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Carbon dioxide utilization in concrete curing or mixing may not produce a net climate benefit

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Abstract

Carbon capture and utilization for concrete production (CCU concrete) is estimated to sequester 0.1 to 1.4 gigatons of carbon dioxide (CO₂) by 2050. However, existing estimates do not account for the CO₂ impact from the capture, transport and utilization of CO₂, change in compressive strength in CCU concrete and uncertainty and variability in CCU concrete production processes. By accounting for these factors, we determine the net CO₂ benefit when CCU concrete produced from CO₂ curing and mixing substitutes for conventional concrete. The results demonstrate that there is a higher likelihood of the net CO₂ benefit of CCU concrete being negative i.e. there is a net increase in CO₂ in 56 to 68 of 99 published experimental datasets. Strategies to increase the net CO₂ benefit from CCU concrete should achieve a high compressive strength from CO₂ curing and mixing and decrease the electricity used in CO₂ curing.

Introduction

The capture and utilization of carbon dioxide (CO₂) to produce economically viable products offers the twin benefit of mitigating climate change and generating economically viable products.¹ Among the portfolio of products that can potentially utilize CO₂, concrete offers several advantages including (i) a thermodynamically favorable reaction mechanism^{2,3} to sequester the CO₂ as calcium or magnesium carbonates^{4,5} in ordinary Portland cement (OPC)^{6,7} (ii) the long term CO₂ sequestration in the form of stable carbonate beyond the life-time of the infrastructure (> 60 years) (iii) the significant sequestration potential due to the overall and expected growth in global cement production to meet increasing demand.⁸ Concrete along with aggregates and chemicals/fuels are end-products with the potential to sequester the maximum amount of CO₂ (in gigatons).^{1,3}

Multiple emerging approaches such as carbonation of recycled concrete aggregates,⁹ CO₂ sequestration in alternative MgO based binders,¹⁰ CO₂ mineralization in industrial waste-derived aggregates and fillers^{11,12} and CO₂ dissolution in mixing water^{13,14} have been investigated for CO₂ utilization in concrete. However, this study focuses on the two approaches of CO₂ mixing and CO₂ curing as they are more extensively analyzed and applied for CO₂ utilization in concrete (supplementary information (SI) Section 2). In CO₂ mixing, high-purity CO₂ is injected into fresh concrete during batching and mixing. The CO₂ binds to the calcium silicate clinker in OPC to form nano-scale CaCO₃ particles.^{15,16} In CO₂ curing, CO₂ is utilized as a curing agent⁵ to accelerate precast concrete fabrication. A review of CO₂ curing and mixing studies reveals that the CO₂ uptake potential in CO₂ curing of precast concrete applications is significantly higher than in CO₂ mixing (SI Figure S6).

A common assumption motivating research and commercial interests in CCU concrete is that CO₂ uptake during curing and mixing¹⁷⁻²⁰ of CCU concrete lowers the CO₂ burden of concrete production. Estimates show that 0.1 to 1.4 gigatons of CO₂ can be utilized in concrete by 2050.^{1,3} However, a literature review (SI Section 13) demonstrates these estimates are not based on a comprehensive assessment which accounts for the change in compressive strength of concrete from CO₂ utilization; the CO₂ impact of capturing, transporting and utilizing CO₂; the CO₂ emissions from compensating for the energy penalty of CO₂ capture and producing supplementary cementitious materials (SCM), which are by-products of coal electricity and pig iron production; and may not always be based on primary experimental data, which is required for a robust life cycle CO₂ assessment.

CO₂ curing can decrease the compressive strength of CCU concrete when compared to conventional concrete. For example, a review of 99 experimental datasets from existing literature shows that CCU concrete has a lower compressive strength than conventional concrete in 31 datasets (SI Section 2 Figure S3). In such cases, CCU concrete would require a greater amount of OPC than conventional concrete to produce the same compressive strength. OPC production is a major source of CO₂ emissions. Therefore, increased OPC content in a concrete formulation leads to an increase in CO₂ emissions from upstream cement production processes, which may outweigh the benefit of the CO₂ captured and used in concrete production.

In addition, the CO₂ impact of CCU concrete can be difficult to generalize due to the lack of consistency in the boundaries and scope of analysis. For example, the energy associated with the capture and transport of CO₂ is included in certain studies²¹ while being excluded from others.^{20,22,23} Moreover, the uncertainty and variability in data and process parameters, which is typical in the early stages of R&D, impacts the environmental assessment of emerging

technologies such as CCU concrete.²⁴⁻²⁸ Life cycle assessments (LCA) of CCU concrete rely on point values for process parameters rather than parameter distributions that provide a more realistic representation of uncertainty and variability.^{16,21} The failure to account for uncertainty in the early stages of technology development can hinder research efforts to address hotspots and increase the CO₂ benefit from CCU concrete.^{25,26} An uncertainty assessment in the early stages of technology development can determine process parameters and inventory items that are the most significant contributors to the CO₂ burden of CCU concrete and, thereby, help identify research strategies that are most effective in addressing the hotspots.

To address these issues, we review 99 datasets from 19 publications to determine the range of potential net CO₂ benefit associated with CCU concrete. The net CO₂ benefit is defined as the difference between the lifecycle CO₂ impact of producing conventional concrete and producing CCU concrete through CO₂ curing or CO₂ mixing. The net CO₂ benefit accounts for the life cycle CO₂ impact of the 13 upstream processes to capture, transport and utilize CO₂ and produce and transport the materials used in concrete. The net CO₂ benefit accounts for any changes in compressive strength when CCU concrete is produced through CO₂ curing or CO₂ mixing. We conduct a sensitivity analysis consisting of a scatter plot analysis and moment independent sensitivity analysis^{25,29,30} to determine the key processes with the most significant influence on the net CO₂ benefit.

Results

Illustrative Example: Net CO₂ Benefit for Dataset 1

We illustrate the interpretation of the results using a single dataset, which helps better understand the findings across the 99 datasets. Consider the dataset of conventional and CCU concrete production in the A1 scenario reported in ³¹ wherein CO₂ is used for curing and OPC is used as

binder. Based on the inventory requirements presented in the dataset, the CO₂ emissions for the 13 processes (Figure 1 and **supplementary information (SI) Table S1**), the total life cycle CO₂ emissions from producing CCU concrete (TOT_{CCU}, Equation 1) and conventional concrete (TOT_{Conv}, Equation 5), and the net CO₂ benefit (Equation 6) are stochastically determined in 10000 Monte Carlo runs.

