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**Assessing the Relative Climate Impact of Carbon dioxide Utilization for Concrete,  
Chemical and Mineral Production**

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## Abstract

Estimates show that 8.9 gigatons of carbon dioxide (CO<sub>2</sub>) can be captured and utilized to produce chemicals, concrete and minerals by 2050. However, it is difficult to compare the climate benefit across different carbon capture and utilization (CCU) pathways to determine the most effective use of captured CO<sub>2</sub>. This challenge is particularly evident when assessing concrete, chemical and mineralization pathways, which represent three very distinct uses of CO<sub>2</sub>. We present a novel climate return on investment (ROI) metric to rank and prioritize CO<sub>2</sub> utilization across 20 concrete, chemical and mineralization CCU pathways based on the realized climate benefit. The greatest climate benefits are observed for CCU for concrete mixing and carbon monoxide production through dry reforming, and formic acid through the hydrogenation of captured CO<sub>2</sub>. Meanwhile, CCU to produce methane, methanol, kerosene, formic acid through electrolytic reduction of CO<sub>2</sub> and dimethyl carbonate are unlikely to generate a climate benefit. 2 CCU concrete pathways, which use CO<sub>2</sub> during concrete mixing, and 2 CCU chemical pathways, which produce formic acid through hydrogenation of CO<sub>2</sub> and carbon monoxide through dry reforming of methane, generate the greatest climate ROI and are the only pathways with a higher likelihood of generating a climate benefit than a climate burden.

## Introduction

Carbon capture and utilization (CCU) is the capture and use of carbon dioxide (CO<sub>2</sub>) emitted from industrial processes to produce commercially viable products. CCU generates a climate benefit through two pathways. First, by preventing atmospheric carbon emissions, and second, by avoiding the climate impacts associated with manufacturing products through the conventional pathway. The potential climate benefit that can be realized by utilizing 8.9 gigatons of CO<sub>2</sub> by 2050<sup>1</sup> has motivated policies to incentivize CCU.<sup>2,3</sup> Decisions to globally scale CCU operations will require guidance on identifying CCU products which maximize the climate benefit from utilizing CO<sub>2</sub>.

Despite advances in the environmental impact assessment of CCU products (Supplementary Information (SI) Section 1), there exists no robust mechanism to compare the relative climate benefit from manufacturing CCU chemicals, concrete and minerals, which are three pathways offering a significant potential to utilize CO<sub>2</sub>.<sup>4</sup> Existing methods to quantify the climate impact of CCU chemicals are not directly applicable to CCU concrete and minerals. For chemical pathways, a mass based (e.g. kg of CCU chemical produced<sup>5</sup>, CCU chemical produced per kg of H<sub>2</sub><sup>6</sup> or CO<sub>2</sub> used<sup>7</sup>) or volume based<sup>8</sup> or energy content based<sup>9</sup> functional unit is used to compare the climate impact of conventional and CCU products. This is a reasonable approach for chemical pathways, since the produced chemical is the same, regardless of the series of reactions used to generate the chemical. Concrete and minerals have more complex physical properties (e.g. compressive strength) that cannot be adequately represented by a simple mass-based comparison. CCU may increase<sup>10</sup> or decrease<sup>11</sup> the compressive strength of concrete and minerals when compared to the conventional process of generating the product. A direct comparison of CCU and conventionally produced concrete or minerals using a mass or volume-

based functional unit is, therefore, not valid. As a result, the climate impact of chemical, concrete and mineralization pathways are usually assessed separately in the literature (SI Supplementary Figure 1).

In addition, there is a significant uncertainty in the inventory required for CCU products as they are emerging technologies.<sup>12</sup> The uncertainty in the inventory can generate uncertainty in the climate impact of a CCU product.<sup>6,7</sup> Despite recommendations,<sup>12-14</sup> there is a lack of a robust assessment to decrease uncertainty and identifying the key inventory contributors (SI Supplementary Figure 1), which is vital in selecting a particular product for CCU and formulating effective research strategies<sup>15</sup> to improve the climate benefit from CCU products. Given these challenges, there is a lack of an objective method to rank CCU concrete, chemical and mineral product based on the greatest climate benefit per unit of CO<sub>2</sub> utilized. A climate ranking will help prioritize R&D towards CCU pathways with the greatest climate benefit and avoiding pathways that incur a significant climate burden and offer little scope for improvement. Here we develop the climate return on investment (ROI) metric, which is the ratio of the net climate benefit generated per functional unit of the CCU pathway to the quantity of CO<sub>2</sub> utilized per functional unit of the CCU pathway. Climate ROI accounts for the life cycle CO<sub>2</sub> emitted from producing the CCU product and the CO<sub>2</sub> avoided by substituting the conventional process used to generate the product. The climate ROI metric is determined stochastically to account for the uncertainty in the climate impact of a CCU product. We conduct a moment independent sensitivity analysis to identify the climate hotspots and potential strategies to increase the climate ROI of CCU pathways.

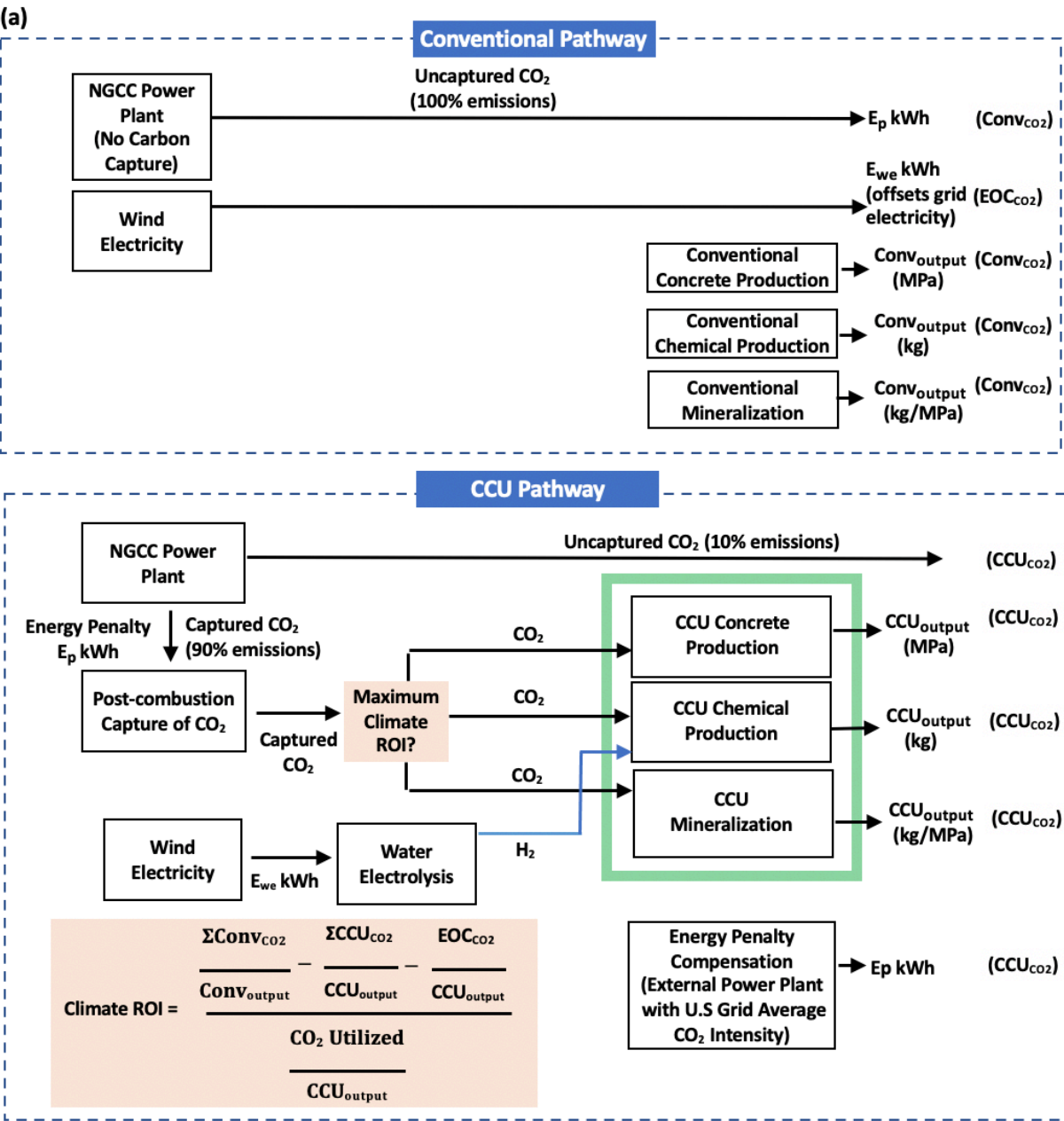
Using the climate ROI metric, we rank 4 concrete, 13 chemical and 3 mineralization pathways. Four CCU pathways involving the production of concrete by mixing the captured CO<sub>2</sub>, carbon

