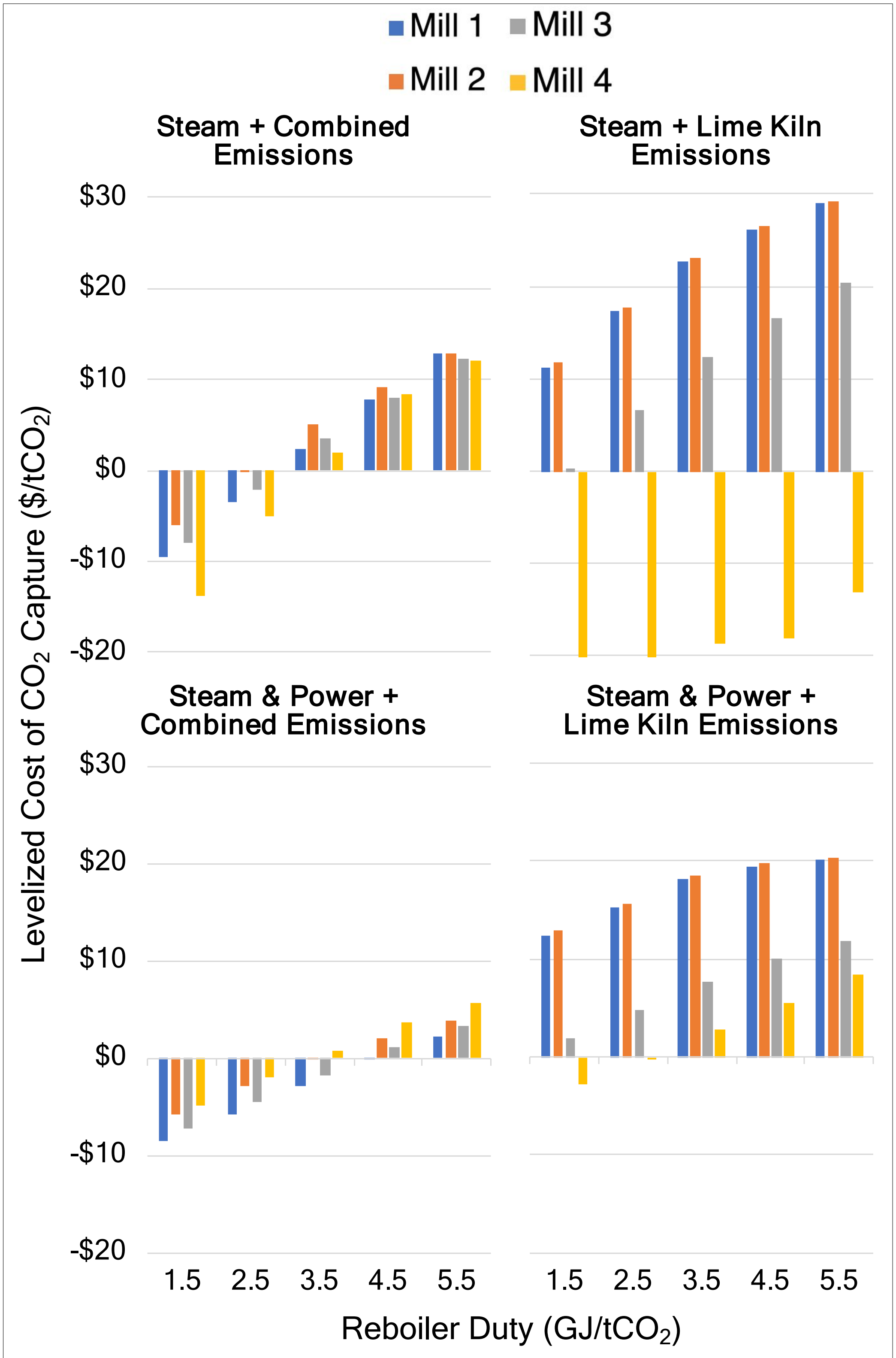

Figure 9. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in reboiler duty. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