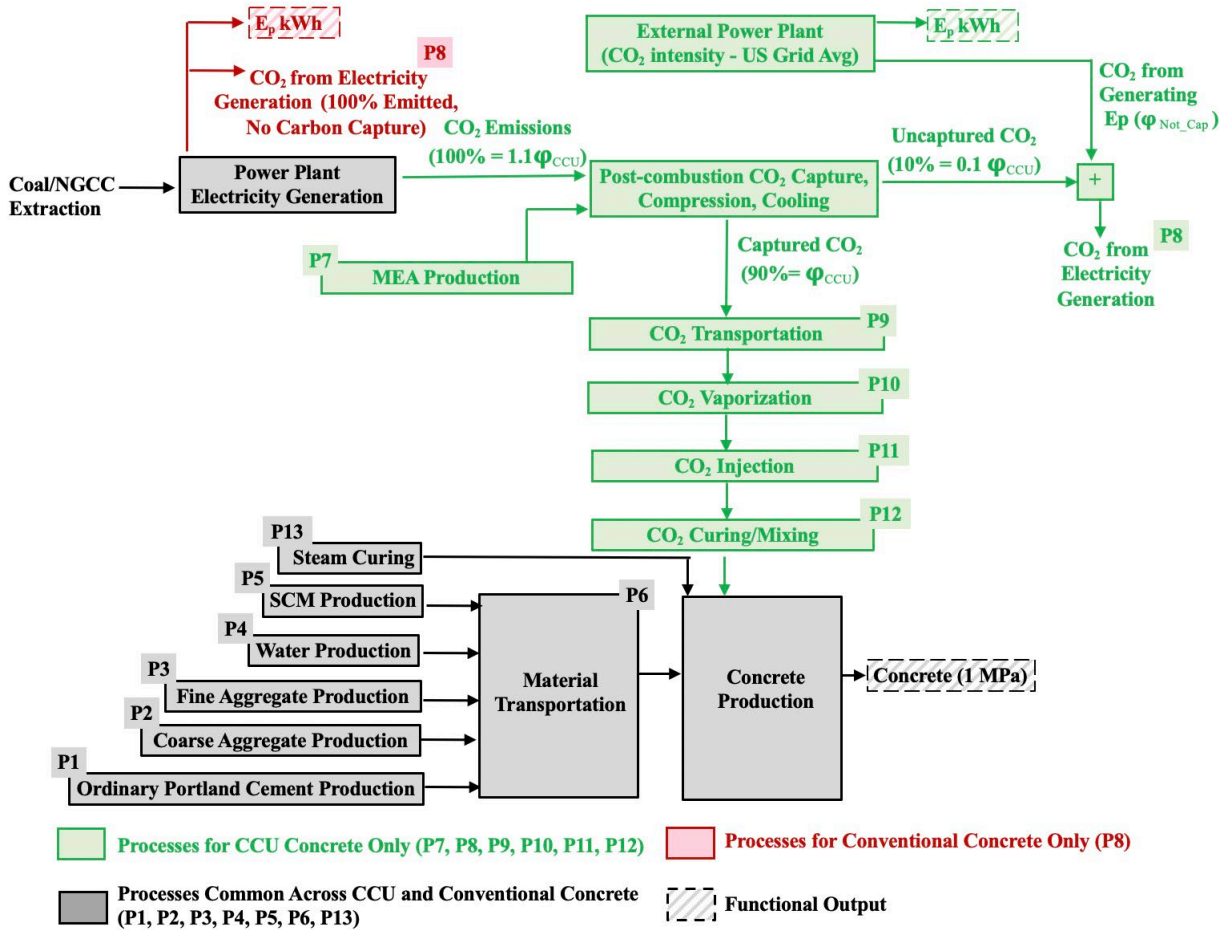


Figure 1 The processes required to produce CCU concrete are highlighted in grey and green. The processes required to manufacture conventional concrete are highlighted in grey and red. The CO₂ emissions, which is utilized in the curing or mixing of CCU concrete (ϕ_{CCU} in kg), is captured from a power plant. The energy penalty from capturing ϕ_{CCU} (E_p kWh) is compensated by an external power plant. When conventional concrete is produced,

there is no carbon capture during electricity generation and the CO₂ from generating E_p in the power plant is completely emitted. The functional unit - 1 m³ of concrete with 1 MPa strength and E_p kWh of electricity - is common across the CCU and conventional concrete production pathways. The CO₂ emissions from each of the CCU and conventional concrete production process is quantified in Equations 1 and 5.

The distribution in Figure 2(a) quantifies the likelihood of the net CO₂ benefit in dataset 1 being positive (right of the y-axis) in 10000 Monte Carlo runs. If the net CO₂ benefit is positive, then the total life cycle CO₂ emissions from producing CCU concrete (TOT_{CCU}) is lower than conventional concrete (TOT_{Conv}). For dataset 1 the likelihood is 0%, which signifies that TOT_{CCU} is greater than TOT_{Conv} in all of the 10000 Monte Carlo runs. Alternately, the distribution for the net CO₂ benefit is always negative which indicates that, on a life cycle basis, producing CCU concrete is more CO₂ intensive than conventional concrete in the 10000 Monte Carlo runs.

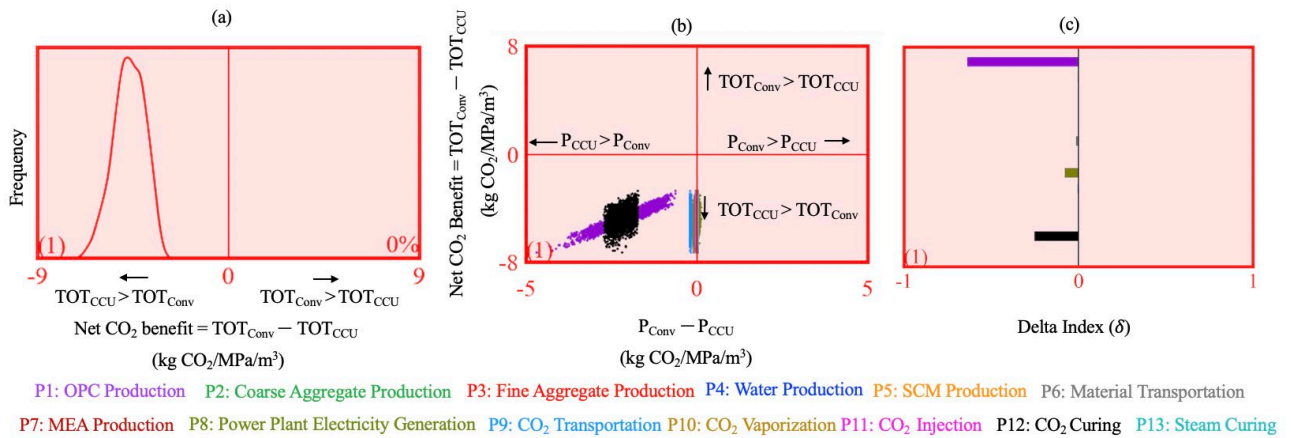


Figure 2 (a) The distribution of the net CO₂ benefit of CCU concrete in 10000 Monte Carlo runs. The net CO₂ benefit is the difference between the total CO₂ emissions from producing conventional concrete (TOT_{Conv}) and CCU concrete (TOT_{CCU}) (b) the scatter plot demonstrating the sensitivity of the net CO₂ benefit to the difference in the CO₂ emissions

from the 13 contributing processes for conventional concrete (P_{Conv}) and CCU concrete (P_{CCU}). Positive values on the y-axis indicates that $TOT_{Conv} > TOT_{CCU}$ (upper right and upper left quadrants), negative values on the y-axis indicate that $TOT_{CCU} > TOT_{Conv}$ (lower right and lower left quadrants), positive values on the x-axis indicate that $P_{Conv} > P_{CCU}$ (upper right and lower right quadrants), negative values on the x-axis indicate that $P_{CCU} > P_{Conv}$ (upper left and lower left quadrants) (c) delta indices to determine the sensitivity of the net CO₂ benefit to the difference between P_{Conv} and P_{CCU} . The number in parenthesis represents the dataset number (from the literature review) for which the results are determined. The red background in (a), (b) and (c) signifies that the net CO₂ benefit is negative (i.e. TOT_{CCU} is greater than TOT_{Conv}) in at least 5000 out of the 10000 Monte Carlo runs (i.e. likelihood greater than 50%). If the background is green, then it signifies that the net CO₂ benefit is positive with a likelihood greater than 50%.

The scatter plot analysis of the 10000 Monte Carlo runs (Figure 2b) determines the key drivers in the relationship between the difference in the CO₂ emissions from the 13 conventional concrete (P_{Conv}) and CCU concrete (P_{CCU}) processes and the net CO₂ benefit. The scatter plot shows the 10000 net CO₂ benefit values on the ordinate and the 10000 values of the difference between the CO₂ emissions from the 13 processes on the abscissa. A visual inspection of the scatter plot reveals a difference in the slopes of the scattered points. A higher slope indicates higher sensitivity of the net CO₂ benefit to the contributing process. The scatter plot for dataset 1 (Figure 2(b)) demonstrates that the difference in the CO₂ emissions from cement used (P1:OPC) has a significant slope. As the difference in CO₂ emissions from OPC production are scattered in the third quadrant, the CO₂ emissions from OPC production for CCU concrete is greater than conventional concrete. Therefore, for dataset 1, the difference between the CO₂ emissions from

149 OPC production is the most important reason for TOT_{CCU} being greater than TOT_{Conv} (i.e. the net
 150 CO_2 benefit being negative).
 151 This finding is confirmed by a moment independent sensitivity analysis,^{25,29,30} which determines
 152 δ indices for each of the 13 processes contributing to the net CO_2 benefit (Figure 2c). The δ
 153 index is a measure of the contribution from the difference in the CO_2 emissions from a
 154 conventional and CCU concrete production process to the probability distribution function of the
 155 net CO_2 benefit. The process with a greater δ index value has a greater contribution to the net
 156 CO_2 benefit than a parameter with a lower δ index value. By convention, if the δ value is
 157 negative then the CO_2 emissions from the process are greater in CCU concrete production than in
 158 conventional concrete production. The difference in the CO_2 emissions from OPC production has
 159 the highest δ value and is, therefore, the most significant contributor to the net CO_2 benefit. The
 160 negative value indicates that the CO_2 emissions from OPC production are greater in CCU
 161 concrete than in conventional concrete. The finding from the sensitivity analysis for dataset 1 can
 162 be attributed to the compressive strength of CCU concrete (16-17.4 MPa) being lower than
 163 conventional concrete (18 – 18.6 MPa). The mean compressive strength of CCU concrete (16.7
 164 MPa) is 9% lower than that of conventional concrete (18.3 MPa). This implies that in dataset 1 a
 165 greater mass of OPC is produced for CCU concrete to achieve the same compressive strength as
 166 conventional concrete. In dataset 1, the OPC produced per MPa for CCU concrete is 24.8
 167 kg/MPa and for conventional concrete is 22.6 kg/MPa.
 168 Using a mean value of 0.948 kg CO_2 /kg OPC for the life cycle CO_2 footprint of OPC (SI Table
 169 S2), the CO_2 emissions from OPC production for CCU concrete is 23.5 kg CO_2 /MPa and for
 170 conventional concrete is 21.4 kg CO_2 /MPa. Therefore, the difference between the CO_2 emissions
 171 from OPC production for CCU and conventional concrete is 2.1 kg CO_2 /MPa, which is greater

than the CO₂ utilized in CCU concrete (1.1 kg CO₂/MPa, SI Section 1). As a result, the life cycle CO₂ emissions from the increased OPC production in CCU concrete is greater than the CO₂ utilized for curing the CCU concrete.

