

# Deep CCS: Moving Beyond 90% Carbon Dioxide Capture

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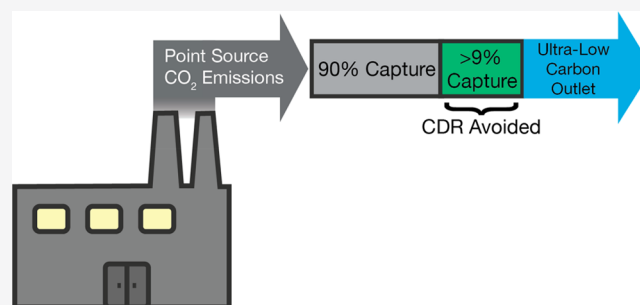
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**ABSTRACT:** The large-scale deployment of carbon capture technologies is expected to play a crucial role in efforts to meet stringent climate targets set forth by the Paris Agreement, but current models rely heavily upon carbon dioxide removal (CDR) strategies for which viability at the gigatonne scale is uncertain. While most 1.5 and 2 °C scenarios project rapid decarbonization of the energy sector facilitated by carbon capture and sequestration (CCS), they generally assume that CCS units can only capture ~90% of the CO<sub>2</sub> in coal and natural gas combustion flues because this was previously considered the optimal condition for aqueous amine scrubbers. In this Perspective, we discuss a small but growing body of literature that examines the prospect of moving significantly beyond 90% capture—a concept we term deep CCS—in light of recent developments in materials and process design. The low incremental costs associated with performing varying degrees of deep CCS suggest that this approach is not only feasible but may also alleviate burdens placed upon CDR techniques facing significant barriers to large-scale deployment. We estimate that rapid deployment of deep CCS in deep decarbonization pathways could avoid more than 1 gigatonne of CO<sub>2</sub> globally each year. The principles of deep CCS could also be applied directly to the CDR strategy of employing bioenergy with CCS, which could lead to a significant alleviation of the land and freshwater burden associated with this technology.

**KEYWORDS:** deep decarbonization, capture rate, carbon dioxide removal



## 1. INTRODUCTION

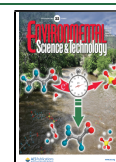
Global anthropogenic CO<sub>2</sub> emissions now exceed 40 Gt/yr,<sup>1</sup> ~75% of which derive from the combustion of fossil fuels.<sup>2</sup> In order to keep average surface temperatures well below 2 °C of preindustrial values, as set forth by the Paris Agreement, integrated assessment model (IAM) scenarios (see [Supporting Information \(SI\) Table S1](#) for a glossary of commonly used terms) have underscored the need for rapid decarbonization of the power generation sector.<sup>3,4</sup> Variable renewable energy technologies are prevalent in these scenarios, although their penetration into the global energy portfolio is still limited by techno-economic and sociopolitical hurdles.<sup>5–12</sup> Principal among these obstacles are the costs associated with the construction of infrastructure for renewable electrical generation and the need to balance inflexible supply with time-varying demand.<sup>13–18</sup> Although many 1.5 and 2 °C IAM scenarios project rapid scale-up of variable renewables by midcentury,<sup>3,19</sup> the growing need to curb anthropogenic CO<sub>2</sub> emissions has motivated the deployment of alternative low-carbon technologies in the near-term.

Carbon capture and sequestration (CCS) is one leading strategy that has been proposed as a means to substantially mitigate CO<sub>2</sub> emissions<sup>20</sup> and refers to a set of technologies that capture CO<sub>2</sub> directly from the flue exhaust of point sources (such as fossil-fueled power plants) and store it underground in geological formations. Currently, the most

commercially mature technology for capturing CO<sub>2</sub> from point sources involves absorbing the CO<sub>2</sub> into aqueous amine solutions.<sup>20</sup> In many cases, existing coal- and natural gas-fired power stations can be retrofitted with carbon capture units, and thus it would not be necessary to overhaul or replace all existing fossil-fuel burning plants to reap the benefits of CCS. Plants that are retrofitted with carbon capture units may also be operated flexibly in response to changes in electrical demand.<sup>18,21–24</sup> While 1.5 and 2 °C IAM scenarios have frequently projected that CCS will be instrumental in minimizing the cost of climate change mitigation,<sup>25</sup> commercial deployment of CCS has not yet reached the scale projected by these scenarios. This disparity is due in part to cost overruns and technical obstacles faced by first-of-a-kind CCS facilities as well as past limited public and private support. Until now, these factors have restricted the deployment of CCS to <50 MtCO<sub>2</sub>/yr globally, with a large portion of these plants performing enhanced oil recovery with the captured CO<sub>2</sub>.<sup>26–32</sup> However, the near tripling of large-scale CCS capacity in