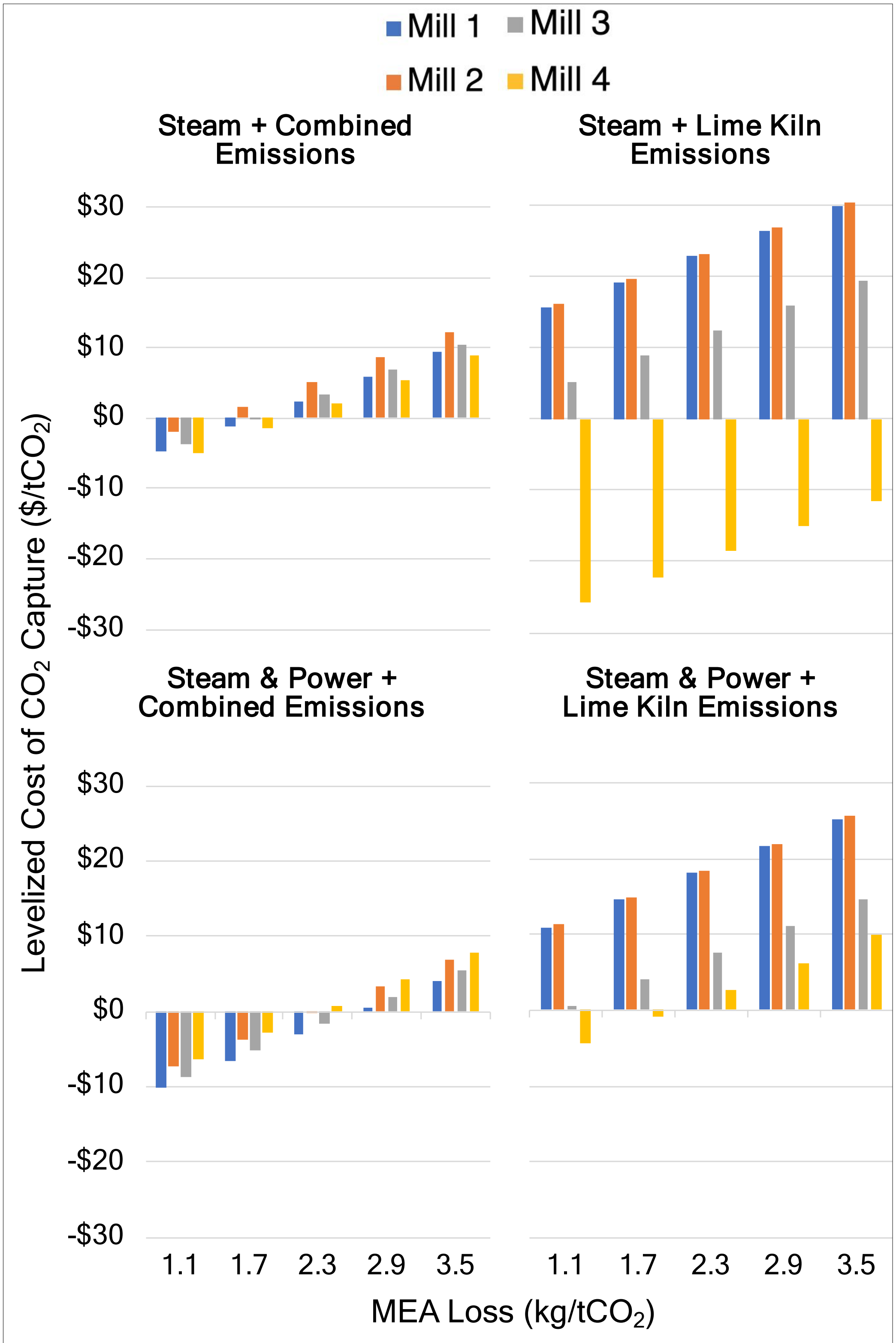

Figure 10. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in rate of monoethanolamine (MEA) loss. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