Increased Binder Use Undermines CO₂ Benefit of CCU Concrete

We extend the analysis conducted for dataset 1 to the remaining 98 datasets. The 99 datasets are divided into 4 categories. Category 1 has 50 datasets wherein CO₂ is used for curing and the binder consists of only OPC. Category 2 has 20 datasets wherein CO₂ is used for curing and the binder consists of a mix of OPC and supplementary cementitious materials (SCM). Category 3 has 8 datasets wherein CO₂ is used for mixing and the binder consists of only OPC. Category 4 has 21 datasets wherein CO₂ is used for mixing and the binder consists of a mix of OPC and SCM.

A visual inspection of the slopes in the scatter plot in Figure 4 and the δ indices in Figure 5 reveal that the net CO₂ benefit is most sensitive to the amount of OPC produced and used in the design mix (e.g. P1 in datasets 1, 2, 3 and 4 in Figure 5), the energy used for CO₂ curing (e.g. P12 in datasets 21, 22 and 23 in Figure 5) and SCM produced and used (e.g. P5 in datasets 51, 52, 53 and 54 in Figure 5).

The plots with the red background in Figure 3, Figure 4 and Figure 5 demonstrate that the net CO₂ benefit is negative (i.e. CCU concrete has higher life cycle CO₂ emissions than conventional concrete) with at least a 50% likelihood in 56 out of the 99 datasets. The compressive strength in CCU concrete decreases due to CO₂ curing when compared to conventionally cured concrete. The OPC and SCM consumed to produce the same compressive strength is greater in CCU concrete than in conventional concrete. Therefore, the results demonstrate the CO₂ burden of increased OPC and SCM consumption for CCU concrete outweighs the benefit of the CO₂ that is

captured and used in CCU curing. Additionally, in category 1 datasets, the electricity use in the CO₂ curing process (SI Table S1, Process 12) is the second key contributor to the increase in the total life cycle CO₂ emissions from CCU concrete (e.g. datasets 46, 47 and 48 in Figure 5). The results in Figure 5 also demonstrate that the life cycle CO₂ emissions from capturing, compressing, transporting and vaporizing CO₂, and the CO₂ emissions from producing fine aggregate, coarse aggregate, water and steam curing are not significant contributors to the net CO₂ benefit. This can be attributed to the mass of CO₂ utilized in concrete being lower than the bulk materials of cement and coarse and fine aggregate (SI Figure S1) and the life cycle CO₂ intensity of coarse and fine aggregate being significantly lower than cement (SI Table S2). To investigate the change in results when CO₂ intensity of OPC production decreases, we determine the net CO₂ benefit of CCU concrete when CO₂ is captured from a cement plant (SI Section 11). The results show that CCU concrete has higher life cycle CO₂ emissions than conventional concrete (i.e. negative CO₂ benefit) in 44 out of the 99 datasets (Figure 4 in SI Section 11) when compared to 56 out of the 99 datasets in the baseline scenario (Figure 5). Therefore, when CO₂ is captured from a cement plant, there is a lower likelihood of CCU concrete producing a negative net CO₂ benefit than when CO₂ is captured from a power plant. The difference in the results can be attributed to the reduced CO₂ intensity of cement production due to CO₂ capture at the cement plant.

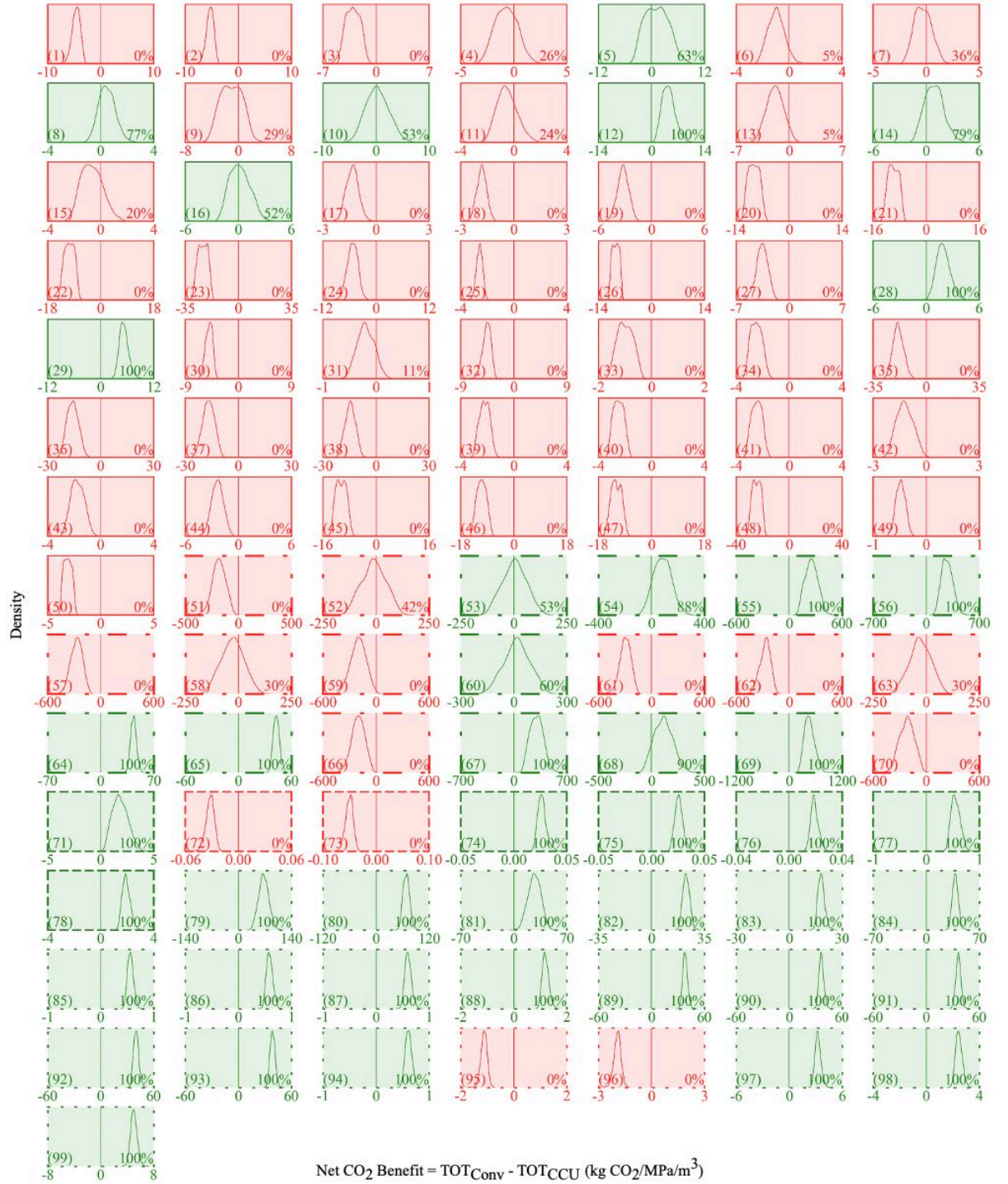
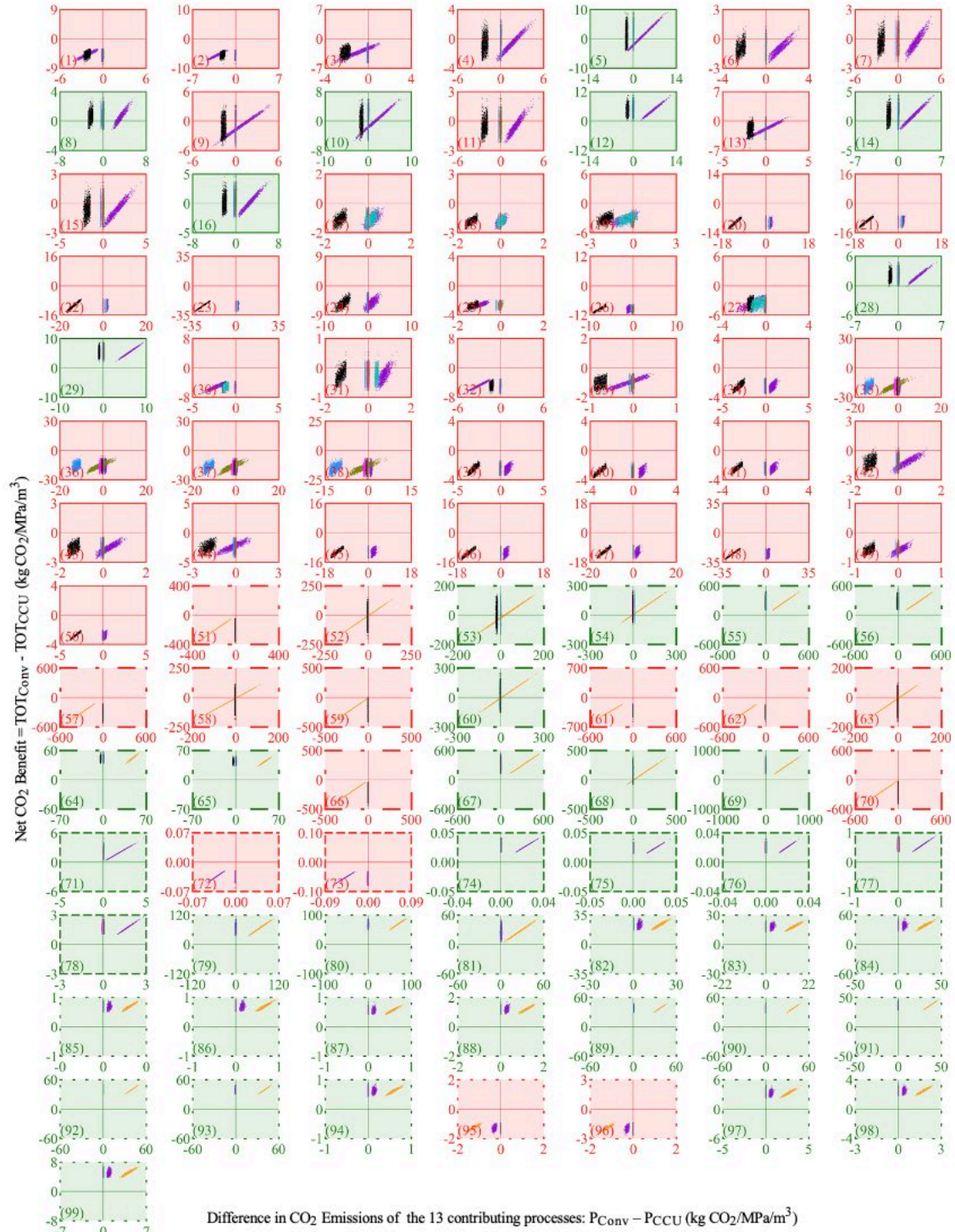