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development between 2017 and 2020 suggests that this technology is now entering a new period where large-scale commercial deployment will soon be feasible.<sup>32</sup>

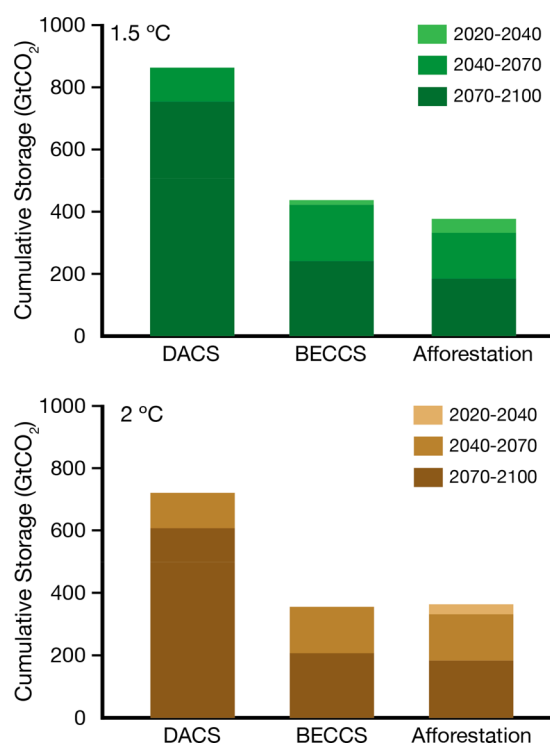
Although CCS can in principle decarbonize a substantial portion of the power generation sector, it is unable to mitigate emissions from disperse sources, such as those derived from the transportation and residential sectors. By contrast, techniques capable of actively removing CO<sub>2</sub> from the air can ameliorate these disperse emissions, and are widely deployed in deep decarbonization pathways.<sup>33–35</sup> These techniques, collectively referred to as carbon dioxide removal (CDR), describe an array of natural and artificial strategies for removing CO<sub>2</sub> from ambient air. Currently, the most commonly explored CDR strategies for large-scale deployment include afforestation/reforestation, bioenergy with CCS (BECCS), and direct air capture with sequestration (DACs), among others.<sup>36–41</sup> Briefly, afforestation describes the process of planting trees on land where there was not recently forestry, whereas reforestation describes the planting of trees on recently deforested land.<sup>42–45</sup> BECCS describes the capture and storage of CO<sub>2</sub> produced from the conversion of biomass or biomass-derived fuels into energy,<sup>46–49</sup> and DACs encompasses the direct removal and storage of CO<sub>2</sub> from ambient air via engineered processes.<sup>50–53</sup> Although these techniques are commonly expected to remove >100 Gt of CO<sub>2</sub> cumulatively by the end of the century (Figure 1),<sup>4,38,54</sup> their feasibility for large-scale deployment remains highly uncertain. For BECCS and afforestation/reforestation, this uncertainty is primarily due to concerns over land and freshwater availability at the levels needed for large-scale CDR and the ecological and ethical risks that arise from the dedication of fertile land for

this purpose.<sup>55</sup> Given these biogeophysical and socioeconomic constraints and the uncertain practical potential of afforestation/reforestation and BECCS, substantial investigation into alternatives, such as DACs, is underway in order to satisfy the global CDR requirement.<sup>37,56</sup> DACs offers some alleviation of the land/water burden because it does not require arable land or nearly as much water as afforestation/reforestation or BECCS. However, commercial DACs operations are currently hampered by substantial costs, primarily arising from the need to regenerate CO<sub>2</sub> from strongly binding capture media.<sup>50,57–59</sup> The direct air capture company Carbon Engineering recently published a conceptual analysis of its pilot plant that estimates an overall levelized cost for DACs in the range \$113–232 per tonne of CO<sub>2</sub> captured, with the lower end of this estimate corresponding to designs for which low-carbon electricity is available at low cost.<sup>60</sup> Although this is a significant improvement upon previous cost estimates for DACs (e.g., \$600/tonne of CO<sub>2</sub>),<sup>52,61</sup> it is still more expensive than the majority of estimates for conventional point-source CCS technologies (e.g., \$36–53/tonneCO<sub>2</sub> for coal CCS and \$48–111/tonneCO<sub>2</sub> for natural gas combined cycle CCS).<sup>62</sup> While there are circumstances under which DACs could become cost-competitive with other decarbonization measures in the near-term,<sup>63,64</sup> most 1.5 and 2 °C IAM scenarios project that fossil CCS will hold a central role in decarbonizing the power generation sector in the next few decades.