monoxide through dry reforming and formic acid through the hydrogenation of captured CO<sub>2</sub> generate the highest climate benefit. The increased use of ordinary Portland cement and supplementary cementitious materials and the electricity required for CO<sub>2</sub> curing are the key drivers of the climate ROI for CCU concrete. Similarly, the use of electricity and heat are the key drivers of the climate ROI for CCU chemicals.

## **Results**

A climate ROI greater than zero indicates that the life cycle CO<sub>2</sub> intensity of manufacturing a product through the CCU pathway is lower than for the conventional pathway. Therefore, CO<sub>2</sub> is avoided on a life cycle basis and a climate benefit is generated by manufacturing the CCU product. The climate ROI of the 20 CCU products that were examined here accounts for three factors, (i) the life cycle CO<sub>2</sub> emissions from the CCU process used to generate a functional unit of the product, (ii) the life cycle CO<sub>2</sub> avoided when the CCU process substitutes the conventional process used to generate a functional unit of the product and (iii) the quantity of CO<sub>2</sub> utilized per functional unit of the CCU product (Figure 1). The first factor accounts for the energy penalty of capturing the CO<sub>2</sub> from a combined cycle natural gas plant, the hydrogen (H<sub>2</sub>) required to produce chemicals in 7 of the 13 chemical pathways, and the life cycle CO<sub>2</sub> footprint of the material and the energy required to convert the captured CO<sub>2</sub> into the end product. We assume the H<sub>2</sub> is generated through the electrolysis of water using wind electricity as it is environmentally preferable to the conventional process of producing H<sub>2</sub> from natural gas.<sup>16</sup> In this scenario, the generation of H<sub>2</sub> incurs an opportunity cost as the wind electricity can instead be supplied to the grid to offset CO<sub>2</sub> intensive electricity. The CO<sub>2</sub> that can potentially be avoided by supplying wind electricity to the grid is the environmental opportunity cost (EOC) of using wind electricity<sup>17</sup> to generate the H<sub>2</sub> in the 7 CCU chemical pathways. For pathways that

do not require H<sub>2</sub> feedstock to generate the CCU product, the EOC is zero. In addition, EOC gradually reduces to zero when the CO<sub>2</sub> intensity of the grid electricity approaches that of wind or other sources of renewable electricity (e.g., photovoltaics).



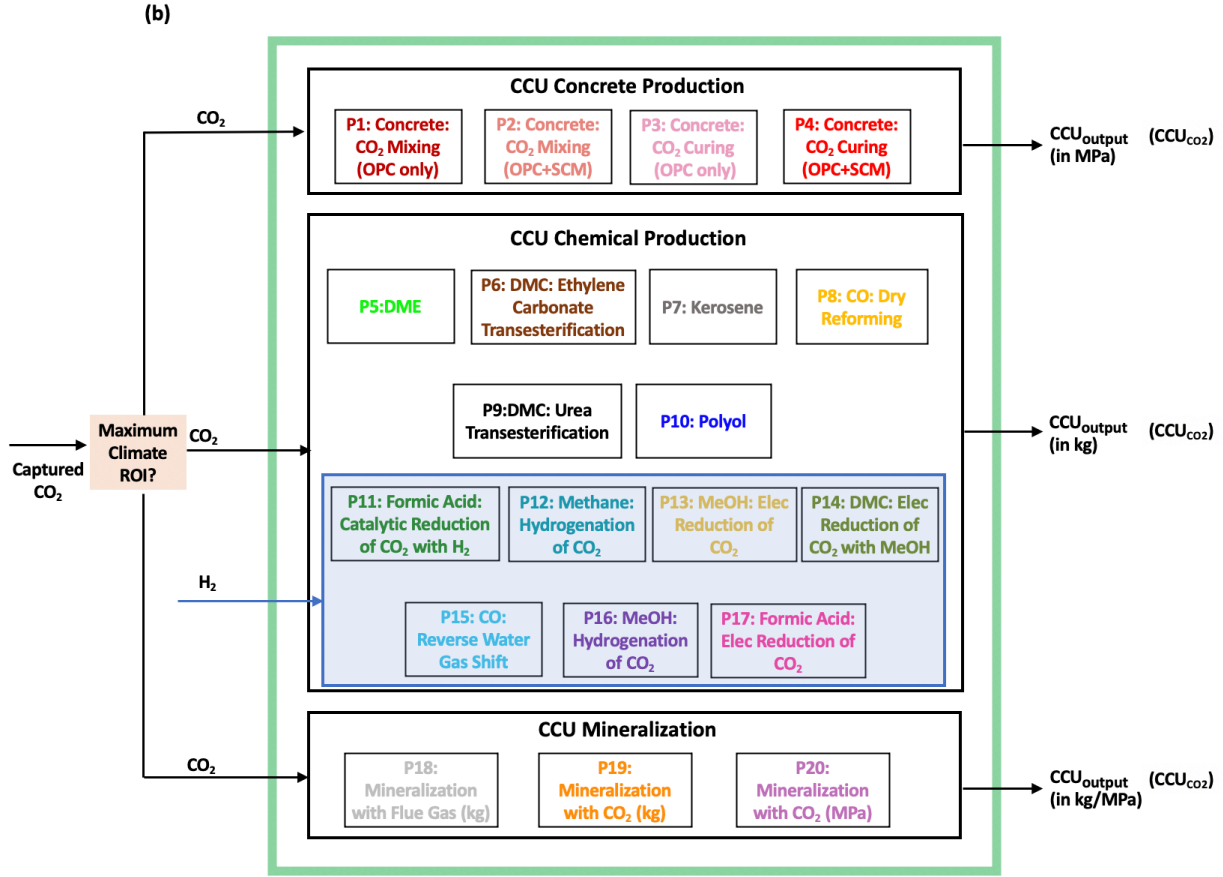


Figure 1 (a) System boundary diagram to determine the climate ROI when CO<sub>2</sub> is utilized across 4 concrete (P1 to P4), 13 chemical (P5 to P17) and 3 mineralization pathways (P18 to P20).  $\Sigma\text{CCU}_{\text{CO}_2}$  and  $\Sigma\text{Conv}_{\text{CO}_2}$  represent the total life cycle CO<sub>2</sub> emitted from producing concrete, chemical and minerals through the CCU and conventional pathways, respectively.  $\Sigma\text{CCU}_{\text{CO}_2}$  and  $\Sigma\text{Conv}_{\text{CO}_2}$  are normalized to the output of the CCU ( $\text{CCU}_{\text{output}}$ ) and the conventional ( $\text{Conv}_{\text{output}}$ ) production pathways.  $\text{CCU}_{\text{output}}$  and  $\text{Conv}_{\text{output}}$  are quantified in compressive strength (MPa) for the concrete pathways, in mass (kg) for the chemical pathways, in mass (kg) for the P18 and P19 mineralization pathways and the compressive strength (MPa) for the P20 mineralization pathway.  $E_p$  is the energy penalty from the post-combustion capture of CO<sub>2</sub>, which is utilized in CCU chemical, concrete and mineralization pathways.  $E_{\text{we}}$  is the wind electricity used to electrolyze water and generate

