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

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1 **Cost and life-cycle greenhouse gas implications of integrating biogas upgrading and**  
2 **carbon capture technologies in cellulosic biorefineries**

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13  
14  
15 **Abstract (200 words)**

16 Gaseous streams in biorefineries have been undervalued and underutilized. In cellulosic  
17 biorefineries, co-produced biogas is assumed to be combusted alongside lignin to generate  
18 process heat and electricity. Biogas can instead be upgraded to compressed biomethane and used  
19 as a transportation fuel. Capturing CO<sub>2</sub>-rich streams generated in biorefineries can also  
20 contribute to greenhouse gas (GHG) mitigation goals. We explore the economic and life-cycle  
21 GHG impacts of biogas upgrading and CO<sub>2</sub> capture and storage (CCS) at ionic liquid-based  
22 cellulosic ethanol biorefineries using biomass sorghum. Without policy incentives, biorefineries  
23 with biogas upgrading systems can achieve a comparable minimum ethanol selling price (MESP)

24 and reduced GHG footprint (\$1.38/liter gasoline equivalent (LGE) and 12.9 gCO<sub>2e</sub>/MJ) relative  
25 to facilities that combust biogas onsite (\$1.34/LGE and 24.3 gCO<sub>2e</sub>/MJ). Incorporating  
26 renewable identification number (RIN) values advantages facilities that upgrade biogas relative  
27 to other options (MESP of \$0.72/LGE). Incorporating CCS increases the MESP, but dramatically  
28 decreases the GHG footprint (-21.3 gCO<sub>2e</sub>/MJ for partial, -110.7 gCO<sub>2e</sub>/MJ for full CCS). The  
29 addition of CCS also decreases the cost of carbon mitigation to as low as \$52-\$78/t CO<sub>2</sub>,  
30 depending on the assumed fuel selling price, and is the lowest-cost option if both RIN and  
31 California's Low Carbon Fuel Standard credits are incorporated.

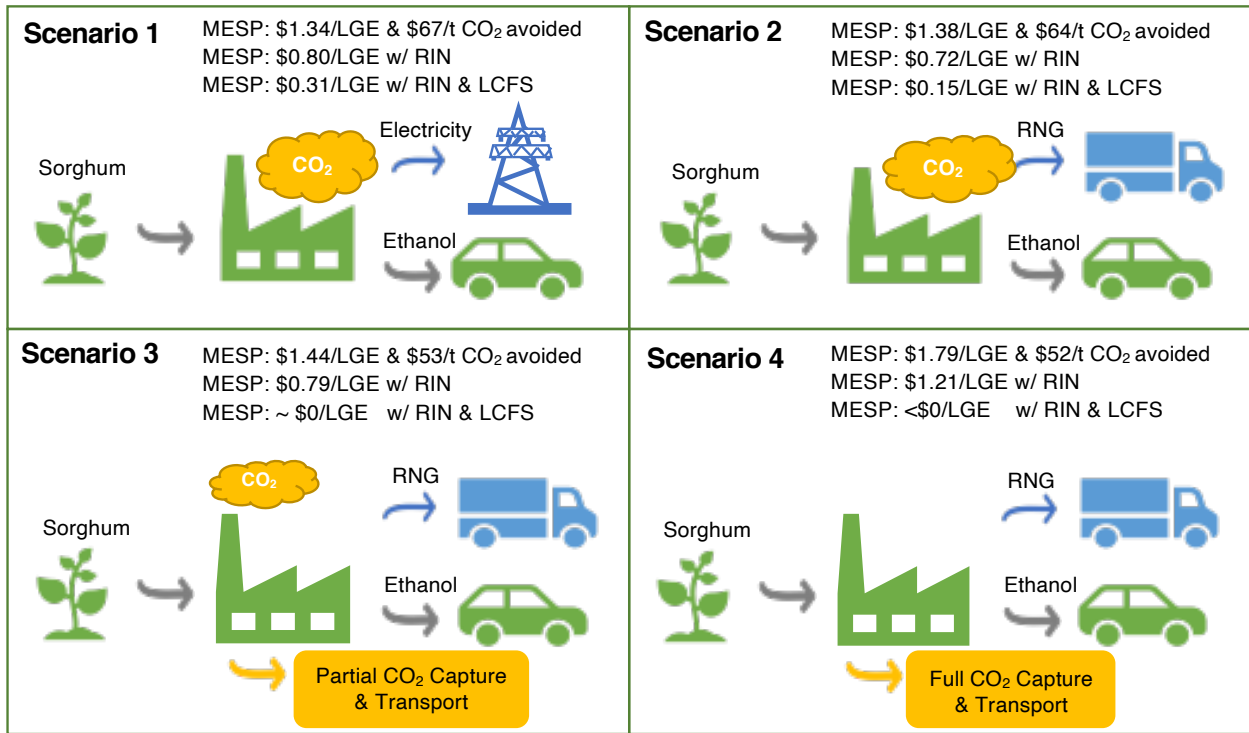
32

### 33 **Keywords**

34 Biogas upgrading, carbon capture and storage (CCS), cellulosic biorefinery, renewable fuel  
35 policy, technoeconomic analysis, life-cycle assessment

36

37



## 41 **Introduction**

42 Cellulosic biofuels have the potential to reduce greenhouse gas (GHG) emissions by around  
43 80% relative to gasoline.<sup>1,2</sup> This is due in part to the heat and electricity generated by combusting  
44 lignin alongside biogas from onsite wastewater treatment, which satisfies the facility's energy  
45 needs and can also result in net power exports to the grid.<sup>3-5</sup> However, these facilities have the  
46 potential to achieve net-negative GHG emissions and contribute to targets for bioenergy with  
47 carbon capture and sequestration (BECCS), which most climate stabilization scenarios rely on to  
48 compensate for difficult-to-decarbonize sectors.<sup>6,7</sup> To meet the target of <2°C of global warming,  
49 the International Panel and Climate Change (IPCC) predicts that 3.6 Gt of biogenic CO<sub>2</sub> annually  
50 must be sequestered via BECCS by 2050.<sup>8</sup> BECCS discussions tend to focus on gaseous streams  
51 from power generation, while studies on capture and utilization of gaseous streams from  
52 advanced biorefineries are limited and tend to focus on microalgae.<sup>9-11</sup> Advanced biorefineries  
53 converting lignocellulosic biomass to fuels result in multiple gaseous streams, the fates of which  
54 have been underexplored. Untreated biogas from the onsite anaerobic digestion (AD) of process  
55 wastewater can be upgraded to biomethane with well-established technologies and injected into  
56 existing natural gas pipelines, used as a feedstock for hydrogen production, or compressed for  
57 use as a transportation fuel (typically referred to as renewable natural gas, or RNG). Biogas  
58 upgrading also results in a concentrated CO<sub>2</sub> waste stream that can be combined with the CO<sub>2</sub>-  
59 rich stream from fermentation and then sequestered or utilized. Additional CO<sub>2</sub> can be captured  
60 from the flue gas in the combined heat and power (CHP) unit. This study focuses on quantifying  
61 the economic and GHG implications of variations on biogas upgrading and CO<sub>2</sub> capture  
62 strategies at ethanol-producing cellulosic biorefineries, including the value of potential policy  
63 incentives.