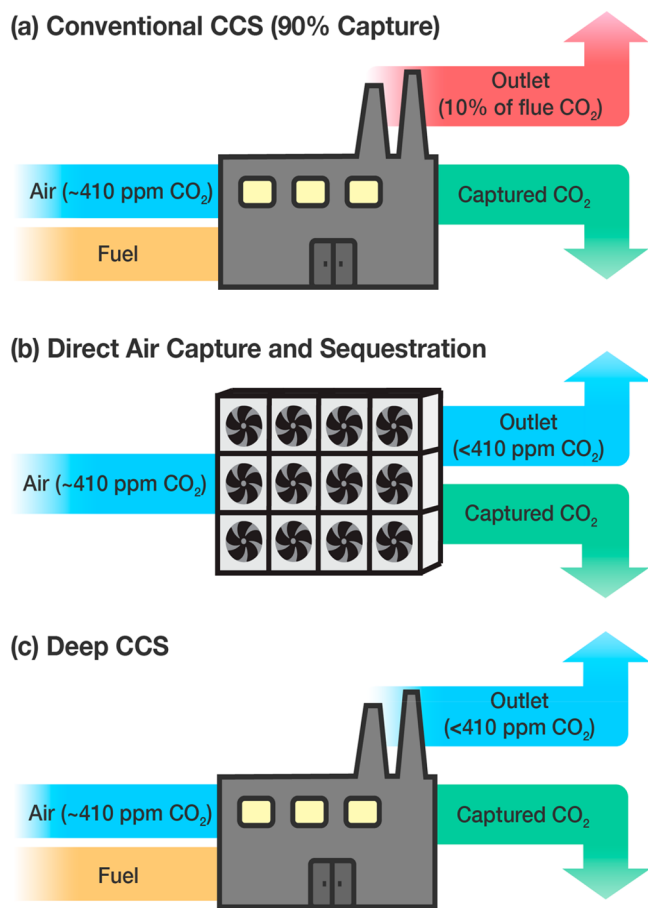
In these scenarios, decarbonization of the power generation sector is typically only partially achieved with CCS. One fundamental reason is that the capture fractions considered in most designs and IAMs are well below the values of ~99% and ~99.7% for natural gas combined cycle (NGCC) and pulverized coal combustion, respectively, that would be needed to achieve outlet CO<sub>2</sub> compositions equal to the ~410 ppm in ambient air (commonly referred to as “carbon-zero” or “carbon neutral” capture fractions). Thus, residual CO<sub>2</sub> emissions still escape fossil fuel-fired units, despite the presence of carbon capture. Although ~90% capture is commonly assumed in designs and models irrespective of plant size or the composition of the flue gas, it is not necessarily the most cost-effective capture fraction for these circumstances (Figure 2a).<sup>69–72</sup> Recent studies<sup>72–75</sup> have demonstrated that pursuing capture significantly beyond 90% in point-source CCS, hereafter referred to as deep CCS, may be associated with only marginal increases in plant capital and operating expense. In this Perspective, we assess the pursuit of such deeper capture targets in light of the global carbon budget and propose that the residual emissions avoided may dramatically alleviate burdens placed upon CDR techniques. We note that our discussion hinges upon the assumption that deep CCS would be implemented only after the successful deployment of conventional CCS. Our hope is that this article will broadly motivate scientists and engineers to consider >90% capture in their investigations of carbon capture materials and process configurations, and that it will encourage the modeling community to move beyond the artificial assumption of 90% in their representations of CCS.

