

# Insights in negative emission technologies 2021

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# Insights in negative emission technologies: 2021

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# Editorial: Insights in negative emission technologies: 2021

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## KEYWORDS

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## Editorial on the Research Topic

### Insights in negative emission technologies: 2021

We are delighted to present *Insights in Negative Emission Technologies: 2021* a Research Topic in *Frontiers in Climate*. These manuscripts showcase new insights, novel developments, current challenges, latest discoveries, recent advances, and future perspectives in the field. Our specific goal was to shed light on the progress made in the past decade in Negative Emission Technologies (NETs), and on its future challenges. We are sure that this article collection will inspire, inform and provide direction and guidance to researchers in the field. We present 9 articles from 36 authors from a wide range of topics including geochemical NETs, ocean-based approaches, biomass energy carbon capture and storage (BECCS), durable carbon utilization, and policy.

The original research piece by [Freer et al.](#) use a bespoke model to interrogate the supply chains of three BECCS case-studies, with specific focus on supply-chain emissions from siting BECCS facilities in the United Kingdom.

This is followed by the perspective of [Nehler and Fridahl](#) who make the case for improvements to EU regulation for BECCS, particularly allowing member states to use negative emissions from BECCS in their climate obligations, the exemption of leakage of biogenic CO<sub>2</sub>, and removing regulatory barriers.

In their review article, [Honegger et al.](#) dive deeply into governance principles of carbon dioxide removal to inform policy development. They provide a review of the concept of governance principles, international governance in the context of climate change mitigation (e.g., policy regime of UN Framework Convention on Climate Change), environmental integrity, fair-share efforts, national siting, and public participation/deliberation. They use this deliberation to explore policy considerations for direct air capture, and suggest “a less holistic perspective on CDR policy proposals focusing on techno-economic factors alone would fail to capture such [interlinked governance principles].”

[Woodall and McCormick](#) explore the application of the “Aines Principle,” proposed previously in a 2020 Lawrence Livermore National Laboratory report ([Sandalow et al., 2021](#)), and named after Associate Editor Roger Aines, states that “at some carbon [removal] price, the revenue generated from CO<sub>2</sub> removal will exceed the revenue generated from energy production from a given bioconversion process.” In two case studies, that threshold was found to be \$130 tCO<sub>2</sub><sup>-1</sup> for municipal solid waste, and about \$200–400 tCO<sub>2</sub><sup>-1</sup> for a range of liquid or hydrogen-based fuel conversion pathways.

While highly ephemeral carbon storage (of say <100 years) might have value for emissions reduction, its utility for negative emissions is limited. It is within this context that actors within the voluntary offset market are navigating, with an apparent need to account for durability within purchases. The perspective from Wenger et al. proposes a ton-year accounting frame work as a tool for comparison.

Storage of CO<sub>2</sub> in mineral-based products within the construction sector offers a scalable, and potentially marketable, vector within existing supply chains, at least this is the basis of the perspective piece by Sick et al.. The authors suggest that multiple gigatonnes of CO<sub>2</sub> may be possible with a market value of around \$1 trillion yr<sup>-1</sup> by 2050.

The oceans are an important environment within the Earth's climate and offers considerable potential for atmospheric CO<sub>2</sub> removal. This potential was explored by a recent report from the National Academies of Science, Engineering and Medicine (Committee on A Research Strategy for Ocean-based Carbon Dioxide Removal and Sequestration, Ocean Studies Board, Division on Earth and Life Studies, and National Academies of Sciences, Engineering, and Medicine, 2022), which call for an acceleration of research. Williamson and Gattuso compliment this with a review of the potential of “blue carbon” ecosystems (e.g., mangrove forests, seagrass meadows, and tidal saltmarshes). While blue carbon ecosystems are important habitats for biodiversity, spatially dense long-term carbon sinks, they have reportedly limited scalability (Committee on Developing a Research Agenda for Carbon Dioxide Removal and Reliable Sequestration, Board on Atmospheric Sciences and Climate, Board on Energy and Environmental Systems, Board on Agriculture and Natural Resources, Board on Earth Sciences and Resources, Board on Chemical Sciences and Technology, Ocean Studies Board, Division on Earth and Life Studies, and National Academies of Sciences, Engineering, and Medicine, 2019), Williamson and Gattuso argue that the uncertainty in how these systems accumulate, store, or transport carbon, makes previous cost estimates similarly uncertain.

Finally, a two-part contribution considers the potential of geochemical-based NETs. The topic is reviewed in detail by Campbell et al., presenting an overview of the reaction chemistries,

implications for resource availability, kinetics, technology function, novel applications of biotechnology, considerations for life-cycle assessment, and an appraisal of the current state of the field. In Part 2, as perspective, Maesano et al. present a roadmap for the research, development, and large-scale deployment of geochemical NETs. They highlight bottlenecks to progress including the technical readiness, social license to operate, demand and supply, human capital, and infrastructure. They also consider actions and opportunities to overcome these limitations and propose a set of near-term priorities for research and development.

Since 2011, NETs have grown from a fringe interest of the climate change mitigation community, to an integral part of proposals to meet climate targets. Manuscripts contained within this Research Topic, *Insights in Negative Emission Technologies: 2021*, offer a broad snapshot of this growing field.

## Author contributions

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

## Conflict of interest

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# Putting Bioenergy With Carbon Capture and Storage in a Spatial Context: What Should Go Where?

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This paper explores the implications of siting a bioenergy with carbon capture and storage (BECCS) facility to carbon emission performances for three case-study supply chains using the Carbon Navigation System (CNS) model. The three case-study supply chains are a wheat straw derived BECCS-power, a municipal solid waste derived BECCS-waste-to-energy and a sawmill residue derived BECCS-hydrogen. A BECCS facility needs to be carefully sited, taking into consideration its local low carbon infrastructure, available biomass and geography for successful deployment and achieving a favorable net-negative carbon balance. On average, across the three supply chains a 10 km shift in the siting of the BECCS facility results in an 8.6–13.1% increase in spatially explicit supply chain emissions. BECCS facilities producing low purity CO<sub>2</sub> at high yields have lower spatial emissions when located within the industrial clusters, while those producing high purity CO<sub>2</sub> at low yields perform better outside the clusters. A map is also generated identifying which of the three modeled supply chains delivers the lowest spatially explicit supply chain emission options for any given area of the UK at a 1 MtCO<sub>2</sub>/yr capture scale.

**Keywords:** BECCS, negative emissions, supply chains, macro-energy systems, GIS, carbon-optimal transportation

## INTRODUCTION

Bioenergy with Carbon Capture and Storage (BECCS) will most likely play an important and critical role in limiting global temperature rise to 1.5 and 2.0°C and reach global net-zero targets by offsetting hard-to-abate residual emissions (IPCC, 2018; CCC, 2019). BECCS is the installation of carbon capture and storage (CCS) technology onto a bioenergy facility where the captured biogenic CO<sub>2</sub> is transported for secure and “permanent” geological storage (Gough et al., 2018; Freer et al., 2021). BECCS as a form of carbon dioxide removal (CDR) has been projected to supply between 43.5 and 96.5 MtCO<sub>2</sub>/yr within the UK by 2050, depending on the assumed scenario within the committee on climate change sixth carbon budget (CCC, 2020a). BECCS is often projected to contribute the majority of negative emissions, with direct air capture and storage (DACS) and other CDR approaches providing the remainder (CCC, 2020a). The majority of nationally determined contributions do not specify what kind of BECCS will be used to create their negative emissions and often claim a generic form of BECCS will achieve their targets. BECCS is not a monolithic technology, and no two facilities are the same.

A realistic scale and footprint of an envisaged medium-sized BECCS supply chain situated in the Humber region of England, which is storing CO<sub>2</sub> offshore in the Endurance saline aquifer is presented in **Figure 1**. The green lines depict the biomass routings, the blue lines depict the CO<sub>2</sub> transportation to the Endurance field and the red line depicts hydrogen routing for national grid injection from the chosen facility site. The deployment of BECCS and other CDR approaches will play a critical role in meeting national Net-Zero commitments through the generation of negative emissions, but the roll out of the technologies has been slow, although has been accelerating in recent years with multiple large scale BECCS projects soon to come online (Norwegian Ministry of Petroleum Energy, 2016a,b; Brevik, 2017; Greenberg et al., 2017; Kemper, 2017; Occidental Petroleum Organisation, 2017; Global CCS Institute, 2018).

The Committee on Climate Change Sixth Carbon Budget has developed six classifications for BECCS being BECCS-Power, BECCS-Waste-to-Energy, BECCS-Manufacturing/Construction, BECCS-Hydrogen, BECCS-Biofuels and BECCS-Biomethane, which is beneficial to predict the future deployment of the different types of BECCS (CCC, 2020b). However, these classifications are restrictive and do not reflect the full spectrum of the types of BECCS. These BECCS classifications fail to capture supply chains that straddle multiple classifications such as municipal solid waste (MSW) to hydrogen BECCS supply chains and does not reflect future BECCS types such as algae/aquatic biomass derived BECCS or new forms of BECCS like alkaline thermal treatment BECCS (Pour et al., 2018; Melara et al., 2020; Zhou and Park, 2020).

BECCS is an incredibly diverse and unique technology that needs to be carefully tailored to its local infrastructure, available biomass, economic environment, energy systems, land management strategies, socio-political context and geography for successful deployment and integration (Forster et al., 2020; Clery et al., 2021). A BECCS facility and supply chain that performs optimally in one location will not operate at the same level in another location (Freer et al., 2021).

This analysis presented here uses the Carbon Navigation System (CNS) model to simulate and evaluate the carbon performances of BECCS supply chains across the UK (Freer et al., 2021). The CNS was developed as a heuristic to aid policymakers and decision making with the sustainable and carbon-efficient deployment of BECCS across the UK. This paper describes the analysis of three BECCS supply chains across the UK, each calibrated to store 1 MtCO<sub>2</sub>/yr as this reflects the scale of upcoming large-scale BECCS facilities, generating carbon performance heatmaps depicting the variation in supply chain performance across different geographies, biomass distributions, access to geological storage and access to low carbon infrastructure. The heatmaps were generated for Scotland, England and Wales, while also focusing within five industrial carbon capture (ICCS) clusters across the UK. The paper will also determine the level of impact on the spatially explicit supply chain emissions by shifting the location of the BECCS facility across the UK. Spatially explicit supply chain emissions are the emissions that vary depending on the location of the energy facility; these include transportation and resource

gathering. This paper also provides a comparative decision map depicting which of the three modeled BECCS supply chains should be chosen to store 1 MtCO<sub>2</sub>/yr for the least amount of spatially explicit supply chain emissions at any location in the UK.

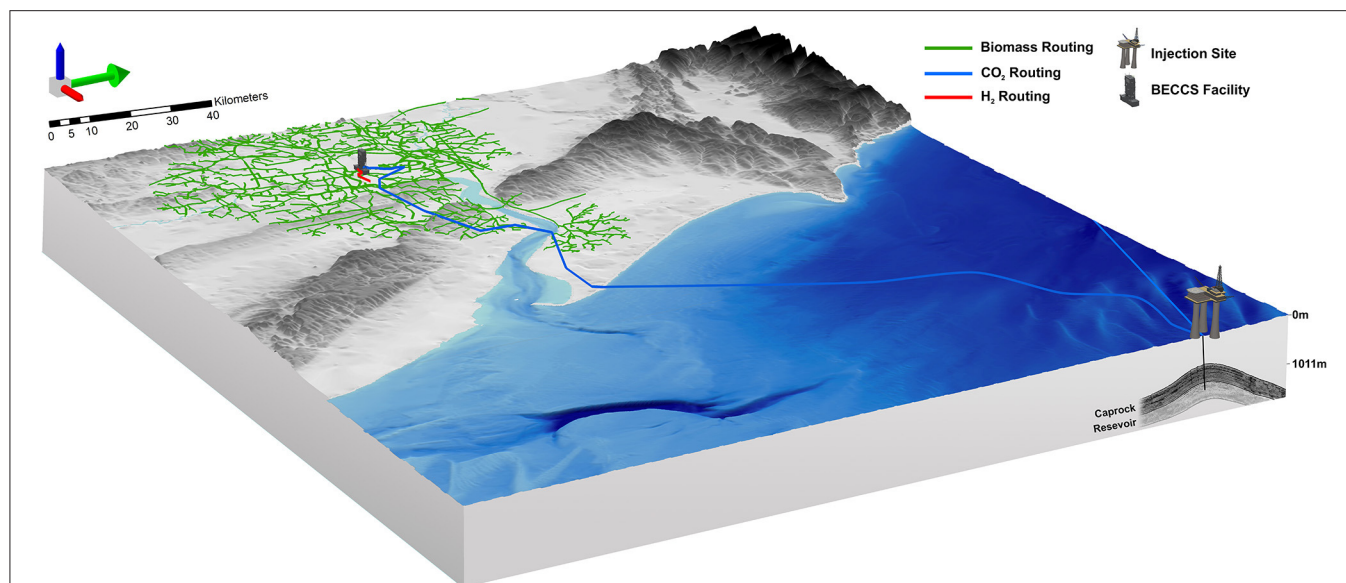
## The Carbon Navigation System Model

The CNS model, developed in Freer et al. (2021), is a carbon-efficient digital twin of the UK's industrial freight transport network for cargo and tanker transportation, which has been used to model BECCS supply chains in a macro-energy system analysis. A digital twin is a virtual representation of assets or networks that allows for the simulation of processes or interactions within a virtual version of the assets or networks to allow for system optimization or learning (Zhang et al., 2021). The CNS model simulates and calculates the spatial explicit supply chain emissions of specific BECCS supply chains to generate high spatial resolution carbon performance heatmaps for the UK. The CNS can search and route for any specific quantity of biomass to any location in the UK, route the produced CO<sub>2</sub> to its most suitable geological storage site and route its energy output to its end-user, all while automatically switching between truck, rail, shipping and pipeline transportation to minimize CO<sub>2</sub> emissions and calculate carbon optimal routings.