hydrogen required to produce the chemical through the CCU pathway. The environmental opportunity cost is the  $\text{CO}_2$  avoided ( $\text{EOC}_{\text{CO}_2}$ ) when the wind electricity, which is used for water electrolysis ( $E_{\text{we}}$ ) in the CCU pathway, is instead used to offset grid electricity when chemicals are produced through the conventional pathway. The post combustion MEA capture system captures 90% of the  $\text{CO}_2$  emissions for utilization and the remaining 10% is emitted. (b) Figure 1(b) presents the 4 concrete, 13 chemicals and 3 mineralization CCU pathways corresponding to the green box in Figure 4(a).  $\text{H}_2$  is required to produce chemicals in the blue box.

### **Illustrative Example: Climate ROI for Concrete and Formic Acid**

An illustrative example, shown in Figure 2, is presented to describe the major concepts used in the analysis and characterize the results. The results in Figure 2 compare the climate ROI of CCU pathway 11 for the production of formic acid through the hydrogenation of the captured  $\text{CO}_2$  (P11: Formic Acid) and CCU pathway 2 for CCU concrete production through  $\text{CO}_2$  mixing (P2: Concrete).

Figure 2(a) demonstrates the difficulties comparing the climate impact of concrete and chemical pathways. For P11: Formic Acid, the functional unit is expressed as 1 kg and is the same regardless of whether Formic Acid is produced through a CCU or conventional pathway since the material properties of formic acid are the same. As a result, changes in performance do not need to be taken into account, with all values of % change in output equal to zero, and the distribution of data resides entirely on the y-axis (green points in Figure 2(a)). Meanwhile, CCU concrete has a different compressive strength than conventional concrete, which must be taken into account when analyzing the climate ROI for concrete. As a result, the amount of material

(e.g. ordinary Portland cement) needed to achieve similar function is variable and the data for concrete % change in output have a range of non-zero values, unlike the chemical pathway.

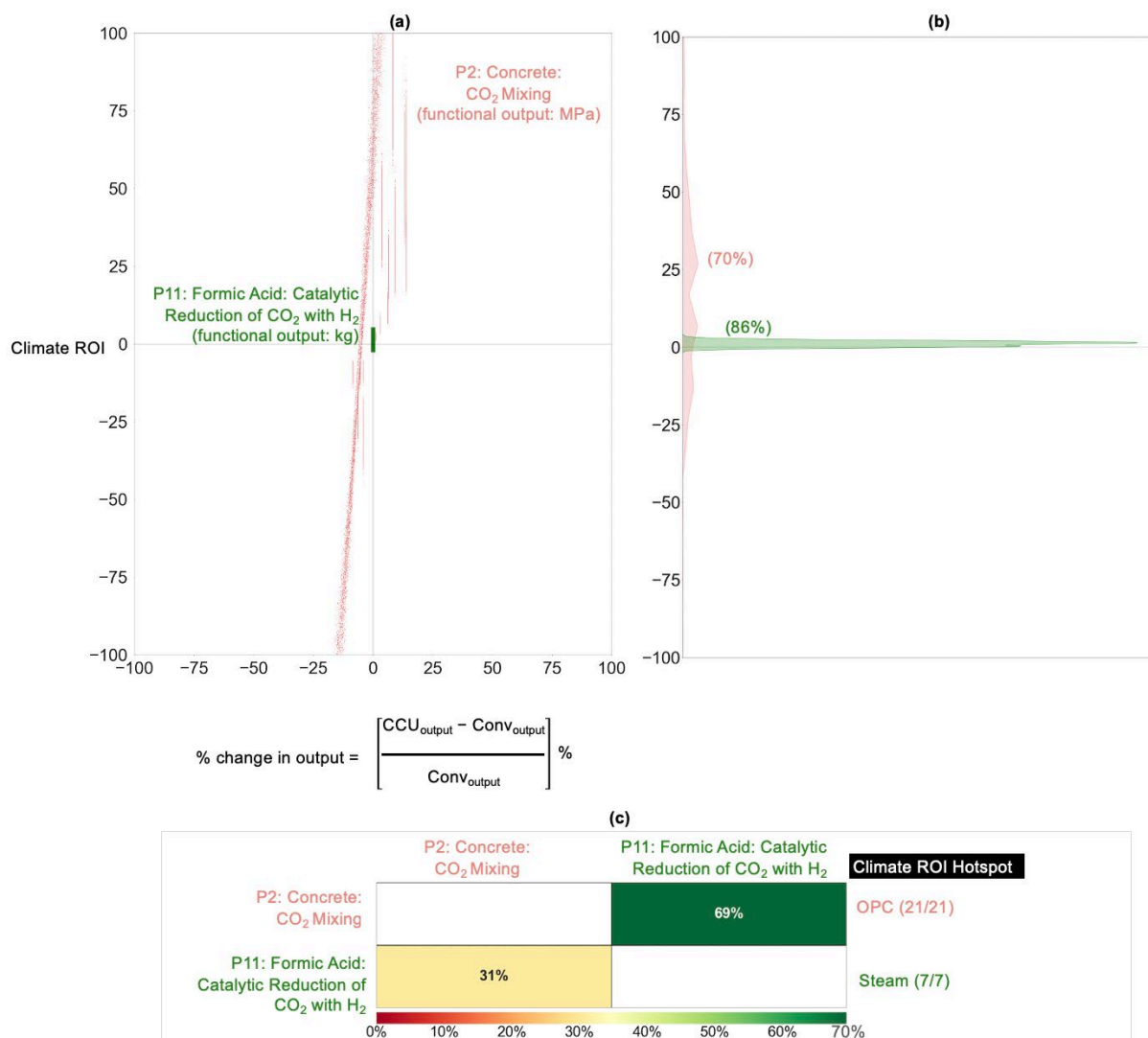


**Figure 2 (a) A scatter plot of the climate ROI of CCU concrete (pathway P2) and CCU formic acid (pathway P11) versus the percentage change in the output.  $\Sigma\text{Conv}_{\text{CO}_2}$  and  $\Sigma\text{CCU}_{\text{CO}_2}$  represent the sum of the life cycle CO<sub>2</sub> emissions from manufacturing P2: Concrete or P11: Formic acid through the conventional and CCU pathway as depicted in Figure 1.  $\text{Conv}_{\text{output}}$  and  $\text{CCU}_{\text{output}}$  represent the compressive strength of the P2: Concrete (in MPa) or the mass of the P11: Formic acid (1 kg) produced through the conventional**

and CCU pathways. If the climate ROI is greater than 0, then CO<sub>2</sub> is avoided when producing the CCU product. If the climate ROI is lesser than 0, then CO<sub>2</sub> is emitted. (b) The density plot of the climate ROI datapoints obtained from the scatter plot. The percentage value is the likelihood of the climate ROI of an individual CCU pathway being greater than zero in 96600 Monte Carlo comparisons. (c) A heatmap presenting the likelihood of the climate ROI of P11: Formic Acid exceeding P2: Concrete in 96600 Monte Carlo comparisons. The column to the right of the heatmap presents the inventory item which is the key driver of the climate ROI.

