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Policy Analysis

Cost and Life-Cycle Greenhouse Gas Implications of Integrating Biogas Upgrading and Carbon Capture Technologies in Cellulosic Biorefineries

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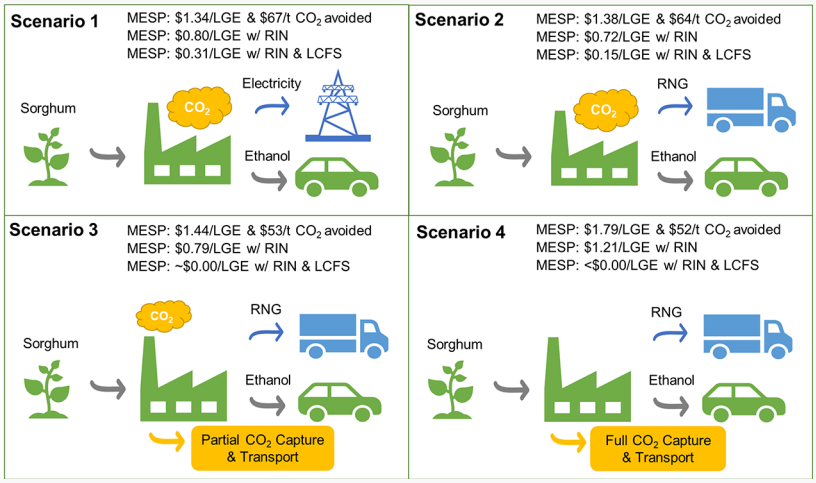

ABSTRACT: Gaseous streams in biorefineries have been undervalued and underutilized. In cellulosic biorefineries, coproduced biogas is assumed to be combusted alongside lignin to generate process heat and electricity. Biogas can instead be upgraded to compressed biomethane and used as a transportation fuel. Capturing CO_2-rich streams generated in biorefineries can also contribute to greenhouse gas (GHG) mitigation goals. We explore the economic and life-cycle GHG impacts of biogas upgrading and CO_2 capture and storage (CCS) at ionic liquid-based cellulosic ethanol biorefineries using biomass sorghum. Without policy incentives, biorefineries with biogas upgrading systems can achieve a comparable minimum ethanol selling price (MESP) and reduced GHG footprint ($1.38/liter gasoline equivalent (LGE) and 12.9 gCO_{2e}/MJ) relative to facilities that combust biogas onsite ($1.34/LGE and 24.3 gCO_{2e}/MJ). Incorporating renewable identification number (RIN) values advantages facilities that upgrade biogas relative to other options (MESP of $0.72/LGE). Incorporating CCS increases the MESP but dramatically decreases the GHG footprint (−21.3 gCO_{2e}/MJ for partial, −110.7 gCO_{2e}/MJ for full CCS). The addition of CCS also decreases the cost of carbon mitigation to as low as $52−$78/t CO_2, depending on the assumed fuel selling price, and is the lowest-cost option if both RIN and California's Low Carbon Fuel Standard credits are incorporated.

INTRODUCTION

Cellulosic biofuels have the potential to reduce greenhouse gas (GHG) emissions by around 80% relative to gasoline.[1,2] This is due in part to the heat and electricity generated by combusting lignin alongside biogas from onsite wastewater treatment, which satisfies the facility's energy needs and can also result in net power exports to the grid.[3−5] However, these facilities have the potential to achieve net-negative GHG emissions and contribute to targets for bioenergy with carbon capture and sequestration (BECCS), which most climate stabilization scenarios rely on to compensate for difficult-to-decarbonize sectors.[6,7] To meet the target of <2 °C of global warming, the International Panel and Climate Change (IPCC) predicts that 3.6 Gt of biogenic CO_2 annually must be sequestered via BECCS by 2050.[8] BECCS discussions tend to focus on gaseous streams from power generation, while studies on capture and utilization of gaseous streams from advanced biorefineries are limited and tend to focus on microalgae.[9−11] The conversion of lignocellulosic biomass to fuels by advanced