64 There are three gaseous streams of interest in cellulosic biorefineries using biological  
65 conversion: the biogas produced during onsite wastewater treatment, the CO<sub>2</sub>-rich waste stream  
66 from fermentation, and flue gas produced during combustion of lignin and other residual solids.  
67 The common assumption that biogas will be combusted onsite for heat and electricity<sup>3,12</sup> is likely  
68 based on outdated market conditions. Competition from wind, solar, and natural gas-fired power  
69 plants on the grid, along with economic incentives for renewable transportation fuels, has made  
70 upgrading biogas to RNG increasingly attractive.<sup>13,14</sup> Untreated biogas produced from AD  
71 consists of a roughly 50/50 mixture of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>) with small  
72 amounts of impurities including hydrogen sulfide, carbon monoxide, oxygen (O<sub>2</sub>) and nitrogen.<sup>15</sup>  
73 In order to inject biomethane into existing pipelines, the quality of biomethane needs to meet  
74 certain standards. Pacific Gas and Electricity (PG&E), one of the largest electric and gas utilities  
75 based in California, requires the gas to have less than 1% CO<sub>2</sub> and 0.1% O<sub>2</sub>.<sup>16</sup> Numerous  
76 technologies have been explored for biogas upgrading, such as pressurized water scrubbing,  
77 pressure swing adsorption, membrane separation, cryogenic separation, and chemical  
78 adsorption.<sup>15,17,18</sup> These processes can produce pipeline quality biomethane, as well as a CO<sub>2</sub>-rich  
79 waste stream that can be captured.

80 In addition to the biogenic CO<sub>2</sub> waste stream resulting from biogas upgrading, biogenic CO<sub>2</sub>  
81 generated during bioconversion of sugars or other intermediates to fuel can be captured for  
82 sequestration or possible utilization.<sup>19</sup> Capturing CO<sub>2</sub> from fermentation, referred to as a pre-  
83 combustion CO<sub>2</sub> capture system, does not require further purification if the biological conversion  
84 process is anaerobic, since the gaseous waste stream is already high-purity (>96% CO<sub>2</sub>).<sup>3</sup> In  
85 contrast to the pre-combustion CO<sub>2</sub> capture system, the post-combustion system is used to  
86 capture CO<sub>2</sub> from flue gas generated during combustion processes at the biorefinery and is more

87 costly because the CO<sub>2</sub> concentration is much lower (~20%), thus requiring separation prior to  
88 sequestration or utilization.<sup>3</sup> Previously published cost estimates for pre-combustion systems and  
89 post-combustion systems are around \$30/t CO<sub>2</sub> and \$70 - \$120/t CO<sub>2</sub>, respectively.<sup>7,20,21</sup>

90 A few prior studies have analyzed the GHG mitigation potential, and in some cases cost  
91 implications, of integrating CCS with bioenergy. Carminati et al. explored the possibility of  
92 integrating CCS in sugarcane based-biorefineries and found that it can be economically viable in  
93 scenarios that include, for example, a carbon tax of \$40–80 USD/t CO<sub>2</sub>.<sup>22</sup> Sagues et al.  
94 investigated the potential for BECCS in the pulp and paper industry, which emits ~116 million  
95 tonnes of biogenic CO<sub>2</sub> each year, and Laude et al. explored CCS integration with sugar beet  
96 bioethanol production in Europe.<sup>23,24</sup> Gelfand et al. quantified the potential for net GHG  
97 emission reductions (including soil organic carbon sequestration) by integrating BECCS with  
98 either biopower generation or ethanol production, both for use in light-duty vehicles and found  
99 that the near-term GHG mitigation potential in these systems could exceed the estimated  
100 sequestration potential for reforestation.<sup>25</sup> Currently, five biorefineries across the world are using  
101 carbon capture and storage (CCS) technologies with an annual capture of 1.5 million tonne of  
102 CO<sub>2</sub> per year, which lags several orders of magnitude behind the IPCC climate change mitigation  
103 target and indicates that current economics and incentive structures do not adequately motivate  
104 the deployment of CCS.<sup>26</sup> However, the question of whether some combination of biogas  
105 upgrading and CO<sub>2</sub> capture is attractive for next-generation cellulosic biofuel facilities has  
106 received scant attention.

107 The main objective of this study is to answer three questions: 1) Is upgrading the biogas co-  
108 product at lignocellulosic biorefineries to RNG advantageous from a cost and GHG standpoint  
109 relative to combusting it onsite? 2) What are the cost and emissions impacts of capturing

110 biogenic CO<sub>2</sub> at lignocellulosic biorefineries with and without policy incentives? 3) What is the  
111 national significance of the RNG production and carbon sequestration potential at biorefineries?