Figure 3 The net CO₂ benefit of CCU concrete production across 99 datasets. The curve in each plot represents the distribution of the net CO₂ benefit, which is the difference between

217 the total CO₂ emissions from producing conventional and CCU, across 10000 Monte Carlo
218 runs. When the net CO₂ benefit is negative in at least 5000 of the 10000 Monte Carlo runs
219 (50% likelihood), the background is red. If the background is green, then it signifies that
220 the net CO₂ benefit is positive with a likelihood greater than 50%. The likelihood is
221 presented as a percentage value in the lower right corner of the plot. The parenthesized
222 value in the lower left corner is the dataset number.

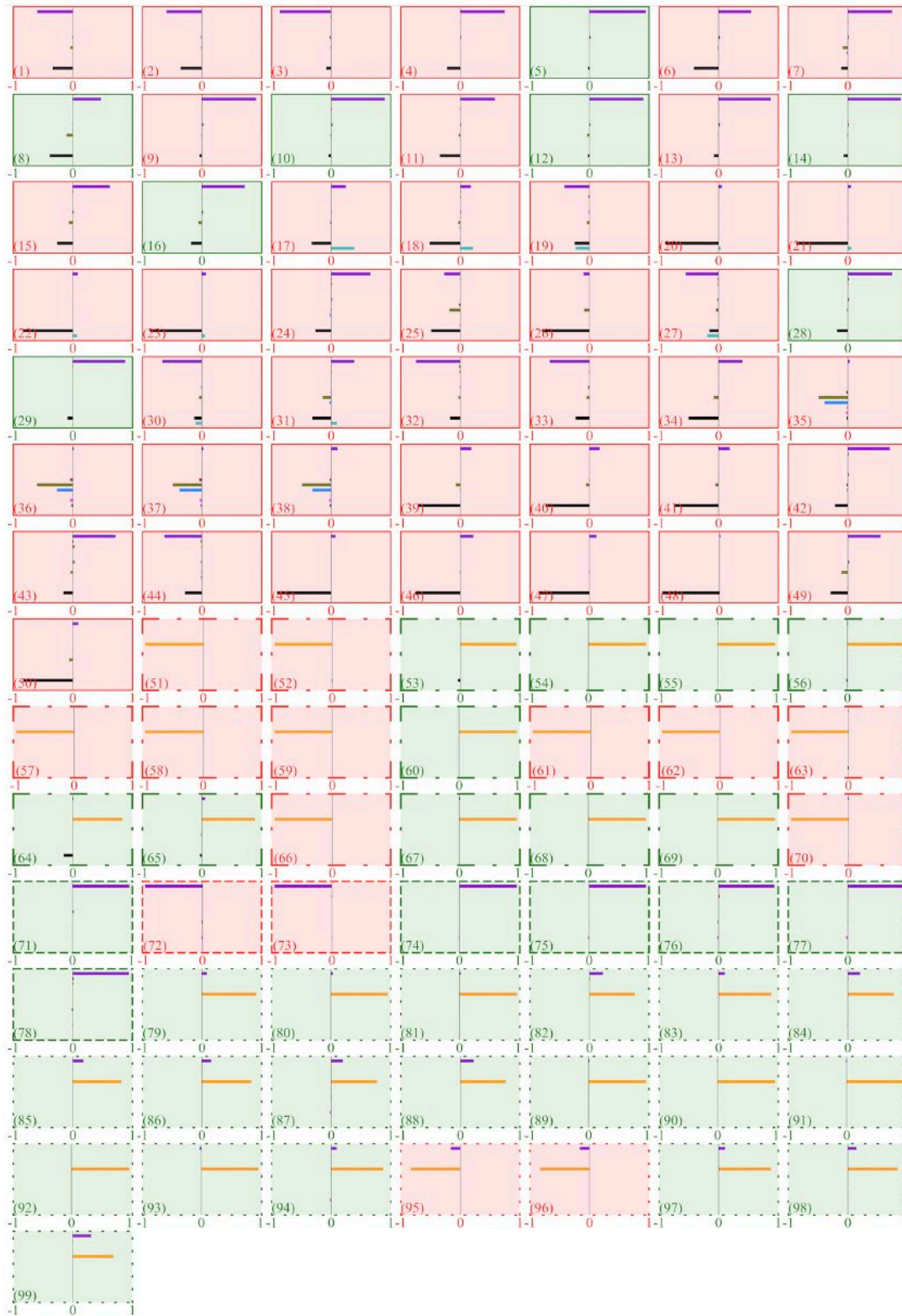
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☐ Category 1: Datasets (1) to (50): CO₂ Use: Curing, Binder: OPC Only
 ☐ Category 3: Datasets (71) to (78): CO₂ Use: Mixing, Binder: OPC Only
☐ Category 2: Datasets (51) to (70): CO₂ Use: Curing, Binder: OPC+SCM
 ☐ Category 4: Datasets (79) to (99): CO₂ Use: Mixing, Binder: OPC+SCM

P1: OPC Production P2: Coarse Aggregate Production P3: Fine Aggregate Production P4: Water Production P5: SCM Production
 P6: Material Transportation P7: MEA Production P8: Power Plant Electricity Generation P9: CO₂ Transportation P10: CO₂ Vaporization
 P11: CO₂ Injection P12: CO₂ Curing P13: Steam Curing

227 **Figure 4 Scatter plot quantifying the influence of the difference between the CO₂ emissions**
228 **from the 13 processes of CCU and conventional concrete production (x-axis) on the net**
229 **CO₂ benefit (y-axis) for the 99 datasets. The parenthesized value in the lower left corner is**
230 **the dataset number. Higher resolution scatter plots for each of the 13 processes can be**
231 **downloaded from Table S12 in the SI.**
232



☐ Category 1: Datasets (1) to (50): CO₂ Use: Curing, Binder: OPC Only
 ☐ Category 3: Datasets (71) to (78): CO₂ Use: Mixing, Binder: OPC Only
☐ Category 2: Datasets (51) to (70): CO₂ Use: Curing, Binder: OPC+SCM
 ☐ Category 4: Datasets (79) to (99): CO₂ Use: Mixing, Binder: OPC+SCM
 P1: OPC Production P2: Coarse Aggregate Production P3: Fine Aggregate Production P4: Water Production P5: SCM Production
 P6: Material Transportation P7: MEA Production P8: Power Plant Electricity Generation P9: CO₂ Transportation P10: CO₂ Vaporization
 P11: CO₂ Injection P12: CO₂ Curing P13: Steam Curing

Figure 5 δ indices quantifying influence of the difference between the CO₂ emissions from the 13 processes of CCU and conventional concrete production on the net CO₂ benefit. A process with a greater δ index value has a greater influence on the net CO₂ benefit than a process with a lower δ index value. The parenthesized value in the lower left corner is the dataset number.