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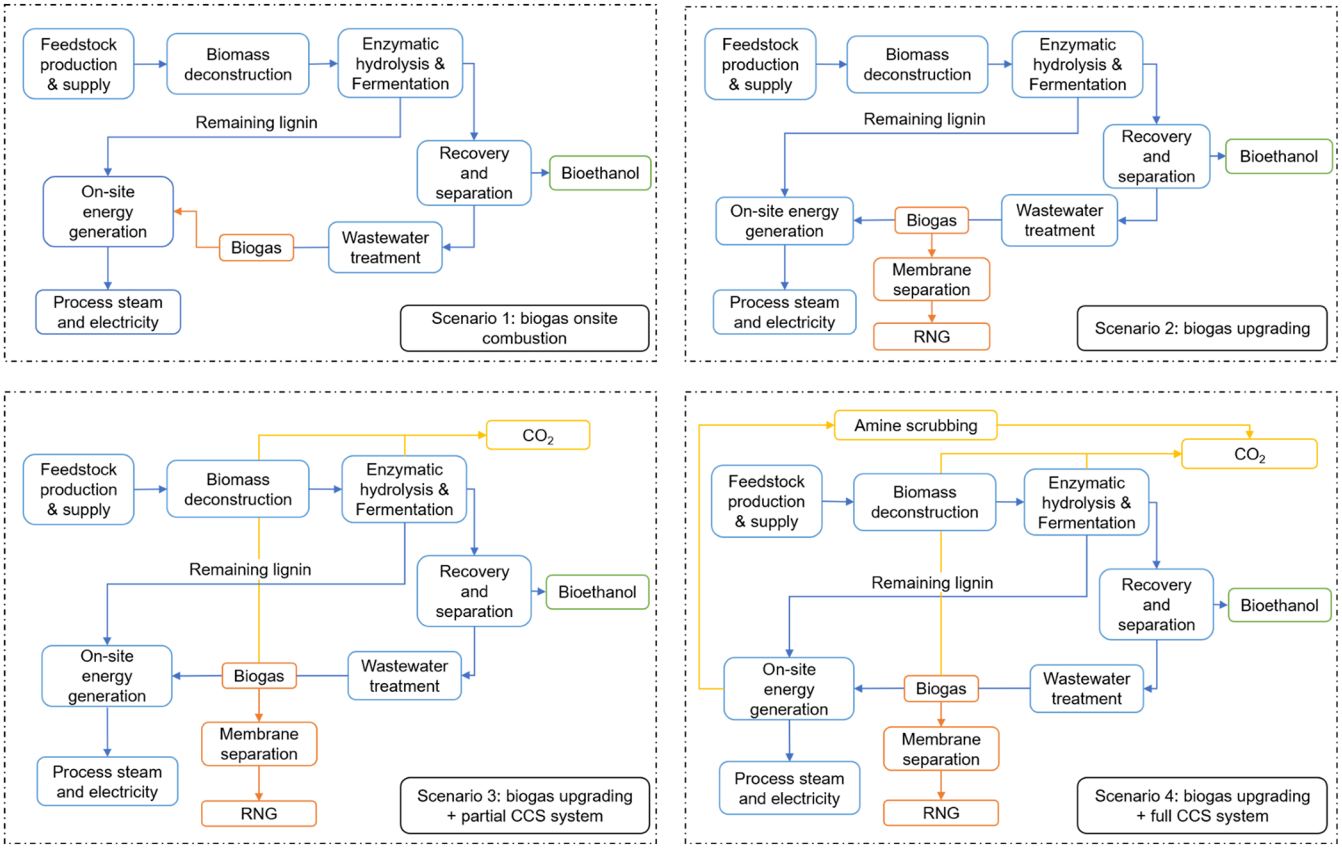

Figure 1. Process flow diagrams of the four scenarios analyzed in this study.

biorefineries results in multiple gaseous streams, the fates of which have been underexplored. Untreated biogas from the onsite anaerobic digestion (AD) of process wastewater can be upgraded to biomethane with well-established technologies and injected into existing natural gas pipelines, used as a feedstock for hydrogen production, or compressed for use as a transportation fuel (typically referred to as renewable natural gas, or RNG). Biogas upgrading also results in a concentrated CO_2 waste stream that can be combined with the CO_2-rich stream from fermentation and then sequestered or utilized. Additional CO_2 can be captured from the flue gas in the combined heat and power (CHP) unit. This study focuses on quantifying the economic and GHG implications of variations on biogas upgrading and CO_2 capture strategies at ethanol-producing cellulosic biorefineries, including the value of potential policy incentives.

There are three gaseous streams of interest in cellulosic biorefineries using biological conversion: the biogas produced during onsite wastewater treatment, the CO_2-rich waste stream from fermentation, and flue gas produced during combustion of lignin and other residual solids. The common assumption that biogas will be combusted onsite for heat and electricity[3,12] is likely based on outdated market conditions. Competition from wind, solar, and natural gas-fired power plants on the grid, along with economic incentives for renewable transportation fuels, has made the upgrading of biogas to RNG increasingly attractive.[13,14] Untreated biogas produced from AD consists of a roughly 50/50 mixture of methane (CH_4) and carbon dioxide (CO_2) with small amounts of impurities including hydrogen sulfide, carbon monoxide, oxygen (O_2) and nitrogen.[15] In order to inject biomethane into existing pipelines, the quality of biomethane needs to meet certain standards. Pacific Gas and Electricity (PG&E), one of the largest electric and gas utilities based in California, requires the gas to have less than 1% CO_2 and 0.1% O_2.[16] Numerous technologies have been explored for biogas upgrading, such as pressurized water scrubbing, pressure swing adsorption, membrane separation, cryogenic separation, and chemical adsorption.[15,17,18] These processes can produce pipeline quality biomethane as well as a CO_2-rich waste stream that can be captured.

In addition to the biogenic CO_2 waste stream resulting from biogas upgrading, biogenic CO_2 generated during bioconversion of sugars or other intermediates to fuel can be captured for sequestration or possible utilization.[19] The CO_2 captured from fermentation, referred to as a precombustion CO_2 capture system, does not require further purification if the biological conversion process is anaerobic, since the gaseous waste stream is already high-purity (>96% CO_2).[3] In contrast to the precombustion CO_2 capture system, the postcombustion system is used to capture CO_2 from flue gas generated during combustion processes at the biorefinery and is more costly because the CO_2 concentration is much lower (~20%), thus requiring separation prior to sequestration or utilization.[3] Previously published cost estimates for precombustion systems and postcombustion systems are around \$30/t CO_2 and \$70–\$120/t CO_2, respectively.[7,20,21]

A few prior studies have analyzed the GHG mitigation potential and, in some cases, the cost implications, of integrating CCS with bioenergy. Carminati et al. explored the possibility of integrating CCS in sugar cane based-biorefineries and found that it can be economically viable in