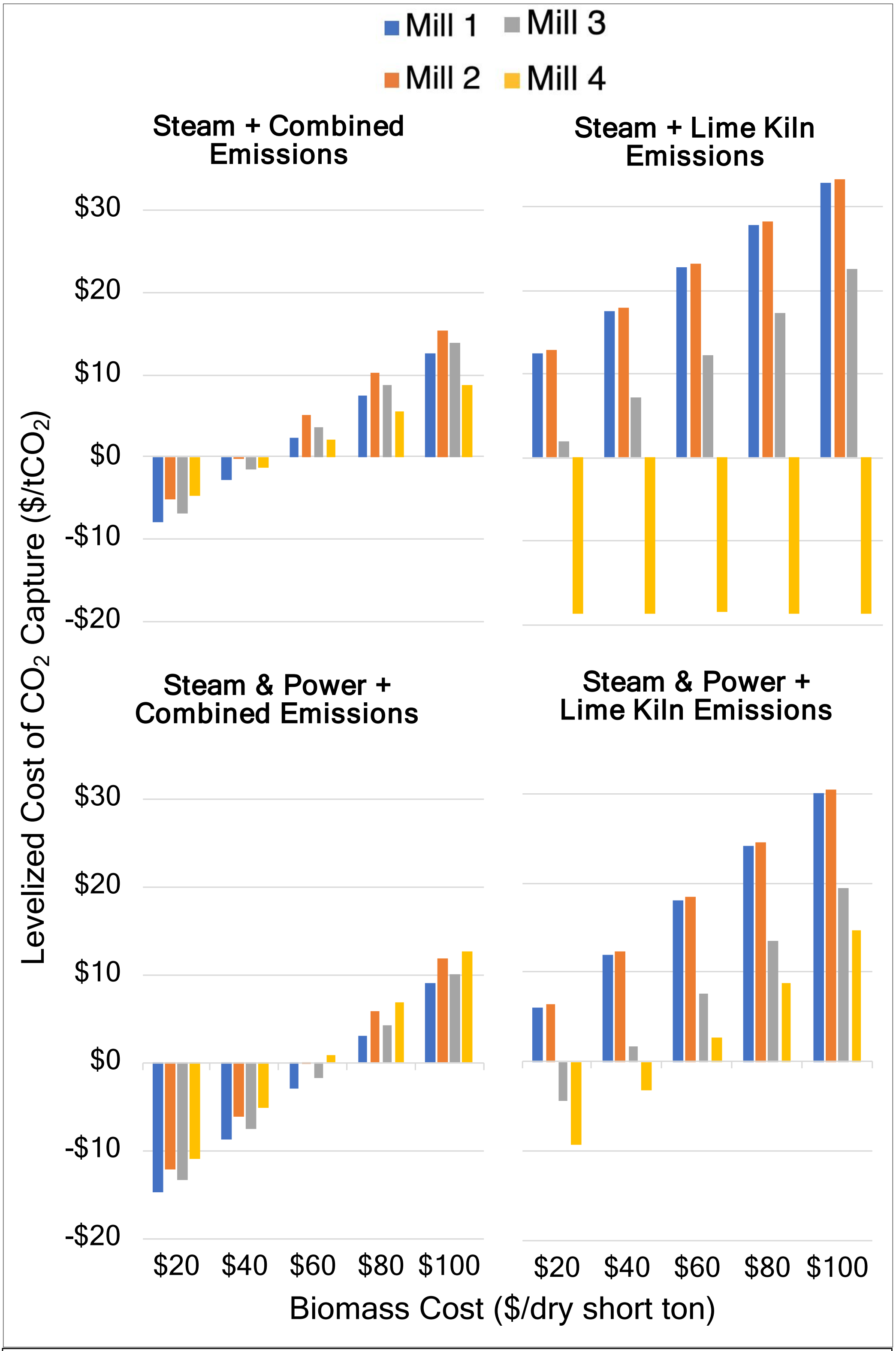

Figure 11. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in biomass cost. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

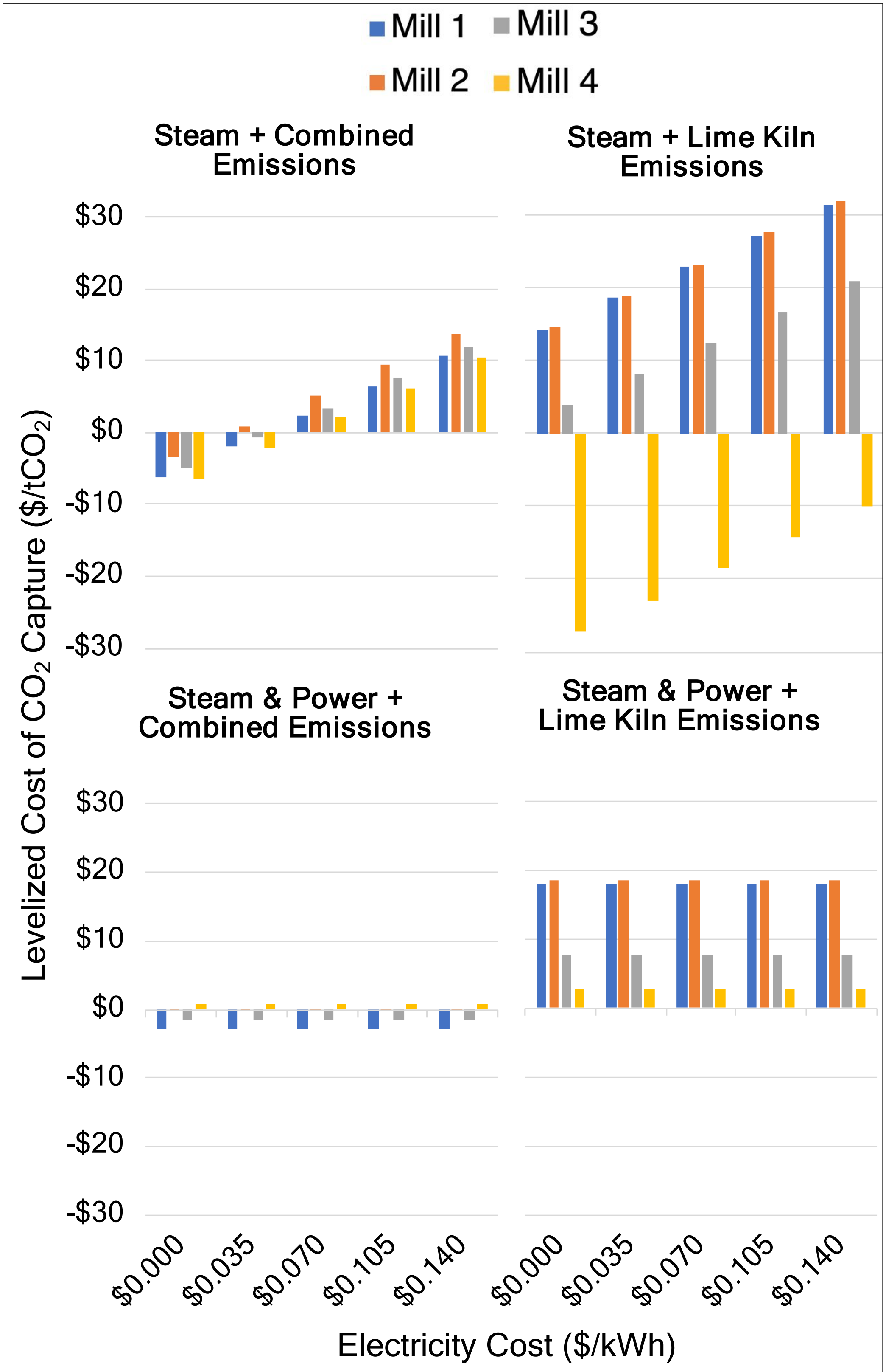

Figure 12. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in electricity cost. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