Results Across CO₂ Use, Binder Type and Allocation Method

Figure 6 summarizes the results depending on whether CO₂ is used for curing or mixing, SCM is used as a binder material or not, and the type of allocation method used to determine the CO₂ emissions from producing the SCM material. Two types of SCMs are used in concrete production - ground granulated blast furnace slag and fly ash. Slag is a by-product of pig-iron production and fly ash is a by-product of electricity generation in coal power plants. As a result, we need a method to allocate total CO₂ emissions from pig-iron production between slag and pig-iron and from coal electricity generation between fly-ash and coal electricity. We use three methods - system expansion (SE), mass-based (MA) and economic value-based allocation (EA) - to allocate and account for the life cycle CO₂ emissions from producing the SCMs (process P5).

The overall results in Figure 6 demonstrate that in 35 (EA in “Overall”) to 43 (SE in “Overall”) of the 99 datasets reported in literature, CCU concrete production has a lower life cycle CO₂ emission than conventional concrete production. In these cases, CCU concrete substituting conventional concrete lowers CO₂ emissions. Negative CO₂ net benefit values are obtained in the remaining 56 to 64 datasets. A similar analysis for CO₂ capture from a NGCC power plant shows that the net CO₂ benefit is negative in 61, 65 and 68 of the 99 datasets when SE, MA and EA are used, respectively (SI Section 10). The overall results demonstrate that the CO₂ benefit of CCU concrete production is negative in 56 to 68 of the 99 datasets depending on whether CO₂ is

captured from a coal or a NGCC power plant and when SE, MA or EA is used. As a result, there is a higher likelihood of the net CO₂ benefit of CCU concrete being negative. Consequently, not all CCU concretes can help realize sequestering 0.1 to 1.4 gigatons/year of CO₂ in concrete by 2050.^{1,3}

The results of the sensitivity analysis are similar across categories 1, 2, 3 and 4 as the production and use of binder material (OPC in Categories 1 and 3 and OPC+SCM in Categories 2 and 4) has the most significant influence on the net CO₂ benefit.

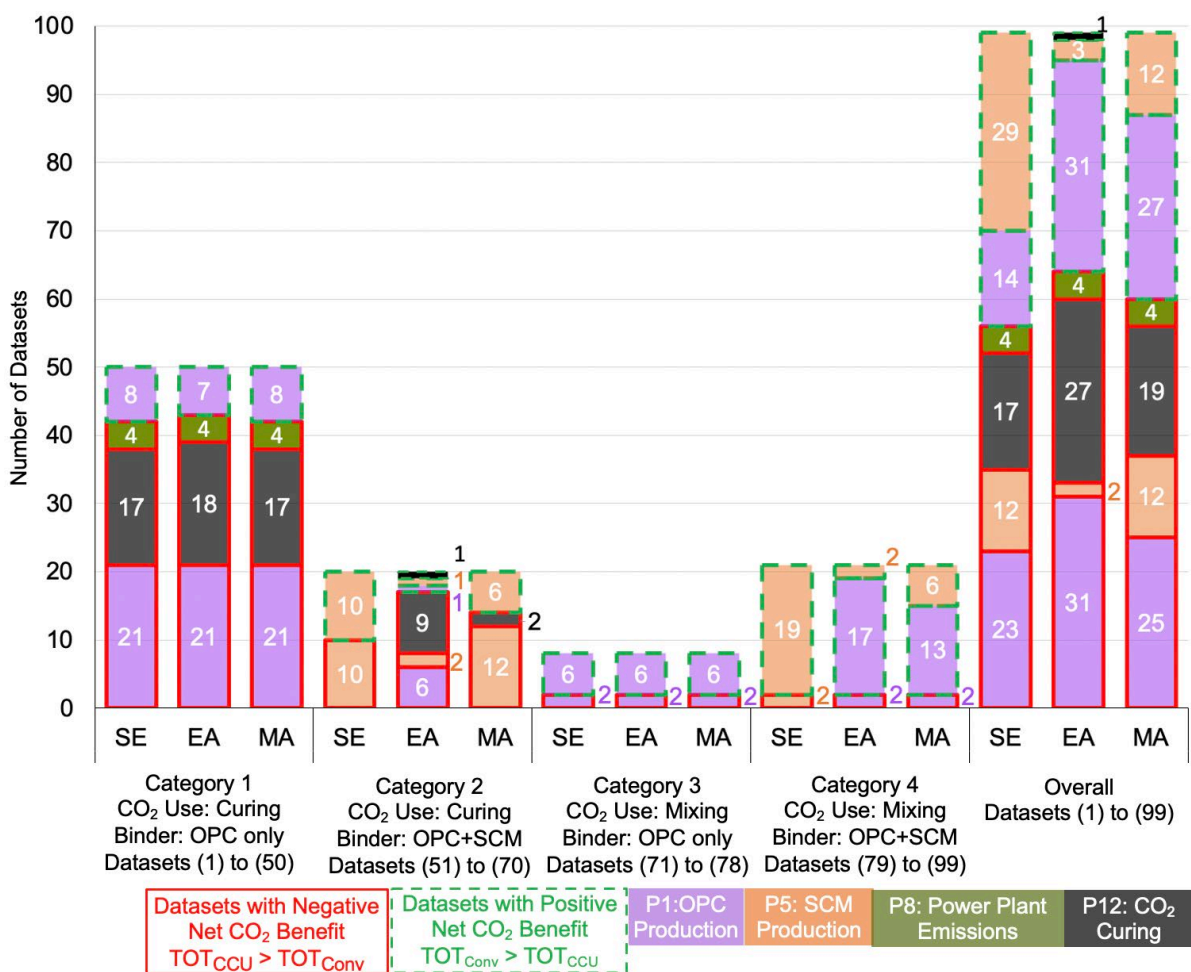


Figure 6 Results providing a break-up of the datasets with positive and negative net CO₂ benefit from CCU concrete and the most significant driver of the net CO₂ benefit of CCU

concrete across category 1, category 2, category 3 and category 4 datasets (SI Section 2).

The results were determined using system expansion (SE), economic value-based (EA) and mass-based (MA) allocation to determine the CO₂ emissions from slag (co-product of iron ore production) and fly ash (co-product of coal electricity generation), which are used as SCM in category 2 and 4 datasets. The individual plots for provided in SI Section 9.

The overall results on the number of datasets in which there is a positive net CO₂ benefit (i.e. CCU is less CO₂ intensive than conventional concrete) is not significantly impacted by the choice of system expansion SE, MA or EA. CCU is less CO₂ intensive than conventional concrete in a minimum of 35 datasets when EA is used and a maximum of 43 datasets when SE is used. However, for category 2 and 4 datasets, the choice of SE, MA or EA impacts the results from the δ indices sensitivity analysis. When SE or MA is used in category 2 datasets, SCM use is a key contributor to the net CO₂ benefit of CCU concrete. When EA is used in category 2 datasets, the CO₂ emissions from the CO₂ curing process is the key contributor. When SE is used in category 4 datasets, SCM use is a key contributor to the difference in the total CO₂ emissions between conventional and CCU concrete. When EA or MA is used in category 4 datasets, OPC use is the key contributor. However, the choice of SE, MA or EA is an artifact of the method of analysis – not actually changing real CO₂ emissions – and, therefore, is not within the purview of engineering strategies to improve processes and decrease the CO₂ emissions from producing CCU concrete.

The findings from this analysis are based on the design mixes, material usage, compressive strength and parameters such as the CO₂ curing duration, water to cement and SCM to cement ratios obtained from the 99 datasets (SI Section 2 Figures 1 to 5). It is important to note that the

findings do not preclude future research (as discussed below) from optimizing the design mixes, curing processes and material properties to increase the net CO₂ benefit from CCU concrete.