112

### 113 **Methods and Data**

114 In this study, we simulate a base-case lignocellulosic biorefinery using a biomass sorghum  
115 feedstock, ionic liquid (cholinium lysinate: [Ch][Lys]) pretreatment, and biological conversion of  
116 pentose and hexose sugars to ethanol as the primary product. The base-case biorefinery does not  
117 capture any CO<sub>2</sub> and combusts biogas and lignin in a CHP unit to produce process heat and  
118 electricity. We then compare the results from the base-case biorefinery against facilities that  
119 upgrade biogas to RNG, as well as facilities that upgrade biogas to RNG and capture CO<sub>2</sub>. We  
120 develop a cost and mass/energy balance for each design to evaluate the impacts on minimum  
121 ethanol selling price (MESP) and the life-cycle GHG emissions. While the numerical cost and  
122 emissions results are specific to the biorefinery configuration we selected for analysis, the goal  
123 of this study is to generate insights on the relative advantages of different biogas and CO<sub>2</sub>  
124 management strategies that can be generalized across many different biochemical biorefinery  
125 configurations, including dilute-acid pretreatment, hot water, and ammonia fiber explosion  
126 (AFEX). Additional information about a range of pretreatment methods can be found in other  
127 studies.<sup>27,28</sup>

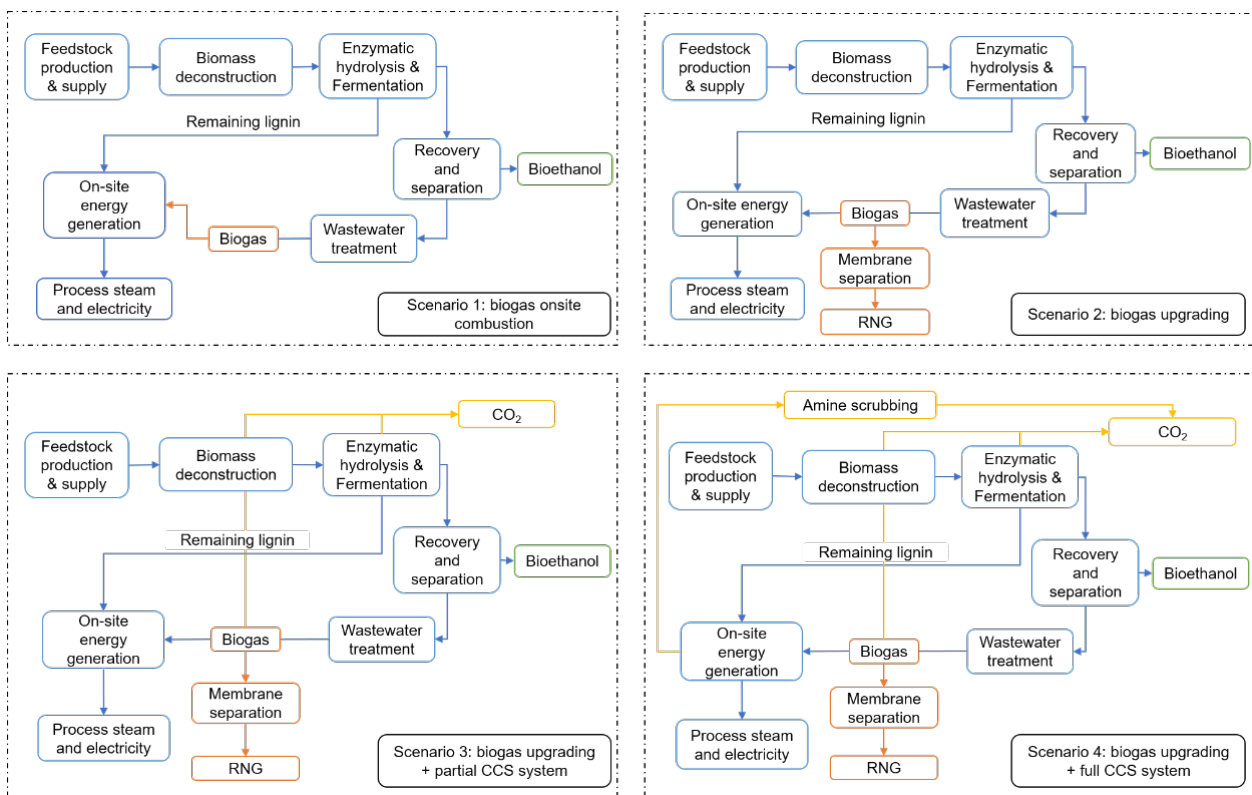
128

### 129 *Scenarios*

130 There are four scenarios representing different levels of investment in gas capture and  
131 upgrading (see Figure 1): Scenario 1 (S1) is a base-case cellulosic biorefinery where biogas is  
132 combusted onsite to generate process heat and power and no CO<sub>2</sub> is captured; Scenario 2 (S2)



133 incorporates biogas upgrading to pipeline-quality RNG for use in place of fossil natural gas with  
 134 separated CO<sub>2</sub> vented to the atmosphere; Scenario 3 (S3) includes biogas upgrading and  
 135 CCS of the separated CO<sub>2</sub> stream from biogas upgrading along with the concentrated CO<sub>2</sub> streams  
 136 from fermentation; Scenario 4 (S4) includes RNG in addition to full CCS of both pre- and post-  
 137 combustion CO<sub>2</sub> (streams from biogas upgrading, fermentation, and boiler). Results presented in  
 138 the main text reflect the use of membrane separation (MS) for CO<sub>2</sub> separation, and we have  
 139 included results for cryogenic separation (CS) in the Supporting Information (SI).



140  
 141 Figure 1. Process flow diagrams of the four scenarios analyzed in this study.

142  
 143 *Biofuel production process*

144 Biomass sorghum is used as a representative feedstock across all scenarios because it is a  
 145 promising bioenergy crop;<sup>29</sup> sorghum also avoids complexities associated with co-product

146 allocation at the farm level, and its costs are similar to those modeled for other potential  
147 bioenergy crops.<sup>30</sup> The average delivered cost of biomass sorghum bales (20% moisture) is  
148 estimated at \$95.0 per dry tonne.<sup>31</sup> After transporting biomass sorghum to the biorefinery's  
149 short-term storage, the feedstock is sent to an integrated high-gravity ionic liquid (IL)  
150 pretreatment process, in which 0.29 kg of [Ch][Lys] is added per kg of biomass. [Ch][Lys] is  
151 chosen due to its compatibility with downstream enzymes and microbes as well as its  
152 effectiveness in biomass depolymerization (~90 wt.% glucose and xylose yield after enzymatic  
153 hydrolysis).<sup>32,33</sup> The pretreated biomass is transferred to the enzymatic hydrolysis and  
154 fermentation section to produce ethanol, which is recovered through a distillation column and  
155 dehydrated using molecular sieves. Lignin and other residual solids are sent to the CHP unit for  
156 combustion. Wastewater is treated and recycled using AD, an aerobic digester, and a clarifier.  
157 The biogas generated in the AD unit is sent to either the onsite combustion section or biogas  
158 upgrading section depending on the scenario. Additional details on process conditions and yields  
159 are included in SI-Table S1, which are also discussed in more detail in previous studies.<sup>32,33</sup>