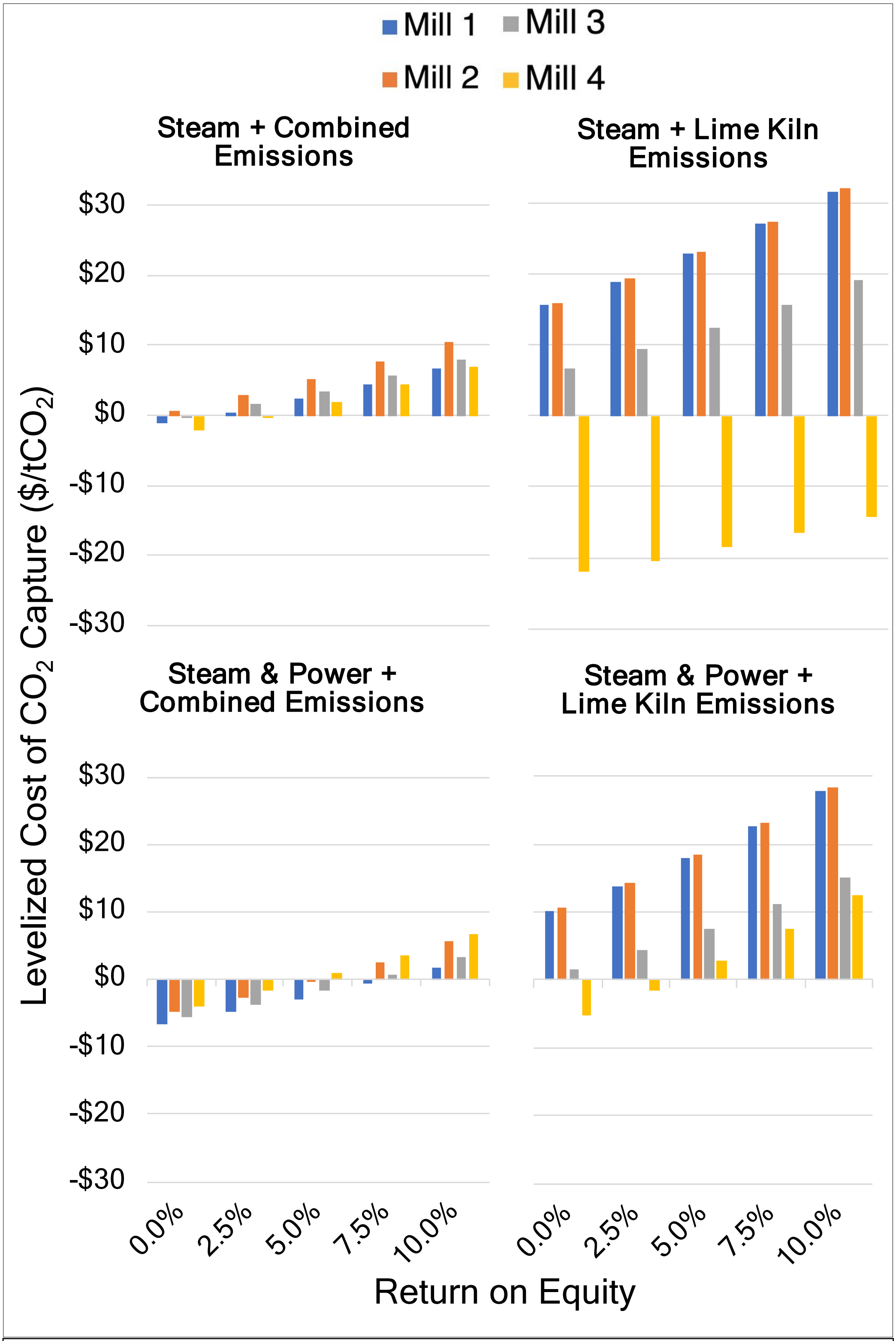

Figure 13. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in return on equity. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