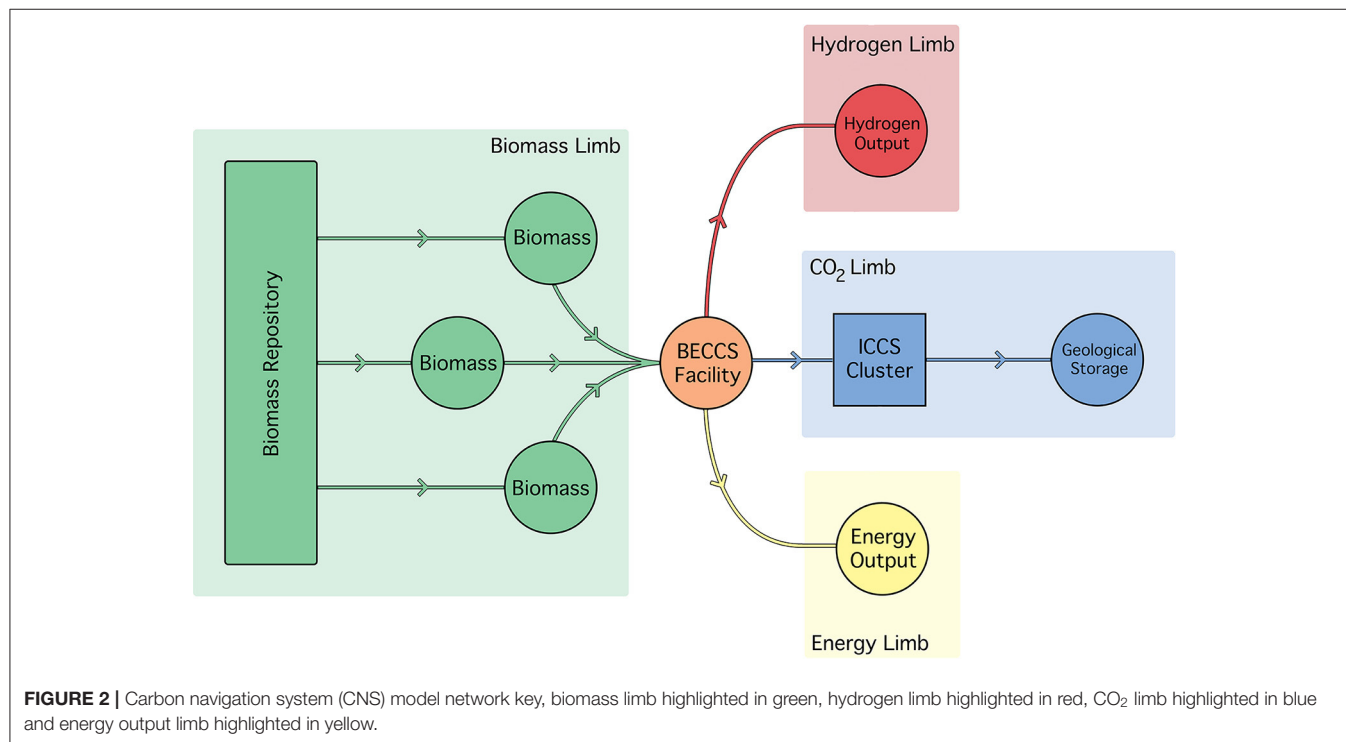
The CNS model's structure was designed so that it could be tailored and customized to analyze any form of BECCS. The model is split into multiple segments, called limbs, which focus on a specific aspect of a BECCS supply chain (Freer et al., 2021). The four limbs of the CNS model are the biomass limb which focuses on biomass transportation, the CO<sub>2</sub> limb which focuses on the transportation and storage of CO<sub>2</sub> via an ICCS cluster, the energy output limb which focuses on the transportation and distribution of the energy produced by the facility (including heat, power and biofuels), and the newly developed hydrogen limb to cover the transportation and distribution of hydrogen fuel across the UK. The hydrogen limb was added to the model due to the increasing potential role hydrogen will play in future energy systems, as the UK hydrogen strategy aims to generate 5 GW of low-carbon hydrogen by 2030 (HM Government, 2021). The four limbs all converge around the BECCS facility, and the structure of the CNS model is presented in **Figure 2**. The data used to build the model networks were extracted and georeferenced from National Infrastructure Planning documents and opensource transportation polylines published by the UK Government (BGS, 2020; HM Government, 2020a; HyNet, 2021b; Net Zero Teesside, 2021). The truck shapefiles were taken from the 2018 OS Open Roads dataset, the rail shapefiles were taken from the 2017 Freight Usage dataset and the shipping routes were taken from the 2016 MMO Anonymised AIS Derived Track Lines dataset (HM Government, 2020a). Within Freer et al., the methods in which the shapefiles were processed, refined, filtered and combined into multiple networks is explained in more detail (Freer et al., 2021).

The scope of the CNS model focuses on the spatially explicit supply chain emissions produced by a BECCS supply chain, being the emissions that vary depending on where the facility





**FIGURE 1 |** BECCS supply chain block diagram, depicting an envisaged 0.5 MtCO<sub>2</sub>/yr BECCS facility operating in the Humber region of England with geological storage of CO<sub>2</sub> in the endurance reservoir *via* pipeline. The diagram is to scale in the horizontal dimension, a vertical exaggeration factor of 10 was applied to the vertical dimension.



**FIGURE 2 |** Carbon navigation system (CNS) model network key, biomass limb highlighted in green, hydrogen limb highlighted in red, CO<sub>2</sub> limb highlighted in blue and energy output limb highlighted in yellow.

is located. The spatially explicit emissions include the gathering of biomass, transportation of biomass, transportation of CO<sub>2</sub>, transportation of hydrogen fuel and the transportation of other energy outputs. The CNS model does not calculate the spatially static supply chain emissions such as biomass drying, biomass grinding, fuel combustion, CO<sub>2</sub> compression, CO<sub>2</sub> injection or

CO<sub>2</sub> remediation approaches as these are better calculated *via* a life-cycle analysis (LCA) (Laude et al., 2011; Pour et al., 2018; Yi et al., 2018; Bello et al., 2020; García-Freites et al., 2021; Lask et al., 2021). The scope of the CNS model has been specially designed to plug into any BECCS LCA to improve the spatial context of the analysis.

## High Spatial Resolution Biomass Mapping

A high-resolution biomass repository was developed alongside the CNS model detailing the spatial distribution and quantities of various biomass types across the UK down to the plot of land resolution. Using the biomass repository, the CNS can accurately search and route specific quantities of biomass to any location in the UK. The biomass repository focuses on agricultural residues and industrial wastes but also includes many of the primary feedstocks used to generate the residues and wastes. A range of biomass remote sensing and mapping techniques were used to create the repository, including the main four mapping techniques below:

- I. The visual assessment of land performed by farmers and experts (Newnham et al., 2010)
- II. Accounting of biomass as it is collected and transported (Ali et al., 2017)
- III. *In-situ* mechanical or electronic sensors collecting readings of biomass as it arrived at a transfer station of facility (Hakl et al., 2012)
- IV. Spectral image analysis taken by satellite and drones (Jones et al., 2020)

## Geospatial BECCS Analysis

The geospatial analysis of BECCS is a growing discipline within the literature as the deployment of the technology accelerates, and the spatial implications on BECCS supply chains need to be identified, evaluated and monitored. Many of the nations worldwide are banking on CDR approaches, predominantly afforestation and BECCS, to generate their negative emissions to offset their hard-to-abate sectors. However, many of these commitments have yet to declare where these CDR approaches will be sited and what are the land-use and operational footprints of these projects.

In particular, many LCAs and Techno-Economic Assessments (TEAs) for BECCS, bioenergy and bio-economy projects do not use realistic spatial data within their studies, where instead, they use simplified distances traveled and distributions of biomass in their study parameters (Laude et al., 2011; Pour et al., 2018; Yi et al., 2018; Bello et al., 2020; García-Freites et al., 2021; Lask et al., 2021). The poor siting of a BECCS facility relative to its biomass, CO<sub>2</sub> infrastructure and energy end-users have severe emissions implications for the project (Freer et al., 2021) and will result in far-reaching fiscal, temporal, environmental and social knock-on effects. The spatial context needs to be better considered and incorporated into studies to avoid these emissions implications and knock-on effects.

The literature is now starting to consider the spatial implications of BECCS as the large-scale deployment of the technology nears. **Figure 3** depicts which of the BECCS studies ranging from LCAs to TEAs and environmental assessments have considered the spatial implications of their projects, graded on the percentage of their methodologies, results, discussions and conclusions explore the spatial implications. The spatial resolution of the studies has also been evaluated and represented in the figure using a symbol hierarchy to demonstrate which studies had the highest resolution. The other focuses of the

studies have also been graded in the same manner to highlight the primary objectives of the studies, including the fiscal, emissions, temporal, environmental and social context of the studies.

## BECCS Geospatial Analysis Context and Resolution

The focus and resolution of BECCS geospatial analysis studies vary drastically from study to study, ranging from studies with almost zero geographic perspective to those with high spatial resolutions. The spatial context of BECCS studies is not typically the primary focus of the analysis, resulting in many using very little spatial data. Many BECCS studies have opted to map the locations of key infrastructure for BECCS, such as CO<sub>2</sub> storage sites and point emission sources, but have not carried out extensive spatial analysis (Baik et al., 2018; da Silva et al., 2018; Albanito et al., 2019; Melara et al., 2020; Sagues et al., 2020).

The spatial resolution of the majority of transnational BECCS studies are limited by the data availability and data quantity, resulting in most transnational BECCS studies opting for a country block spatial resolution with values assigned to each country with no differentiation of geospatial data within the country blocks (Fajardy and Mac Dowell, 2018; Negri et al., 2021). The Rosa et al. (2021) trans-national study currently has the highest spatial resolution of all trans-national BECCS studies by using 1 km blocks rather than country blocks.

Some BECCS studies have opted for large spatial blocks, ~40–50 km, to differentiate the distribution of resources and infrastructure (Zhang et al., 2019; Gabrielli et al., 2020), but the most common form of spatial resolution used in BECCS studies are the local authority blocks. The use of local authority blocks offers an acceptable level of resource and infrastructure differentiation within a country while not producing extremely large quantities of data. Local authority blocks are most commonly used for countries with large land areas, such as the US and China (Baik et al., 2018; Dolan et al., 2020; Xing et al., 2021).

The spatial resolution of BECCS studies has increased over time with the development and popularization of high spatial resolution mapping of the technology. Most high spatial resolution studies opt to use small spatial blocks, ranging from 1 to 2 km, to differentiate resources within the countries (Donnison et al., 2020; Krause et al., 2020; Rosa et al., 2021). The Freer et al. (2021) study is currently the only geospatial analysis BECCS study that has achieved a higher spatial resolution down to the plot of land. This study does have a higher spatial resolution than the Rosa et al. study, but the Rosa et al. study covers a much larger study area for Europe compared to only the UK in the Freer et al. study.

## BECCS Geospatial Analysis Study Areas and Scale

The study areas of geospatial BECCS studies have been primarily focused within the northern hemisphere. The majority of the studies focus on the UK and the US due to a high level of data availability, political will and abundance of pre-existing infrastructure compatible with BECCS (Sanchez and Callaway, 2016; Baik et al., 2018; Fajardy and Mac Dowell, 2018; Albanito et al., 2019; Zhang et al., 2019; Dolan et al., 2020; Donnison et al., 2020; Melara et al., 2020; Sagues et al., 2020; Freer et al., 2021).





FIGURE 3 | Geospatial analysis of BECCS literature review dashboard.

Studies focusing on Europe and the countries within are becoming more frequent as the potential for trans-national collaborations develop and mature (Gabrielli et al., 2020; Krause et al., 2020; Negri et al., 2021; Rosa et al., 2021). Geospatial BECCS studies focusing within Asia are not well represented in the literature, with only one study focusing on China (Xing et al., 2021). This lack of representation is also apparent for countries within the Middle East, South America, Indo-Australasia and Africa, showing a major research gap.

There has been a clear progression of the scale of analysis within geospatial BECCS studies. The first studies focused on smaller regional evaluations, which then developed into national evaluations. The next stage of evaluation will be the analysis of multiple collaborating countries, which can be seen in the publication of trans-national BECCS studies in recent years.

### BECCS Biomass Types

The most researched type of biomass feedstocks in BECCS geospatial analysis studies are energy crops including miscanthus, switchgrass, willow short rotation coppicing (SRC) and poplar SRC (Fajardy and Mac Dowell, 2018; Albanito et al., 2019; Dolan et al., 2020; Donnison et al., 2020; Negri et al., 2021; Xing et al., 2021). However, these studies do not specify where exactly the biomass is or will be grown, as they use generalized blocks. There are fewer studies involving other biomass types such as agricultural residues including wheat straw, corn stover and woody residues

(Sanchez and Callaway, 2016; Fajardy and Mac Dowell, 2018; Krause et al., 2020; Negri et al., 2021; Xing et al., 2021) or industrial wastes including MSW, forestry residues, wastewater treatment, pulp, paper and manure (da Silva et al., 2018; Zhang et al., 2019; Gabrielli et al., 2020; Melara et al., 2020; Sagues et al., 2020; Freer et al., 2021; Rosa et al., 2021). This paper addresses some of these gaps by specifically considering opportunities for utilizing waste and residues in BECCS facilities.

### Other BECCS Context

Early BECCS analyses rarely focused on spatial context. However, more recent studies have recognized the integral and critical role of spatial context in the sustainable development and deployment of the technology, but a paradigm shift toward the incorporation of the spatial context of BECCS starting to play an integral and critical role in the sustainable development and deployment of the technology.

The majority of geospatial BECCS studies tend to focus more on the fiscal and emissions contexts of BECCS due to nations exploring the logistics and operations of deploying BECCS (Sanchez and Callaway, 2016; Baik et al., 2018; Fajardy and Mac Dowell, 2018). Only a few studies focus on the environmental and temporal contexts and very few focusing on the social context (Zhang et al., 2019; Gabrielli et al., 2020; Melara et al., 2020; Sagues et al., 2020; Freer et al., 2021; Negri et al., 2021; Rosa et al., 2021; Xing et al., 2021). There is a clear gap in the research focusing on the spatial aspects of BECCS with a social framing, exploring the spatial variations in public perception, ethics and governance of BECCS.

## BECCS Supply Chain Tethering

Supply chain tethering, a concept developed in Freer et al. (2021), is a quantifiable and comparable metric of understanding which limb of a supply chain produces the most CO<sub>2</sub> emissions and is used to inform decision making on how to best target carbon emissions saving measures. For example, a BECCS supply chain can be 6.04 times more tethered to its CO<sub>2</sub> infrastructure compared to its biomass infrastructure, indicating that the CO<sub>2</sub> emissions produced in CO<sub>2</sub> transportation are 6.04 times larger than the CO<sub>2</sub> emissions produced in biomass transportation. Knowing the scale of tethering for a BECCS facility will allow prioritization of carbon saving measures, as in the example case, measures targeting CO<sub>2</sub> transportation emissions would have greater overall savings compared to measures targeting emissions from biomass transportation.

Some styles of BECCS may also be more tethered to certain aspects of the supply chain, as the sugar beet derived bioethanol BECCS supply chain in the Freer et al. study was more tethered to its biomass infrastructure (Freer et al., 2021). However, due to no two BECCS supply chains operating the same way, not all BECCS supply chains are more tethered to their biomass than their CO<sub>2</sub> or energy output and not all supply chains are more tethered to its CO<sub>2</sub>. This difference in tethering is also apparent when a BECCS supply chain is shifted location, as in one location the supply chain may be more tethered to its CO<sub>2</sub> infrastructure, while in another location it may be more equally tethered to its CO<sub>2</sub> and biomass infrastructure. Each supply chain must be evaluated on a case-by-case basis and, depending on the locations of the supply chain, exists on a spectrum of tethering between the limbs of the supply chains.