## 2. TECHNOECONOMIC BENEFITS OF DEEP CCS

Deep CCS may offer an inexpensive route toward deep decarbonization of the fossil-fueled energy sector because it requires only minor adjustments to conventional (~90% capture) designs. Although the operating expense per tonne of CO<sub>2</sub> avoided in deep CCS will be larger than in conventional



**Figure 1.** Cumulative carbon dioxide removal for 1.5 °C (upper) and 2 °C (lower) scenarios. Data are from the Grantham Institute TIMES Integrated Assessment Model in ref 54. Until recently, integrated assessment modeling of technological CDR other than BECCS has been uncommon.<sup>63–68</sup>



**Figure 2.** (a) In conventional CCS, typically only  $\sim 90\%$  of the  $\text{CO}_2$  from the power plant exhaust is captured. (b) The outlet  $\text{CO}_2$  composition resulting from DACS must necessarily be less than the inlet composition ( $\sim 410$  ppm of  $\text{CO}_2$ ). (c) With deep CCS, it is possible to achieve an outlet composition of  $< 410$  ppm of  $\text{CO}_2$ . However, note that there may be circumstances where it is more feasible to pursue outlet compositions  $\geq 410$  ppm in deep CCS. Because deep CCS generally describes capture significantly beyond 90%, it can yield carbon positive, carbon negative, or net carbon zero/carbon neutral outlets.

CCS due to the need to overcome incremental kinetic and thermodynamic limitations, the increase in minimum separation work per mole of  $\text{CO}_2$  captured upon moving from 90% to  $\sim 100\%$  capture is relatively small, namely  $\sim 0.48$  and  $\sim 0.57$   $\text{kJ/molCO}_2$  for pulverized coal combustion and NGCC, respectively (all calculations are performed at a temperature of  $25^\circ\text{C}$ ; for more details, see SI Section 2). The energetic palatability of deep CCS arises from the logarithmic dependence of the minimum separation work on the composition of the inlet stream (SI Figure S2), which is  $\sim 11\text{--}15\%$   $\text{CO}_2$  for coal and  $\sim 4\text{--}6\%$  for NGCC flues. By contrast, the energy needed to compress the additional  $\sim 10\%$  of  $\text{CO}_2$  scales linearly with the amount of  $\text{CO}_2$  captured (provided that the purity of the desorbed stream is unchanged upon moving from 90% to  $\sim 100\%$  capture), and is only  $\sim 12$   $\text{kJ/molCO}_2$ . Although entropic irreversibilities prohibit practical attainment of these thermodynamic minima for separation and compression,<sup>76,77</sup> this simple calculation demonstrates that the operating expense for deep CCS may closely approach that for conventional CCS if incremental efficiency losses can be minimized upon exceeding  $\sim 90\%$  capture.

These incremental efficiency losses can indeed be kept small in aqueous amine scrubbers, for which the operating expense is dominated by the reboiler duty for solvent regeneration. Higher capture fractions can be achieved by increasing the interfacial area of the absorber and by modulating the column hydrodynamics with increased liquid:gas ratios, although absorber intercooling can reduce the liquid flow rates needed. Provided that mass transfer limitations are not prohibitive, the rich loading is unlikely to vary substantially with capture fraction, and the reboiler duty will scale with the increased solvent flow.<sup>21,23,78</sup> Rochelle and colleagues recently demonstrated that 99.1% capture from coal combustion flue can be accomplished with only a 20% increase in solvent flow rate and less than 5% increase in the reboiler duty with the advanced flash stripper design, both relative to 90% capture.<sup>74</sup> A recent modeling study from Jiang et al.<sup>73</sup> further demonstrated that the advanced flash stripper design can enable 99.7% capture from coal combustion flue gas with an increase in the cost of  $\text{CO}_2$  avoided by only  $\$2.6/\text{tonne}$  and an increase in the levelized cost of electricity by only  $\$8.1/\text{MWh}$  (both relative to 90% capture). Even without extensive thermal integration of the scrubbing system, capture fractions exceeding 99% can be achieved with minimal added cost, but thermal degradation of the amine solutions in the high-temperature stripper remains a critical concern.<sup>79–81</sup>

Many of the amine solvents that have shown promise for fossil CCS are thermodynamically capable of capturing  $\text{CO}_2$  at compositions well below 410 ppm. However, mass transfer limitations arising from these ultradilute  $\text{CO}_2$  compositions mandate large absorption interfacial areas with associated capital expense, imposing a practical upper limit on the capture fraction. In the same study referenced above from Jiang et al.,<sup>73</sup> the authors found that approximately half of the absorber column is needed to achieve just 3.2% of the overall  $\text{CO}_2$  capture. In scrubbers treating NGCC flue gas, which is more dilute in  $\text{CO}_2$  than coal flue gas, the interfacial areas needed to achieve a 410 ppm outlet may become unfeasibly large and may dominate the incremental capital expense. Considering that many 1.5 and  $2^\circ\text{C}$  IAM scenarios project that NGCC with CCS will occupy a substantial portion of the global energy portfolio in 2100,<sup>19</sup> efforts should be directed toward the development and testing of materials capable of performing low-cost deep CCS from NGCC flues.<sup>82</sup>