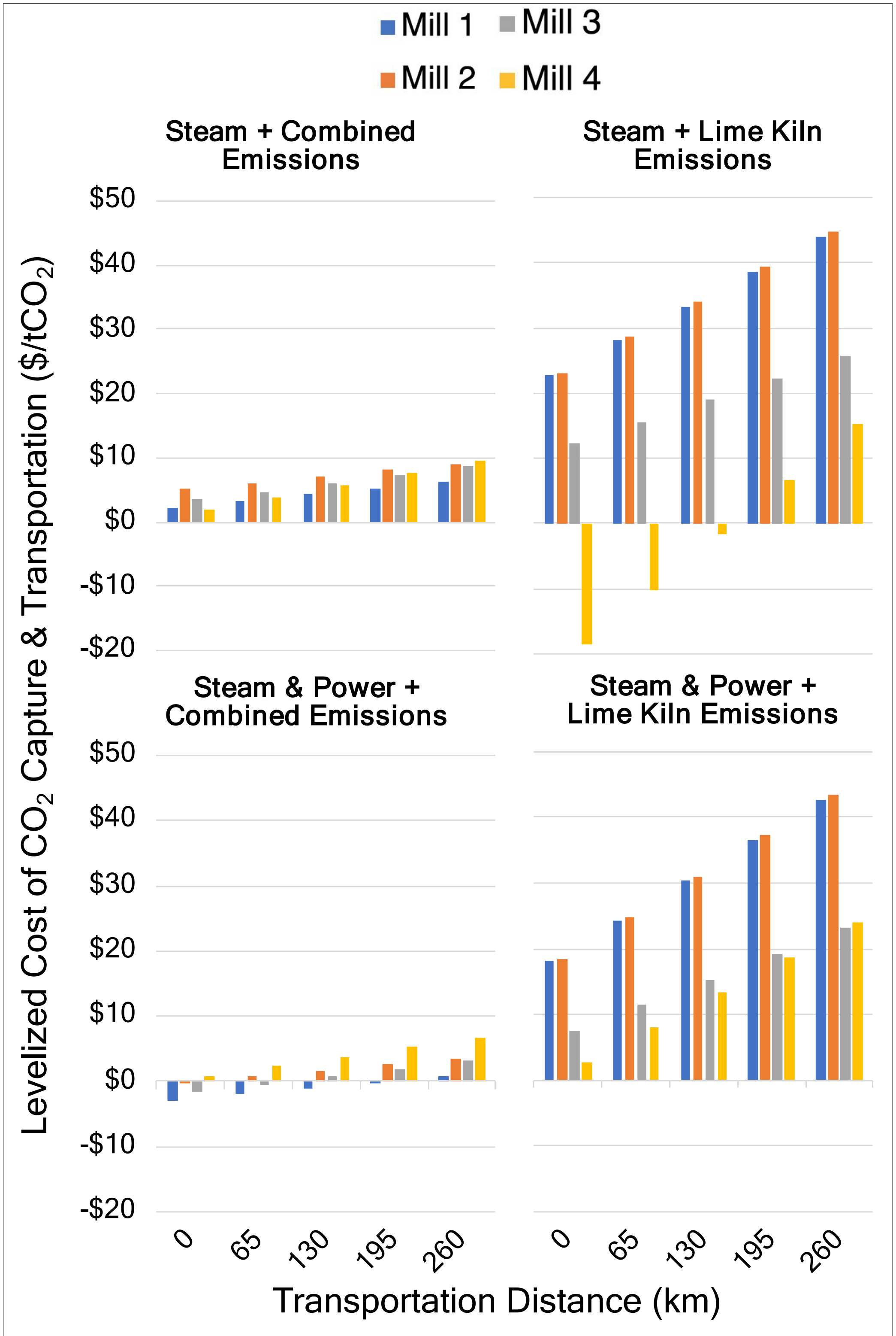

Figure 14. Sensitivity analysis showing levelized costs of CO_2 capture ($/t$CO_2$) with variation in transportation distance. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