## METHODOLOGY

### BECCS Supply Chain Descriptions

Three BECCS supply chains were selected and applied through strict criteria to maximize the impact and real-world applications of the results. To be included, supply chains must: utilize a UK domestic feedstock; the biomass types must cover a range of spatial distributions; similar supply chains should be mentioned in industrial and governmental reports; must use a mixture of carbon capture technologies; must cover a range of different types of BECCS; and one must generate hydrogen fuel. Based on a literature search, the following three supply chains met all of the criteria and have been used in this analysis (Muresan et al., 2013; Moreira et al., 2016; Mendiara et al., 2018; Pour et al., 2018; Tagomori et al., 2019; Agrawal and Rao, 2020; Emenike et al., 2020; Ghiat et al., 2020; Patzschke et al., 2020; García-Freites et al., 2021):

1. The “MSW supply chain,” evaluates the direct combustion of the organic fraction of municipal solid waste (MSW) for electricity generation with fitted post-combustion monoethanolamine carbon capture technology. This supply chain best represents the BECCS-Waste-to-Energy classification of BECCS.
2. The “Wheat Straw supply chain,” evaluates the combustion of wheat straw in an oxygen-rich environment fitted

with oxy-fuel carbon capture technology for electricity generation. This supply chain best represents the BECCS-Power classification of BECCS.

3. The “Sawmill Residue supply chain,” evaluates the co-generation of electricity and hydrogen derived from the gasification of sawmill residues converted into pellets through the process of integrated gasification combined cycle (IGCC) fitted with pre-combustion carbon capture technology. This supply chain best represents the BECCS-Hydrogen classification of BECCS.

Block flow diagrams for each of the three supply chains are presented in **Figure 4**, depicting the types of biomass used, energy output generated and CO<sub>2</sub> transportation *via* an ICCS cluster for offshore geological storage. Each block flow diagram illustrates potential geological storage in either the Goldeneye, Endurance, or Hamilton reservoirs.

### BECCS Supply Chain Biomass Mapping

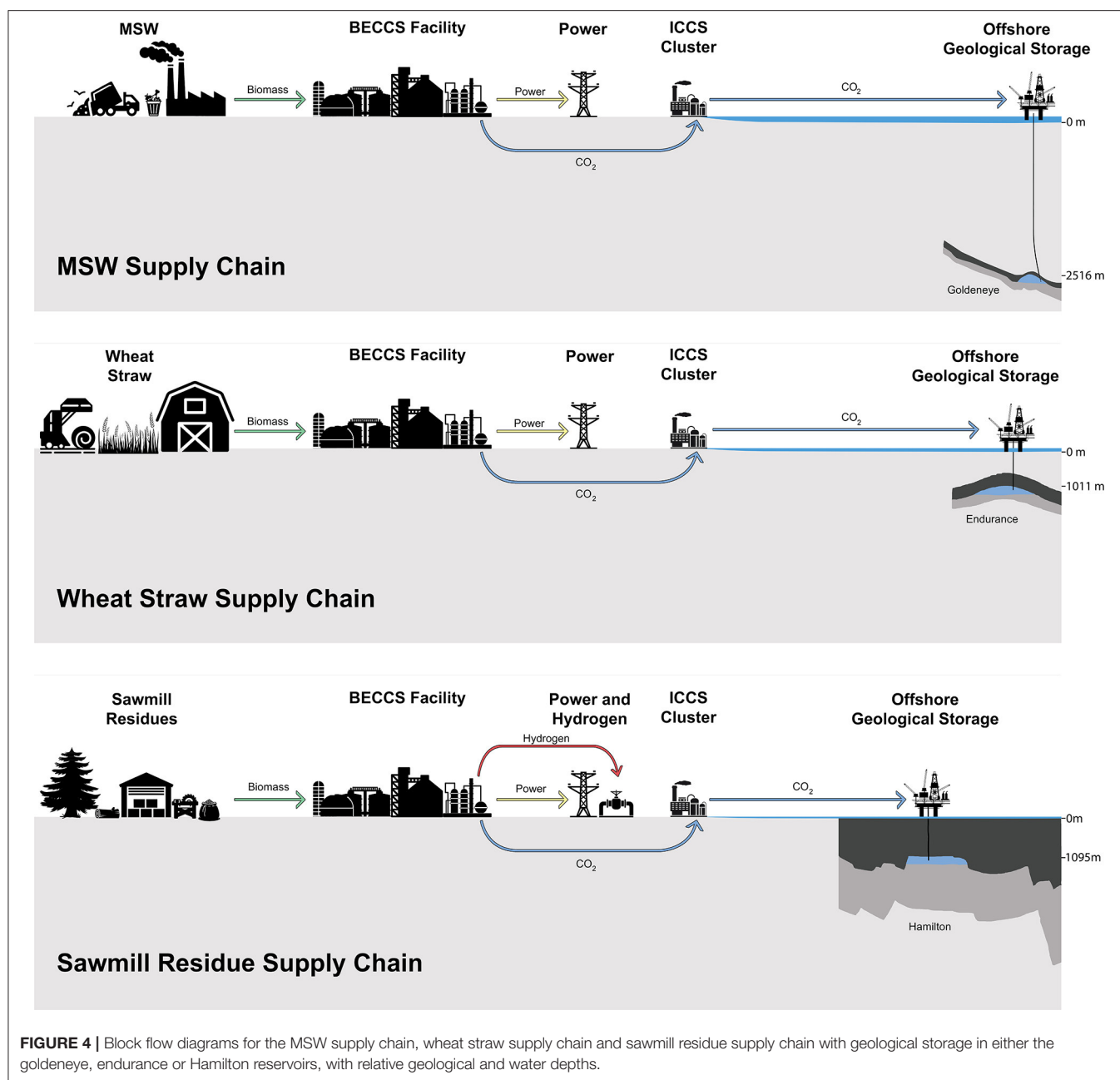
Different methods could be used to map the supply chains, each with different relative strengths and weaknesses in relation to the quality of data recording, scale and resolution; consequently a variety of mapping methods has been used to map the organic fraction of MSW, wheat straw, and sawmill residue.

The spatial distributions for the organic fraction of MSW, wheat straw and sawmill residue are presented in **Figure 5**.

The mapping of the organic fraction of MSW used waste generation data recorded by local authorities. The amount of biomass generated at each waste facility/site was allocated and integrated into the biomass repository. The supply chain taps into the organic fraction of MSW sent for incineration and landfill waste stream, as this waste redirection offers better utilization of the resource and targets the base resources of the waste management hierarchy for greater biomass availability.

The mapping of the wheat straw used a spectral image analysis approach. The base data for the current distribution of wheat straw locations in the UK was taken from the UKCEH Land Cover Plus Crops dataset for 2020 (UKCEH, 2019). From this dataset, the wheat straw plots of land were extracted and had their areas measured. A wheat straw yield was applied to each polygon, which is presented in **Table 1**. The total wheat straw was then verified against reported DEFRA census data to verify the results (DEFRA, 2020), of which they did match.

The mapping of the sawmill residue was performed using a combination of biomass accounting and satellite image analysis. The data availability for the UK forestry industry is very high. However, most of the data has been anonymised, making it difficult to allocate amounts of biomass to specific locations. This anonymising issue was bypassed by using satellite imagery to measure the area of the lumber yards at each of the sawmills in the UK. The amount of timber processed at eight sawmills was found and assigned to their respective sawmill lumber yard (UK Forestry Commission, 2021). These eight data points were then used to determine the amount of roundwood produced per m<sup>2</sup> of lumber yard and used to determine the amount of roundwood produced at all the other sawmills. The total amount of roundwood was then verified



against the total anonymised data from the forestry commission to verify the results, of which they did match. Sawmill residue yields for sawdust and chipwood were then applied to each site.

## Carbon Navigation System Model Parameters

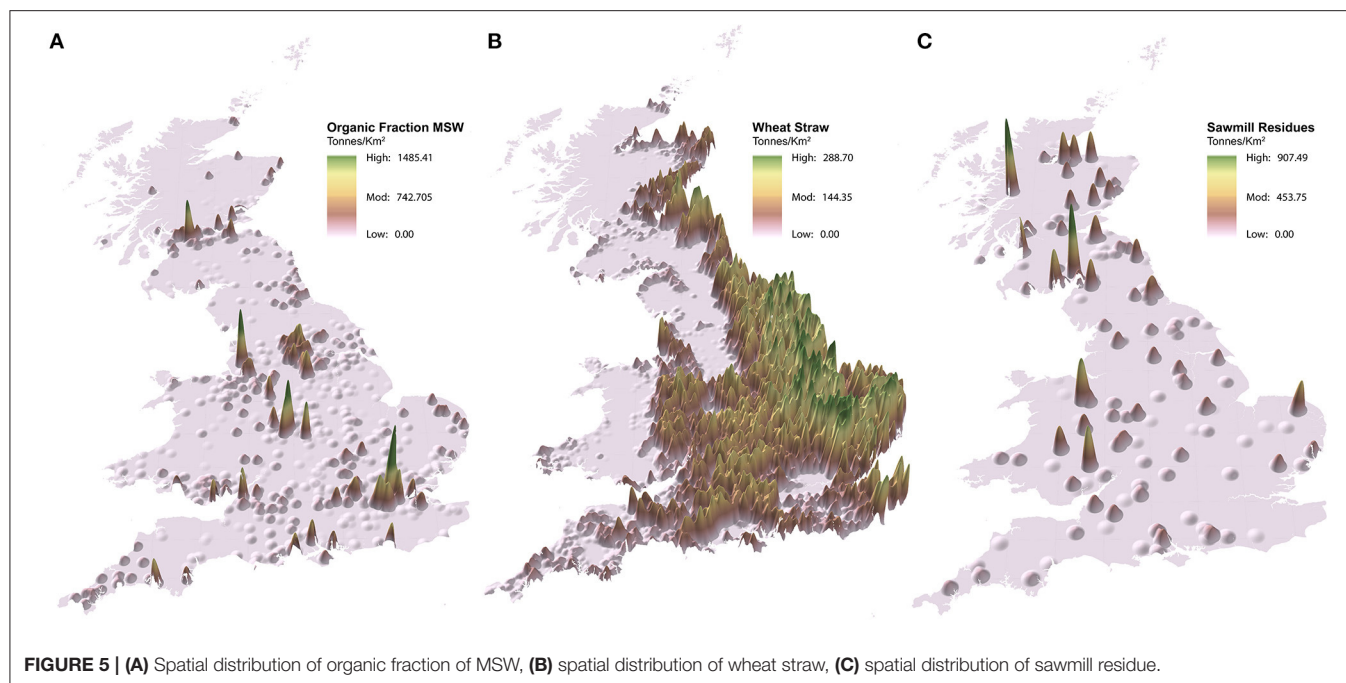
The three supply chains have all been calibrated and standardized to store 1 MtCO<sub>2</sub>/yr and produce an energy output in the form of power or power and hydrogen. A 1 MtCO<sub>2</sub>/yr storage scale was selected as this reflects the scale of upcoming large-scale BECCS facilities, although future analysis will focus on the performance

of smaller-scale facilities. The parameters of the supply chains are presented in **Table 1**.

## Carbon Navigation System Infrastructure CO<sub>2</sub> Transportation Infrastructure

The UK has one of the fastest developing CCS infrastructure networks in the world, with plans rapidly evolving as the first geological injection, scheduled for 2024 (Acorn, 2017a; CCC, 2019; OGC, 2019; Pale Blue Dot, 2019; ZeroCarbonHumber, 2019; HyNet, 2020; BEIS, 2021).

The UK has adopted an ICCS cluster approach with five ICCS clusters under development and chosen due to their proximity



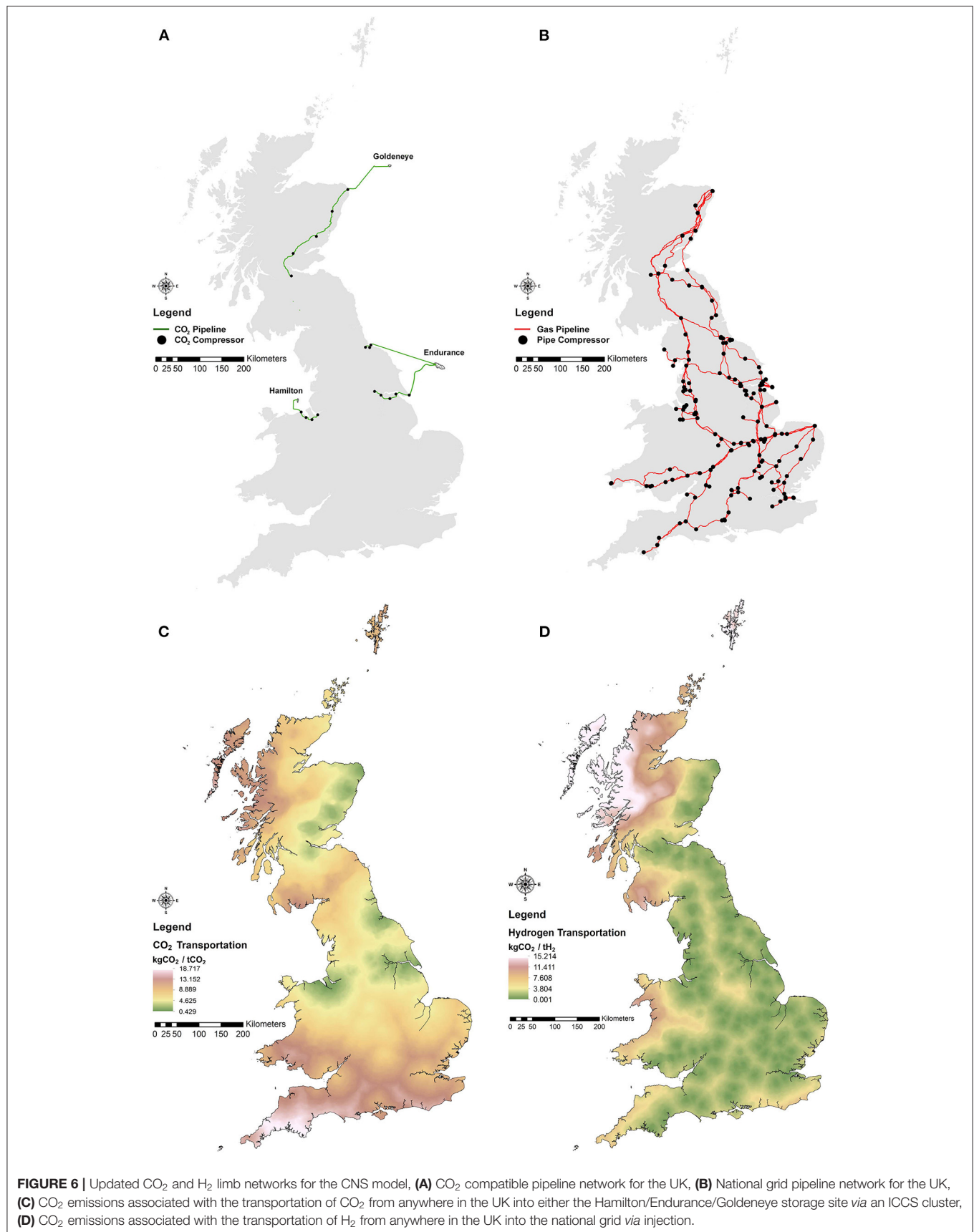
**TABLE 1 |** MSW supply chain, wheat straw supply chain and sawmill residue supply chain inputs and output parameters.