The superior diffusional kinetics and high volumetric capacities of porous adsorbents make them excellent candidates in this context, as the overall land footprint of deep CCS units utilizing these materials can be kept fairly small.<sup>20</sup> Modular designs may also be possible, wherein conventional (i.e.,  $\sim 90\%$  capture) contactors can be appended with deeper capture units in a multistage configuration that alleviates some of the financial risk in adopting deep CCS. Indeed, these deeper capture units could be financed by incentives and regulatory frameworks aimed at residual emissions reductions at a later time. Such an approach might enable a reduction in incremental capital and operating expenses, given that smaller contactors could be used for the higher concentration separation upstream and thermodynamic tuning of the stages could be optimized to minimize heat transfer inefficiencies. The immense diversity of amine-functionalized adsorbents renders them particularly well suited for this application, because the capture material (and hence the Gibbs energy of adsorption) can be varied at different stages to achieve the desired capture fraction, assuming kinetic



limitations are not governing.<sup>83–85</sup> In particular, amine-appended metal–organic frameworks exhibiting cooperative adsorption could enable precise control over the adsorption enthalpies in each stage, which may lead to considerable energy savings for the overall unit, especially if direct-contact steam stripping is used for regeneration.<sup>86–90</sup> Research into these adsorbents is only now emerging, and will benefit from the development of new models and technoeconomic analyses to establish realistic cost and energy considerations that will guide their development for deep CCS. For additional discussion of potential sorbent materials and process configurations for deep CCS, see SI Section 3.

Given that deep CCS will produce outlet streams with CO<sub>2</sub> concentrations similar to that in ambient air, the deployment of materials and process configurations in deep CCS may synergistically inform cost reductions for DACS through technological learning.<sup>27,91,92</sup> We anticipate that for suitable coal and NGCC units, the cost of implementing deep CCS will be lower than that for DACS for a number of reasons. For example, unless wind currents can be harnessed effectively,<sup>93</sup> DACS requires pressurization of the inlet air with the aid of large fans operating with low-carbon electricity (Figure 2b). In the ~1 MtCO<sub>2</sub>/yr design from Carbon Engineering, 61 kWh/tonne CO<sub>2</sub> of electricity is needed to support an air velocity of 1.4 m/s in a fan with inlet area of 45 000 m<sup>2</sup>.<sup>60</sup> In contrast, deep CCS does not require sizable fans, because it draws directly from a pressurized flue. Another requirement for direct air capture plants is that they operate at ambient temperature, given the impracticality of heating large quantities of ambient air. Therefore, for a temperature-swing process, large amounts of sensible heat would be required to regenerate the sorbents, which typically have large heat capacities and/or low thermal conductivities. On the other hand, the temperature of a deep CCS inlet can be controlled with direct contact coolers that are already employed commercially. The above-ambient sorption temperatures that could be utilized in deep CCS would require less sensible heat relative to DACS and may also enable a stark reduction in water coadsorption in solids, due to the exponential dependence of relative humidity on temperature.<sup>94,95</sup>

We note that DACS also offers advantages that are otherwise inaccessible with deep CCS. For example, direct air capture can ameliorate legacy emissions from unmitigated fossil fuel combustion. Additionally, DACS operations are inherently more flexible in their geographical placement than CCS technologies, and as such the cost of transporting and storing CO<sub>2</sub> may be lower for DACS than for CCS operations. Additionally, direct air capture does not require pretreatment of the incoming air to protect the capture media from toxic gases (e.g., SO<sub>x</sub> and NO<sub>x</sub>), which can be particularly problematic for coal capture units.<sup>96,97</sup> Although these economic benefits are unlikely to surmount the intrinsic cost differential between DACS and deep CCS, there are scenarios where the large-scale deployment of DACS may be justified in the near-term.<sup>63,64,98</sup> An improved understanding of all factors influencing deployment of DACS relative to deep CCS is direly needed in conjunction with research into materials and process configurations devoted to each approach.