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scenarios that include, for example, a carbon tax of $40−80 USD/t CO_2.[22] Sagues et al. investigated the potential for BECCS in the pulp and paper industry, which emits ∼116 million tonnes of biogenic CO_2 each year, and Laude et al. explored CCS integration with sugar beet bioethanol production in Europe.[23,24] Gelfand et al. quantified the potential for net GHG emission reductions (including soil organic carbon sequestration) by integrating BECCS with either biopower generation or ethanol production, both for use in light-duty vehicles, and found that the near-term GHG mitigation potential in these systems could exceed the estimated sequestration potential for reforestation.[25] Currently, five biorefineries across the world are using carbon capture and storage (CCS) technologies with an annual capture of 1.5 million tonne of CO_2 per year, which lags several orders of magnitude behind the IPCC climate change mitigation target and indicates that current economics and incentive structures do not adequately motivate the deployment of CCS.[26] However, the question of whether some combination of biogas upgrading and CO_2 capture is attractive for next-generation cellulosic biofuel facilities has received scant attention.

The main objective of this study is to answer three questions: (1) Is upgrading the biogas coproduct at lignocellulosic biorefineries to RNG advantageous from a cost and GHG standpoint relative to combusting it onsite? (2) What are the cost and emissions impacts of capturing biogenic CO_2 at lignocellulosic biorefineries with and without policy incentives? (3) What is the national significance of the RNG production and carbon sequestration potential at biorefineries?

METHODS AND DATA

In this study, we simulate a base-case lignocellulosic biorefinery using a biomass sorghum feedstock, ionic liquid (cholinium lysinate: [Ch][Lys]) pretreatment, and biological conversion of pentose and hexose sugars to ethanol as the primary product. The base-case biorefinery does not capture any CO_2 and combusts biogas and lignin in a CHP unit to produce process heat and electricity. We then compare the results from the base-case biorefinery against facilities that upgrade biogas to RNG as well as facilities that upgrade biogas to RNG and capture CO_2. We develop a cost and mass/energy balance for each design to evaluate the impacts on minimum ethanol selling price (MESP) and the life-cycle GHG emissions. While the numerical cost and emissions results are specific to the biorefinery configuration we selected for analysis, the goal of this study is to generate insights on the relative advantages of different biogas and CO_2 management strategies that can be generalized across many different biochemical biorefinery configurations, including dilute-acid pretreatment, hot water, and ammonia fiber explosion (AFEX). Additional information about a range of pretreatment methods can be found in other studies.[27,28]

Scenarios. There are four scenarios representing different levels of investment in gas capture and upgrading (see Figure 1): Scenario 1 (S1) is a base-case cellulosic biorefinery where biogas is combusted onsite to generate process heat and power and no CO_2 is captured. Scenario 2 (S2) incorporates biogas upgrading to pipeline-quality RNG for use in place of fossil natural gas with separated CO_2 vented to the atmosphere. Scenario 3 (S3) includes biogas upgrading to RNG and CCS of the separated CO_2 stream from biogas upgrading along with the concentrated CO_2 streams from fermentation. Scenario 4 (S4) includes RNG in addition to full CCS of both pre- and postcombustion CO_2 (streams from biogas upgrading, fermentation, and boiler). Results presented in the main text reflect the use of membrane separation (MS) for CO_2 separation, and we have included results for cryogenic separation (CS) in the Supporting Information (SI).

Biofuel Production Process. Biomass sorghum is used as a representative feedstock across all scenarios because it is a promising bioenergy crop;[29] sorghum also avoids complexities associated with coproduct allocation at the farm level, and its costs are similar to those modeled for other potential bioenergy crops.[30] The average delivered cost of biomass sorghum bales (20% moisture) is estimated at $95.0 per dry tonne.[31] After transporting biomass sorghum to the biorefinery's short-term storage, the feedstock is sent to an integrated high-gravity ionic liquid (IL) pretreatment process in which 0.29 kg of [Ch][Lys] is added per kg of biomass. [Ch][Lys] is chosen due to its compatibility with downstream enzymes and microbes as well as its effectiveness in biomass depolymerization (∼90 wt % glucose and xylose yield after enzymatic hydrolysis).[32,33] The pretreated biomass is transferred to the enzymatic hydrolysis and fermentation section to produce ethanol, which is recovered through a distillation column and dehydrated using molecular sieves. Lignin and other residual solids are sent to the CHP unit for combustion. Wastewater is treated and recycled using AD, an aerobic digester, and a clarifier. The biogas generated in the AD unit is sent to either the onsite combustion section or biogas upgrading section depending on the scenario. Additional details on process conditions and yields are included in SI-Table S1, which are also discussed in more detail in previous studies.[32,33]

Biogas Upgrading Process. Biogas upgrading via MS is a relatively mature technology and is widely used in commercial applications.[34] MS is less energy- and capital-intensive than alternative upgrading technologies such as cryogenic distillation and water scrubbing; however, it demands multiple-stage separation to reach a high purity of CH_4.[34−36] In a single-step MS process, no more than 95% of CH_4 can be recovered.[35] Due to the purity requirement for gas pipeline injection (>96%),[35] multistep gas permeation processes are used in this study (see SI-Figure S2). In this process, untreated biogas leaving AD at a pressure of 0.11 MPa (1.1 bar) is first compressed to 2 MPa (20 bar). The compressed gas is filtered at ambient temperature to remove any liquids before it it conveyed to the membrane separation unit. The retentate, mostly CH_4, can be directly injected into an existing pipeline at 4 MPa (40 bar).[35] In this study, a hollow fiber membrane is used in gas permeation because of its higher effective surface area per unit volume.[35] The selectivity of CO_2/CH_4 (ratio of permeabilities) is assumed to be 15.6 with a membrane cost of $125 per m^2 and membrane life of 5 years, as reported by a private-owned biogas upgrading plant in South Africa.[37] Methane loss on the permeable side is assumed to be 5%.[35] The purity of final RNG is estimated to be 99%.