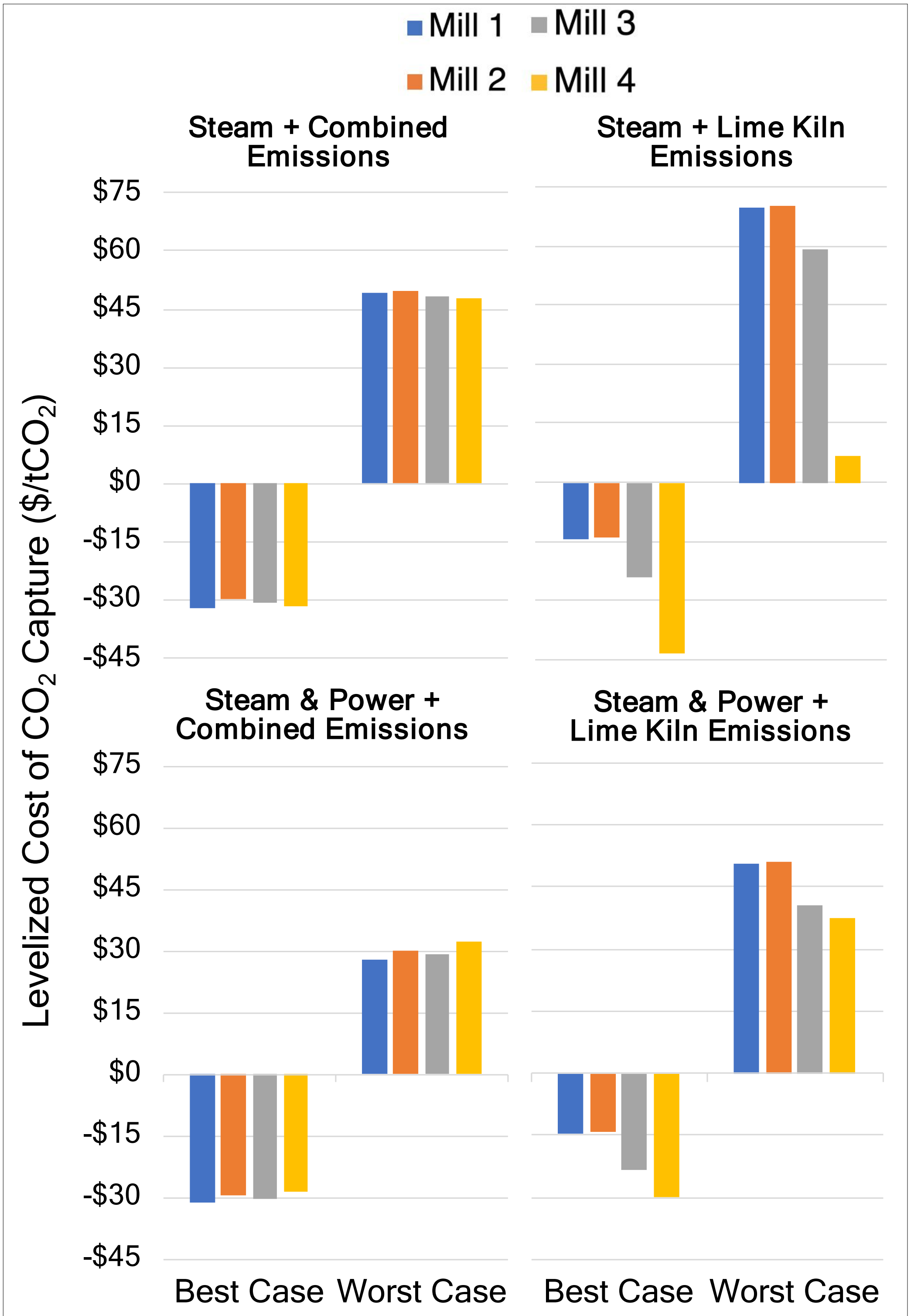

Figure 15. Levelized costs of CO_2 capture ($/t$CO_2$) for best and worst case scenarios. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

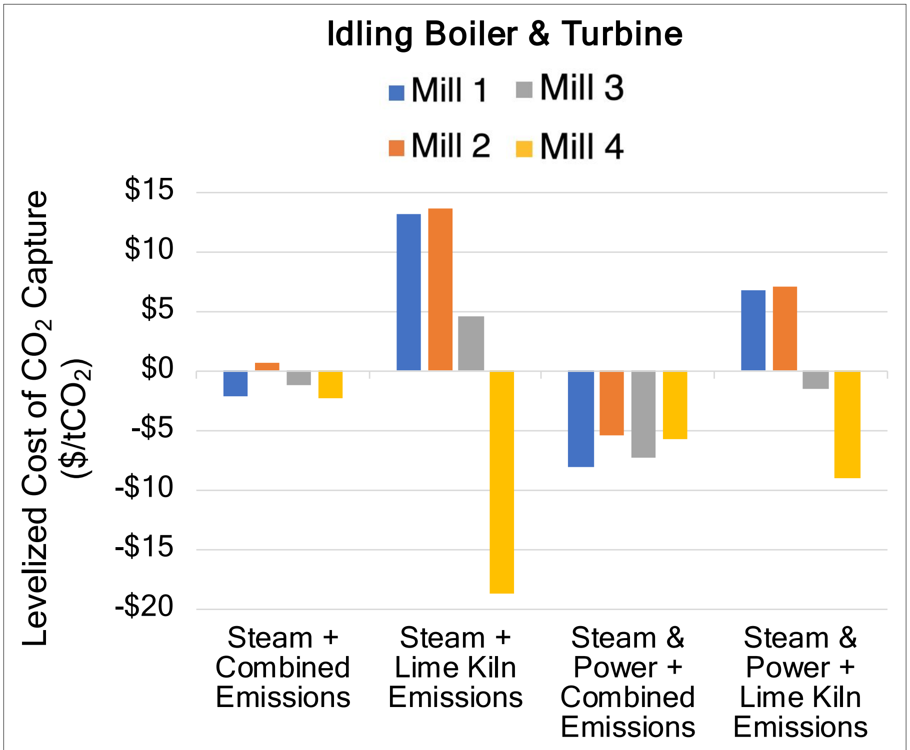

Figure 16. Levelized costs of CO_2 capture ($/t$CO_2$) for baseline scenarios with existing boiler and turbogenerator subsystems. Year of cost estimation: 2026. 45Q tax credit: $50/t$CO_2$.

Table 8. Levelized costs of capturing CO_2 from major operations at all sites (year 2026). To account for the 45Q tax credit, a $50/t$CO_2$ reduction in cost is incorporated in each cost estimate.

Operation	Min.	Max.	Avg.	Std. Dev.
Lime Kiln	$2.1	$5.0	$4.6	$0.6
Multi-Fuel Boiler	$1.6	$12.7	$8.1	$3.6
Recovery Boiler	$1.2	$5.9	$4.0	$1.1
Combined	-$0.6	$12.7	$6.4	$4.6