### 3. THE PROSPECT OF NEGATIVE EMISSIONS

Conservation of mass requires that the CO<sub>2</sub> composition of a DACS outlet be less than the inlet air composition. DACS and deep CCS both obey the same thermodynamic limit of 100%

capture, and thus while it is commonly thought that fossil CCS is at best carbon neutral (~410 ppm outlet), there is no fundamental reason why a deep CCS exhaust cannot be carbon negative with respect to the inlet air supply (Figure 2c). However, even for a ~ 0 ppm of CO<sub>2</sub> deep CCS outlet, the negative emissions achieved inside the plant boundaries (which we define as starting at the point of fuel combustion and ending at the exhaust of the CO<sub>2</sub> capture unit) will be small compared to the current and anticipated lifecycle emission rates for the coal and natural gas supply chains in the United States (SI Section 7). As such, deep CCS should not be viewed as a CDR technique, but the pursuit of sub-410 ppm outlet compositions can still offset a portion of the upstream emissions and is warranted if the incremental cost can be kept low. Because the proportion of CO<sub>2</sub> in NGCC flue streams is smaller than that in coal flue gas, it may be more suitable to target these sub-410 ppm levels in NGCC combustors (SI Section 6). With regards to process design, the weak mass-transfer driving forces associated with capturing CO<sub>2</sub> from sub-410 ppm streams will likely require use of capture media with rapid diffusional kinetics (e.g., certain porous adsorbents).<sup>72</sup>

BECCS is one proposed strategy where lifecycle negative emissions can theoretically be achieved, and which may additionally benefit from the pursuit of deeper capture targets. The extent of negative emissions achievable in BECCS is intertwined with direct/indirect land use changes and the carbon footprint of the biomass.<sup>99–106</sup> Achieving deeper capture targets in the BECCS unit could synergistically reduce the amount of biomass that must be processed upstream to achieve a fixed negative emission rate, albeit at some reduction in energy efficiency due to the need to overcome the increased parasitic load.<sup>48,107</sup> Consider the direct firing of European *Miscanthus* as a representative energy-dedicated crop. In order to remove 1 MtCO<sub>2</sub> equivalents per year (MtCO<sub>2</sub>e/yr) on a lifecycle basis, we estimate that achieving a 99.5% capture fraction with deep BECCS requires 3900 fewer hectares of arable land relative to 90% capture (a savings of ~10%), without taking into account land use changes that could further magnify these reductions (see SI Section 4). Additionally, since the freshwater demand for BECCS is expected to be dominated by losses due to evapotranspiration during biomass cultivation, the transition from 90% to 99.5% capture could save more than 15 Mth<sub>2</sub>O/yr, or ~10% of the freshwater needed for 90% capture. Although the additional parasitic load incurred in achieving this deeper capture target would decrease electrical production, this inefficiency is ameliorated in part by the embodied energy saved in consuming less biomass. The incremental parasitic load associated with deep BECCS is expected to decrease over time with the development of new technology, bringing the lifecycle energy efficiency of deep BECCS closer to that for conventional BECCS. Co-generation of bioenergy with a low-carbon fuel, such as natural gas, may close this efficiency gap without substantially reducing the lifecycle carbon negativity.<sup>108,109</sup> Ultimately, the calculations described here are order-of-magnitude estimates obtained using the average multiplicative factors detailed in SI Section 4, and are intended to motivate further research into deep BECCS. Detailed investigations of the technoeconomics and food–energy–water implications of deep BECCS are needed in order to more accurately assess the practical potential of this concept.