Description	Unit	MSW	Wheat straw	Sawmill residue	References
Biomass yield	Tons/Hectare	–	8.5	–	DEFRA, 2019, 2020
Biomass availability	%	44	28	19	DECC, 2012, 2014; Cadent Gas Ltd, 2017; Fantini, 2017; Townsend et al., 2018; Scarlat et al., 2019; UK Forestry Commission, 2021
Lower heating value	MJ/kg	16.8	17.8	15.9	ECN Biomass Energy Efficiency, 2021
Hydrogen : Electricity ratio	–	–	–	1.3:1	Chiesa et al., 2005; IEA, 2008; Moldenhauer et al., 2020
CO <sub>2</sub> captured	tCO <sub>2</sub> /MWh	0.90	1.01	0.73	Pour et al., 2018; Agrawal and Rao, 2020; Emenike et al., 2020; García-Freites et al., 2021
CO <sub>2</sub> capture efficiency	%	90	92	90	Pour et al., 2018; Agrawal and Rao, 2020; Emenike et al., 2020; García-Freites et al., 2021
Days operations per Year	Days	300	300	300	Pour et al., 2018; Agrawal and Rao, 2020; Emenike et al., 2020; García-Freites et al., 2021
Biomass tonnage	Mt/yr	0.24	0.20	0.31	–
Energy capacity	MW	154	138	83	–
Hydrogen tonnage	ktH <sub>2</sub> /yr	–	–	23.11	–
CO <sub>2</sub> tonnage	MtCO <sub>2</sub> /yr	1.00	1.00	1.00	–

to gas terminals for offshore geological storage, pre-existing infrastructure and in the catchment of multiple CO<sub>2</sub> emitters. The five ICCS clusters are in Scotland, Teesside, North-West England, Humberside, and South Wales. The current UK CCS strategy is to establish four low carbon clusters by 2030 and one net-zero cluster by 2040 (BEIS, 2018; HM Government, 2018). The locations of the ICCS clusters are presented in **Figure 6**.

The Teesside, North West and Humberside ICCS clusters have opted to construct new CO<sub>2</sub> compatible pipelines for offshore geological storage of CO<sub>2</sub> (OGC, 2019; ZeroCarbonHumber, 2019; HyNet, 2020), while the Scottish cluster will repurpose the feeder 10 and offshore pipelines for CO<sub>2</sub> transportation (Acorn, 2017a,b; Pale Blue Dot, 2019). Due to the lack of proximal geological storage to the South Wales cluster, the cluster has opted





to transport CO<sub>2</sub> *via* ship to be integrated into the North West cluster.

The UK has access to a large potential of secure offshore geological storage sites in the North Sea and the east Irish Sea, with theoretical storage capacities estimated to be between 61.7 and 75.6 GtCO<sub>2</sub> (Pale Blue Dot, 2020). The most suitable storage sites in the North Sea in terms of reservoir thickness, permeability, storage capacity, level of containment risk, CAPEX + OPEX costs, level of seismic surveys, CO<sub>2</sub> injection rates, caprock viability and level of pre-existing infrastructure are Captain X, Goldeneye, Forties 5, Bunter Enclosure 36, Endurance, Viking A, Hewett, Hamilton and Morecambe formations (Jin et al., 2012; Pale Blue Dot, 2016, 2020; Williams et al., 2016; Akhurst et al., 2017; HM Government, 2018). This analysis assumes an early level of development and deployment of geological storage, resulting in the use of the Goldeneye, Endurance and Hamilton storage sites in the analysis, with Monte Carlo theoretical storage capacities not exceeding 50% of estimates (P50) of 37, 248, and 122 MtCO<sub>2</sub>, respectively (Pale Blue Dot, 2020). The Goldeneye and Hamilton reservoirs are depleted gas fields, while the Endurance reservoir is a saline aquifer.

This study reflects the current UK CCS strategy by focusing on England, Scotland and Wales. This research can be rolled out to cover Northern Ireland and the potential Londonderry shipping ICCS cluster (BEIS, 2020), but has not been included due to limitations and barriers such as grid reference projection, the integration of island infrastructure, potential tariffs and politics within Ireland and implications of CO<sub>2</sub> storage with the IMO London Protocol (Dixon, 2019).

The UK CO<sub>2</sub> pipeline networks and the carbon transportation costs of CO<sub>2</sub> storage in the Goldeneye, Endurance and Hamilton reservoirs from anywhere in the UK are presented in **Figure 6**.

### *H<sub>2</sub> Transportation Infrastructure*

Hydrogen has many advantages as a clean energy vector due to its high energy density, potential for combustion products without pollutants, renewability, and its possible use in transport, industrial and domestic applications (Rashid et al., 2015; Felgenhauer et al., 2016; Zhou and Park, 2020). Hydrogen can be transported by ship or pipeline, with straightforward modifications to existing infrastructure, with onshore and offshore storage potential in the UK and could substitute fossil fuels in various applications including industrial processes, heat and transport (Kuo and Wu, 2015; Gerboni, 2016; Gondal, 2016; Dawood et al., 2020; ETIP Bioenergy, 2021; HM Government, 2021; Yu et al., 2021). It has been estimated that the deployment of hydrogen to phase out fossil fuels in the UK could result in a 0.6–2.3% emissions reduction by 2030 (FCH, 2020). The UK Hydrogen strategy has also set ambitions to generate 5 GW of low carbon hydrogen by 2030 (HM Government, 2021). Although, uncertainties remain in the magnitude and extent of the UK hydrogen economy (Calver et al., 2022). Hydrogen production is projected to play a substantial part in the pursuit of net-zero and plays a larger role in the UK's ten-point plan (HM Government, 2020b). A phased deployment of hydrogen supply is predicted for the UK, starting with hydrogen derived from a fossil fuel source with CCS (blue hydrogen) with the objective to switch to

hydrogen derived from renewable sources (green hydrogen) (HM Government, 2021; UK Parliament, 2021).

Bio-hydrogen is an example of a third-generation biofuel and may be formed *via* many technologies, including steam reformation, gasification, fermentation, microbial fuel cells, and algal cultivation (ETIP Bioenergy, 2021). The carbon balance of bio-hydrogen has the potential to be carbon neutral, but when combined with CCS, a form of BECCS, has the potential to produce a net-negative carbon balance (Zhou and Park, 2020).

Hydrogen has the potential to be blended with natural gas in a national grid. HyNet in the northwest of England has proposed to inject hydrogen into the national grid at a 20% volume, with the goal of reaching 100% in the future (HyNet, 2020). Other projects such as H<sub>2</sub> Aberdeen are seeking to develop a hydrogen economy through hydrogen fuel HGVs, hydrogen district heating and national gas grid replacement (Aberdeen City Council, 2015). In line with these initiatives, the hydrogen produced by the Sawmill Residue supply chain case study was directed for national grid injection at 20% volume. The national grid pipelines and the carbon transportation costs of H<sub>2</sub> injection into the national grid from anywhere in the UK is presented in **Figure 6**.

### *Carbon Navigation System Infrastructure Upgrade*

The CCS infrastructure in the UK is quickly evolving toward the goal of efficiently and permanently storing CO<sub>2</sub> in the offshore subsurface. The Teesside and Humberside clusters have recently formed a collaboration named the “Northern Endurance Partnership,” which plans to introduce a new CO<sub>2</sub> compatible pipeline directly connecting the Teesside cluster to the Endurance reservoir (Zero Carbon Humber, 2020). The CNS model has been updated to incorporate this new pipeline into all calculations. The updated CO<sub>2</sub> pipeline network is presented in **Figure 6**.

### **Carbon Navigation System Cost Matrix and Emissions Profiles**

The CNS model uses a standardized carbon-based cost matrix, kgCO<sub>2</sub>/ton-km, to minimize the spatially explicit CO<sub>2</sub> emissions produced by the supply chain. This analysis assumes that all transportation is fossil fuel derived and uses the same fossil fuel emissions profile and transport switching costs used in Freer et al. (2021). This analysis also assumes that empty return journeys do take place within the supply chains.

The three modeled BECCS supply chains all produce power as an energy output, producing negligible spatially explicit supply chain emissions from the transportation of the power through the grid. Due to this, the energy output limb of the CNS was not used for the three supply chains. The emissions produced *via* the construction of new powerlines to rural areas fall out of scope for this analysis, but it is important to note that there will be additional carbon costs for connecting BECCS-Power in rural areas at a rate of 6.3 ktCO<sub>2</sub>/km (Harrison et al., 2010).

### **Biomass Data Processing**

The CNS model is capable of searching for specific quantities of biomass through the processing of biomass into standardized “Biomass Packages” through a named process of either splitting or bundling (Freer et al., 2021). Splitting the biomass points



involves taking a singular biomass location representing a large quantity of biomass and splitting the singular point into multiple stacked points, each representing 27.45 tons of biomass (truck capacity with applied loading factor). Bundling the biomass is applied when a singular biomass point represents <27.45 tons of biomass and is combined with other biomass points to constitute the standardized 27.45 tons of biomass while integrating the added carbon cost of gathering the biomass points together. The process of splitting and bundling the biomass data is further explained in Freer et al. (2021).

The biomass data within the MSW supply chain and Sawmill Residue supply chain were split, while the biomass data within the Wheat Straw supply chain were split and bundled due to the distribution of high and low yield locations.

### Biomass Availability and Availability Distribution

Biomass availability considered in this study is the percentage of total biomass production that is collatable and of high enough quality for energy production (Welfle et al., 2014a,b). Due to the lack of data accessibility for biomass availability for BECCS, bioenergy biomass availability was used as a proxy. The biomass availability percentages for each supply chain have been taken from the literature specific to each biomass type and are presented in **Table 1** (DECC, 2012, 2014; Cadent Gas Ltd, 2017; Fantini, 2017; Townsend et al., 2018; Scarlat et al., 2019; UK Forestry Commission, 2021). The chosen biomass availability percentages are representative of the current biomass strategy but may be subject to change with future policies and climate impacts and would require future work to determine the impact of a variable biomass availability on BECCS supply chains.

An equal distribution of biomass availability was also assumed due to the lack of spatially explicit reported data for biomass use and competition distribution. Future work will be required to determine the spatial distribution of biomass availability but would require a fully transparent database of biomass users for the entire UK at high spatial resolution.

### Batch Processing of the Supply Chains

Each of the supply chains was simulated over a range of locations to create the carbon performance heatmaps. The CNS model calculates the routings for one location, dissolves the routings into the point, and then automatically moves onto the next locations. Each of the supply chains were simulated over 219,878 locations to generate the heatmaps, resulting in 6.01 billion total routings between the three supply chains. Each of the supply chains also perform drastically differently for the same location. **Figure 7** depicts the routings for the three supply chains from the same location, which access the Endurance storage site via the Teesside ICCS cluster, showing the range in the footprints of the supply chains.

Each of the simulated BECCS facility emissions for the supply chains were dissolved and joined to their respective point

locations, where an inverse distance weighted interpolation was applied to generate the carbon performance heatmaps.

## RESULTS AND DISCUSSION

This analysis evaluates the carbon performances of three modeled BECCS supply chains across the UK using the CNS model. The carbon performance heatmaps for the MSW, Wheat Straw and Sawmill Residue supply chains is presented in **Figure 8**.