The probability density plot (Figure 2b) is obtained from the datapoints in Figure 2(a) and reveals that the climate ROI of P11: Formic Acid and P2: Concrete have an 86% and 70% likelihood of being greater than zero, respectively. It is important to note that the probability density plot and percentage values assess the climate ROI of the two CCU pathways individually.

In contrast, the heatmap analysis (Figure 2 (c)) is a comparative assessment of the climate ROI of the two CCU pathways. The heatmap demonstrates that in a pairwise comparison across 96600 Monte Carlo runs, the climate ROI of P2: Concrete exceeds that of P11: Formic Acid in 66650 runs. As a result, there is a 69% likelihood of the climate ROI of P2: Concrete exceeding that of P11: Formic Acid. Alternately, there is a 31% likelihood of the climate ROI of P11: Formic Acid being higher than that of P2: Concrete. By convention, a pathway with a greater likelihood of generating a higher climate ROI is placed above an alternate pathway with a greater likelihood of generating a lower climate ROI. As a result, P2: Concrete is the top row and P11: Formic Acid is the bottom row of the heatmap.

A moment-independent sensitivity analysis identified that the ordinary Portland cement (OPC) used in CCU concrete production is the key driver of the climate ROI of P2: Concrete in all the 21 datasets (SI Section 2), which were considered for the climate ROI analysis. The OPC used is normalized to the compressive strength, which is the functional unit for the climate ROI analysis of CCU concrete production. CO<sub>2</sub> mixing may decrease the compressive strength of the CCU concrete when compared to conventionally produced concrete.<sup>11</sup> As a result, more OPC is required in the mix design, to achieve the same compressive strength for CCU concrete. As OPC is the most CO<sub>2</sub> intensive material used in the mix design of concrete,<sup>18,19</sup> the increased use of OPC to compensate for the loss in compressive strength makes CCU concrete more climate intensive than conventional concrete. As a result, an R&D agenda focused on increasing the compressive strength when CO<sub>2</sub> is used in the mixing of concrete, and thereby, decreasing the OPC consumed on a per MPA basis, will be the most effective in increasing the climate ROI of P2: Concrete.

### **Positive Climate ROI versus Greater Potential for Improvement**

The results in Figure 2(c) demonstrate the importance of distinguishing between pathways with a higher likelihood of a positive climate ROI and pathways with a greater potential for improvement in climate ROI. For example, the climate ROI of P11: Formic Acid has an 86% likelihood (Figure 2(b)) of being positive in contrast to the 70% for P2: Concrete. However, the climate ROI for P2: Concrete has a wider distribution than P11: Formic Acid and spans both positive and negative ROI values. On the one hand, this indicates that there is a higher certainty that the climate benefits of P11: Formic Acid are likely to be positive, albeit with a fairly modest climate benefit. On the other hand, there is a greater opportunity to achieve significantly larger carbon benefits through P2: Concrete, but close attention needs to be paid to choosing

formulations that have better carbon profiles and focused R&D efforts are required to ensure that there are carbon benefits rather than increased emissions. As identified in the results of Figure 2(c), R&D targeted on increasing the compressive strength when CO<sub>2</sub> is used in the mixing of concrete can decrease OPC consumed per MPa and significantly increase the climate ROI of P2: Concrete.

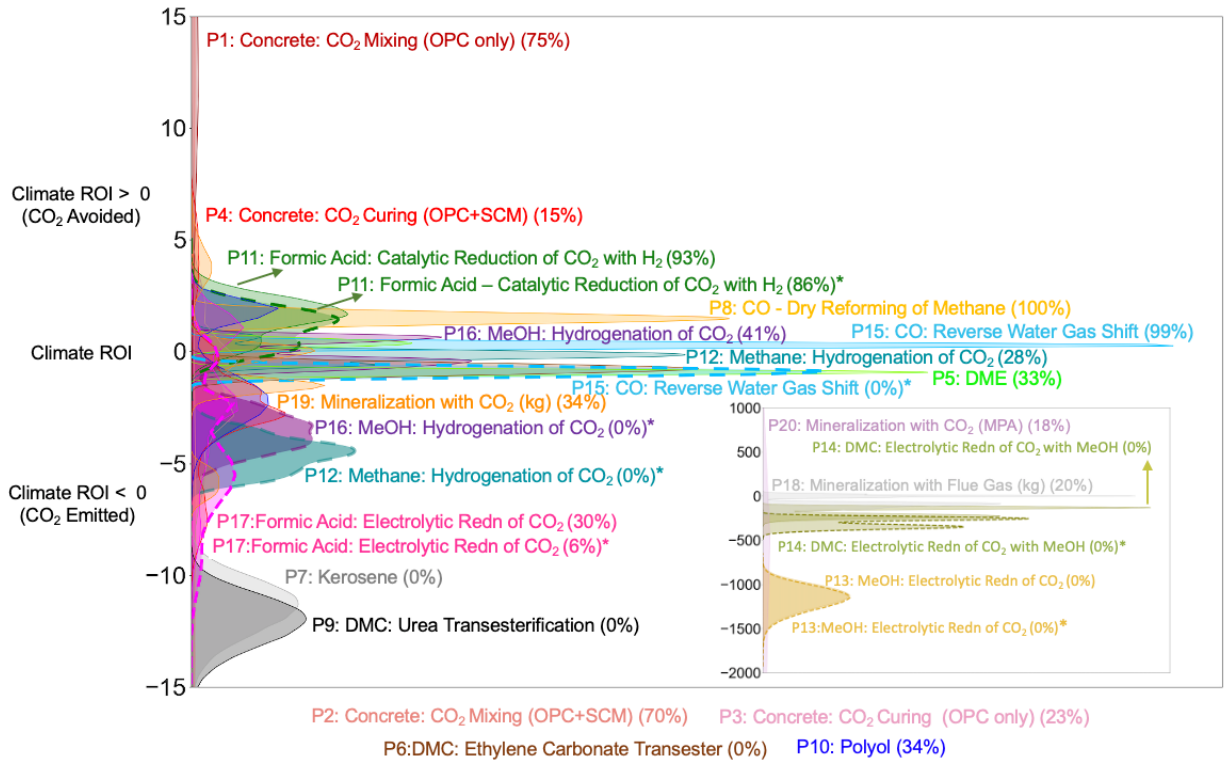


**Figure 3 The climate ROI for 4 concrete (P1 to P4), 13 chemical (P5 to P17) and 3 mineralization (P18 to P20) CCU pathways. If the climate ROI is greater than 0, then CO<sub>2</sub> is avoided when producing the CCU product. If the climate ROI is lesser than 0, then CO<sub>2</sub> is emitted. The percentage values indicate the likelihood of a CCU pathway having a climate ROI greater than 0 in 96600 Monte Carlo runs. The CCU pathways with a dashed curve or an asterisk require wind electricity and, therefore, incur an environmental opportunity cost (EOC). The climate ROI distributions for 4 pathways – P2, P3, P6 and**

**P10 – are masked by distributions of other pathways. The percentage values for these four pathways are presented below the graph.**

### **Promising CCU Pathways and Strategies to Improve Climate ROI**

Four CCU pathways – P1: Concrete, P2: Concrete P8: CO, P11: Formic Acid\* - have a likelihood of 75%, 70%, 100% and 86% of having a positive climate ROI (Figure 3). After accounting for the environmental opportunity cost, the four pathways are the only pathways with a greater likelihood of generating a climate benefit than a climate burden (i.e., >50% likelihood of generating a positive climate ROI). Moreover, the four CCU pathways constitute the top four rows in the heatmap (Figure 4(a)) and, therefore, have a significantly higher likelihood of producing a greater climate ROI than the remaining CCU pathways. This is illustrated by the 69% to 100% likelihood in the first four rows between column 5 to 20 in the heatmap (Figure 4(a)).