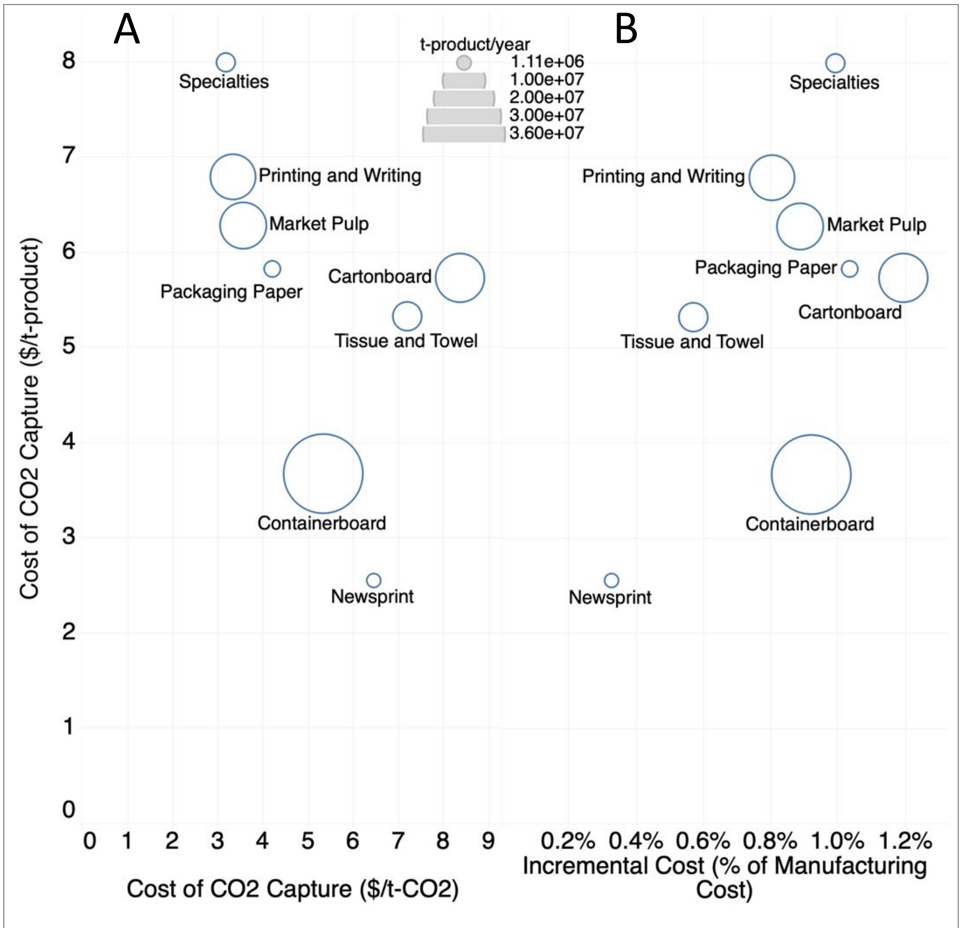

Figure 17. A) Costs of capturing combined CO_2 emissions on the basis of product mass ($/tproduct) and CO_2 mass ($/t$CO_2$). B) Costs of capturing combined CO_2 emissions on the basis of product mass and the resultant incremental cost in manufacturing cost expressed as percent increase in manufacturing cost. Year of cost estimates is 2026. To account for the 45Q sequestration tax credit, a $50/t$CO_2$ reduction in cost is incorporated in each cost estimate.

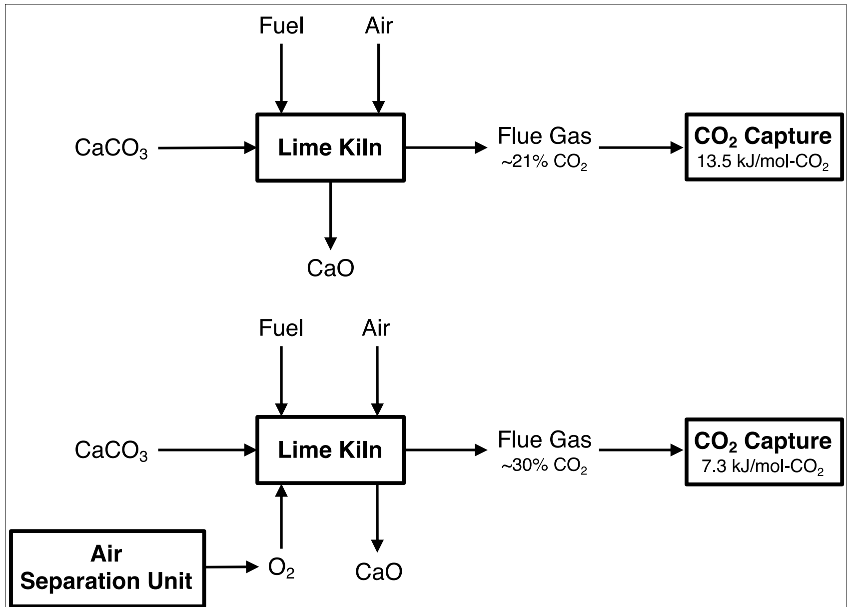

Figure 18. Process flow diagrams demonstrating the potential for process innovation to reduce costs of CO_2 capture at existing Kraft pulping mills that utilize oxygen delignification through partial oxy-fuel combustion of lime kilns

A

C = carbon in biomass

1. $2NaOH + C \rightarrow Na_2CO_3 + H_2O$
2. $Na_2CO_3 + Ca(OH)_2 \rightarrow 2NaOH + CaCO_3$
3. $CaCO_3 \rightarrow CaO + CO_2$
4. $CaO + H_2O \rightarrow Ca(OH)_2$