160

### 161 *Biogas upgrading process*

162 Biogas upgrading via MS is a relatively mature technology and is widely used in commercial  
163 applications.<sup>34</sup> MS is less energy- and capital-intensive than alternative upgrading technologies  
164 such as cryogenic distillation and water scrubbing; however, it demands multiple-stage of  
165 separation to reach a high purity of CH<sub>4</sub>.<sup>34-36</sup> In a single-step MS process, no more than 95% of  
166 CH<sub>4</sub> can be recovered.<sup>35</sup> Due to the purity requirement for gas pipeline injection (>96%),<sup>35</sup> multi-  
167 step gas permeation processes are used in this study (see SI-Figure S2). In this process, untreated  
168 biogas leaving AD at a pressure of 0.11 Mpa (1.1 bar) is first compressed to 2 Mpa (20 bar). The

169 compressed gas is filtered at ambient temperature to remove any liquids before traveling to the  
170 membrane separation unit. The retentate, mostly CH<sub>4</sub>, can be directly injected into an existing  
171 pipeline at 4 Mpa (40 bar).<sup>35</sup> In this study, a hollow fiber membrane is used in gas permeation  
172 because of its higher effective surface area per unit volume.<sup>35</sup> The selectivity of CO<sub>2</sub>/CH<sub>4</sub> (ratio  
173 of permeabilities) is assumed to be 15.6 with a membrane cost of \$125 per m<sup>2</sup> and membrane life  
174 of 5 years, as reported by a private-owned biogas upgrading plant in South Africa.<sup>37</sup> Methane  
175 loss on the permeable side is assumed to be 5%.<sup>35</sup> The purity of final RNG is estimated to be  
176 99%.

177

### 178 *Carbon capture and storage (CCS)*

179 Pre-combustion CO<sub>2</sub> capture only requires gas compression and dehydration (see SI-Figure  
180 S3) because of the relatively highly concentrated CO<sub>2</sub> generated from fermentation (~96% CO<sub>2</sub>)  
181 and upgrading processes (~87% CO<sub>2</sub>). In the post-combustion CO<sub>2</sub> capture system, amine  
182 scrubbing is employed, given its long history in separating CO<sub>2</sub> from other gaseous streams such  
183 as natural gas and hydrogen.<sup>38</sup> The absorber requires 30 wt.% monoethanolamine (MEA) loading  
184 (0.3 kg MEA per kg CO<sub>2</sub> input), of which 90% is recycled.<sup>38,39</sup> Afterwards, water is condensed,  
185 leaving pure CO<sub>2</sub> (99%) stored at 4 Mpa (40 bar).<sup>38</sup> Once CO<sub>2</sub> is captured in biorefineries, we  
186 assume it will be transported to geological storage sites. The transportation cost has been  
187 estimated to be \$12/t CO<sub>2</sub> removed based on a new report published by Lawrence Livermore  
188 National Laboratory.<sup>40</sup> Geologic storage cost of CO<sub>2</sub> is around \$8/t CO<sub>2</sub> of net injected.<sup>41</sup> A 90%  
189 CO<sub>2</sub> capture rate is investigated in this study. Input process parameters can be found in SI-Table  
190 S1. We have not attempted to incorporate CO<sub>2</sub> upgrading to fuels or chemicals in this study.

191 However, a utilization route may be economically and environmentally favorable, depending on  
192 the process and target product.<sup>19</sup>

193

#### 194 *Technoeconomic analysis*

195 All technoeconomic models are developed in *SuperPro Designer v11*. We assume the  
196 biorefinery operates for 8,410 hours per year and the plant life is 30 years. The capacity of the  
197 biorefinery is 2,000 dry tonne of biomass sorghum per day. The unutilized biomass, mainly  
198 lignin, and biogas generated from the anaerobic treatment of wastewater are sufficient to meet  
199 the facility's heat and power demands in every scenario. We assume that untreated biogas  
200 produced in the anaerobic digester is used to fulfill the onsite heat and power demand in the  
201 biorefinery first, with excess biogas upgraded to RNG. After performing mass and energy  
202 balances, the discounted cash flow analysis is conducted using a 10% discount rate. The MESP  
203 is reached when the net present value of the project equals zero, holding all other parameters  
204 constant. In this study, MESP for each scenario is reported in both costs per liter of gasoline  
205 equivalent (\$/LGE) and costs per gallon of gasoline equivalent (\$/GGE), adjusted based on the  
206 higher heating value (HHV). To explore the impact of key uncertain parameters, we generated  
207 sensitivity bars using baseline, maximum, and minimum values. We also conducted a single-  
208 point sensitivity analysis using the minimum and maximum values. Ranges for each input  
209 parameter can be found in SI-Table S1. All costs are reported based in 2019 dollars. Additional  
210 assumptions are consistent with the landmark National Renewable Energy Laboratory report on a  
211 dilute-acid route to converting corn stover to ethanol<sup>3</sup> and previous studies.<sup>31,42</sup>

212

213 *Life-cycle greenhouse gas inventory*

214 We use a hybrid process-based/physical units-based input-output model to conduct the life-  
215 cycle greenhouse gas inventory for each scenario. This hybrid LCA approach has been widely  
216 used in assessing environmental impacts of biorefineries in prior LCA studies.<sup>1,43–45</sup> Background  
217 data are generated from various sources including Ecoinvent, GREET, the U.S. LCI database,  
218 and peer-reviewed literature and documented in an input-output table. The system boundary  
219 includes all stages as described in the *Biofuel production process* section, including upstream  
220 emissions from sorghum cultivation, harvesting, and transportation to biorefinery. Mass and  
221 energy balances used in the life cycle inventory are obtained directly from the process  
222 simulations models developed in *SuperPro Designer*. The carbon footprint of delivered biomass  
223 sorghum was calculated based on nutrient inputs (N, P and K fertilizers), herbicides, and fuel  
224 required for biomass harvesting and transportation (SI-Tables S2). We also assume that 1.15% of  
225 N applied in fertilizer is released as N<sub>2</sub>O as a result of microbial nitrification/denitrification  
226 processes in the soil.<sup>46</sup> After the biomass sorghum is harvested, it is dried down in the field,  
227 baled, and transported to the biorefinery directly. We assume the transportation distance from  
228 field to biorefinery is 64.4 km (40 miles), which is sufficient to collect the biomass sorghum with  
229 a yield of 10 tonne per acre and land utilization of 10%. Major data inputs are summarized in the  
230 SI-Tables S3–S5.