#### 4. THE POTENTIAL FOR DEEP IMPACT

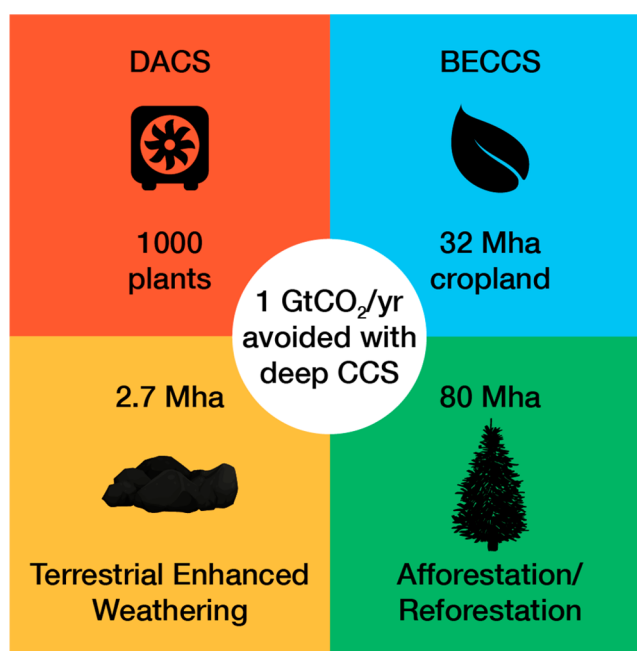
All of the IAM scenarios featured in the 27th Energy Modeling Forum required cumulative sequestration of at least 600 GtCO<sub>2</sub> by 2100 to meet 2 °C warming targets, but to the best of our knowledge, none of these scenarios considered capture fractions significantly beyond 90% during peak deployment, or the period of time during which the maximum number of plants are outfitted with fossil CCS.<sup>20,110–118</sup> Indeed, while 90% capture is not a strict limit in IAMs, it is frequently informed by technoeconomic assessments developed for first-generation CCS designs.<sup>70,71</sup> Given the stringent carbon budgets of 1.5 °C scenarios, the residual emissions from conventional fossil CCS units could necessitate an accelerated transition to renewables or other technologies with lower lifecycle emissions.<sup>19,25,119,120</sup> Alternatively, the rapid development and commercialization of deep CCS may provide a smoother transition toward renewables and, by minimizing residual emissions from the fossil-fueled power generation sector, may obviate a substantial amount of CDR over the century.<sup>35,121</sup>

Working with published results from the IAM MESSAGE GLOBIOM,<sup>122</sup> we estimate that replacing existing fossil CCS units with deep CCS units worldwide could avoid an additional ~1.8 GtCO<sub>2</sub>/yr in the 2 °C scenario at peak deployment. In the 1.5 °C scenario, which projects lower overall deployment of CCS, the CO<sub>2</sub> avoided is reduced to only ~770 Mt/yr (SI Section 5). However, if cost reductions over the next ~40 years can enable the implementation of deep CCS in existing coal and NGCC units for which conventional CCS is currently considered uneconomical, the impact of deep CCS in the 1.5 °C scenario can be significantly augmented. Indeed, if deep CCS is ultimately recognized as a technique for reducing the need for CDR, the number of coal and NGCC plants deemed suitable for CCS may exceed current projections. We emphasize that deep CCS should not be regarded as a replacement for CDR, because there are circumstances where CDR can provide economic, social, and/or environmental benefits that are otherwise inaccessible with deep CCS.<sup>36,53,123–125</sup> However, in some cases, deep CCS has the potential to serve as a viable and cost-effective alternative to replace certain CDR strategies that currently face significant technical or economic hurdles. Note that the above analysis represents at best an order-of-magnitude estimate of the impact of deep CCS in these particular scenarios, and ultimately, the detailed incorporation of deep CCS into IAM frameworks will be needed in order to determine the circumstances under which CDR can truly be avoided by deep CCS.<sup>126</sup>

In an initial evaluation of potential emissions avoided from retrofitting coal and NGCC fleets with deep CCS, we focused on plants in the United States, which in 2017 generated 14.5% of global combustion CO<sub>2</sub> emissions.<sup>2</sup> Working with 2018 data from the Energy Information Administration,<sup>127,128</sup> we identified plants potentially suitable for deep CCS retrofit by categorizing operational coal and NGCC units by electrical production and age. Assuming conservative fuel-based parasitic loads of 13% for NGCC and 21% for coal,<sup>62</sup> we estimate that a combined total of 128 MtCO<sub>2</sub>/yr of emissions can theoretically be avoided. Due to the assumptions made in this analysis (see SI Section 6), this calculation should be regarded as an order-of-magnitude estimate of impact. More accurate assessments can be achieved with plant-specific

considerations and improved parametrizations of CCS technoeconomic models to account for deep capture targets.<sup>22,129</sup>

As discussed above, although deep CCS can achieve negative emissions inside the plant boundaries, these removals are likely to be outweighed by supply chain emissions upstream that are bounded by extraction, transportation, and processing of the fuel. These supply chain emissions include fugitive (unintentional) and vented (intentional) emissions from coal mines, natural gas reservoirs, pipelines, railcars, ships, and process equipment.<sup>130–135</sup> However, the increment between 90 and ~100% capture *can* be carbon negative if supply chain emissions can be kept small. Even if this increment is not carbon negative, it could still alleviate dependences on some of the least viable CDR strategies projected by current 90% model scenarios, so long as the residual emissions captured exceed the supply chain emissions (Figure 3). Working with