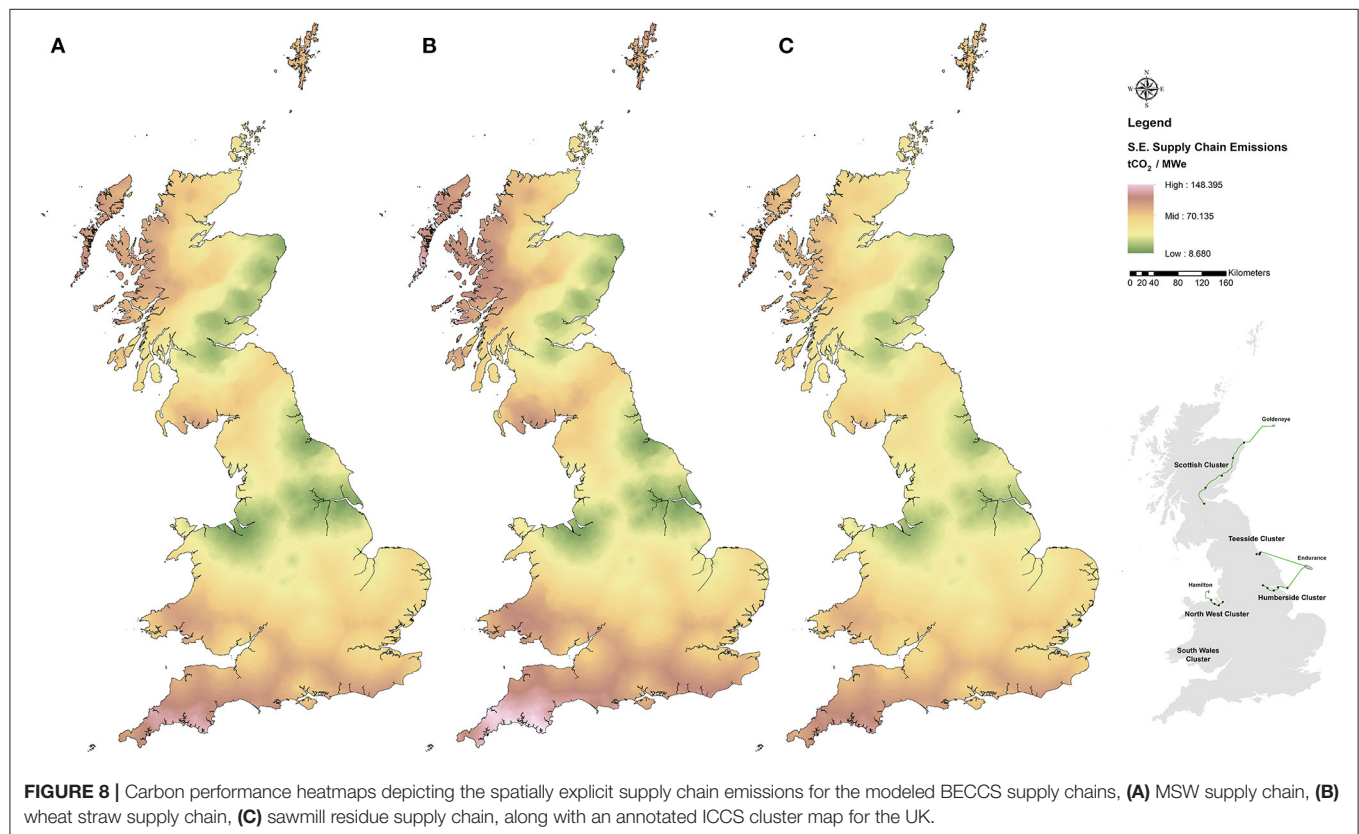
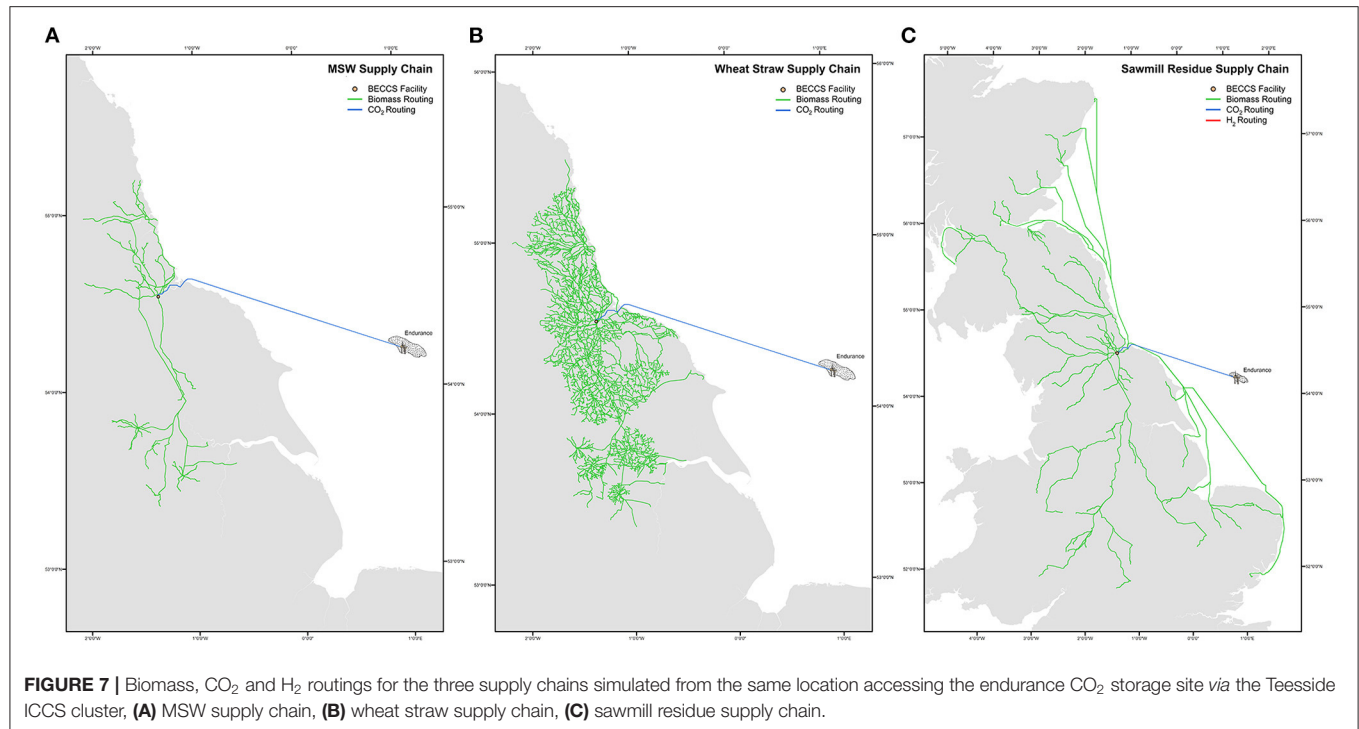
### BECCS Supply Chains Carbon Performance Areas

Across all three supply chains, the areas of highest performance (areas that have produced the lowest amount of supply chain transportation emissions) are concentrated around the ICCS clusters that use a dedicated CO<sub>2</sub> pipeline. Compared to the other clusters, the South Wales cluster underperforms due to the large and carbon-intensive distance required to transport CO<sub>2</sub> for geological storage. The decarbonization of the shipping or rail industry would greatly benefit the carbon performance of the area. Additionally, the conversion of an oil platform in the Irish sea into a CO<sub>2</sub> catchment hub with a dedicated pipeline connected to the geological storage sites in the area would benefit the South Wales cluster as well as Northern Ireland, Ireland and the rural west coast of the England and Scotland.

The siting location with the highest performance for the MSW and Sawmill Residue supply chains is Connah's Quay (located within the North West ICCS cluster) due to its strong access to CO<sub>2</sub> infrastructure, available biomass, low carbon infrastructure and energy end-user. Contrarily, the highest performing siting location for the Wheat Straw supply chain is in Barrow-Upon-Humber (located within the Humberside ICCS cluster) as this location has greater access to available biomass. Areas surrounding low carbon transport switching locations, such as shipping ports and rail terminals, have elevated performance due to the increased access to less carbon-intensive transport methods. The least optimal location for all three supply chains is Kingsbridge (30 km east of Plymouth) due to it being the furthest distance in the UK from one of the modeled geological storage sites. Great efforts in decarbonization would have to be made to improve the carbon performance of the area. This is not to say that BECCS is not possible in this location, but rather these three types of BECCS do not perform well in this location. Another type of BECCS, perhaps a high purity low yield manure derived anaerobic digester BECCS would perform better in this area but would still require major decarbonization of transportation.

### Carbon Impact of Shifting BECCS Siting

The optimization of the siting of a BECCS facility is critical in minimizing the positive emissions in the projects carbon balance to achieve a favorable net-negative carbon balance. It may not be possible to site a BECCS facility in its most carbon-optimal location due to competition, in terms of land and available infrastructure/resources, with other sectors or potentially other BECCS facilities. The siting of a BECCS facility is extremely important to maximize its carbon performance.



The shifting of the siting location of each supply chain results in drastic changes in their carbon performance. For each of the supply chains, multiple radial lines of points every

10 m perpendicular to the transition from green to orange to red/white regions of the carbon performance maps were used to extract the values, and the average percentage change in

**TABLE 2 |** Percentage increase in spatially explicit supply chain emissions for the MSW, wheat straw and sawmill residue supply chains from shifting the siting of the facilities from a green region to an orange region and from a green region to a red/white region.

Supply chains	Green region to orange region ( $\Delta\%$ )	Green region to red/white region ( $\Delta\%$ )
MSW	672.9	1,345.9
Wheat straw	804.8	1,609.6
Sawmill residue	298.8	597.5

carbon performance from green regions to orange regions and green regions to red/white regions were recorded. These average percentage changes showcase the percentage increase in supply chain emissions for each of the supply chains if they were moved from a high performance area (green region) to a moderate performance area (orange region) and to a low performance area (red/white region).

Shifting the siting of the BECCS facilities from a green region of the performance maps to an orange region results in a 298.8–804.8% increase in spatially explicit supply chain emissions, while shifting from a green zone to a red/white zone results in a 597.5–1,609.6% increase. A breakdown of the percentage changes in shifting from a green region to an orange and red/white region by each of the supply chains is presented in **Table 2**. Dramatically shifting the location of the BECCS facility in such a manner will make achieving a favorable net-negative carbon balance very difficult. On average, a 10 km shift in the siting of a BECCS facility will result in between an 8.6 and 13.1% increase in spatially explicit supply chain emissions.

## BECCS Supply Chain Tethering

The carbon performance heatmaps for the supply chains look similar as all three of the modeled supply chains in this analysis are more tethered to their CO<sub>2</sub> infrastructure than to their biomass and hydrogen, indicating that greater carbon savings would be made by focusing decarbonization efforts on CO<sub>2</sub> transportation rather than the other limbs of the supply chains. However, the level and scale of tethering varies dramatically between the supply chains and where the BECCS facility is located.

Whether or not a BECCS supply chain will be more tethered to its biomass or its CO<sub>2</sub> infrastructure depends heavily on its geography, access to low carbon infrastructure, access to available biomass, distribution of its biomass and the ratios of transported tonnages. A generalized trend can be identified for the tethering of different styles of BECCS. BECCS supply chains which produce an initial high purity CO<sub>2</sub> stream at low yields will generally be more tethered to its biomass [CPER, Decatur Illinois, Occidental White Energy, Lamberton, Plainview and Goldfield (BusinessWire, 2019; Global CCS Institute, 2021; Summit Carbon Solutions, 2021; U.S. Securities Exchange Commission, 2021)], while BECCS supply chains which produce an initial low purity CO<sub>2</sub> stream at high yields will generally be more tethered to its CO<sub>2</sub> infrastructure [Drax, Stockholm Exergi, Copenhill,

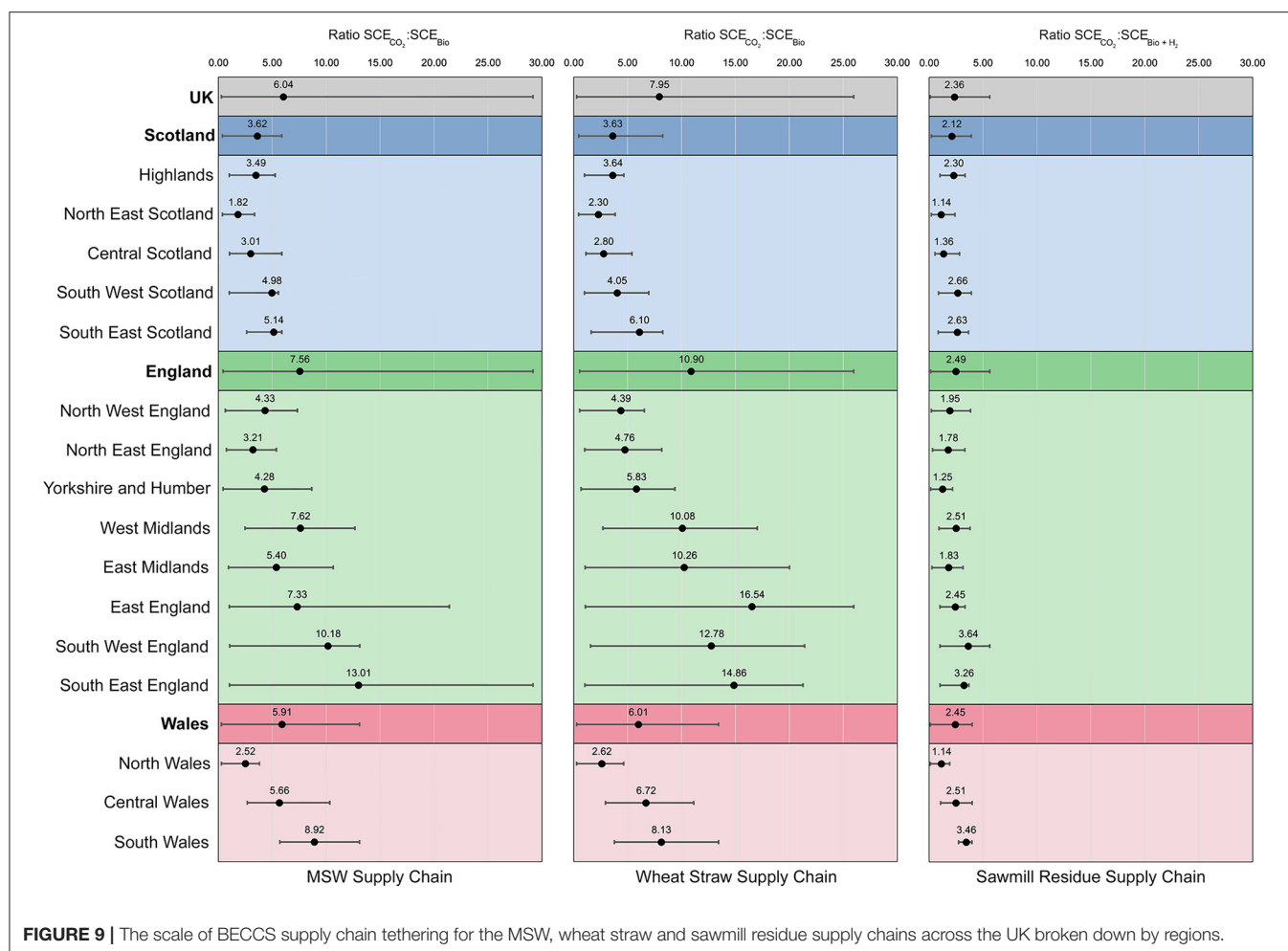
Mikawa and Project Bright (Drax, 2019; Toshiba, 2020; Global CCS Institute, 2021; HyNet, 2021a; Stockholm Exergi, 2021)]. Although each supply chain would have to be tailored and evaluated on a case-by-case basis and depending on their relative access to available biomass and geological storage would exist on a sliding scale of tethering. Generally, high purity low yield BECCS supply chains will perform more carbon optimally in more rural areas with strong access to biomass, while low purity high yield BECCS supply chains will perform more carbon optimally in more industrial areas with strong access to geological storage sites.

The level and scale of tethering can be quantified by taking the ratio between pairs of each supply chain limb (CO<sub>2</sub> limb, Biomass limb and H<sub>2</sub> limb), revealing how many more times the supply chain is tethered to a specific limb. The scales of tethering for the modeled supply chains broken down by regions in the UK is presented in **Figure 9**. On average across the entire UK, the MSW supply chain is 6.04 times more tethered to its CO<sub>2</sub> infrastructure compared to the other limbs, the Wheat Straw supply chain is 7.95 times more tethered and the Sawmill Residue supply chain is 2.36 times more tethered.