231 We consider the U.S. average grid mix as the source of electricity in this study because, even  
232 if the final fuel is sold in California to take advantage of LCFS credits, it is likely that facilities  
233 relying on biomass sorghum will be located in states with lower-cost agricultural land. Using a  
234 California average grid mix would reduce the GHG offset credit for electricity exports, further  
235 incentivizing the RNG scenarios. The RNG produced from biogas upgrading is assumed to

236 replace compressed natural gas (CNG) for the purposes of reporting net GHG emissions.  
237 However, because RNG sold as a transportation fuel for trucks is considered to offset diesel from  
238 the perspective of California's LCFS program, we use a diesel offset credit when calculating  
239 LCFS credits. Uncertainty analysis for the life-cycle GHG emissions captures a +/- 10%  
240 variation in each input parameter and the impact on net emissions if RNG is credited for  
241 offsetting fossil natural gas rather than diesel fuel.<sup>47</sup>

242

## 243 **Results and Discussion**

244 Our analysis explored the relative economic and life-cycle GHG impacts of shifting from a  
245 more commonly considered lignocellulosic biorefinery configuration, in which biogas generated  
246 during onsite wastewater treatment is combusted for heat and electricity and all CO<sub>2</sub> streams are  
247 vented to the atmosphere (referred to as S1), to strategies that arguably have greater GHG  
248 emissions reduction potential in the long term. These scenarios include upgrading biogas to RNG  
249 (S2), upgrading biogas to RNG with capture and transport of CO<sub>2</sub>-rich streams from  
250 fermentation and biogas upgrading (S3), and upgrading biogas to RNG with capture and  
251 transport of all major CO<sub>2</sub> streams (S4). Each scenario was modeled in detail with *SuperPro*  
252 *Designer* using a representative lignocellulosic biorefinery that converts biomass sorghum to  
253 ethanol via IL pretreatment, enzymatic saccharification, and fermentation. We present results  
254 with and without policy incentives to show the impact of the RIN values and LCFS credits,  
255 which are important drivers of investments in bioenergy production.<sup>47</sup>

256

257 *Biorefineries with biogas upgrading*

258 Figure 2 shows the MESP for each scenario, with and without policy incentives. As shown in  
259 Figure 2, MESP in S1 (biogas onsite combustion) is \$1.34/LGE (\$5.08/GGE) and in S2 (biogas  
260 upgrading to RNG), MESP increases to \$1.38/LGE (\$5.23/GGE). Absent any policy  
261 intervention, there is a relatively small difference in the MESP between the base case in which  
262 all biogas is combusted onsite (S1) and the scenario where excess biogas is upgraded to RNG  
263 and injected into pipelines (S2). In S2, ~65% of the biogas must be combusted onsite to generate  
264 steam needed for the facility, leaving only 35% for upgrading and sale into the market as RNG.  
265 Given expected increases in renewable power generation through 2050, and resulting decreases  
266 in the carbon intensity and marginal electricity generation costs<sup>48</sup>, this result should be  
267 considered conservative and the relative advantage of S2 will likely increase in the long term.  
268 However, there are costs and an energy penalty associated with biogas upgrading; this strategy  
269 increases total costs by \$6.3 million. Additionally, 0.32 kWh of electricity is required per Nm<sup>3</sup> of  
270 biomethane based on our calculations, which is within the previously reported range of 0.25 to  
271 0.43 kWh/Nm<sup>3</sup> reported for MS in previous studies.<sup>18,49</sup> With highly selective membranes, the  
272 energy consumption in MS has the potential to be less than 0.22 kWh/Nm<sup>3</sup>.<sup>18</sup> The annual revenue  
273 from biomethane sales in S2, assuming at a natural gas commodity price of \$0.11/Nm<sup>3</sup>  
274 (\$3/MMBTU)<sup>50</sup>, is ~ \$2.6 million. Summing amortized capital expenditures (CAPEX) and  
275 operating cost (OPEX), the upgrading cost of biomethane for S2 is calculated to be \$0.18/Nm<sup>3</sup>.  
276 This cost is largely dependent on IL cost, feedstock supply cost, IL recovery rate, and the  
277 methane loss, which alter the resulting MESP. If methane loss increases from 5 to 20%, the  
278 resulting MESP increases from \$1.38/LGE to \$1.44/LGE for S2 (SI-Figure S1). Other studies  
279 reported a higher production cost for pressure swing adsorption, water scrubbing, and physical

280 scrubbing than MS.<sup>35,51</sup> Ji et al. further suggested that by adopting an energy- and cost-effective  
281 ionic liquid technology, the CAPEX could decrease by 10% relative to other processes, including  
282 MS considered in this study.<sup>51</sup>

283 If the MS biogas upgrading system is combined with CCS, the results indicate that full CCS  
284 (S4) leads to a much higher MESP than the pre-combustion CCS scenario (S3). S3, where only  
285 concentrated CO<sub>2</sub> is captured, can be implemented for a relatively modest increase in MESP  
286 (\$1.44/LGE or \$5.44/GGE), while the pre- and post-combustion CCS system (S4) results in an  
287 MESP of \$1.79/LGE (\$6.77/GGE), as shown in Figure 2. The full CCS system (S4) containing  
288 both pre- and post-combustion carbon capture is capital-intensive, accounting for ~\$0.43/LGE  
289 compared to partial CCS (S3) containing only pre-combustion system of ~\$0.03/LGE. In S4,  
290 ~90% of the untreated biogas needs to be combusted onsite to fulfill steam demand of the  
291 facility, leaving 10% for upgrading to RNG. The amount of CO<sub>2</sub> captured from fermentation,  
292 biogas upgrading, and the boiler is about 17 t/h, 1.9 t/h and 65 t/h, respectively. Post-combustion  
293 carbon capture requires larger upfront investments relative to pre-combustion or oxy-fuel  
294 combustion systems due to the large quantity of the lean-CO<sub>2</sub> mixture, which requires large-scale  
295 process equipment.<sup>45</sup> The carbon capture costs with pre- and post-combustion CCS are about  
296 \$22/t CO<sub>2</sub> and \$63/t CO<sub>2</sub>, respectively. For comparison, typical carbon capture costs estimated  
297 for fermentation off-gas or pre-combustion systems are around \$30/t CO<sub>2</sub>; in the post-  
298 combustion scenario, this cost could be in the range of \$60 - \$90/t CO<sub>2</sub> for large-scale  
299 industries.<sup>7,52-54</sup> Although this study does not consider possible utilization of captured CO<sub>2</sub>, a  
300 new report released by the California Energy Commission demonstrated that converting CO<sub>2</sub>  
301 removed from RNG into dimethyl ether could increase the competitiveness of RNG in the  
302 marketplace, depending on the hydrogen feed price.<sup>55</sup>