Strategies to Improve the Net CO₂ Benefit of CCU Concrete

An R&D agenda focused on the following items, which are within the control of the CCU concrete production process, can increase the net CO₂ benefit.

- (i) Ensure increase in compressive strength from CO₂ curing: A key priority is to determine a CO₂ curing protocol that consistently increases compressive strength of CCU concrete. An increase in compressive strength implies that a smaller quantity of carbon intensive binder material is used in CCU concrete to achieve the same compressive strength as conventional concrete (i.e. lower quantity of OPC or SCM is consumed on a kg per MPa basis). Fine tuning the curing process such as duration of the pre-hydration and post-carbonation water compensation are promising candidates to restore the reduction in 28-day compressive strength observed for CO₂-cured concrete.³²⁻³⁴ For example, a longer duration of pre-hydration may enhance 28-day compressive strength but decreases CO₂ uptake at early age. Further investigations are needed for enhancing consistency of CO₂-cured concrete production and for implementing the laboratory strategies in field applications.
- (ii) Decrease the CO₂ emissions from the CO₂ curing process: Electricity use, which is the key contributor to the CO₂ emitted during the CO₂ curing process, can be lowered by streamlining the curing process. Future research can investigate and standardize promising options such as natural drying or waste heat drying for pre-curing of CO₂-cured concrete³⁵ with the end goal of accelerating adoption in industry.

(iii) Improve understanding on the impact of CO₂ curing on durability: The findings of this research are based on the compressive strength property of CCU concrete, which is limited from a lifecycle perspective. Prior studies show that construction and repair frequencies are key drivers in determining concrete life cycle CO₂ impacts.^{36,37} Therefore, the effect of CCU on concrete durability must be considered when analyzing life cycle CO₂ emissions. Preliminary lab scale studies demonstrate that CO₂ curing improves durability related parameters such as permeability, sorptivity and sulfate and acid resistance.^{22,38-40} However, the variability in the curing conditions and the design mixes used in the studies should be accounted for to ensure that durability gains can be consistently realized when CO₂ curing of concrete is adopted at a commercial scale. Future work can prioritize standardizing the CO₂ curing protocol (e.g. the steam curing time, pre-hydration time, post-hydration time) and study the resulting durability impact on different design mixes (e.g. use of different SCMs), with the overall goal of identifying optimal curing conditions and design mixes to maximize durability. This applies to ready-mix concrete and general precast applications with end-products such as masonry units,^{32,35} pipes²² and pavers.⁴¹ In addition, CO₂ curing for reinforced concrete needs further investigation due to the possibility of increased risk of steel reinforcement corrosion led by concrete carbonation. Moreover, CCU can be potentially synergized with established strategies for concrete crack width control, e.g., engineered cementitious composites with microfiber reinforcement, to further promoting concrete durability.^{42,43}

The system boundary (Figure 1) assumes that the CO₂ captured from the power plant is used for CCU concrete production without any intermediate storage. In practice, the total CO₂ captured from a power plant may be significantly greater than the maximum utilization capacity at a CCU

concrete production plant. In such cases, the excess captured CO₂ may be temporarily stored for future utilization in CCU concrete production or routed towards other utilization pathways. Given that CO₂ utilization is an emerging field and in the early stages of commercialization, there is a lack of time-sensitive data on how the captured CO₂ feedstock is either temporarily stored or immediately allocated to other utilization pathways. As a result, a system boundary that incorporates the time-sensitive utilization of CO₂ captured from a power plant is beyond the scope of this work and is a topic for future research.

The transport of CO₂ through pipelines has a lower CO₂ impact than road-based transport using semi-trailer trucks,⁴⁴ which is modeled in this analysis. To quantify the maximum possible gains from shifting to a less carbon intensive mode of CO₂ transport, we conduct a scenario analysis with the optimistic assumption that the CO₂ impact of CO₂ transportation is zero (SI Section 12). Despite this optimistic assumption of zero-carbon CO₂ transport, CCU concrete has a lower CO₂ impact than conventional concrete in 44 of the 99 datasets, which is similar to the 43 of the 99 datasets obtained in the baseline scenario (SI Figure S10 versus Figure 5). As a result, a shift from road to pipeline based CO₂ transport will not impact the findings from this analysis.

This analysis focusses on the use of pure CO₂ and two approaches of CO₂ utilization - curing and mixing - as they are more extensively investigated (e.g. 99 datasets used in this study) than alternate approaches such as concrete curing with flue gas,^{21,45-47} carbonation of recycled concrete aggregates,⁹ CO₂ sequestration in alternative MgO based binders,¹⁰ and CO₂ dissolution in mixing water.^{13,14} The increased availability of experimental data is necessary to robustly quantify the net CO₂ benefit of CCU concrete and account for the impact of data uncertainty and process variability on the results. For example, further experimental research can generate data on the variability in the compressive strength of flue-gas cured concrete properties when the 2-

week curing time is reduced⁴⁷ and the CO₂ concentration in the flue gas is varied.⁴⁵ With increased availability of inventory and process data from future experimental research, the life cycle approach presented in this study can be extended to quantify the net CO₂ benefit of CCU concrete produced from flue gas and other alternate approaches of CO₂ utilization. The impact of CO₂ curing or CO₂ mixing on the lifetime behaviors and geography specific factors are important practical considerations when using CCU concrete in commercial applications. The lifetime behaviors are impacted by the variations in the geographical sources of raw materials, mix type, product type and the service environment in which the concrete is deployed. For example, the impact of CO₂ curing on concrete lifespan would differ substantially between concrete with and without steel reinforcement, due to the heightened steel corrosion⁴⁸ caused by the CO₂-induced pH reduction.⁴⁹ Additionally, geography specific factors such as sulfate-rich soils,⁵⁰ cold regions,³⁹ or acidic environments²² can impact the durability of CCU concrete. The findings of this study can be further complemented by future research which quantifies the impact of variations in the lifetime and the geography specific factors on the net CO₂ benefit of CCU concrete.

Methods

Literature Review to Categorize CO₂ Use in Concrete

We conducted a literature review to obtain the 99 datasets from 19 studies presenting life cycle material and energy inventory data and process parameters for the production of CCU and conventional concrete. The literature review identified the 19 studies^{16,19,22,23,31-33,35,38,40,51-59} as they were the only ones to report the following three items (i) the design mix consisting of the energy and material inventory required for the production of conventional and CCU concrete (SI Section 2). The energy and material inventory are required to determine the life cycle CO₂

impact of producing conventional and CCU concrete; (ii) the quantity of CO₂ used in mixing or curing of concrete. This is required to determine the life cycle CO₂ impact of capturing, transporting and utilizing the CO₂ used in producing CCU concrete; and (iii) the compressive strength of CCU and conventional concrete at the end of 28 days, which helps account for the change in the material property between conventional and CCU concrete. The 28-day compressive strength is among the most widely used technical parameters for assessing concrete quality, categorizing concrete mix designs⁶⁰ and forms the basis for concrete structural design^{61,62} and is, therefore, chosen as the functional property based on which conventional and CCU concrete are compared. Based on whether CO₂ is used in CCU concrete for curing or mixing and if SCM was used in the design mix, the 99 datasets were organized into four categories.

- (i) Category 1: CO₂ is used in the curing of concrete and only OPC is used as the cementitious material in the design mix.^{22,31,33,38,40,56-59} This category contains 50 datasets.
- (ii) Category 2: CO₂ is used in the curing of concrete and a combination of OPC and SCM is used as the cementitious material in the design mix.^{23,32,35,55} This category contains 20 datasets.
- (iii) Category 3: CO₂ is used in the mixing of concrete and only OPC is used as the cementitious material in the design mix.^{16,19,51} This category contains 8 datasets.
- (iv) Category 4: CO₂ is used in the mixing of concrete and a combination of OPC and SCM is used as the cementitious material in the design mix.^{16,51-54} This category contains 21 datasets.

SCM took the form of either ground granulated blast furnace slag, which is a by-product of the pig-iron production,⁶³ or fly ash, which is a by-product of electricity generation in coal power plants.

Functional Unit

The use of CO₂ during mixing or curing changes the compressive strength of CCU concrete when compared to concrete produced through conventional mixing or curing. In addition, an energy penalty (E_p kWh) is incurred for CCU concrete in power plants due to the energy associated with capturing the CO₂, which is used in the curing or mixing of CCU concrete (ϕ_{CCU} , kg CO₂). E_p is not incurred when conventional concrete is produced since there is no CO₂ capture. Therefore, the net CO₂ benefit of substituting CCU concrete for conventional concrete should account for the CO₂ impact from the change in the compressive strength and E_p , which is incurred in power plants only when CO₂ is captured.