B

C = flue gas

1. $2NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O$
2. $Na_2CO_3 + Ca(OH)_2 \rightarrow 2NaOH + CaCO_3$
3. $CaCO_3 \rightarrow CaO + CO_2$
4. $CaO + H_2O \rightarrow Ca(OH)_2$

Figure 19. Chemical reactions involved in A) Kraft pulping of biomass and B) calcium looping CO_2 capture

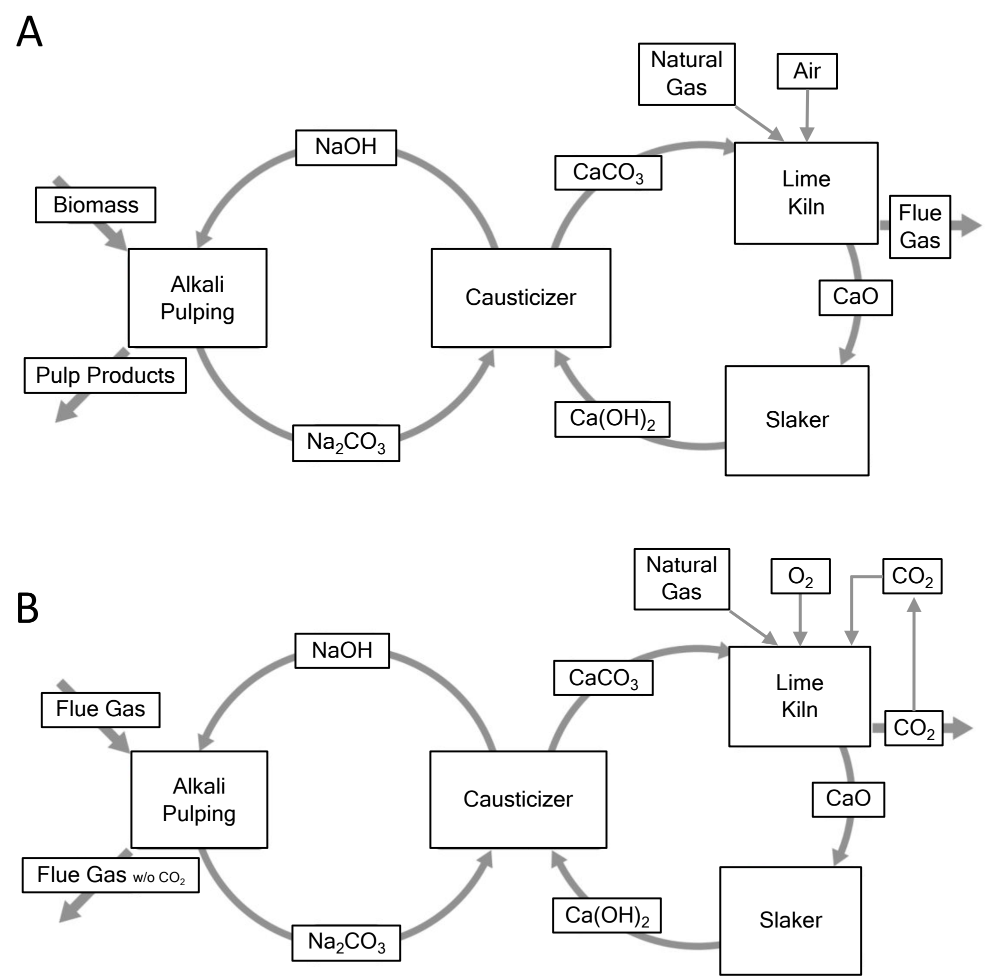

Figure 20. A) Generalized process flow diagram of the Kraft pulping process highlighting the alkaline chemistry used for biomass pulping. B) Generalized process flow diagram of the Kraft pulping process being utilized for calcium looping CO_2 capture.

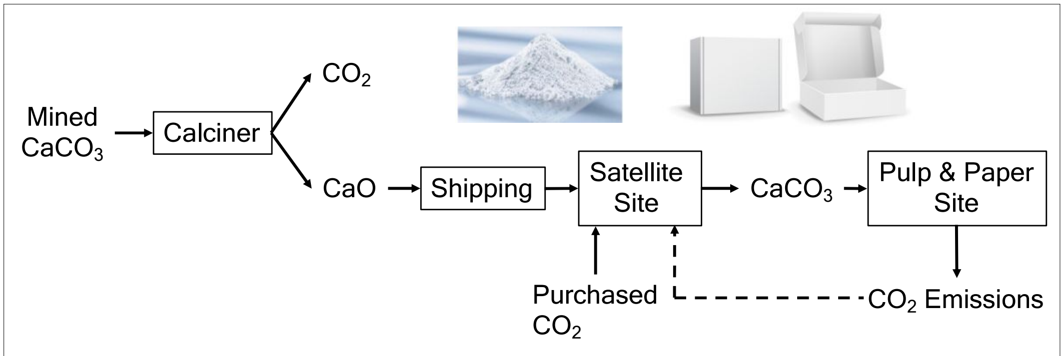

Figure 21. Generalized flow diagram showing how calcium carbonate filler is made for pulp and paper products