Carbon Capture and Storage (CCS). Precombustion CO_2 capture only requires gas compression and dehydration (see SI-Figure S3) because of the relatively highly concentrated CO_2 generated from fermentation (∼96% CO_2) and upgrading processes (∼87% CO_2). In the postcombustion CO_2 capture system, amine scrubbing is employed, given its long history in separating CO_2 from other gaseous streams such as natural gas and hydrogen.[38] The absorber requires 30 wt % monoethanolamine (MEA) loading (0.3 kg MEA per kg CO_2 input) of

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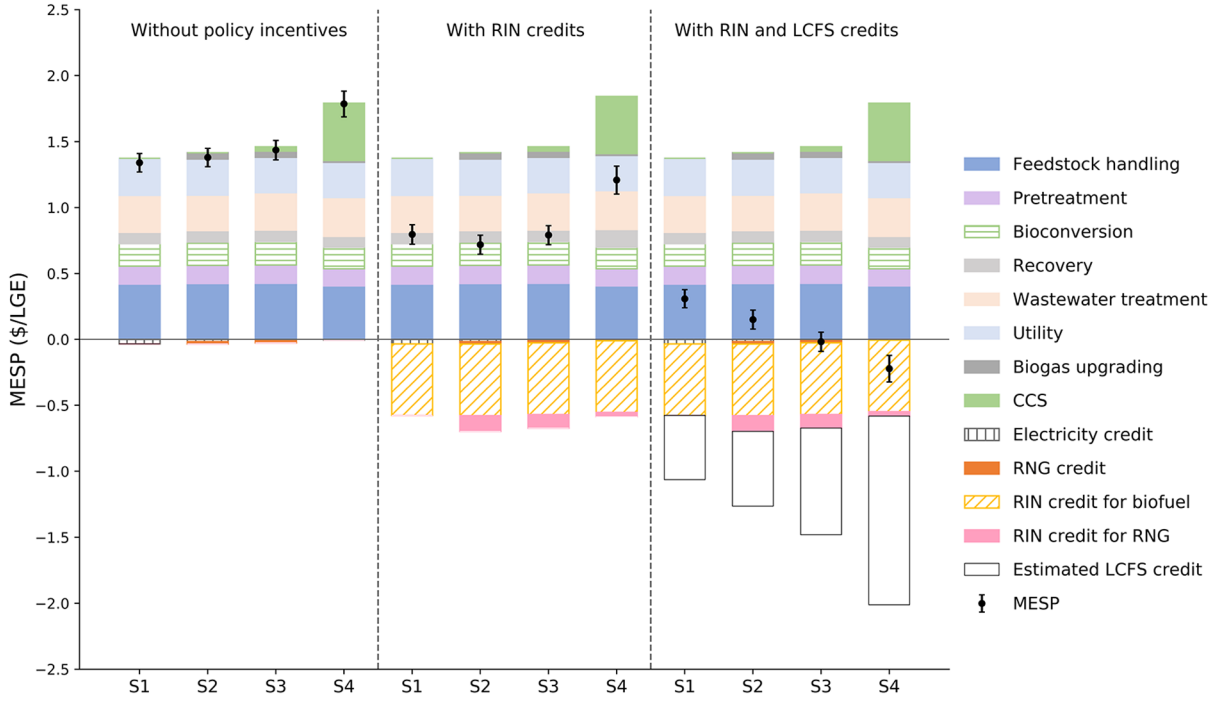

Figure 2. Technoeconomic analysis results of biorefineries combining biogas upgrading and carbon capture and storage (CCS) with and without policy incentives. S1: biorefinery with biogas onsite combustion. S2: integrated biorefinery with biogas upgrading via membrane separation (MS). S3: integrated biorefinery with biogas upgrading via MS and partial CCS (precombustion). S4: integrated biorefinery with biogas upgrading via MS and full CCS (pre- and postcombustion). MESP: minimum ethanol selling price.

which 90% is recycled.[38,39] Afterward, water is condensed, leaving pure CO_2 (99%) stored at 4 MPa (40 bar).[38] Once CO_2 is captured in biorefineries, we assume it will be transported to geological storage sites. The transportation cost has been estimated to be \$12/t CO_2 removed based on a new report published by Lawrence Livermore National Laboratory.[40] Geologic storage cost of CO_2 is around \$8/t CO_2 of net injected.[41] A 90% CO_2 capture rate is investigated in this study. Input process parameters can be found in SI-Table S1. We have not attempted to incorporate CO_2 upgrading to fuels or chemicals in this study. However, a utilization route may be economically and environmentally favorable, depending on the process and target product.[19]