303

304 *Cost of carbon mitigation and impact of policy incentives*

305 If the primary goal of these biorefineries is to mitigate GHG emissions, it is possible to  
306 determine which scenario is most cost-effective on a per tonne of CO<sub>2e</sub> basis. This cost of carbon  
307 mitigation calculation is dependent on the assumed selling price for cellulosic ethanol, so we  
308 include two scenarios: (1) an MSEP equal to the target fuel selling price of \$1.00/LGE (\$2.50/gal  
309 ethanol), as set by the U.S. DOE<sup>56</sup>, and (2) an MESP equivalent to the 1940-2020 historical  
310 average U.S. gasoline rack sales price of \$0.61/LGE (\$1.53/gal ethanol). If ethanol sells for  
311 \$1.00/LGE (\$2.50/gal ethanol), the mitigation costs per tonne CO<sub>2e</sub> avoided are \$67 (for S1), \$64  
312 (for S2), \$53 (for S3), and \$52 (for S4) (see SI-Figure S6). If cellulosic ethanol sells for the  
313 historical average gasoline rack price, the GHG mitigation costs for S1 through S4 are \$143,  
314 \$131, \$99, and \$78/t CO<sub>2e</sub>, respectively. The results indicate that the biorefineries with pre- and  
315 post-combustion CCS are most cost-effective at mitigating GHGs. These costs are within the  
316 Interagency Working Group's established range for the social cost of CO<sub>2e</sub>, which they estimated  
317 at an average value of \$42/t CO<sub>2e</sub> and a maximum of CO<sub>2e</sub> of \$123/t in 2020 assuming a discount  
318 rate of 3.0%.<sup>57</sup>

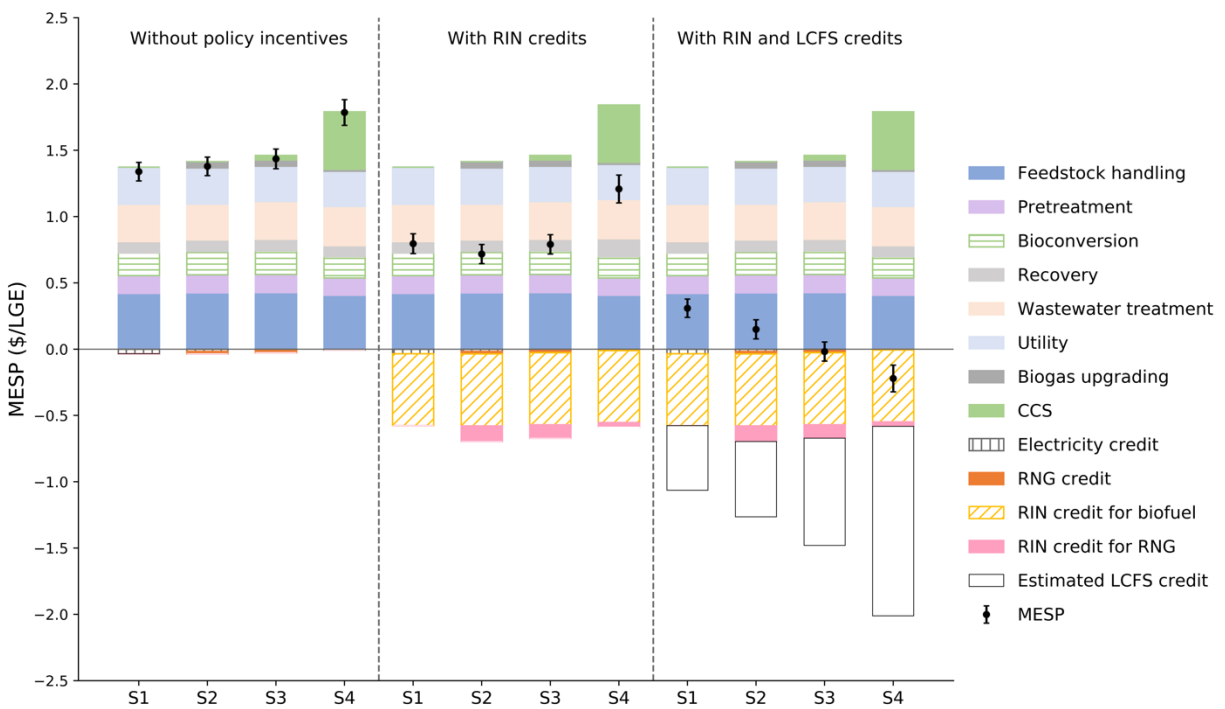
319 Another approach to assessing the relative merits of these strategies is to update each MESP  
320 with the estimated value of policy incentives. Although policy-based economic incentives are  
321 outside the control of researchers and biorefinery operators, they are important drivers in  
322 industry decision-making. RINs and LCFS credits are the two most relevant sources of economic  
323 incentives in this case; RINs can be applied to both the ethanol and co-produced RNG (if the  
324 RNG is sold for use as a transportation fuel) and LCFS credits can be applied to ethanol as a  
325 replacement for gasoline in light-duty vehicles and biomethane as a substitute for diesel fuel in

326 trucks. Both ethanol and RNG produced from cellulosic biomass generate D3 RINs. After RIN  
327 values are incorporated into our analysis (see Figure 2), the base case (S1) MESP of \$1.34/LGE  
328 (\$5.08/GGE) is reduced to \$0.80/LGE (\$3.02/GGE). Including RIN credits for ethanol and RNG  
329 in S2 results in an even more substantial drop in MESP, from \$1.38/LGE (\$5.23/GGE) to  
330 \$0.72/LGE (\$2.72/GGE). We find that the fluctuation of RIN price in past years has an important  
331 impact on the MESP. With the lowest RIN price (\$0.47 per RIN)<sup>58</sup>, the MESP for S2 becomes  
332 \$0.79/LGE (\$3.06/GGE). However, the MESP could drop to \$0.58/LGE (\$2.18/GGE) in S2 with  
333 the highest historical price of \$2.96 per RIN.<sup>58</sup>