The moment independent sensitivity analysis identifies the use of OPC as the most significant contributors to the climate ROI of P1: Concrete and P2: Concrete. As explained in an earlier section, R&D towards increasing the compressive strength will decrease the OPC consumed per MPa and produce a significant increase in the climate ROI when CO<sub>2</sub> is used during concrete mixing. Decreasing the magnitude and the CO<sub>2</sub> intensity of the energy used can significantly improve the climate ROI of P8: CO and P11: Formic Acid.



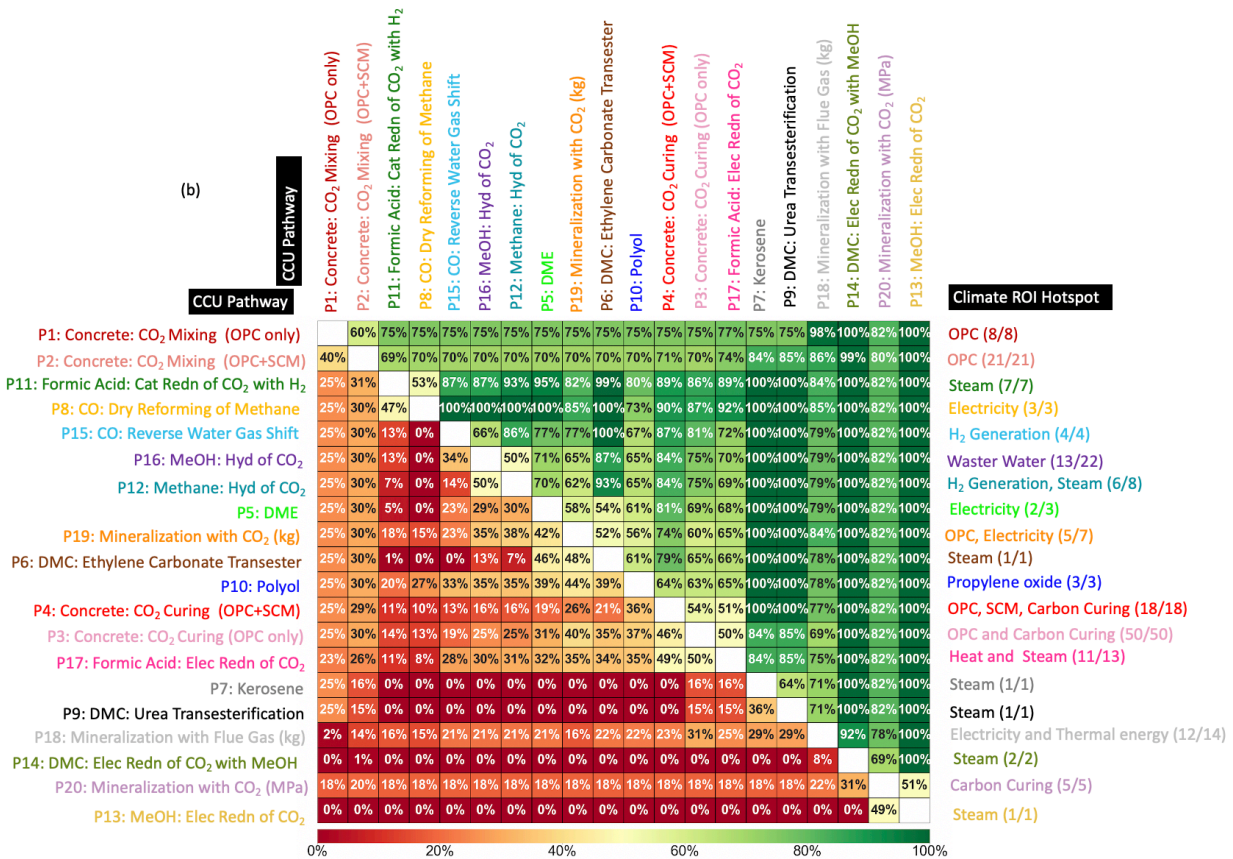


**Figure 4 Comparative assessment of the climate ROI for the 4 concrete (P1 to P4), 13 chemical CCU pathways (P5 to P17) and 3 mineralization pathways (P18 to P20). Figure (a) accounts for the environmental opportunity cost (EOC) of using wind-generated electricity when quantifying the climate ROI for the 7 CCU chemical pathways which require hydrogen as a feedstock (indicated with an asterisk) and (b) does not account for the EOC. Rows at the top of the heatmap have a greater likelihood of generating a higher climate ROI than the rows at the bottom. Each cell in the heatmap quantifies the likelihood of the climate ROI of a CCU pathway in a row being greater than the CCU pathway in a column in 96600 Monte Carlo runs. The climate ROI hotspots, which are the parameters most significantly influencing the climate ROI of a CCU pathway, are presented on the**

right of the heatmap. The parentheses to the right of the climate ROI hotspot indicate the fraction of datasets in which the inventory item is the hotspot.

Four chemical CCU pathways - P7: Kerosene, P9: DMC: Urea Transesterification, P13: MeOH\* and P14: DMC\*, - have a 0% likelihood of a positive climate ROI (Figure 3). Despite utilizing CO<sub>2</sub>, the life cycle CO<sub>2</sub> emissions in the CCU pathway ( $\Sigma\text{CCU}_{\text{CO}_2}$ ) is greater than the conventional pathway ( $\Sigma\text{Conv}_{\text{CO}_2}$ ) and, therefore, the four CCU pathways generate an increase in climate burden on a lifecycle basis. Figure 4(a) shows that the 4 pathways (P7, P9, P13\*, P14\*) are among the bottom 6 rows of the heatmap. Along with the four pathways, two other pathways - P18: Mineralization and P20: Mineralization - are at the bottom of the heatmap (Figure 4(b)), with a lower relative climate ROI benefit when compared to other CCU pathways.

When the EOC is not accounted for in 3 CCU pathways (P12: Methane, P16: MeOH and P17: Formic Acid), the likelihood of a positive climate ROI is 28%, 41%, and 30%, respectively, (Figure 3). However, including the EOC decreases the likelihood of a positive climate ROI for P12: Methane\*, P16: MeOH\*, and P17: Formic Acid\* to 0%, 0%, and 6%, respectively (Figure 3). The low climate ROI of the three pathways after including the EOC is further illustrated by the results in Figure 4 (b) wherein P12: Methane\*, P16: MeOH\*, and P17: Formic Acid\* rank only above the bottom 6 pathways discussed previously.

In summary, based on the cell values between rows 12 to 20 and between columns 1 to 4 in Figure 4 (b), the 9 pathways at the bottom of the heatmap (Figure 4 (b)) have a 0 to 30% likelihood of exceeding the climate ROI of the pathways in the top 4 rows. In addition, the pathways in the 9 bottom rows in the heatmap (Figure 4 (b)) have 0 to 20% likelihood of generating a positive climate ROI (Figure 3). As a result, utilizing CO<sub>2</sub> in the 9 bottom pathways

forgoes an opportunity to realize a greater climate benefit by utilizing CO<sub>2</sub> in the pathways in the top 4 rows in the heatmap (Figure 4 (b)).