As a result, we use a functional unit of concrete with 1 MPa compressive strength and 1 m³ of volume and E_p kWh of electricity.

The functional unit accounts for the change in compressive strength and ensures consistency by normalizing the materials and energy consumed for producing 1 m³ of CCU and conventional concrete to 1 MPa of compressive strength. The inclusion of E_p kWh of electricity in the functional unit accounts for the difference in CO₂ emissions from electricity generation without CO₂ capture in the conventional concrete pathway and with CO₂ capture in the CCU concrete pathway. E_p is determined based on the mass of CO₂ captured from the power plant (SI Table S1 Process 8).

CCU Concrete Production – System Boundary and CO₂ Emissions

The literature review revealed that the total life cycle CO₂ emissions from producing CCU concrete is the sum of the CO₂ emissions from 13 key processes required to capture, transport and utilize CO₂ and produce the materials required in the design mix of concrete (Figure 1).

The expression used to determine the total life cycle CO₂ emissions from producing CCU concrete based on the CO₂ emissions from the 13 processes is presented in Equation 1. The 13 expressions within parenthesis in Equation 1 correspond to the CO₂ emissions from the 13 processes (Figure 1).

$$\begin{aligned} \text{TOT}_{\text{CCU}} = & (\varphi_{\text{C}} * \text{C}_{\text{CCU}}) + (\varphi_{\text{CA}} * \text{CA}_{\text{CCU}}) + (\varphi_{\text{FA}} * \text{FA}_{\text{CCU}}) + (\varphi_{\text{W}} * \text{W}_{\text{CCU}}) + (\varphi_{\text{SCM}} * \text{SCM}_{\text{CCU}}) \\ & + (\text{D}_{\text{M}} * \varphi_{\text{TM}} * \text{M}_{\text{Conv}}) + (\varphi_{\text{CCU}} * \varphi_{\text{MEA}}) + (\text{Alloc}_{\text{elec}} * \varphi_{\text{Not Cap}} + \varphi_{\text{Avg}} * \text{E}_{\text{p}}) \\ & + (\varphi_{\text{CCU}} * (1+2\text{T}_{\text{w}}) * \text{D}_{\text{CO}_2} * \varphi_{\text{T}}) + (\varphi_{\text{CCU}} * \varphi_{\text{Vap}}) + (\varphi_{\text{CCU}} * (\varphi_{\text{Inj}} + (1-\eta))) + (\varphi_{\text{CO}_2_ \text{Cur}}) \\ & + (\varphi_{\text{Stm_Cur}}) \end{aligned} \quad \text{Equation 1}$$

Process 1 to 4 – Ordinary Portland cement (C), coarse aggregate (CA), fine aggregate (FA), and water (W) production: The CO₂ impact is the product of (i) the life cycle CO₂ emissions from producing the material (φ_{C} , φ_{FA} , φ_{CA} and φ_{W} in kg CO₂/kg material) and (ii) and the mass of material used in the design mix normalized to the compressive strength of CCU concrete (C_{CCU} , CA_{CCU} , FA_{CCU} and W_{CCU} in kg material/MPa/m³). The material used and the compressive strength are obtained from the literature review (SI Section 2) and φ_{C} , φ_{FA} , φ_{CA} and φ_{W} are obtained from the ecoinvent database (SI Table S2).

Process 5 – SCM production: SCM_{CCU} represents the mass of SCM used in the design mix normalized to the compressive strength of CCU concrete (in kg material/MPa/m³).

Slag and fly ash, which are co-products of iron-ore production and electricity generation from coal, are used as SCM in the design mix of concrete. Three methods - system expansion (SE),

economic value-based allocation (EA) and mass-based allocation (MA) – are widely used in LCA to determine the CO₂ emissions of co-products being generated by a single system. In SE, the CO₂ emissions from producing a required mass of slag is determined by expanding the system to include the production of a corresponding mass of iron-ore (based on a ratio of iron-ore to slag, SI Section 4). In the case of MA and EA, the total CO₂ emission from a process producing both iron-ore and slag is allocated between iron-ore and slag based on the mass and economic value of the co-products, respectively (SI Sections 5 and 6). To explore variability in the CO₂ emissions from CCU concrete production based on the allocation method, this analysis applies the three methods when determining the CO₂ emissions for slag and fly ash.

The CO₂ impact of slag (Φ_{SCM_slag} in kg CO₂/kg slag) is determined from Equation 2

$$\Phi_{SCM_slag} = Alloc_{slag} * 7.7 * \Phi_{IO} \quad \text{Equation 2}$$

The value of $Alloc_{slag}$ is 1, 0.008 or 0.11 when SE, MA or EA is chosen, respectively (SI Sections 4, 5 and 6).

Φ_{IO} is the life cycle CO₂ emissions from producing 1 kg of iron ore and is 2.2 kg CO₂/kg iron ore (SI Section 4).

When fly ash is used as the SCM, the CO₂ impact per kg of fly ash (Φ_{SCM_ash} in kg CO₂/kg fly ash) is determined from Equation 3

$$\Phi_{SCM_ash} = Alloc_{ash} * 22.7 * \Phi_{Elec_Coal} * \alpha_{Cap} \quad \text{Equation 3}$$

The value of $Alloc_{ash}$ is 1, 0.02 or 0.06 when SE, MA or EA is chosen, respectively (SI Sections 4, 5 and 6). Φ_{Elec_Coal} , which is the life cycle CO₂ emission from producing 1 kWh of coal electricity, is 1.25 kg CO₂/kWh (SI Section 4). α_{Cap} is 0.1 if CO₂ is captured at a coal plant and

467 used in CCU concrete production. α_{Cap} is 1 if there is no carbon capture at a coal plant i.e. when
 468 CO₂ is captured from a combined cycle natural gas plant and used in CCU concrete production.
 469 Process 6 - Material Transportation: The CO₂ emissions from material transport is the product of
 470 the 5 materials used in the design mix (M_{CCU} in kg/MPa/m³), the CO₂ intensity of the mode of
 471 transportation used (φ_M in kg CO₂ per kg-km) and the distance over which the materials are
 472 transported (D_M in km). M_{CCU} represents C_{CCU} , F_{ACCU} , CA_{CCU} , W_{CCU} and SCM_{CCU} from
 473 processes 1 to 5. D_M values for road, rail, ocean and barge transport are obtained from the
 474 national average values for the US concrete industry (SI Section 7).⁶⁰ φ_M for the four
 475 transportation modes are obtained from the Ecoinvent database (SI Section 7).
 476 Process 7 – Monoethanolamine (MEA) Production: The CO₂ impact of carbon capture is the
 477 product of the mass of CO₂ which is captured and used in the curing or mixing of CCU concrete
 478 (φ_{CCU} , kg CO₂) and the life cycle CO₂ emissions from producing a monoethanolamine (MEA)
 479 post-combustion CO₂ capture system (φ_{MEA}). φ_{MEA} is obtained from the literature review of 21
 480 studies^{44,64-83} (SI Section 3).
 481 MEA systems are considered as they capture CO₂ with a high efficiency (90%),^{64,65,84} capture
 482 CO₂ from dilute concentrations,⁸⁵ are retrofittable to power plants currently in operation and are
 483 a commercially mature technology.^{86,87} The power sector accounts for 28% of the overall CO₂
 484 emissions in the U.S.⁸⁸ and is, therefore, a good candidate for carbon capture. As a result, we
 485 consider CO₂ capture from power plants. Post-combustion capture is considered as it more
 486 commonly deployed than oxy-fuel and pre-combustion systems.^{65,85} The reader can refer to^{65,85}
 487 for further details on the underlying physical principles of carbon capture using MEA, which is
 488 beyond the scope of this work.

Process 8 – Power plant electricity generation: When CCU concrete is produced, the total CO₂ emissions from the power plant is the sum of two components.

$$(\text{Alloc}_{\text{elec}} * \varphi_{\text{Not Cap}} + \varphi_{\text{Avg}} * E_p)$$

$\text{Alloc}_{\text{elec}}$ quantifies the allocation of CO₂ emissions from a coal power plant between the co-products of electricity and fly ash, which is used as SCM in concrete production in certain datasets. $\text{Alloc}_{\text{elec}}$ is 0.98 or 0.94 as economic or mass allocation allocates 0.02 and 0.06 of the total CO₂ emissions from the coal power plant to the co-product of fly ash (SI Sections 5 and 6). $\text{Alloc}_{\text{elec}}$ is 1 when electricity is sourced from a combined cycle natural gas power plant or when system boundary expansion is used (instead of economic or mass allocation). $\varphi_{\text{Not Cap}}$ accounts for the 10% of CO₂ which is not captured as the capture efficiency of the MEA system is 90%.^{64,65,84} The second component accounts for the CO₂ emissions from compensating for the energy penalty (E_p in kWh), which is incurred when CO₂ is captured from a power plant. The second component is the product of E_p and the CO₂ intensity of the electricity used to compensate for E_p (φ_{Avg} in kg CO₂/kWh).