The difference between the supply chain tethering across the modeled supply chains is driven by its access to available biomass, the distribution of its biomass, access to geological CO<sub>2</sub> storage and access to its energy end-user. The MSW and Wheat Straw supply chains follow similar trajectories in their tethering as both have a well-distributed and plentiful biomass source across the UK, resulting in a wide range in the scale of supply chain tethering. The Sawmill Residue supply chain follows the same trajectory of tethering but has been skewed toward a more equal balance in supply chain tethering due to its concentrated and clustered biomass distribution and the level of access to its energy end-user. The sawmill residue across the UK are not as well distributed as the other modeled biomass types and tend to be far away from the ICCS clusters, resulting in further and more carbon-intensive biomass transportation than the other supply chains. The Sawmill Residue supply chains must also incorporate hydrogen transportation into the national grid into the tethering, while the other two supply chains do not. Other low purity high yield BECCS supply chains with concentrated biomass distributions, such as energy crop derived BECCS projects, will be expected to have similar and more equal scales of tethering to the Sawmill Residue supply chain rather than the other modeled supply chains.

Within a BECCS supply chain the scale of tethering varies greatly and exists on a spectrum depending on if the facility is located within an ICCS cluster or in a more rural location.

BECCS facilities located within ICCS clusters that have access to a CO<sub>2</sub> pipeline tend to be more equally tethered to its biomass and CO<sub>2</sub> but are still overall more tethered to its CO<sub>2</sub> infrastructure. The level of tethering within 50 km of pipeline access for the Scottish, Teesside, North West and Humber clusters and 50 km from the shipping ports for the South Wales cluster are presented in **Table 3**. Indicating that a more balanced approach in decarbonizing the supply chain limbs would result in the most carbon savings while still having a slight focus on the CO<sub>2</sub> limb. However, since the South Wales



**FIGURE 9 |** The scale of BECCS supply chain tethering for the MSW, wheat straw and sawmill residue supply chains across the UK broken down by regions.

cluster lacks a CO<sub>2</sub> pipeline, the facilities within the cluster are more tethered to its CO<sub>2</sub> infrastructure than the UK average due to the large shipping distances to store CO<sub>2</sub> within the Hamilton reservoir. On average, the MSW, Wheat Straw and Sawmill Residue supply chains are 8.92, 8.13, and 3.46 times more tethered to its CO<sub>2</sub> infrastructure within the South Wales cluster, respectively. Indicating that greater carbon savings within the supply chains would be made if decarbonization efforts were focused on decarbonizing shipping within the cluster compared to the rest of the supply chain.

Due to the lack of proximal access to CO<sub>2</sub> storage sites, the scale of tethering for more rural areas of the UK are heavily tethered to its CO<sub>2</sub> infrastructure for the modeled supply chains. On average, the MSW, Wheat Straw and Sawmill Residue supply chains are 3.94, 3.64, and 2.12 times more tethered to their CO<sub>2</sub> infrastructure in the Highlands, respectively. The supply chains are 5.66, 6.72, and 2.51 times more tethered within Central Wales and 13.01, 14.86, and 3.26 times more tethered in South-East England. The decarbonization of shipping and rail transportation would dramatically improve the carbon performances in these more rural areas around the UK, allowing for the wide dissemination of BECCS across the UK and not only siting the facilities within the ICCS clusters. A general

trend across all supply chains is that the further south the BECCS facility is placed, the more tethered the facility is to its CO<sub>2</sub> infrastructure. This is due to the majority of geological storage sites and CO<sub>2</sub> compatible pipelines across the UK being in the country's northern regions, resulting in larger emissions accessing this infrastructure from the south.

## BECCS Supply Chain Performances Within ICCS Clusters

The carbon performances of the modeled BECCS supply chains differ drastically within the ICCS clusters. The MSW, Wheat Straw and Sawmill Residue supply chains carbon performance maps focusing within each of the ICCS clusters is presented in **Figure 10**. The highest performance areas within the clusters are concentrated around the CO<sub>2</sub> pipeline compressors. In contrast, for South Wales the highest performance areas are concentrated around the shipping ports and rail terminals since the cluster lack a CO<sub>2</sub> compatible pipeline and proximal access to geological storage.

The MSW and Wheat Straw supply chains outperform the Sawmill Residue supply chain in all ICCS clusters with a CO<sub>2</sub> pipeline due to the comparatively increased access to available biomass, energy end-users and low carbon infrastructure.



**TABLE 3 |** Supply chain tethering for MSW, wheat straw and sawmill residue broken down by ICCS clusters.

	MSW		Wheat straw		Sawmill residue	
	Mean	Range	Mean	Range	Mean	Range
Scottish	2.71	0.36–5.89	2.43	0.46–5.49	1.11	0.20–2.16
Teesside	2.19	0.75–4.13	3.49	1.02–7.02	0.95	0.31–1.61
North West	2.81	0.28–6.13	2.48	0.26–4.78	0.84	0.11–1.52
Humberside	3.72	0.43–8.25	5.42	0.67–8.99	0.93	0.15–1.48
South Wales	9.37	5.70–13.08	8.52	3.77–14.11	3.52	2.74–3.99

Notably, the MSW supply chain performs at a higher level along the entire length of the Feeder 10 pipeline in the Scottish cluster due to the abundance of available biomass, while the Wheat Straw supply chain performs at a higher level at the northern end of the pipeline due to the distribution of wheat straw being greater in the Northeast of Scotland. Contrarily, the Sawmill Residue supply chain outperforms both the MSW and Wheat Straw supply chains within the South Wales cluster due to the moderate access to sawmill residue and lack of large enough quantities of MSW and wheat straw to store the required 1 MtCO<sub>2</sub>/yr.

## Comparative BECCS Supply Chain Decision Mapping

A decision map is a map that can choose between a selection of options under a specific set of criteria at a high spatial resolution to clearly show what should go where under the set criteria. The main objective of this decision map is to provide a heuristic to help inform stakeholders such as policymakers, NGOs, academia or industry in determining where to locate BECCS facilities.

A comparative BECCS supply chain decision map was generated for the three modeled supply chains, indicating for a given area of the UK which of the three supply chains captured 1 MtCO<sub>2</sub>/yr for the lowest amount of spatially explicit supply chain emissions per MWe. The decision map is presented in **Figure 11**.

The phrasing of the question when generating a decision map is critical. A decision map showing which supply chains should be chosen to capture 0.5 MtCO<sub>2</sub>/yr for the lowest amount of spatially explicit supply chain emissions per MWe would show vastly different results than 1 MtCO<sub>2</sub>/yr. This is due to the smaller operational footprints of the supply chains as less distance is needed to be traveled to meet the biomass quotas to capture 0.5 MtCO<sub>2</sub>/yr compared to 1 MtCO<sub>2</sub>/yr.

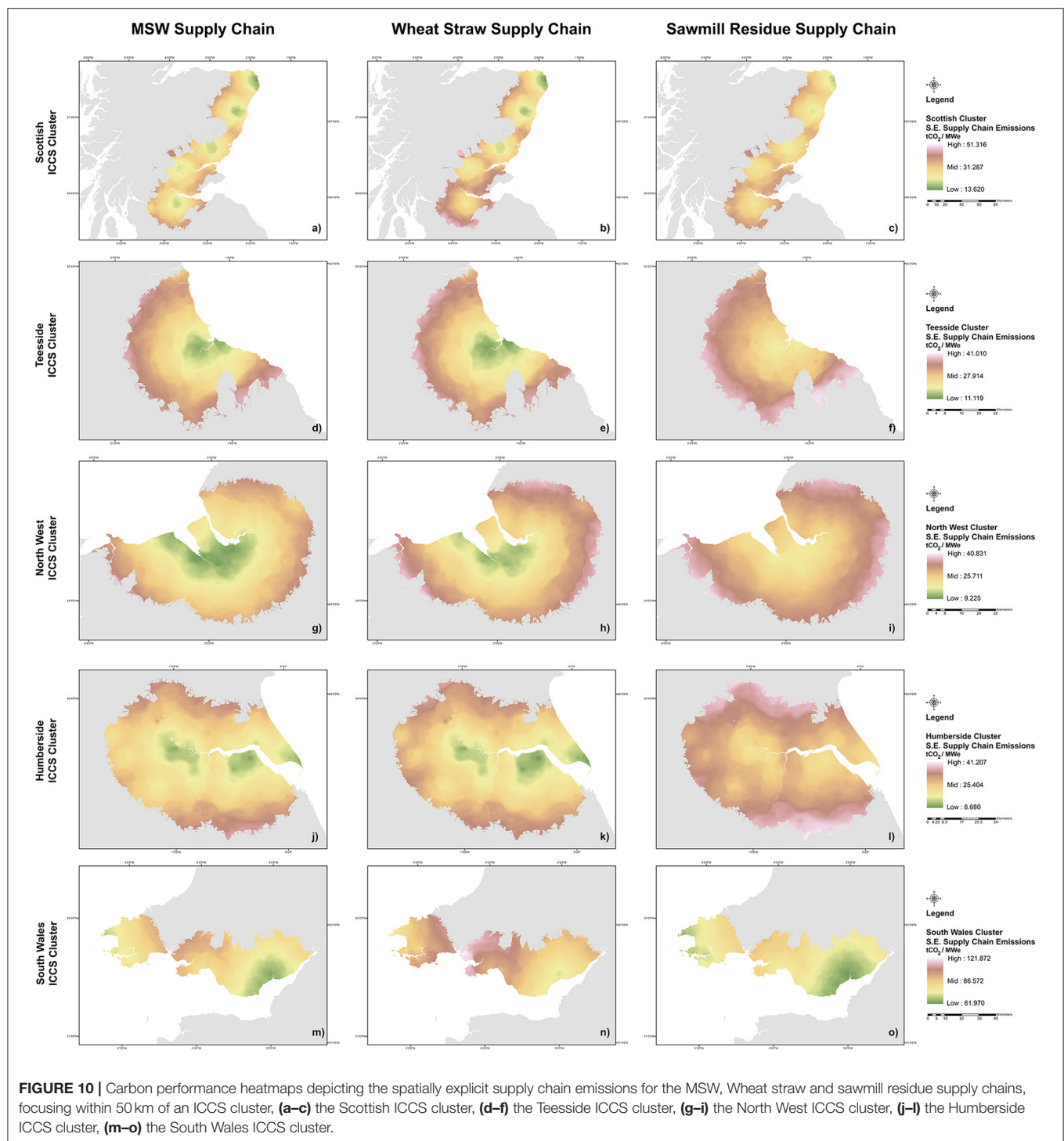
The selection process deciding which supply chain is chosen is based on access to biomass, energy end-users and access to low carbon infrastructure. The MSW and Wheat Straw supply chains were selected for each location based on their biomass and low carbon infrastructure access. In comparison, the Sawmill Residue supply chain selection was based on its biomass, energy end-user and low carbon infrastructure access.

There are four main bodies of choice for the three supply chains across the UK. The Wheat Straw supply chain is the better choice across Teesside, the east side of Humberside and the coastal areas of the East Midlands and East England. The MSW supply chain is the better choice across North Wales, NW England, Yorkshire, the west side of Humberside and Scotland's

east coast. The MSW supply chain has not been selected for South Wales, but it may have been chosen if the decision map was recalibrated for a smaller BECCS facility rather than 1 MtCO<sub>2</sub>/yr. The Sawmill Residue supply chain is best located in more isolated rural areas such as Southwest England, Cumbria, Scottish Highlands, the Outer Hebrides, Orkney and Shetlands.

Notably, there are a series of small zones within the decision map which identify one of the supply chains within another supply chain zone. This is most apparent for the Greater London area, where the MSW supply chain performs best within wider surroundings dominated by the Sawmill Residue. These zones are termed “enclaves” and are driven by areas with increased levels of access to biomass, energy end-users and particularly low carbon infrastructure compared to their surrounding areas. The Greater London enclave is selected due to the abundance of MSW and excellent access to shipping and rail terminals compared to the areas surrounding Greater London. There are eight groupings of enclaves across the UK, with the majority based in England, one in Scotland and none in Wales. There are more enclaves in England compared to Scotland and Wales due to the widespread distribution of low carbon transport switching locations. England has distinct areas of dense and sparse infrastructure which allow the decision map to form enclaves, while the infrastructure in Scotland and Wales is concentrated along the east coast and north and south coast, respectively, which makes enclaves less likely. The enclaves in England are driven by increased access to shipping ports and rail terminals, while the enclaves in Scotland are driven by increased access to the feeder 10 pipeline.

The information conveyed by the decision map has the potential to help inform policymakers and industry on how to best place BECCS facilities across the UK, but much more research will be needed to cover all aspects and contexts of BECCS deployment. The decision map only focuses on the carbon emissions for the modeled supply chains. Further research will be needed to ensure that BECCS facilities are right-sized into its local infrastructure, available biomass, economic environment, energy systems, land management strategies, geography, and socio-political context. The decision map has been calibrated for 1 MtCO<sub>2</sub>/yr, but many locations may be better suited for larger or more likely smaller facilities. Future research into determining the optimal BECCS facility scale for any location in the UK will be greatly needed to ensure a successful and sustainable deployment of BECCS. Future work will also be performed on the decision map to capture other constraints such as biomass availability volatility, transportation costs, integration

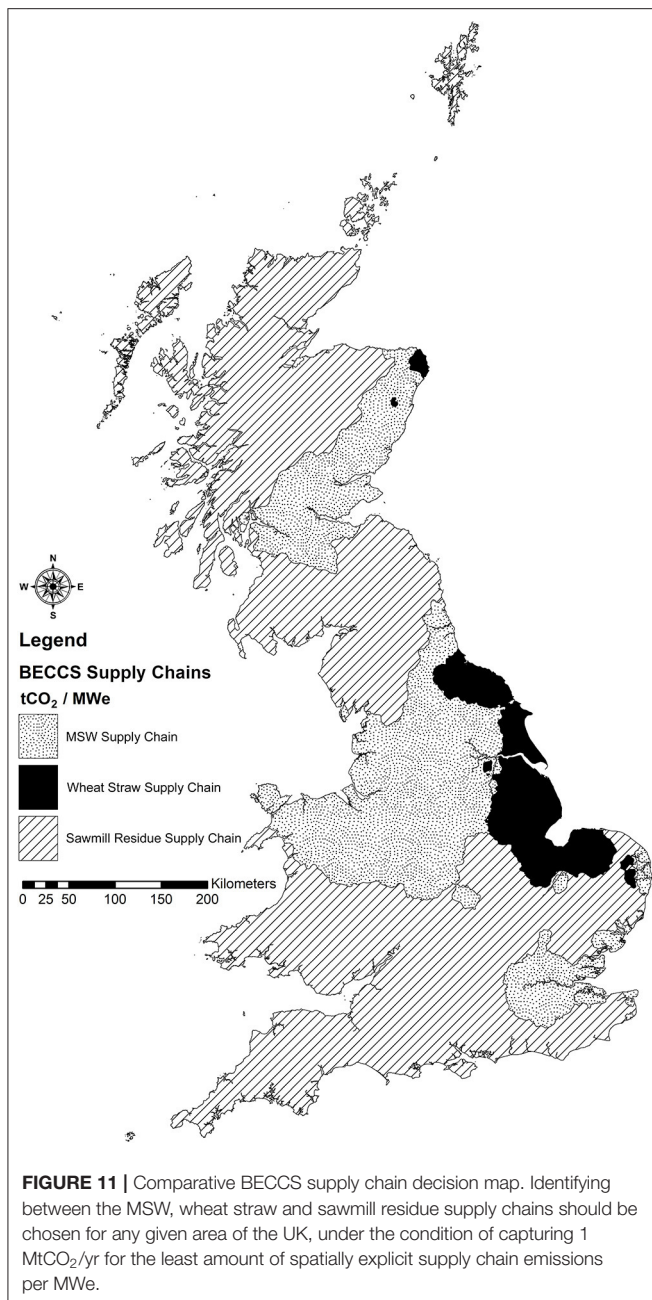


of renewable fuels, rapidly evolving  $\text{CO}_2$  infrastructure and static supply chain emissions.