Seven CCU pathways – P3: Concrete, P4: Concrete, P5: DME, P6: DMC, P10: Polyol, P15: CO\* and P19: Mineralization - have a 0 to 34% likelihood of their climate ROI being greater than zero (Figure 3). While offering a higher climate ROI than the pathways in the bottom 9 rows of the heatmap (Figure 4(b)), the seven pathways offer a lower climate ROI than the top 4 CCU pathways as the likelihood values in the cells between columns 1 to 4 and between rows 5 to 11 range between 0 and 31%. As a result, utilizing CO<sub>2</sub> in the top 4 pathways offers a significantly higher climate benefit than the 7 CCU pathways.

The moment independent sensitivity analysis identifies the CO<sub>2</sub> curing process and the use of binder materials (OPC and supplementary cementitious materials (SCM)) as the climate ROI hotspots in P3: Concrete and P4: Concrete. Decreasing the energy intensity of the CO<sub>2</sub> curing process will produce a significant increase in the climate ROI of P3: Concrete. As explained earlier, increasing the compressive strength with CO<sub>2</sub> curing will decrease the quantity of OPC and SCM required on a per MPa basis for CCU concrete and, thereby, ensure a significant increase in the climate ROI of P3: Concrete and P4: Concrete.

### **Scenario Analysis to Assess the Impact of Energy Transition Trends**

In addition to strategies which are specifically designed to address the hotspots and improve the climate ROI of individual CCU pathways (Figure 4(b)), we expect three energy transition trends - a decrease in the EOC of using renewable electricity for H<sub>2</sub> generation and a transition to low-carbon electricity and heat sources - to impact the climate ROI of the 20 CCU pathways in the future. The results from the scenario analysis, which is discussed below, quantify the impact of

the three energy transition trends on the relative climate ROI performance of the 20 CCU pathways.

1. **Decreasing the environmental opportunity cost of H2 generation:** Under certain scenarios, the EOC can be decreased to generate a corresponding increase in the climate ROI (Equation 1) of the 7 CCU chemical pathways, which require RE use for H2 generation. EOC can be decreased by utilizing RE dedicated for H2 generation (e.g., through captive RE plants at the site of electrolysis) and not for offsetting fossil electricity on the grid. Additionally, excess RE, which is curtailed,<sup>20,21</sup> can instead be stored and used for H2 generation. In these two optimistic scenarios, the EOC is zero as RE is not offsetting grid electricity.

A comparison of the heatmap analysis in Figure 4(a), which accounts for the EOC, and Figure 4(b), which does not account for the EOC (i.e. the EOC is zero), illustrates the change in the climate ROI rankings of the CCU pathways with the maximum possible decrease in the EOC (i.e. from baseline EOC to zero EOC). The results in Figure 4(b) show that only three additional candidates - P12: Methane, P15: CO and P16: Methanol - have a significant increase in the climate ROI rankings. P12: Methane, P15: CO and P16: Methanol, which were ranked 8, 12 and 13 in the baseline scenario (Figure 4(a)), are ranked 5, 7 and 6 when the EOC is zero (Figure 4(b)) and become increasingly attractive candidates to increase the climate benefit of CO2 utilization.

2. **Transition to low carbon electricity sources in CCU production processes:** As demonstrated by the hotspot analysis, energy sources (electricity, heat and steam) are significant drivers of the climate ROI of CCU chemical and concrete pathways (Figure 4 (a)). Therefore, a transition to less carbon intensive sources of electricity<sup>22,23</sup> can improve the

climate ROI of CCU concrete, chemicals and minerals. To simulate a transition to low-carbon electricity sources, we assume the CO<sub>2</sub> intensity of the grid, which is used to power the CCU production processes (e.g., electricity used in concrete curing in P4: Concrete), is equal to that of wind electricity which has the lowest CO<sub>2</sub> footprint among electricity sources.<sup>24,25</sup>

The results show that P18: Mineralization, P3: Concrete, P16: MeOH\* and P12: Methane\* witness the most improvements in the climate ROI rankings when wind electricity (SI Supplementary Figure 2) replaces electricity with a CO<sub>2</sub> footprint equal to the average of the US grid regions in the baseline scenario (Figure 4 (b)). P18: Mineralization, P3: Concrete, P16: MeOH\* and P12: Methane\*, which were ranked 17, 11, 12 and 13 in the baseline scenario (Figure 4 (b)), have an improved ranking of 2, 7, 8 and 9, respectively.

3. **Transition to low carbon heat sources in CCU production processes:** A similar scenario analysis for low-carbon heat shows that P17: Formic Acid\*, P7: Kerosene and P13: MeOH\* (SI Figure 2) witness the greatest improvement in climate ROI rankings when compared to the baseline scenario (Figure 4 (a)). P17: Formic Acid\*, P7: Kerosene and P13: MeOH\*, which were ranked 14, 15 and 20 in Figure 4 (a) in the main paper, are now ranked 7, 10 and 17 in Supplementary Figure 3 in the SI.

It is important to note that P1: Concrete, P2: Concrete, P11: Formic Acid, P8: CO, which are the pathways with the highest climate benefit in the baseline scenario (Figure 4 (b)), continue to offer a high climate ROI in the zero EOC and low-carbon electricity and heat scenarios. This is an important finding which demonstrates that the four CCU pathways are well suited to leverage a transition to zero EOC and low carbon electricity and heat sources and generate the maximum climate benefits.

## Methods

### Material and Energy Inventory Data to Determine Climate ROI

We obtained the material and energy inventory data, which is required to determine the climate ROI (*Figure 1*), from 194 datasets consisting of 99, 69 and 26 datasets focusing on CCU concrete production, chemical production and mineralization, respectively (SI Section 2). The 99 datasets for concrete include four categories wherein CO<sub>2</sub> is utilized for (i) curing of concrete when only OPC is used as the binder, (ii) curing of concrete when a mix of OPC and SCM is used as the binder, (iii) mixing of concrete when only OPC is used as the binder, and (iv) mixing of concrete when a mix of OPC and SCM is used as the binder. The 99 datasets (SI Section 2) were obtained from 19 studies.<sup>10,11,26-42</sup> The 19 studies were shortlisted based on the criteria that they reported the 28-day compressive strength for both conventional and CCU concrete; the mass of OPC, SCM (if used), water, fine aggregate and coarse aggregate used in the design mix; the steam curing duration for both conventionally produced and CCU concrete; the CO<sub>2</sub> curing duration for CCU concrete production; and the CO<sub>2</sub> utilized in CCU concrete production. The material inventory (e.g. OPC, fine and coarse aggregates) and steam curing duration are required to determine the total life cycle CO<sub>2</sub> emissions from producing conventional and CCU concrete ( $\Sigma\text{ConvCO}_2$  and  $\Sigma\text{CCUCO}_2$  in *Figure 1*). The carbon curing duration is required to determine  $\Sigma\text{CCUCO}_2$ . The compressive strength is required to determine the life cycle CO<sub>2</sub> emitted in producing 1 MPa of conventional and CCU concrete. The CO<sub>2</sub> utilized is required to determine the climate ROI ('CO<sub>2</sub> Utilized' in Equation 1). We leverage a recently published review article<sup>43</sup> to identify the 69 datasets for the production of the 13 chemicals through CCU. The material and energy inventory required to produce the 13 chemicals through the CCU pathway were obtained by reviewing the literature sources cited in <sup>43</sup>

(SI Section 2). The life cycle CO<sub>2</sub> emissions from producing the chemical through the CCU pathway ( $\Sigma\text{CCU}_{\text{CO}_2}$  in **Figure 1**) was determined from the inventory requirements (SI Section 4). The life cycle CO<sub>2</sub> emissions from producing the chemical through the conventional pathway ( $\Sigma\text{Conv}_{\text{CO}_2}$  in **Figure 1**) was determined from the Ecoinvent database using the SimaPro software (SI Section 5).