Technoeconomic Analysis. All technoeconomic models are developed in *SuperPro Designer v11*. We assume the biorefinery operates for 8410 h per year and the plant life is 30 years. The capacity of the biorefinery is 2000 dry tonne of biomass sorghum per day. The unutilized biomass, mainly lignin, and biogas generated from the anaerobic treatment of wastewater are sufficient to meet the facility's heat and power demands in every scenario. We assume that untreated biogas produced in the anaerobic digester is used to fulfill the onsite heat and power demand in the biorefinery first, with excess biogas upgraded to RNG. After performing mass and energy balances, the discounted cash flow analysis is conducted using a 10% discount rate. The MESP is reached when the net present value of the project equals zero, holding all other parameters constant. In this study, MESP for each scenario is reported in both costs per liter of gasoline equivalent (\$/LGE) and costs per gallon of gasoline equivalent (\$/GGE), adjusted based on the higher heating value (HHV). To explore the impact of key uncertain parameters, we generated sensitivity bars using baseline, maximum, and minimum values. We also conducted a single-point sensitivity analysis using the minimum and maximum values. Ranges for each input parameter can be found in SI-Table S1. All costs are reported based in 2019 dollars. Additional assumptions are consistent with the landmark National Renewable Energy Laboratory report on a dilute-acid route of converting corn stover to ethanol[3] and previous studies.[31,42]

Life-Cycle Greenhouse Gas Inventory. We use a hybrid process-based/physical units-based input-output model to conduct the life-cycle greenhouse gas inventory for each scenario. This hybrid LCA approach has been widely used in assessing environmental impacts of biorefineries in prior LCA studies.[1,43−45] Background data were generated from various sources including Ecoinvent, GREET, the U.S. LCI database, and peer-reviewed literature and documented in an input-output table. The system boundary includes all stages as described in the *Biofuel production process* section, including upstream emissions from sorghum cultivation, harvesting, and transportation to biorefinery. Mass and energy balances used in the life cycle inventory are obtained directly from the process simulations models developed in *SuperPro Designer*. The carbon footprint of delivered biomass sorghum was calculated based on nutrient inputs (N, P, and K fertilizers), herbicides, and fuel required for biomass harvesting and transportation (SI-Tables S2). We also assume that 1.15% of N applied in fertilizer is released as N_2O as a result of microbial nitrification/denitrification processes in the soil.[46] After the biomass sorghum is harvested, it is dried down in the field, baled, and transported to the biorefinery directly. We assume the transportation distance from field to biorefinery is 64.4 km (40 miles), which is sufficient to collect the biomass sorghum with a yield of 10 tonne per acre and land utilization of 10%. Major data inputs are summarized in the SI-Tables S3−S5.

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We consider the U.S. average grid mix as the source of electricity in this study: even if the final fuel is sold in California to take advantage of LCFS credits, it is likely that facilities relying on biomass sorghum will be located in states with lower-cost agricultural land. Using a California average grid mix would reduce the GHG offset credit for electricity exports, further incentivizing the RNG scenarios. The RNG produced from biogas upgrading is assumed to replace compressed natural gas (CNG) for the purposes of reporting net GHG emissions. However, because RNG sold as a transportation fuel for trucks is considered to offset diesel from the perspective of California's LCFS program, we use a diesel offset credit when calculating LCFS credits. Uncertainty analysis for the life-cycle GHG emissions captures a ± 10% variation in each input parameter, and the impact on net emissions if RNG is credited for offsetting fossil natural gas rather than diesel fuel.[47]

RESULTS AND DISCUSSION

Our analysis explored the relative economic and life-cycle GHG impacts of shifting from a more commonly considered lignocellulosic biorefinery configuration, in which biogas generated during onsite wastewater treatment is combusted for heat and electricity and all CO_2 streams are vented to the atmosphere (referred to as S1), to strategies that arguably have greater GHG emissions reduction potential in the long term. These scenarios include upgrading biogas to RNG (S2), upgrading biogas to RNG with capture and transport of CO_2-rich streams from fermentation and biogas upgrading (S3), and upgrading biogas to RNG with capture and transport of all major CO_2 streams (S4). Each scenario was modeled in detail with *SuperPro Designer* using a representative lignocellulosic biorefinery that converts biomass sorghum to ethanol via IL pretreatment, enzymatic saccharification, and fermentation. We present results with and without policy incentives to show the impact of the RIN values and LCFS credits, which are important drivers of investments in bioenergy production.[47]

Biorefineries with Biogas Upgrading. Figure 2 shows the MESP for each scenario, with and without policy incentives. As shown in Figure 2, MESP in S1 (biogas onsite combustion) is \$1.34/LGE (\$5.08/GGE) and in S2 (biogas upgrading to RNG), MESP increases to \$1.38/LGE (\$5.23/GGE). Without any policy intervention, there is a relatively small difference in the MESP between the base case in which all biogas is combusted onsite (S1) and the scenario where excess biogas is upgraded to RNG and injected into pipelines (S2). In S2, ∼65% of the biogas must be combusted onsite to generate steam needed for the facility, leaving only 35% for upgrading and sale into the market as RNG. Given the expected increases in renewable power generation through 2050 and resulting decreases in the carbon intensity and marginal electricity generation costs,[48] this result should be considered conservative and the relative advantage of S2 will likely increase in the long term. However, there are costs and an energy penalty associated with biogas upgrading; this strategy increases total costs by \$6.3 million. Additionally, 0.32 kWh of electricity is required per Nm^3 of biomethane based on our calculations, which is within the previously reported range of 0.25 to 0.43 kWh/Nm^3 reported for MS in previous studies.[18,49] With highly selective membranes, the energy consumption in MS has the potential to be less than 0.22 kWh/Nm^3.[18] The annual revenue from biomethane sales in S2, assuming at a natural gas commodity price of \$0.11/Nm^3 (\$3/MMBTU),[50] is ∼\$2.6 million. Summing amortized capital expenditures (CAPEX) and operating cost (OPEX), the upgrading cost of biomethane for S2 is calculated to be \$0.18/Nm^3. This cost is largely dependent on IL cost, feedstock supply cost, IL recovery rate, and the methane loss, which alter the resulting MESP. If methane loss increases from 5 to 20%, the resulting MESP increases from \$1.38/LGE to \$1.44/LGE for S2 (SI-Figure S1). Other studies reported a higher production cost for pressure swing adsorption, water scrubbing, and physical scrubbing than MS.[35,51] Ji et al. further suggested that by adopting an energy- and cost-effective ionic liquid technology, the CAPEX could decrease by 10% relative to other processes, including MS considered in this study.[51]