It is important to highlight that this study only considers the spatially explicit supply chain emissions for each supply chain and does not include the spatially static emissions such as biomass drying, biomass grinding, fuel combustion,  $\text{CO}_2$  compression,  $\text{CO}_2$  injection and  $\text{CO}_2$  remediation. The study

also does not explore land-use change implications such as soil carbon sequestration, erosion control and consequences of climate change. This is particularly important for cereal straw derived BECCS projects like the Wheat Straw supply chain in this study. Further collaboration with LCAs, sustainability studies, economic feasibility studies and stakeholder engagement studies will be needed to ensure a sustainable and successful





integration of BECCS into our energy generation, land management strategies, waste management strategies and socio-economic frameworks.

If the three modeled BECCS supply chains were to be rolled out across the UK, assuming the biomass availability used in this analysis, the theoretical output of the MSW supply chain would be 6.53 MtCO<sub>2</sub>/yr, the theoretical output of the Wheat Straw supply chain would be 12.39 MtCO<sub>2</sub>/yr and the theoretical output of the Sawmill Residue supply chain would be 3.79 MtCO<sub>2</sub>/yr. This would constitute 43.5% of the required amount of BECCS to meet Net-Zero by 2050 under the UK BECCS

requirements in the Committee on Climate Change Sixth Carbon Budget balanced pathway scenario. The roll-out of these supply chains would not be as one mega facility, but rather disseminated as a series of smaller facilities that have been tailored and calibrated to be successfully integrated into their geographic, economic, environmental and social context. The realistic output of these supply chains may be less as these roll-outs calculations assume 100% utilization of available biomass, which may not be economically viable, particularly in more rural areas of the country. The theoretical outputs also assume no reduction in the level of competition with other sectors, particularly for MSW, and no reduction in biomass availability from zero-waste campaigns and policies.

Future analysis will also have to account for the impact of climate change on the supply of biomass for BECCS as some representative concentration pathways could see a 46% decrease in biomass yield (IPCC, 2021). The realistic outputs of the supply chains may also be increased through mixing the feedstocks with other sources or through importation, but this may reduce the calorific value of the fuels, reduce the supply of biomass for other countries and result in unwanted additional transportation emissions. Although, there are potential limitations in scaling up bioenergy and BECCS facilities to the required level to meet Net-Zero from what is currently available due to limited biomass supply and limited operational timeframes. These limitations may be circumvented with the recent advances in pre-treatment techniques and dry biomass storage systems to prolong the operational timeframes of the facilities and secure biomass supply (Victorin et al., 2020; Wendt and Zhao, 2020). Future analysis exploring the implications of multiple smaller decentralized BECCS facilities with reduced operational timeframes may also reveal potential pathways for BECCS deployment.

## CONCLUSION

The work produced in this paper using the CNS model has the potential to aid the successful and sustainable deployment of BECCS across the UK. BECCS is an incredibly diverse and unique technology that needs to be carefully tailored to its local infrastructure, available biomass, economic environment, energy systems, land management strategies, socio-political context and geography for successful deployment and integration. Each BECCS facility must be evaluated on a case-by-case basis, and some supply chain designs perform better in some locations than others. The findings from this paper can be replicated for any other BECCS supply chain or simulated in any other country. The findings produced also have the potential to act as a useful heuristic for policymakers and industry when planning where to site specific BECCS projects.

The shifting of a BECCS facility will result in a drastic increase in spatially explicit supply chain emissions, where for the modeled supply chains for each 10 km shift in facility location results between an 8.6 and 13.1% increase in emissions. The optimal placement of a BECCS facility will play a significant role in achieving a favorable net-negative carbon balance and should be considered in the inception of each and every project.

A BECCS project may be more tethered to its CO<sub>2</sub> infrastructure or biomass and, as a general rule, a BECCS project producing an initial high purity low yield CO<sub>2</sub> stream will be overall more tethered to its biomass, and a project producing an initial low purity high yield CO<sub>2</sub> stream will be overall more tethered to its CO<sub>2</sub> infrastructure. All three of the modeled supply chains were more tethered to its CO<sub>2</sub> infrastructure, but the scale of tethering varies greatly and exists on a spectrum depending on if the facility is located within an ICCS cluster or in a more rural location. Locations within ICCS clusters tend to be more equally tethered to both its biomass and CO<sub>2</sub> infrastructure, with a slight preference for the CO<sub>2</sub> infrastructure, and more rural areas tended to be more strongly tethered to its CO<sub>2</sub> infrastructure; Indicating that low purity high yield BECCS project will perform at higher levels within ICCS clusters and high purity low yield BECCS projects will perform at higher levels in more rural areas.

This paper has also introduced a BECCS decision map indicating for any given area which of the modeled BECCS supply chains should be chosen to best store 1 MtCO<sub>2</sub>/yr for the least amount of spatially explicit supply chain emissions. The decision map reveals that the modeled supply chains have a performance proclivity for certain areas of the UK and that one BECCS supply chain that operates optimally in one location cannot be transplanted or replicated into a new location without tailoring

the supply chain to its new geographic, economic, environmental and social context.

## DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

## AUTHOR CONTRIBUTIONS

MF created and calibrated the model and software, performed the statistical analysis, and wrote the first draft of the manuscript. MF, CG, AW, and AL-L wrote sections of the manuscript. All authors contributed to conception and design of the study, contributed to manuscript revision, read, and approved the submitted version.

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# Regulatory Preconditions for the Deployment of Bioenergy With Carbon Capture and Storage in Europe

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Paris-compatible climate scenarios often consider bioenergy with carbon capture and storage (BECCS) as an important technology for carbon dioxide removal (CDR). Although the main barrier to BECCS deployment is often associated with lack of economic policy incentives, unfavorable regulations can also impede investments. Over the past decade, the regulatory conditions at the UN and on the EU level have developed to be more permissive toward BECCS. For instance, CDR accounting guidelines have been developed by the UN, and the EU CCS Directive regulates responsibilities regarding storage of CO<sub>2</sub>. However, several existing regulations still cause deployment hurdles. Taking a European viewpoint, this perspectives article takes stock of recent regulatory developments and provides a discussion on legal acts that need to be reformed in order to facilitate BECCS deployment. Although the European trend is characterized by developing a regulatory regime that is more supportive of BECCS, we identify three areas for further improvement: (1) allowing EU Member States to use negative emissions from BECCS to comply with their obligations under the legislative pillars that underpin the EU's climate objectives; (2) amending the CCS Directive to exempt physical leakage of biogenic CO<sub>2</sub>, attributable to sustainably sourced biomass, from the requirement to surrender emission allowances in the EU ETS or, if BECCS has been economically rewarded, the penalty for leakage should correspond to the level of the reward; and (3) pushing to erase the last few barriers due to multilateral regimes, such as clarifying whether BECCS is covered by the geoengineering moratorium maintained by the UN Convention on Biological Diversity. These proposed reforms would further improve the regulatory preconditions for BECCS deployment in the EU.

**Keywords:** bioenergy with carbon capture and storage (BECCS), carbon dioxide removal (CDR), negative emissions, European Union (EU), climate policy, regulation

## INTRODUCTION

The pathways for restraining global warming to 1.5°C rely on carbon dioxide removal (CDR) to offset emissions. In several climate change scenarios, bioenergy with carbon capture and storage (BECCS) is a key CDR mitigation technology (IPCC, 2018) that could be used to offset residual emissions in sectors that are difficult to fully mitigate, such as agriculture, construction, and



heavy industries (Fuss et al., 2014; Honegger et al., 2021b). While the technical conditions for capturing and storing CO<sub>2</sub> exist, BECCS is still under development. If deployed, BECCS has the potential to achieve negative emissions by capturing and storing biogenic CO<sub>2</sub> that, in turn, has been sequestered from the atmosphere through photosynthesis (Fuss et al., 2018). However, a gap remains between the current lack of BECCS deployment and its theoretical potential (e.g., Geden et al., 2018). Although some countries have started drafting CDR policies (Schenuit et al., 2021) and planned projects and promising pilots exist, few are currently operating at scale. Tamme and Beck (2021) argue that the commercialization gap is primarily explained by high costs and lack of infrastructure for CO<sub>2</sub> transport and storage. In addition, Fridahl et al. (2020a) argue that the gap can, to a large extent, be explained by a lack of policy incentives. On the other hand, every year of non-deployment of CDR techniques like BECCS and DACCS will incur extra costs to meet the European CO<sub>2</sub> targets (Galán-Martín et al., 2021).

By emphasizing the need to address barriers to BECCS if it is to contribute to EU climate action, this perspective article will focus on regulatory preconditions for BECCS deployment. While noting the severe lack of economic policy incentives to commercialize BECCS, this article will instead primarily focus on regulations that can create an administrative burden and thus extra transaction costs or other types of barriers that can hinder the deployment of the entire BECCS chain, from capture, to transportation, to storage of biogenic CO<sub>2</sub>. Applying a European viewpoint, this article provides a discussion on how several multilateral and EU regulatory barriers have recently been lowered or erased and points out the most crucial remaining barriers to BECCS deployment. The article ends with policy recommendations to address and eliminate the remaining regulatory barriers to BECCS deployment.

## BECCS Falls Between the Cracks of the Three EU Climate Policy Pillars

First and foremost, with current accounting rules negative emissions from BECCS cannot be used to achieve the EU's climate objectives. While it is possible for EU Member States to report negative emissions from BECCS, such negative emissions cannot be used to comply with obligations under any of the three legislative pillars designed to deliver the EU's economy-wide objectives for 2030 and 2050, i.e., the EU Emissions Trading System (EU ETS), the Effort Sharing Regulation (ESR), and the Land-Use, Land Use Change, and Forestry (LULUCF) Regulation. In other words, negative emissions from BECCS can be recognized for information purposes but not to achieve Member States' obligations, and thus the current EU regulations do not encourage Member States to incentivize BECCS within their jurisdictions.

It should be noted that the European Commission recently proposed a target to remove 5 MtCO<sub>2</sub> in 2030 using technological CDR (EU, 2021c). Even if this is a rather modest target, and even if it is likely possible for the EU to report such removal as contributing to achieving the EU's Nationally Determined Contribution to the UNFCCC's Paris Agreement, the current

regulations would not allow the EU Member States, in which the engineered CDR method would be deployed, to use the resulting negative emissions to comply with EU-internal obligations. Addressing this shortcoming is vital for incentivizing BECCS deployment in the EU.

Regarding the EU ETS, it establishes a cap-and-trade system in the EU with the aim to cost-effectively reduce greenhouse gas emissions (EU, 2003, 2018a). The EU ETS sets a cap on total emissions allowed from all facilities that are covered by the system. Each year, the facilities must surrender tradable emission allowances corresponding to their actual emissions, thus creating an incentive to reduce emissions. In 2021, the European Commission proposed reforms to strengthen the system in line with the new and more ambitious economy-wide 2030 EU target, i.e., to reduce emissions by at least 55% by 2030 compared to 1990 levels (EU, 2021a). However, the current EU ETS Directive does not acknowledge *negative* emissions from facilities that apply BECCS. While the EU ETS acknowledges biogenic CO<sub>2</sub> emissions from facilities covered by the Directive, for example, biomass co-fired powerplants, the emission factor for biomass is set to zero. Consequently, if such facilities capture and store biogenic CO<sub>2</sub>, the stored CO<sub>2</sub> will have to be accounted for as zero rather than negative emissions. As such, negative emissions from BECCS cannot be used to compensate for the need to surrender allowances. The climate benefit of biogenic CO<sub>2</sub> stored permanently could, in principle, offset an emission pulse of an equal amount of fossil CO<sub>2</sub>, at least if the removal and the emissions were to occur at about the same time (Zickfeld et al., 2016; Fridahl et al., 2020b). As is, BECCS is not allowed to offset fossil emissions, as clarified by the European Commission in answer to a question on this topic posed by the Norwegian government (EC, 2020). The European Commission confirms that there is a lack of legal support in the EU ETS to allow stored biogenic CO<sub>2</sub> to be recorded as a negative emission at the capture facility (EC, 2020). The Commission pointed out that incentives for BECCS should be created in other ways (EC, 2020), yet have also noted elsewhere that "CCS-biomass projects with a clear and verifiable climate benefit could potentially benefit from recognition pursuant to Article 24a of the revised EU ETS Directive" (EU, 2021c, p. 16). Article 24a does not, however, imply an inclusion of BECCS in the EU ETS, and it instead mandates the European Commission to issue allowances or credits from emission reduction projects in Member States provided that the sources are not already covered by the EU ETS. In other words, Article 24 provides a basis to establish an offsetting system *external* to the EU ETS. Rickels et al. (2021) also present a comprehensive analysis of various possibilities for reforming the EU ETS to incentivize BECCS, including how emission allowances could be linked to negative emission credits without eroding incentives to phase out fossil emissions. The fact that biogenic emissions are generally recognized in the EU ETS, even if accounted for as carbon neutral, opens the door for allocating allowances for free to BECCS facilities. Yet, if such an approach were to be implemented, it should be noted that the EU ETS does not cover installations that exclusively use biomass, which would lower the potential of BECCS negative emissions (Rickels et al., 2021). In the Commission's proposal

for EU ETS reforms (EU, 2021a), emitters of biogenic CO<sub>2</sub> from biomass of unsustainable origin—as defined in the Renewable Energy Directive (EU, 2018c) with proposed amendments to strengthen the criteria (EU, 2021d)—would have to surrender emission allowances. If eventually approved as EU law, this would generate incentives for BECCS fueled with *unsustainably* sourced biomass, i.e., incentives to use BECCS for emissions reductions rather than removal. As is, neither the current nor the proposed revised EU ETS Directive would provide economic incentives for generating negative emissions using BECCS.