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

342 If only RIN values are included, S2 offers the lowest MESP. The RIN credits for ethanol and  
343 RNG are not impacted by the inclusion of CCS because the resulting emissions reduction does  
344 not alter their code (D3). However, full CCS becomes economically preferable once LCFS  
345 credits are introduced, because the value of GHG mitigation exceeds the theoretical cost of CCS  
346 (Figure 2). In the partial CCS scenario (S3), ~23 t CO<sub>2</sub>/h are sequestered at the facility and the  
347 net GHG emissions are negative (-21.3 gCO<sub>2e</sub>/MJ), resulting in annual LCFS credits worth  
348 ~\$115 million. For the full CCS scenario (S4), each facility captures ~83 t CO<sub>2</sub>/h and the net

349 GHG emissions are estimated to be  $-109 \text{ gCO}_2\text{e}/\text{MJ}$  of ethanol, earning LCFS credits worth  
 350  $\sim\$203$  million annually, which reduced the MESP by  $\sim\$1.43/\text{LGE}$  ( $\$5.42/\text{GGE}$ ). Similar to  
 351 California, the state of Oregon has also implemented a clean fuels program (CFP) aiming to  
 352 lower the transportation-related carbon intensity.<sup>59</sup> Average CFP credit ranged from  $\sim\$127$  to  
 353  $\$165/\text{t CO}_2$  in 2019 with an annual average credit of  $\$148/\text{t CO}_2$ .<sup>60</sup> The calculated MESP under  
 354 Oregon's CFP (using the average credit) are around  $\$0.19/\text{LGE}$  and  $\$0.15/\text{LGE}$  for partial and  
 355 full CCS scenarios, respectively. These results indicate that biorefineries with biogas upgrading  
 356 and CCS systems could be cost-competitive with petroleum refineries with current policy  
 357 incentives.



358

359 Figure 2. Technoeconomic analysis results of biorefineries combining biogas upgrading and  
 360 carbon capture and storage (CCS) with and without policy incentives. S1: biorefinery with  
 361 biogas onsite combustion. S2: integrated biorefinery with biogas upgrading via membrane  
 362 separation (MS). S3: integrated biorefinery with biogas upgrading via MS and partial CCS (pre-

363 combustion). S4: integrated biorefinery with biogas upgrading via MS and full CCS (pre- and  
364 post-combustion). MESP: minimum ethanol selling price.

365

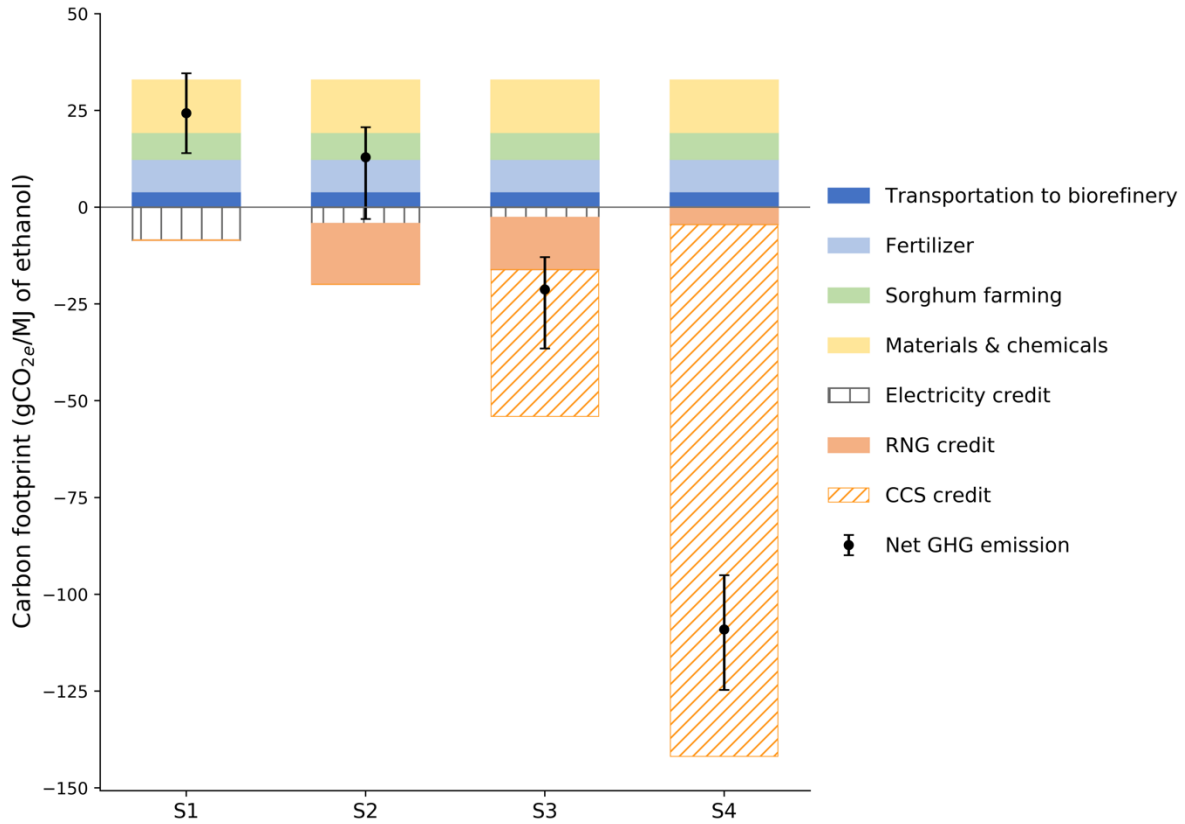
366 *Life-cycle greenhouse gas emissions*

367 Net GHG emissions results for each scenario considered in this study are shown in Figure 3.

368 Regardless of the specific scenario, biomass sorghum production and supply are the largest  
369 contributors to the overall GHG emissions resulting in ~19 gCO<sub>2e</sub>/MJ of ethanol. Cai et al. found  
370 similar results for the biomass sorghum-based ethanol production system where biomass  
371 production is responsible for about 50% of total GHG emissions.<sup>61</sup> Export of excess electricity  
372 results in a GHG offset credit of approximately 8.1 gCO<sub>2e</sub>/MJ for the base case scenario (S1) and  
373 ~3.7 gCO<sub>2e</sub>/MJ for the biogas upgrading scenario (S2). Biogas upgrading to RNG (S2) could  
374 help reduce the GHG emission by 15.7 gCO<sub>2e</sub>/MJ assuming the RNG displaces diesel fuel use  
375 for operating medium- or heavy-duty vehicles (Figure 3). Adding CCS results in net negative  
376 GHG emissions per unit of ethanol produced. Utilizing only pre-combustion CCS (S3) does not  
377 appreciably increase onsite energy and achieves a net GHG footprint of -21.3 gCO<sub>2e</sub>/MJ. Using a  
378 pre- and post-combustion CCS system (S4) results in a net GHG footprint of -111 gCO<sub>2e</sub>/MJ.