E_p is quantified as follows

$$E_p = \varphi_{\text{CCU}} * [(\text{heat}_{\text{ccu}} * \text{hte} * 0.277) + E_{\text{pump}} + E_{\text{liq}}] \quad \text{Equation 4}$$

φ_{CCU} is the mass of CO₂, which is captured from a power plant and utilized in CCU concrete production. heat_{ccu} represents the heat required to regenerate the MEA (2.7 to 3.3 MJ/kg CO₂, SI Table S5), which could have alternately been used to generate electricity in the power plant.^{70,89-91} hte is the heat to electricity factor (0.09 to 0.25, SI Table S5), which is used to determine the electricity equivalent of heat_{ccu} . E_{pump} is the electricity required to power the pumps and fans in

510 the carbon capture unit (16.6 to 30.6×10^{-3} kWh/kg CO₂, SI Table S5) and E_{liq} is the electricity
 511 required to liquify the captured CO₂ (0.089 kWh/kg CO₂, SI Section 3).
 512 This analysis follows the standards recommended by the National Energy Technology
 513 Laboratory (NETL)⁹² to determine the CO₂ intensity of the electricity used to compensate for the
 514 energy penalty. NETL recommends that the energy penalty is compensated through an external
 515 electricity source, which is representative of the grid-mix of the region in which the analysis is
 516 carried out.⁹² φ_{Avg} varies between 0.38 and to 0.56 kg CO₂/kWh, which represents the lower and
 517 upper limit of the average CO₂ intensity of electricity generated in the different grid regions in
 518 the US in 2020.⁹²
 519 Process 9 - CO₂ Transportation: This analysis assumes that the captured CO₂ is transported in a
 520 semi-trailer truck (SI Section 2 “CO₂ Transportation”) as it is necessary to supply the CO₂ from
 521 the site of capture to geographically disperse concrete curing or mixing facilities, which are
 522 primarily accessible by road.²¹ The CO₂ emissions from transporting CO₂ is the product of the
 523 total weight (φ_{CCU} and the tare weight), the distance over which the transport occurs (D_{CO_2} in km)
 524 and the CO₂ intensity of transportation emissions of a semi-trailer truck ($\varphi_T = 112$ g CO₂ per ton
 525 km, SI Table S11). The transport of 1 kg of CO₂ necessitates the transport of an additional tare
 526 weight (T_w) of 0.4 kg in the onward trip to the CCU concrete production facility (SI Section 3).
 527 In the return trip, we account for the CO₂ emissions from the transport of only the tare weight.
 528 As a result, T_w equals 0.8 . We assume D_{CO_2} to be 810 km, which is equal to the longest distance
 529 by which CO₂ can be transported in the U.S.⁹³
 530 Processes 10 and 11 - Vaporization and injection of CO₂: After transportation, the liquified CO₂
 531 needs to be vaporized to a gaseous state and injected into the concrete sample for curing or
 532 mixing.⁹⁴ The CO₂ emissions from vaporizing (φ_{vap}) and injecting CO₂ (φ_{inj}) is the product of

533 ϕ_{CCU} (kg CO₂), ϕ_{Avg} (kg CO₂/kWh) and the electricity required to vaporize (5.3×10^{-3} kWh/kg
 534 CO₂, SI Section 3) and inject CO₂ (37×10^{-3} kWh/kg CO₂),¹⁶ respectively. η is the CO₂
 535 absorption efficiency and represents the portion of the total CO₂ which is absorbed during
 536 mixing or curing of concrete (datasets 71 to 99). η varies between 50% to 85% during
 537 mixing.^{16,19,52} For curing, η is equal to 1 (i.e. 100% absorption) as the curing datasets (datasets 1
 538 to 70) report CO₂ utilized as the ratio of the mass of CO₂ absorbed to the mass of cement.
 539 Processes 12 and 13 - CO₂ and steam curing: The CO₂ emissions from CO₂ curing of the
 540 concrete sample ($\phi_{CO_2_Cur}$) is the product of ϕ_{CCU} (kg CO₂), ϕ_{Avg} (kg CO₂/kWh), the electrical
 541 power requirements of the curing chamber ($P_{CO_2_Cur} = 38.8$ kW/m³ of concrete)^{35,95} and the
 542 duration of curing ($t_{CO_2_Cur}$ in hours, SI Section 2), which is determined from the literature
 543 review.^{38,96} $\phi_{CO_2_Cur}$ is normalized to the compressive strength of the concrete sample. In some
 544 datasets, a combination of steam and CO₂ curing is used for the production of CCU concrete. In
 545 this case, the analysis includes the CO₂ emissions from steam curing of CCU concrete. The CO₂
 546 emissions from steam curing (ϕ_{Stm_Cur}) is the product of CO₂ intensity of steam curing (39.55 kg
 547 CO₂/m³/hour, SI Table S8) and the duration of steam curing (t_{stm_Cur} in hours), which is
 548 determined from the literature (SI Table S1 Process 13). ϕ_{Stm_Cur} is normalized to the
 549 compressive strength of the concrete sample.
 550 When CO₂ is used for mixing of concrete (datasets in category 3 and 4), the CO₂ emissions from
 551 CO₂ and steam curing are assumed to be zero as CO₂ curing of concrete is not conducted.
 552 **Conventional Concrete Production CO₂ Emissions**
 553 The total life cycle CO₂ emissions from producing conventional concrete (TOT_{Conv}) are similarly
 554 quantified in Equation 5.

$$\begin{aligned} \text{TOT}_{\text{Conv}} = & (\varphi_{\text{C}} * \text{C}_{\text{Conv}}) + (\varphi_{\text{CA}} * \text{CA}_{\text{conv}}) + (\varphi_{\text{FA}} * \text{FA}_{\text{conv}}) + (\varphi_{\text{W}} * \text{W}_{\text{conv}}) + (\varphi_{\text{SCM}} * \text{SCM}_{\text{conv}}) \\ & + (\text{E}_{\text{p}} * \varphi_{\text{Pow_plant}} * \text{Alloc}_{\text{elec}}) + \varphi_{\text{Stm_Cur}} + (\text{D}_{\text{M}} * \varphi_{\text{TM}} * \text{M}_{\text{Conv}}) \end{aligned} \quad \text{Equation 5}$$

($\text{E}_{\text{p}} * \varphi_{\text{PP}} * \text{Alloc}_{\text{elec}}$) quantifies the CO₂ emissions from generating E_{p} kWh of electricity in a power plant without carbon capture. $\varphi_{\text{Pow_Plant}}$ is the CO₂ intensity of electricity generated in a coal or NGCC plant (kg CO₂/kWh, [SI Table S1](#)).

Net CO₂ Benefit and Sensitivity Analysis

The difference between the TOT_{CCU} (Equation 1) and TOT_{Conv} (Equation 5) determines the net CO₂ benefit from CCU concrete substituting conventional concrete.

$$\text{Net CO}_2 \text{ Benefit} = \text{TOT}_{\text{Conv}} - \text{TOT}_{\text{CCU}} \quad \text{Equation 6}$$

TOT_{CCU} and TOT_{Conv} are driven by the CO₂ emissions from the 13 processes, which, in turn, are impacted by the uncertainty and variability in the underlying parameters ([SI Table S1](#)).

In the scatter plot analysis, 10000 values are stochastically generated for the material and inventory items and the parameters for the 13 processes, which are obtained from the dataset (ranges and relationships presented in [SI Table S1](#)). The stochastically generated values are applied in Equations 1, 5 and 6 to determine the CO₂ emissions from the 13 processes for conventional and CCU concrete and the net CO₂ benefit. The net CO₂ benefit is plotted on the ordinate. The difference between the CO₂ emissions for each of the 13 contributing processes in conventional and concrete is plotted on the abscissa.

To further verify the results, this analysis conducts a moment independent sensitivity analysis^{25,29,30,97} to determine the process (from the 13 processes) having the most impact on the net CO₂ benefit. The moment independent sensitivity analysis determines an δ index for each of the 13 processes. The δ index quantifies the relative contribution of each of the 13 processes to

the probability distribution function of the net CO₂ benefit. The moment independent sensitivity analysis offers methodological advantages as it accounts for the correlation between the input parameters for the 13 processes and is applicable when the input parameters and the output are not linearly related.⁹⁸ This study determines the δ indices over 10000 Monte Carlo runs based on the approach presented in Wei, Lu and Yuan.⁹⁷

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