If the MS biogas upgrading system is combined with CCS, the results indicate that full CCS (S4) leads to a much higher MESP than the precombustion CCS scenario (S3). S3, where only concentrated CO_2 is captured, can be implemented for a relatively modest increase in MESP (\$1.44/LGE or \$5.44/GGE), while the pre- and postcombustion CCS system (S4) results in an MESP of \$1.79/LGE (\$6.77/GGE), as shown in Figure 2. The full CCS system (S4) containing both pre- and postcombustion carbon capture is capital-intensive, accounting for ∼\$0.43/LGE compared to partial CCS (S3) containing only precombustion system of ∼\$0.03/LGE. In S4, ∼90% of the untreated biogas needs to be combusted onsite to fulfill steam demand of the facility, leaving 10% for upgrading to RNG. The amount of CO_2 captured from fermentation, biogas upgrading, and the boiler is about 17, 1.9, and 65 t/h, respectively. Postcombustion carbon capture requires larger upfront investments relative to precombustion or oxy-fuel combustion systems due to the large quantity of the lean-CO_2 mixture, which requires large-scale process equipment.[45] The carbon capture costs with pre- and postcombustion CCS are about \$22/t CO_2 and \$63/t CO_2, respectively. For comparison, typical carbon capture costs estimated for fermentation off-gas or precombustion systems are around \$30/t CO_2; in the postcombustion scenario, this cost could be in the range of \$60−90/t CO_2 for large-scale industries.[7,52−54] Although this study does not consider possible utilization of captured CO_2, a new report released by the California Energy Commission demonstrated that conversion of CO_2 removed from RNG into dimethyl ether could increase the competitiveness of RNG in the marketplace, depending on the hydrogen feed price.[55]

Cost of Carbon Mitigation and Impact of Policy Incentives. If the primary goal of these biorefineries is to mitigate GHG emissions, it is possible to determine which scenario is most cost-effective on a per tonne of CO_{2e} basis. This cost of carbon mitigation calculation is dependent on the assumed selling price for cellulosic ethanol, so we include two scenarios: (1) an MSEP equal to the target fuel selling price of \$1.00/LGE (\$2.50/gal ethanol), as set by the U.S. DOE,[56] and (2) an MESP equivalent to the 1940−2020 historical average U.S. gasoline rack sales price of \$0.61/LGE (\$1.53/gal ethanol). If ethanol sells for \$1.00/LGE (\$2.50/gal ethanol), the mitigation costs per tonne CO_{2e} avoided are \$67 (for S1), \$64 (for S2), \$53 (for S3), and \$52 (for S4) (see SI-Figure S6). If cellulosic ethanol sells for the historical average gasoline rack price, the GHG mitigation costs for S1 through S4 are \$143, \$131, \$99, and \$78/t CO_{2e}, respectively. The results indicate that the biorefineries with pre- and postcombustion CCS are most cost-effective at mitigating GHGs. These costs are within the Interagency Working Group's established range

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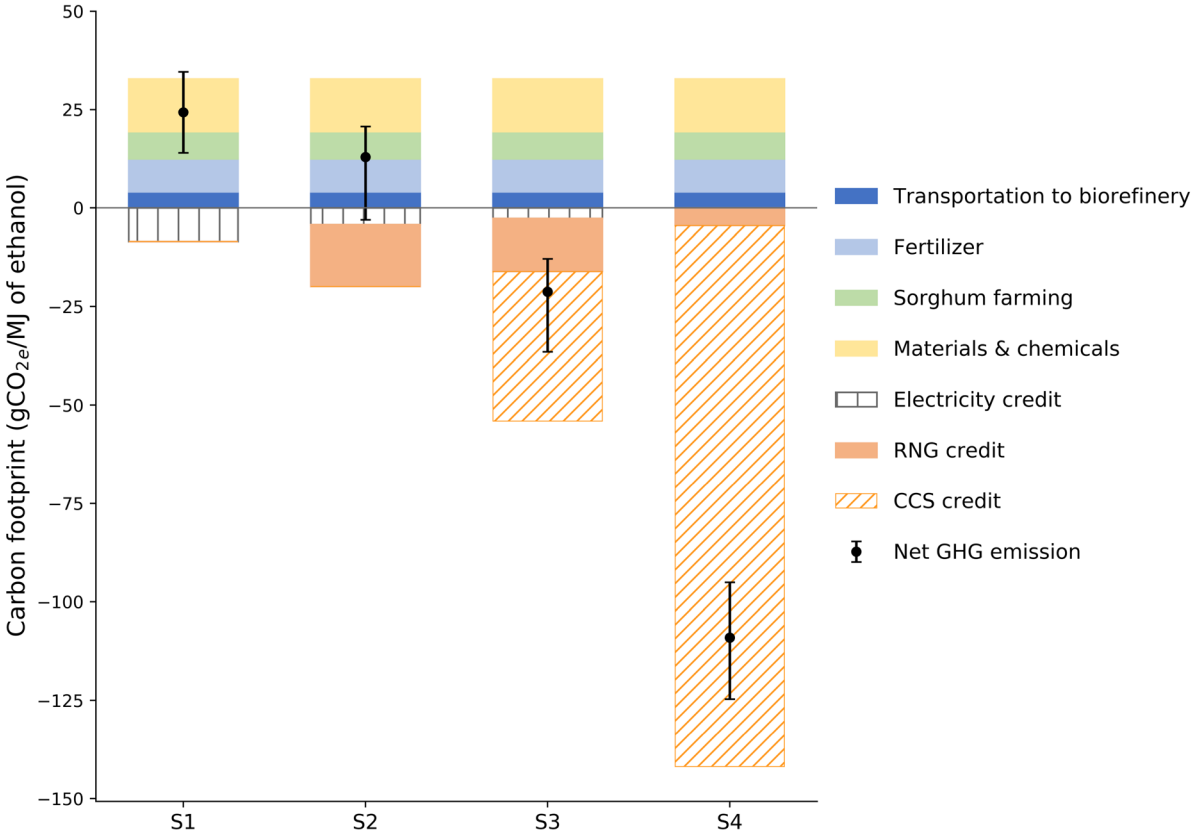