379 This is consistent with previous reported GHG emission reduction in maize stover-based ethanol  
380 vehicle from 20 gCO<sub>2e</sub>/MJ (without CCS) to -99 gCO<sub>2e</sub>/MJ (with CCS).<sup>25</sup> As shown in Figure 3,  
381 the RNG credit in S4 is considerably smaller than for S3 because onsite energy demand increases  
382 and thus less biogas is available for upgrading and export. A clear takeaway from these results is  
383 that, although using CCS to capture and store concentrated CO<sub>2</sub> streams from fermentation and  
384 biogas upgrading can be implemented for modest costs and energy penalties, the magnitude of

385 carbon captured in that case is considerably smaller than what can be captured in post-  
386 combustion CCS.



387

388 Figure 3. Life-cycle greenhouse gas (GHG) emissions for different scenarios. S1: biogas onsite  
389 combustion. S2: integrated biorefinery with biogas upgrading via membrane separation (MS).  
390 S3: integrated biorefinery with biogas upgrading via MS and partial CCS. S4: integrated  
391 biorefinery with biogas upgrading via MS and full CCS. Uncertainty bars capture variation of  $\pm$   
392 10% of input parameters. Uncertainty bars for S2-S4 also include variation in the biomethane  
393 offset credit.

394

396 Beyond the question of GHG emissions mitigation potential at a single facility, it is worth  
397 exploring the national-scale relevance of such a strategy. Cui et al.<sup>62</sup> developed a scenario based  
398 on retrofitting existing corn ethanol biorefineries and constructing a limited number of new  
399 cellulosic biorefineries across the U.S., relying on current corn stover availability and potential  
400 new production of biomass sorghum. They found that, among the existing 214 corn-based  
401 biorefineries in the U.S., with a maximum of 10% conversion of pastureland and cropland to  
402 sorghum field, 82 existing biorefineries (including 36 corn stover-based and 46 sorghum-based  
403 biorefineries) could be retrofitted and additional 71 new facilities could be built to accept  
404 biomass sorghum as the feedstock to produce cellulosic ethanol.<sup>62</sup> The total increase in annual  
405 production in this case would be 17 billion gallons, just over the original RFS 2022 cellulosic  
406 biofuel production target and equivalent to 12% of US gasoline consumption. Integrating biogas  
407 upgrading and CCS systems in these 117 potential cellulosic biorefineries would result in around  
408 3.5 billion Nm<sup>3</sup> of additional RNG production per year. For context, total natural gas production  
409 in the U.S. is 0.87 trillion Nm<sup>3</sup> in 2018 according to the U.S. Energy Information  
410 Administration<sup>63</sup> and this is projected to increase to 1.27 trillion Nm<sup>3</sup> by 2050.<sup>48</sup> When these  
411 potential cellulosic biorefineries are fully established, ~82 Mt of CO<sub>2</sub> could be avoided annually  
412 in the full CCS system and ~22 Mt CO<sub>2</sub> per year if partial CCS system is employed. This CO<sub>2</sub>  
413 reduction contributes 0.6 - 1.9% of the IPCC BECCS goal of 3.6 Gt CO<sub>2</sub> per year by 2050 set by  
414 the IPCC<sup>8</sup>. The total CO<sub>2</sub> sequestration potential from this conservative scenario with 117  
415 facilities is limited, but a more aggressive biorefinery build-out strategy could easily double or  
416 triple the sequestration potential.

418 *Limitations and future work*

419 This study aims to provide some insights into the economics and emissions mitigation  
420 potential associated with biogas upgrading and CCS at biorefineries, but a key limitation is the  
421 uncertainty in how captured CO<sub>2</sub> will be sequestered. The system boundary for this study ends  
422 after CO<sub>2</sub> is transported by pipeline to a potential market or sequestration site, but the manner in  
423 which the CO<sub>2</sub> is used/disposed could either increase or decrease system-wide costs and net  
424 emissions. Availability of appropriate CO<sub>2</sub> storage reservoirs will vary by location, as will the  
425 ease and cost of CO<sub>2</sub> pipeline permitting and installation.<sup>54</sup> For instance, Sanchez et al. explored  
426 some opportunities for deploying CCS in existing biorefineries and they concluded that a carbon  
427 sequestration credit of at least \$60/t CO<sub>2</sub> and a large scale CO<sub>2</sub> pipeline network of 6,900 km in  
428 the U.S. could enable annual sequestration of 30 Mt CO<sub>2</sub>.<sup>7</sup> Bui et al. reviewed new carbon  
429 capture technologies and reported that chemical looping and ionic-liquid based CCS systems are  
430 potentially attractive options.<sup>54</sup> Other future technological improvements not captured in our  
431 study may be more efficient biogas upgrading systems; we select MS as a well-understood  
432 representative process and RNG as the target product, but there will likely be further  
433 improvements that reduce costs, energy demand, and possibly produce other value-added  
434 products. This study could be used as a reference case for further work aiming to evaluate the  
435 costs and environmental impacts of promising technologies in such integrated biorefineries.

436 Our analysis suggests that, even with current technologies, upgrading biogas to renewable  
437 fuel, and implementing CCS for some or all major CO<sub>2</sub> streams is likely to be advantageous from  
438 a climate and cost perspective. Future research that enables more efficient and higher-value  
439 utilization of these gaseous streams will enable a more efficient and carbon-negative  
440 bioeconomy.

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454

455 **Conflict of interest**

456 The authors declare no conflict of interest.

457

458 **Supporting Information**

459 The Supporting Information is available free of charge on the ACS Publications website.  
460 The Supporting Information includes tables of input parameters used for technoeconomic  
461 modeling and uncertainty analyses, sensitivity analysis results, input parameters for the life cycle  
462 assessment, process flow diagrams, and additional cost and greenhouse gas emissions results.

463



465

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