In contrast to the concrete pathways (P1 to P4), where CO<sub>2</sub> is directly incorporated into concrete through mixing or curing without producing a new material, the CO<sub>2</sub> utilization in the mineralization pathways (P18 to P20) produces minerals which can substitute for commercial materials (e.g., aggregates in concrete). CO<sub>2</sub> in flue gas or CO<sub>2</sub> in purified form is used in the carbonation of alkaline waste materials (e.g., steel slag) to generate minerals.<sup>44-50</sup> Furthermore, the studies on CCU mineralization datasets either report or do not report the compressive strength of the CCU mineral. To account for these differences, we include three pathways for CCU mineralization. Pathway 18 (P18 in **Figure 1**), includes 14 datasets for the production of a functional unit of 1 kg of the mineral by utilizing flue gas containing CO<sub>2</sub> (i.e., unpurified CO<sub>2</sub>). Pathway 19 (P19 in **Figure 1**), includes 7 datasets for the production of 1 kg of the mineral from purified CO<sub>2</sub>. Pathway 20 (P20 in **Figure 1**), includes 5 datasets for the production of 1 MPa of the mineral from purified CO<sub>2</sub>.

The 26 datasets for mineralization (SI Section 2) were obtained from studies reporting (i) the energy and inventory items required to produce the mineral through the conventional and CCU pathways; (ii) the quantity (when 1 kg is used as the functional unit) or the 28-day compressive strength (when 1 MPa is used as the functional unit) for both conventionally produced and CCU concrete; and (iii) the quantity of CO<sub>2</sub> utilized (either in the captured and purified form or in the flue gas).

**CO2 Capture:** The CO<sub>2</sub> is assumed to be captured from a combined cycle natural gas power plant as the electricity sector contributes to 28% of the total CO<sub>2</sub> emissions in the US<sup>51</sup> and is, therefore, a promising candidate for CO<sub>2</sub> capture. Moreover, combined cycle natural gas power plants are the most commonly deployed fossil fuel-based electricity generation source in the US.<sup>52</sup> The CO<sub>2</sub> was assumed to be captured using a post-combustion monoethanolamine (MEA) capture system as it the most widely used and commercially mature carbon capture technology<sup>53,54</sup> and has a high CO<sub>2</sub> capture efficiency (>90%).<sup>55-57</sup> The energy and material required to capture CO<sub>2</sub> from a combined cycle natural gas plant using post-combustion MEA system is obtained from <sup>58</sup> (SI Section 6).

**Energy Penalty of CO2 Capture:** The capture of CO<sub>2</sub> using a post-combustion MEA system from a power plant generates an energy penalty ( $E_p$  in kWh) as heat, which would have been used for electricity generation, is instead utilized to regenerate MEA and release the captured CO<sub>2</sub>.<sup>59-62</sup> To compensate for the energy penalty when capturing CO<sub>2</sub>, we use the recommendation from NETL.<sup>63</sup> NETL recommends that the energy penalty is compensated by an external power plant generating electricity with an average CO<sub>2</sub> intensity equal to that of the region in which the CO<sub>2</sub> capture occurs. As we conduct this analysis for the US, we assume the external power plant compensating for the energy penalty has a CO<sub>2</sub> intensity varying uniformly between 0.38 and 0.56 kg CO<sub>2</sub>/kWh, which is the average CO<sub>2</sub> intensity of electricity in the various grid regions in the US (SI Supplementary Table 9).<sup>63</sup>

**H2 Generation:** We assume that H<sub>2</sub>, which is required to convert the captured CO<sub>2</sub> into chemicals in the 7 CCU pathways, is generated by electrolyzing water using wind electricity. The CO<sub>2</sub> footprint of generating H<sub>2</sub> using wind electricity is significantly lower than the conventional route of H<sub>2</sub> production from fossil fuels and, thereby, is an attractive pathway to

decrease the life cycle climate impact of CCU chemical pathways requiring H<sub>2</sub>.<sup>16</sup> Moreover, electrolysis of water is the only technology available at a commercial scale, which is less climate intensive than H<sub>2</sub> generation from fossil fuels.<sup>16,64,65</sup> The material and energy inventory data required to produce H<sub>2</sub> using an alkaline electrolyser is obtained from<sup>66</sup> and detailed in **SI Section 7**.

#### **Determining the life cycle CO<sub>2</sub> footprint of the material and energy used in CCU**

**production:** The life cycle inventory datasets corresponding to the material and energy used in the production of CCU and conventional concrete, chemicals and minerals, H<sub>2</sub>, and the MEA capture system were obtained from the Ecoinvent database (**SI Supplementary Tables 4, 5, 7 and 8**). The SimaPro LCA software<sup>67</sup> was used to apply the global warming mid-point impact category from the TRACI impact assessment method<sup>68</sup> and quantify the life cycle CO<sub>2</sub> footprint corresponding to the life cycle inventory datasets (**SI Sections 3, 4, 5, 6 and 7**).

#### **System Boundary and Functional Unit to Determine Climate ROI**

The system boundary and the processes included within the scope of the analysis to determine the climate ROI is presented in **Figure 1**. As explained earlier, the system boundary to quantify for the climate ROI accounts for the material and energy required to produce conventional and CCU concrete and chemicals, the energy penalty of CO<sub>2</sub> capture, the upstream processes for capturing CO<sub>2</sub> and H<sub>2</sub> generation from water electrolysis, and the EOC of renewable electricity used for H<sub>2</sub> generation. Given that the production of CCU chemical and concrete are emerging technologies, there is no data on the geo-spatial location of commercial scale production plants. As a result, we assume the CO<sub>2</sub> capture and utilization facilities are co-located and there is no transport of CO<sub>2</sub>.

To determine the climate ROI, we use a functional unit (FU) of 1 kg for CCU chemicals (pathways P5 to P17) and minerals (pathways P18 and P19) and 1 MPa of compressive strength for CCU concrete (pathways P1 to P4) and minerals (pathway P20). We choose a FU of 1 kg as it is commonly used to compare the environmental impact of chemicals produced through CCU and conventional pathways.<sup>43</sup> We choose the 28-day compressive strength as it is a commonly used standard to categorize the performance of concrete<sup>69</sup> and minerals and is reported extensively in studies comparing the performance of CCU and conventional concrete and minerals.<sup>10,11,26-42,44-50</sup> More importantly, by normalizing the life cycle CO<sub>2</sub> emission on a per MPa basis, the analysis accounts for the change in compressive strength between CCU and conventional concrete. When datasets comparing conventional and CCU mineralization processes do not report the compressive strength of the mineral produced, we use the functional unit of 1 kg of the mineral.

**Climate ROI Metric:** This analysis uses a novel climate ROI metric to quantify the net climate per kg of CO<sub>2</sub> utilized across chemical, concrete and mineral production. To the best of our knowledge, this the first metric to enable a comparison of the climate benefit when CO<sub>2</sub> is utilized for chemical, concrete and mineral production. Climate ROI is defined as the

$$\text{Climate ROI} = \frac{\frac{\Sigma \text{Conv}_{\text{CO}_2}}{\text{Conv}_{\text{output}}} - \frac{\Sigma \text{CCU}_{\text{CO}_2}}{\text{CCU}_{\text{output}}} - \frac{\text{EOC}_{\text{CO}_2}}{\text{CCU}_{\text{output}}}}{\frac{\text{CO}_2 \text{ utilized}}{\text{CCU}_{\text{output}}}} \quad \text{Equation 1}$$

‘CCU<sub>CO<sub>2</sub></sub>’ and ‘Conv<sub>CO<sub>2</sub></sub>’ are the life cycle CO<sub>2</sub> emissions (kg) from producing the chemicals, concrete or minerals through the CCU and conventional pathway, respectively. ‘EOC’ is the environmental opportunity cost (kg CO<sub>2</sub>) of utilizing wind electricity to electrolyze water and generate the H<sub>2</sub> feedstock required for 7 CCU chemical pathways (blue box in Figure 1(b)).