Figure 3. Life-cycle greenhouse gas (GHG) emissions for different scenarios. S1: biogas onsite combustion. S2: integrated biorefinery with biogas upgrading via membrane separation (MS). S3: integrated biorefinery with biogas upgrading via MS and partial CCS. S4: integrated biorefinery with biogas upgrading via MS and full CCS. Uncertainty bars capture variation of ±10% of input parameters. Uncertainty bars for S2−S4 also include variation in the biomethane offset credit.

for the social cost of CO_{2e}, which they estimated at an average value of \$42/t CO_{2e} and a maximum of CO_{2e} of \$123/t in 2020 assuming a discount rate of 3.0%.[57]

Another approach to assessing the relative merits of these strategies is to update each MESP with the estimated value of policy incentives. Although policy-based economic incentives are outside the control of researchers and biorefinery operators, they are important drivers in industry decision-making. RINs and LCFS credits are the two most relevant sources of economic incentives in this case; RINs can be applied to both the ethanol and coproduced RNG (if the RNG is sold for use as a transportation fuel) and LCFS credits can be applied to ethanol as a replacement for gasoline in light-duty vehicles and biomethane as a substitute for diesel fuel in trucks. Both ethanol and RNG produced from cellulosic biomass generate D3 RINs. After RIN values are incorporated into our analysis (see Figure 2), the base case (S1) MESP of \$1.34/LGE (\$5.08/GGE) is reduced to \$0.80/LGE (\$3.02/GGE). The inclusion of RIN credits for ethanol and RNG in S2 results in an even more substantial drop in MESP, from \$1.38/LGE (\$5.23/GGE) to \$0.72/LGE (\$2.72/GGE). We find that the fluctuation of RIN price in past years has an important impact on the MESP. With the lowest RIN price (\$0.47 per RIN),[58] the MESP for S2 becomes \$0.79/LGE (\$3.06/GGE). However, the MESP could drop to \$0.58/LGE (\$2.18/GGE) in S2 with the highest historical price of \$2.96 per RIN.[58]

Biofuels in California can generate LCFS credits, in addition to RINs, if the biofuel can achieve a lower life-cycle carbon intensity relative to the petroleum-based fuel being replaced. Biomass sorghum-based ethanol has the potential to reduce GHG emissions by ∼70% relative to gasoline (93 gCO_{2e}/MJ), as shown in Figure 3. This figure does not include indirect land use change (iLUC), which has not yet been quantified as part of LCFS for biomass sorghum and remains uncertain (as is true for other dedicated biomass crops, such as switchgrass, Miscanthus, and energy cane). The coupling of both LCFS (assuming no iLUC emissions) and RIN credits reduces the MESP to \$0.31/LGE (\$1.17/GGE) for S1 and \$0.15/LGE (\$0.57/GGE) for S2.

If only RIN values are included, S2 offers the lowest MESP. The RIN credits for ethanol and RNG are not impacted by the inclusion of CCS because the resulting emissions reduction does not alter their code (D3). However, full CCS becomes economically preferable once LCFS credits are introduced, because the value of GHG mitigation exceeds the theoretical cost of CCS (Figure 2). In the partial CCS scenario (S3), ∼23 t CO_2/h are sequestered at the facility and the net GHG emissions are negative (−21.3 gCO_{2e}/MJ), resulting in annual LCFS credits worth ∼\$115 million. For the full CCS scenario (S4), each facility captures ∼83 t CO_2/h and the net GHG emissions are estimated to be −109 gCO_{2e}/MJ of ethanol, earning LCFS credits worth ∼\$203 million annually, which reduced the MESP by ∼\$1.43/LGE (\$5.42/GGE). Similar to California, the state of Oregon has also implemented a clean

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fuels program (CFP) aiming to lower the transportation-related carbon intensity.[59] Average CFP credit ranged from ~$127 to $165/t CO_2 in 2019 with an annual average credit of $148/t CO_2.[60] The calculated MESPs under Oregon's CFP (using the average credit) are around $0.19/LGE and $0.15/LGE for partial and full CCS scenarios, respectively. These results indicate that biorefineries with biogas upgrading and CCS systems could be cost-competitive with petroleum refineries with current policy incentives.