**Figure 3.** We estimate that deep CCS can avoid ~1 GtCO<sub>2</sub>/yr of residual emissions globally, which translates to 80 Mha of forest (with a mean carbon uptake of 3.4 tCeq/ha/yr), 2.7 Mha of land for terrestrial enhanced weathering (assuming a potential of 0.1 GtCeq/yr/Mha), 32 Mha of cropland for BECCS with an energy-dedicated crop (assuming a mean carbon uptake of 8.6 tCeq/ha/yr), or 1000 DACS plants (assuming a net CDR rate of 1 MtCO<sub>2</sub>/yr per plant).<sup>56,60,140</sup>

lower bound emission factors of 12.4 kgCO<sub>2</sub>e/MWh (derived in SI Section 7) and 3.6 gCO<sub>2</sub>e/MJ for the coal<sup>136</sup> and natural gas<sup>137</sup> supply chains, respectively, we find that the incremental carbon intensity for a 100 ppm deep CCS outlet is -1.3 MtCO<sub>2</sub>e/yr (coal) and -2.3 MtCO<sub>2</sub>e/yr (NGCC) for the plants considered for retrofit in the United States. In comparison, one of the DACS designs from Carbon Engineering requires 8.81 GJ of natural gas per tonne of CO<sub>2</sub> captured,<sup>60</sup> and our calculations show that the associated supply chain emissions reduce the effective CDR rate from 966 to 931 ktCO<sub>2</sub>/yr, again assuming the emission factor of 3.6 gCO<sub>2</sub>e/MJ from ref 137.<sup>137</sup> Given that the net carbon intensities of deep CCS and DACS are expected to be highly sensitive to upstream emissions, it will be imperative to reduce

these emissions to maximize the overall impact of both technologies.<sup>71,138,139</sup>

## 5. OUTLOOK

Targeting CO<sub>2</sub> capture fractions significantly beyond 90% is technoeconomically feasible and could reduce the need for large amounts of costly or unfeasible CDR if implemented at scale in the near term. The technological learning gained from commercializing deep CCS could synergistically inform designs for DACS and deep BECCS, alleviating some of the uncertainty in accessing large-scale technological CDR. We note that the principles of deep CCS can be extended in some cases (e.g., iron and steel manufacturing) to the industrial sector, which in 2017 was responsible for 24% of global combustion CO<sub>2</sub> emissions.<sup>2,141–146</sup> Additionally, we highly encourage reparametrization of IAMs to more thoroughly investigate the potential of deep CCS/deep BECCS for various socioeconomic and technological assumptions, particularly given that the artificial assumption of ~90% capture may necessitate a shift to suboptimal power generation mixes in deep decarbonization pathways.<sup>25,98</sup> Although materials capable of performing deep CCS already exist, continued improvements in process efficiency are warranted to bring the intrinsic costs of deep CCS even closer to those of conventional CCS, especially as natural gas continues to occupy a larger portion of the global energy portfolio.<sup>82,147</sup> Although the incremental costs associated with performing deep CCS can be kept low relative to the base cost of conventional CCS, widespread deployment of deep CCS will be predicated on the successful implementation of conventional CCS.

### ■ ASSOCIATED CONTENT

#### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.0c07390>.

Computational details and additional information (PDF)

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#### Author Contributions

All authors have given approval to the final version of the manuscript.

#### Notes

The authors declare the following competing financial interest(s): J.R.L. has a financial interest in and serves on the board of directors of Mosaic Materials, a start-up company working to commercialize metal-organic frameworks for gas separations.

#### Biographies



Jeffrey R. Long is a Professor of Chemistry and Chemical & Biomolecular Engineering at the University of California, Berkeley and a Faculty Senior Scientist in the Materials Sciences Division at Lawrence Berkeley National Laboratory. His research involves the synthesis and physical characterization of new materials with potential applications in sustainable energy and environmental remediation, and his 360 publications have received more than 76 000 citations. He cofounded and directs the start-up company Mosaic Materials, which is developing metal-organic frameworks for low-energy carbon dioxide separations, with particular focus on its direct removal from air.



Simon Weston is a Senior Research Associate at the Corporate Strategic Research Laboratories of ExxonMobil Research and Engineering and dedicated to creating new materials for application in molecular separations, storage and catalysis. Although originally trained as an organic chemist, he has a multidisciplinary background in coordination chemistry, inorganic chemistry, high throughput experimentation, informatics, and CO<sub>2</sub> capture research. He has led several research teams bridging industrial and academic work with a focus on how scientific fundamentals can lead us to better materials for a given application.



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