‘CO2 Utilized’ represents the quantity of CO2 (in kg) which is captured and utilized to produce a chemical, concrete or mineral. ‘CCU<sub>output</sub>’ and ‘Conv<sub>output</sub>’ represent the functional output from the CCU and conventional process. If the climate ROI of CO2 utilization in chemical production is being determined, then CCU<sub>output</sub> and Conv<sub>output</sub> is quantified in kilograms. For concrete or mineral production, CCU<sub>output</sub> and Conv<sub>output</sub> are quantified through the compressive strength (in MPa). If the compressive strength of the mineral produced through the CCU and conventional pathway is not reported by a dataset, then CCU<sub>output</sub> and Conv<sub>output</sub> is quantified in kilograms. CCU<sub>output</sub> and Conv<sub>output</sub> are obtained from the dataset.

Conv<sub>CO2</sub> is determined by

$$\Sigma \text{Conv}_{\text{CO}_2} = \text{Conv\_Output}_{\text{CO}_2} + (\text{NGCC}_{\text{CO}_2} \times E_p) \quad \text{Equation 2}$$

Conv\_<sub>Output</sub><sub>CO2</sub> represents the CO2 emissions from producing the chemical (1 kg), concrete (in MPa) or the mineral (1 kg or in MPa) through the conventional process. Conv\_<sub>Output</sub><sub>CO2</sub> is determined from ecoinvent datasets in SimaPro (SI Sections 3, 4 and 5). NGCC<sub>CO2</sub> is the life cycle CO2 intensity of generating electricity in a combined cycle natural gas plant (SI Section 8). The second term in parenthesis represents the generation of E<sub>p</sub> (in kWh) without carbon capture along with the production of the chemical, concrete or mineral in the conventional pathway (Figure 1(a)).

CCU<sub>CO2</sub> is determined by

$$\Sigma \text{CCU}_{\text{CO}_2} = \text{CCU\_Output}_{\text{CO}_2} + (E_{\text{we}} \times \text{Wind}_{\text{CO}_2}) + (\text{CO}_2 \text{ utilized} \times \text{MEA}_{\text{CO}_2}) + (0.1 \times \text{NGCC}_{\text{CO}_2} \times E_p) + (\text{Grid}_{\text{CO}_2} \times E_p) \quad \text{Equation 3}$$

The first term represents the CO2 emissions from producing a chemical (1 kg), concrete (in MPa) or mineral (in MPa or kg) through the CCU pathway and is obtained by characterizing the

material and energy inventory reported in the literature dataset for the global warming impact category in SimaPro (SI Sections 3 and 4).

The second term accounts for the CO<sub>2</sub> emissions from generating H<sub>2</sub> using wind electricity.  $E_{we}$  is the wind electricity used to generate the H<sub>2</sub>, which is used to produce the CCU chemical, and  $Wind_{CO_2}$  is the CO<sub>2</sub> intensity of wind electricity (kg CO<sub>2</sub>/kWh) (SI Section 8). If H<sub>2</sub> is not required for generating the CCU product, then the second term is zero.

$MEA_{CO_2}$  represents the life cycle CO<sub>2</sub> emissions from manufacturing the post-combustion MEA carbon capture system (per kg of CO<sub>2</sub> captured, SI Section 6).

The fourth term accounts for the 10% of CO<sub>2</sub> that is not captured as the capture efficiency of the post-combustion MEA system is 90%.<sup>55-57</sup>

The final term accounts for the CO<sub>2</sub> emissions from compensating for the energy penalty ( $E_p$ ) through an external power plant.  $Grid_{CO_2}$  varies uniformly between 0.38 and 0.56 kg CO<sub>2</sub>/kWh, which is the average CO<sub>2</sub> intensity of electricity in the various grid regions in the US.<sup>63</sup>

$EOC_{CO_2}$  is determined by

$$EOC_{CO_2} = E_{we} \times (Grid_{CO_2} - Wind_{CO_2}) \quad \text{Equation 4}$$

**Monte Carlo Analysis to Account for Uncertainty:** It is important to note that the Climate ROI metric is impacted by the uncertainty in the life cycle CO<sub>2</sub> intensity of the - CO<sub>2</sub> capture process (SI Section 6); H<sub>2</sub> generation process (SI Section 7); conventional process used to produce chemicals (SI Section 5); and the energy and material required to manufacture the product through the CCU and conventional pathway (SI Sections 3 and 4). To investigate the impact of the uncertainty, we conduct a Monte Carlo analysis and determine the Climate ROI for the CCU chemical, concrete and mineralization pathways over 96600 runs. In each Monte Carlo run, we stochastically generate the value of the material and energy inventory items (SI Section 2) and

the CO<sub>2</sub> intensities of the life cycle inventory data based on the mean and standard deviation provided in the SI (SI Sections 3, 4, 5, 6 and 7) and determine the corresponding value of the climate ROI (Equation 1).

The stochastically determined Climate ROI values from the Monte Carlo analysis are used to generate the density plot (Figure 3), compare the climate ROI of the various CCU chemical, concrete and mineralization pathways in the heatmap analysis (Figure 4) and conduct the moment independent sensitivity analysis to identify the key drivers of the climate ROI (Figure 4).

**Heatmap Analysis to Compare the Climate ROI of Chemical and Concrete Pathways:** The heatmap analysis conducts a pairwise comparison of the 96600 climate ROI samples of each of the 20 CCU pathways taken two at a time. In each of the 96600 pairwise comparisons, we determine a count, which represents the number of times the climate ROI of the first CCU pathway exceeds that of the second pathway. The count helps determine the likelihood (in percentage) of the first CCU pathway having a greater climate ROI than the second in the 96600 Monte Carlo runs. The likelihood is presented in the heatmap (Figure 4).

**Moment Independent Sensitivity Analysis to Determine the Key Drivers of the Climate ROI:** We conducted a moment independent sensitivity analysis<sup>15,70-72</sup> to determine the material and energy inventory items, which are the most significant drivers of the climate ROI of a CCU pathway (climate ROI hotspots). The moment independent sensitivity analysis calculates an  $\delta$  index value for each of the material and energy inventory items required to produce the CCU product and, thereby, contributes to the climate ROI of the CCU product. The inventory item with the highest  $\delta$  index value is the most significant contributor to the probability distribution function of the climate ROI. The  $\delta$  index value is determined for the inventory items reported in

each of the 69 datasets for the 13 CCU chemical pathways, 99 datasets for the 4 CCU concrete pathways and the 26 datasets for the 3 CCU mineralization pathways (**Figure 4**). The  $\delta$  index values are determined through custom code in Python and is based on the approach presented in Wei, Lu and Yuan.<sup>72</sup>

#### **Data Availability**

The datasets utilized in this analysis are included in section 2 of the supplementary information file.

#### **Code Availability**

The code used in the analysis can be accessed on <sup>73</sup>

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DR conducted the research presented in this manuscript at the CSS, UMich. DR is currently affiliated with the National Renewable Energy Laboratory (NREL).

#### **Author Contributions**

DR, GAK and SAM designed the research. DR reviewed the literature, collated the datasets, formulated the climate ROI metric, conducted the analysis, wrote the Python code and developed the first draft of the manuscript. GAK, SAM and VS contributed to improving the analysis, enhancing the visual presentation of the results and revising the manuscript.

#### **Competing Interests**

The authors declare no competing interests.

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