Life-Cycle Greenhouse Gas Emissions. Net GHG emissions results for each scenario considered in this study are shown in Figure 3. Regardless of the specific scenario, biomass sorghum production and supply are the largest contributors to the overall GHG emissions resulting in ~19 gCO_2/MJ of ethanol. Cai et al. found similar results for the biomass sorghum-based ethanol production system where biomass production is responsible for about 50% of total GHG emissions.[61] Export of excess electricity results in a GHG offset credit of approximately 8.1 gCO_{2e}/MJ for the base case scenario (S1) and ~3.7 gCO_{2e}/MJ for the biogas upgrading scenario (S2). Biogas upgrading to RNG (S2) could help reduce the GHG emission by 15.7 gCO_{2e}/MJ assuming the RNG displaces diesel fuel use for operating medium- or heavy-duty vehicles (Figure 3). Adding CCS results in net negative GHG emissions per unit of ethanol produced. Utilizing only precombustion CCS (S3) does not appreciably increase onsite energy and achieves a net GHG footprint of −21.3 gCO_{2e}/MJ. Using a pre- and postcombustion CCS system (S4) results in a net GHG footprint of −111 gCO_{2e}/MJ. This is consistent with previous reported GHG emission reduction in maize stover-based ethanol vehicle from 20 gCO_{2e}/MJ (without CCS) to −99 gCO_{2e}/MJ (with CCS).[25] As shown in Figure 3, the RNG credit in S4 is considerably smaller than for S3 because onsite energy demand increases and thus less biogas is available for upgrading and export. A clear takeaway from these results is that although using CCS to capture and store concentrated CO_2 streams from fermentation and biogas upgrading can be implemented for modest costs and energy penalties, the magnitude of carbon captured in that case is considerably smaller than what can be captured in postcombustion CCS.

National-Scale Energy and Emissions Impacts. Beyond the question of GHG emissions mitigation potential at a single facility, it is worth exploring the national-scale relevance of such a strategy. Cui et al.[62] developed a scenario based on the retrofitting of existing corn ethanol biorefineries and construction of a limited number of new cellulosic biorefineries across the U.S., relying on current corn stover availability and potential new production of biomass sorghum. They found that among the existing 214 corn-based biorefineries in the U.S., with a maximum of 10% conversion of pastureland and cropland to sorghum field, 82 existing biorefineries (including 36 corn stover-based and 46 sorghum-based biorefineries) could be retrofitted and additional 71 new facilities could be built to accept biomass sorghum as the feedstock to produce cellulosic ethanol.[62] The total increase in annual production in this case would be 17 billion gallons, just over the original RFS 2022 cellulosic biofuel production target and equivalent to 12% of US gasoline consumption. Integrating biogas upgrading and CCS systems in these 117 potential cellulosic biorefineries would result in around 3.5 billion Nm^3 of additional RNG production per year. For context, total natural gas production in the U.S. is 0.87 trillion Nm^3 in 2018 according to the U.S. Energy Information Administration[63] and this is projected to increase to 1.27 trillion Nm^3 by 2050.[48] When these potential cellulosic biorefineries are fully established, ~82 Mt of CO_2 could be avoided annually in the full CCS system and ~22 Mt CO_2 per year if partial CCS system is employed. This CO_2 reduction contributes 0.6−1.9% of the IPCC BECCS goal of 3.6 Gt CO_2 per year by 2050 set by the IPCC.[8] The total CO_2 sequestration potential from this conservative scenario with 117 facilities is limited, but a more aggressive biorefinery build-out strategy could easily double or triple the sequestration potential.

Limitations and Future Work. This study aims to provide some insights into the economics and emissions mitigation potential associated with biogas upgrading and CCS at biorefineries, but a key limitation is the uncertainty in how captured CO_2 will be sequestered. The system boundary for this study ends after CO_2 is transported by pipeline to a potential market or sequestration site, but the manner in which the CO_2 is used/disposed could either increase or decrease system-wide costs and net emissions. Availability of appropriate CO_2 storage reservoirs will vary by location as will the ease and cost of CO_2 pipeline permitting and installation.[54] For instance, Sanchez et al. explored some opportunities for deploying CCS in existing biorefineries and they concluded that a carbon sequestration credit of at least $60/t CO_2 and a large scale CO_2 pipeline network of 6900 km in the U.S. could enable annual sequestration of 30 Mt CO_2.[7] Bui et al. reviewed new carbon capture technologies and reported that chemical looping and ionic-liquid based CCS systems are potentially attractive options.[54] Other future technological improvements not captured in our study may be more efficient biogas upgrading systems; we select MS as a well-understood representative process and RNG as the target product, but there will likely be further improvements that reduce costs, energy demand, and possible produce other value-added products. This study could be used as a reference case for further work aiming to evaluate the costs and environmental impacts of promising technologies in such integrated biorefineries.

Our analysis suggests that even with current technologies, the upgrading of biogas to renewable fuel and implemention of CCS for some or all major CO_2 streams are likely to be advantageous from a climate and cost perspective. Future research that enables more efficient and higher-value utilization of these gaseous streams will enable a more efficient and carbon-negative bioeconomy.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c02816.

Tables of input parameters used for technoeconomic modeling and uncertainty analyses, sensitivity analysis results, input parameters for the life cycle assessment, process flow diagrams, and additional cost and greenhouse gas emissions results (PDF)

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Notes

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