The ESR supplements the EU ETS by setting a target for emissions reductions in the non-trading sectors and by allocating individual commitments to the EU Member States (EU, 2009a, 2018b, 2020a). Neither the current ESR regulation (EU, 2018b) nor the recently proposed ESR revision to strengthen the 2030 target from 30 to 40% compared to 2005 levels (EU, 2021b) allow an accounting for negative emission via BECCS. Analogous to the lack of recognizing BECCS in the EU ETS, the fact that the ESR does not allow negative emission via BECCS to reach national reduction targets constitutes a barrier to BECCS deployment.

The LULUCF Regulation does in fact already recognize some forms of CDR (forest sinks and harvested wood products). It requires the EU Member States to maintain their existing LULUCF sinks (the no-debit rule), and the European Commission is proposing to further increase the volume of aggregated, obligatory removal by 2030, to expand the regulation to managed land by 2026, and to extend the regulation to all non-CO<sub>2</sub> emissions and CDR from agriculture from 2031 onwards, including emissions from soils and from other agricultural sources, e.g., enteric fermentation, manure management, liming, and use of fertilizers (EU, 2021e). A prime focus of BECCS in many scenarios underpinning mid-century net-zero targets is to offset residual emissions in agriculture (Geden et al., 2019; Buylova et al., 2021). Thus, moving agriculture from the ESR to the LULUCF Regulation would strengthen the case for recognizing BECCS in the LULUCF Regulation. Allowing BECCS to be used to offset residual emissions from agriculture could alleviate pressure on forestry to increase the carbon sink. This could be an important service provided by BECCS because demand for wood-based products is likely to increase with an increasing need to find substitutes for fossil-intensive products. While there are possible trade-offs between using biomass from forests as a fuel and generating an increasing carbon sink in forestry, the technical potential for BECCS in the pulp and paper industry as well as biofuel production is likely to continue to be high in the foreseeable future. Realizing this potential could help to achieve the no-debit obligation under the LULUCF Regulation if BECCS were to be acknowledged in this sector. However, this would require the use of sustainably sourced biomass in BECCS facilities or else the climate benefit of BECCS would be undermined. At the same time, it should be noted that the BECCS supply chain spans several sectors, including the energy and industrial sectors. Thus, deciding to recognize BECCS under the LULUCF Regulation is not straightforward. If BECCS is included in the LULUCF Regulation, this may raise questions about consequential amendments regarding the existing flexibility between LULUCF and ESR and may require

an evaluation of the need to adopt new flexibility between the LULUCF Regulation and the EU ETS.

### Accounting Guidelines for BECCS Are Improving Rapidly

The IPCC's accounting framework to account for negative emissions from BECCS—even if BECCS cannot be used to fulfill EU Member States' commitments or achieve joint objectives—has developed rapidly in the last couple of years. The international accounting guidelines developed by the IPCC, used as a basis for agreeing on accounting rules in the UNFCCC and the EU (EU, 2009b; UNFCCC, 2019a,b), constitute a favorable basis for allocating the storage of biogenic CO<sub>2</sub> as negative emissions. However, the accounting framework needs to be further developed to enable transparent, internationally comparable reporting of negative emissions in order to incentivize CDR, including BECCS. As stressed by Tamme and Beck (2021), accounting is important to verify that CDR is achieved. Sustainability safeguards, for example, are not well developed. In the IPCC guidelines, it is assumed that emissions (and removal) associated with biomass harvest (and growth) are reported in the LULUCF sector. With this assumption, point source emissions from processes using biomass should be accounted for as climate neutral because the biomass-related emissions and removal should already be accounted for. It follows, therefore, that if BECCS is deployed to remove and store a point source emission pulse, the zero emission baseline means that this stored emission pulse should be reported as a negative emission. From a system perspective, however, the climate benefit of BECCS would be undermined by unsustainably sourced biomass. Therefore, it is positive that further clarity on monitoring and accounting is in the policy pipeline. Three examples of this are noteworthy, namely the strengthened sustainability criteria under the Renewable Energy Directive, the proposal that emitters of biogenic CO<sub>2</sub> have to surrender allowances under the EU ETS unless the biomass meets the new sustainability criteria defined in the Renewable Energy Directive, and the inclusion of managed land under the LULUCF Regulation from 2026 onwards and agriculture from 2031 onwards. The European Commission is also developing a proposal for a carbon removal certification scheme that is expected to be completed within a few years (EU, 2020c). The certification scheme will tentatively be proposed in the last quarter of 2022 (EU, 2021f) and will constitute a key building block of the circular economy action plan (EU, 2020c). It will most likely allow comparing the removal quality of different types of carbon removal, which, as noted by Fridahl et al. (2020b), is necessary to guide political discussions on prioritizations among CDR options and to standardize more refined carbon decay functions in the accounting of various CDR methods, analogous to the agreed half-times of different types of harvested wood products.

### New CDR Market on the Horizon

Based on the proposed certification scheme, the European Commission has also signaled an intent to support the voluntary, or even establish a new market, for CDR in the land sector,

starting with what it refers to as “nature-based” removal (EU, 2021c,e). It seems likely that such a market will be proposed to serve under the no-debit rule in the LULUCF Regulation so that land-based CDR will be allowed to compensate for land-use, forestry, and agriculture emissions much like forest sinks are already allowed to compensate for forest emissions sources.

The certification scheme, and new and more refined accounting of CDR, would make various CDR methods comparable, allowing one to weigh the value of engineered CDR against nature-based CDR, i.e., it would make different types of CDR fungible. As such, the proposed CDR market could be expanded beyond nature-based approaches to also include engineered CDR. Integrating BECCS into this market could be facilitated by regulating it under the same legislative pillar, i.e., LULUCF. This has the potential to strengthen cost-efficiency by diversifying supply and increasing liquidity and to improve preconditions for achieving the EU’s ambition to become a net greenhouse gas sink after 2050. On the other hand, the aspiration of the EU to become net negative beyond 2050 necessitates what Geden et al. (2019) refers to as a comprehensive CDR approach, requiring a substantially higher amount of removal than the net-zero target. A comprehensive CDR approach with a higher EU ambition and a focus on long-term provision of CDR after 2050 is likely to entail a change in political priority and governance. This would mean that the focus on methods to achieve CDR would have to expand, which could translate into encountering new and as yet unidentified regulatory barriers.

It should be stressed, however, that the European Commission has not provided any clear guidance on how it envisages the scope for expanding a new nature-based CDR market to include BECCS and other engineered CDR, nor how such an expanded market could be linked to a regulatory home for BECCS among the legislative pillars that underpin the climate objectives of the EU. This regulatory unclarity, heavily linked to the issue of creating economic incentives for BECCS, requires the full attention of the European Commission in order to scale up BECCS deployment.

## Sustainability Safeguards, the Taxonomy for Investments, and State Aid Rules

The UN Convention on Biological Diversity (UNCBD) is a global treaty aimed at preserving biological diversity and to assure the sustainable use of components of biological diversity. In 2010, the parties to the CBD decided on a moratorium on geoengineering, which stated that because there is no transparent global control mechanism for geoengineering, no climate-related geoengineering activities that might affect biodiversity should take place before sufficient scientific knowledge exists to justify such activities, a decision that was reaffirmed by the UNCBD in 2016 (CBD, 2010, 2016). However, in the moratorium the capture and storage of CO<sub>2</sub> from fossil fuels are excluded. Moreover, the moratorium contains no definition of geoengineering, and thus it is hard to interpret if BECCS is covered, yet the CCS exemption does not refer to BECCS explicitly. In addition, the general confusion on how geoengineering is defined and interpreted in the context of CDR further complicates this dispute (Honegger

et al., 2021a). The ambiguity of how to interpret and apply the moratorium may introduce uncertainty that can translate into an investment barrier for BECCS. Although it is important to clarify the definition and how the moratorium is to be interpreted, the moratorium likely constitutes a relatively low barrier compared to other legal obstacles, not least because the legal force of the UNCBD decision is disputed (Fridahl et al., 2020a).

A future global or European market for negative emissions could lead to increased demand for biomass, which may have negative consequences for biodiversity. However, the Renewable Energy Directive (EU, 2018c) states that biofuels must meet specific sustainability criteria as well as criteria for reducing greenhouse gas emissions from a life cycle perspective in order to be considered renewable. The proposed amendments to the Renewable Energy Directive to strengthen the sustainability criteria include provisions such as applying existing land criteria for biomass from agriculture to biomass from forests, avoiding sourcing biomass from diverse forests and peatlands, and requiring Member States to design biomass support schemes to avoid the use of high-quality roundwood for the production of electricity, biofuels, and heat (EU, 2021d). The European Commission is currently also working on an implementation regulation to guide actors in how to demonstrate that their use of forest biomass meets the new sustainability criteria for renewable bioenergy, as well as to update and strengthen the biomass sustainability criteria (EU, 2021d).

In June 2020, the EU adopted an investment taxonomy, a framework aiming to make it easier for investors to identify environmentally sustainable investments through a standardized classification system (EU, 2019, 2020b). To classify CCS, including BECCS, as sustainable, it is necessary to verify that the total physical leakage of CO<sub>2</sub> from capture to storage does not exceed 0.5%. In addition, the operator needs to comply with ISO standards for geological storage of CO<sub>2</sub> (EU, 2020b). For CO<sub>2</sub> capture to be classified as a sustainable activity, the regulation states that the transport and storage must also meet the required sustainability criteria (EU, 2020b). Further, the EU guidelines on state aid (EU, 2022) provide regulatory conditions that facilitate state investments for the deployment of BECCS beyond 2021. However, the limited time horizon for state aid, capping aid approval to 10 years, could complicate aid schemes targeting BECCS whose technical lifetimes often extend well beyond 10 years (Fridahl and Lundberg, 2021).

## CO<sub>2</sub> Transport and Storage Regulation

Geological sub-seabed storage is regulated in several international treaties. The London Protocol, originating in the International Maritime Organization’s London Convention, aims at limiting marine pollution (IMO, 1996). The London Protocol prohibits all types of dumping of all types of waste or other substances in international waters and in the territorial sea of the constituent Parties, but sub-seabed storage of CO<sub>2</sub> has been exempted since 2006 (IMO, 2006). The Oslo-Paris (OSPAR) Convention, also aimed at limiting marine pollution, adjusted its rules in line with the London Protocol in 2007 (OSPAR Commission, 1992). While the OSPAR Convention



and the London Protocol have been amended to allow sub-seabed storage, the Convention on the Protection of the Marine Environment of the Baltic Sea Area (the Helsinki Convention) does not allow sub-seabed storage (HELCOM, 1992). While sub-seabed storage is allowed by the EU CCS Directive, the Helsinki Convention has legal superiority. Therefore, if not harmonized with the CCS Directive, the Helsinki Convention will constitute a barrier to geological storage in the Baltic Sea. For the most part, however, sub-seabed storage is now allowed in EU territories.

However, access to storage sites often requires cross-border transport, and the London Protocol prohibits export of CO<sub>2</sub> intended for sub-seabed storage. Although an amendment to the London Protocol was agreed on in 2009, which allows for the export of CO<sub>2</sub> (IMO, 2009), several ratifications are still missing and must be obtained before the amendment's entry into force clause can be fulfilled. In anticipation of more ratifications, the Parties to the London Protocol have adopted a resolution that allows provisional application of the 2009 amendment (IMO, 2019). This enables CO<sub>2</sub> export, but the provisional application requires a bilateral agreement between the states concerned.

The CCS directive enables and provides rules for geological storage (EU, 2009b). In addition, as emphasized by Fridahl et al. (2020a), it offers a secure arena and a clear base for planning BECCS investments. Even so, there are important issues to be resolved. For instance, all physical leakage of geologically stored CO<sub>2</sub> requires the surrendering of EU ETS allowances. This applies irrespective of the origin of the CO<sub>2</sub> because the CCS Directive does not distinguish between fossil and biogenic CO<sub>2</sub>. If biogenic CO<sub>2</sub> is emitted into the atmosphere at a point source, it is generally accounted for as carbon neutral and therefore not associated with a cost under various pricing mechanisms such as the EU ETS or national carbon taxes. However, if it is captured, stored geologically, and then leaks, the leaked CO<sub>2</sub> is associated with a cost. If in the future a policy is adopted to establish economic incentives that reward BECCS, then it also makes sense that physical leakage of biogenic CO<sub>2</sub> should be associated with a cost. It is not obvious, however, that this cost should correspond to the allowance price in the EU ETS. Perverse incentives to catch and release biogenic CO<sub>2</sub> might be created if the reward is higher than the penalty. If the reward is lower than the penalty, this creates an investment risk that would increase the cost of BECCS.

Another related issue with the CCS Directive is that domestic sub-seabed storage in areas close to territories outside the EU might be hindered if a geological storage site extends to territories outside the EU. One example is a possible geological storage site in the southern Baltic Sea that includes Russian territory (SOU, 2020).

As noted by Fridahl and Lehtveer (2018), deployment of BECCS technology can differ between regions due to resources and technical conditions. Similarly, differences in existing infrastructure and access to geological storage may affect the existence of possible regulatory barriers. Countries lacking opportunities for safe storage of CO<sub>2</sub> are reliant on CO<sub>2</sub> storage in other regions. For these countries, legal obstacles to the cross-border transport and storage of CO<sub>2</sub> need to be harmonized. Moreover, access to existing infrastructure for CO<sub>2</sub> transport and

storage would increase interest among investors and operators to build new capture facilities (Tamme and Beck, 2021). This highlights the need to determine how bilateral agreements between the country in which the capture facility is located and possible storage countries might be formulated. According to the London Protocol, the agreements must include stated consent for the activity and an allocation of responsibilities. In order to reduce the administrative burden of agreeing to such contracts, it ought to be possible to share experience and provide basic templates for treaty design.

Finally, it is worth recognizing that the existing EU regulation requires the operator of capture facilities to also be responsible for leakage during transport. Operators of capture facilities are expected to subcontract transport of CO<sub>2</sub>, meaning that operators of capture facilities are responsible for leakage during transportation despite their lack of control over the transport. This requires, as noted by Jordal et al. (2022), contractual arrangements between the capture and transport operators regarding responsibilities, which may increase the transaction costs for BECCS. However, the European Commission has proposed to amend the EU ETS Directive so that transport operators will become responsible for any leakage during transportation (EU, 2021a), a reform that would simplify subcontracts for capture facilities that want to implement the full BECCS technology chain. Tamme and Beck (2021) also underline the Trans-European Energy Networks Regulation, which lists development of transport infrastructure for captured CO<sub>2</sub> as a priority area for the EU.

## POLICY RECOMMENDATIONS

The lack of policy leverage to compensate for the high costs of CO<sub>2</sub> capture, transport, and storage infrastructure are crucial for BECCS commercialization. However, regulatory preconditions also have an important role in filling the commercialization gap. Over the past decade, regulatory conditions for BECCS have become more permissive. For instance, accounting guidelines have been developed by the IPCC, and several multilateral agreements have been amended to allow CO<sub>2</sub> export and sub-seabed storage. Several other regulatory reforms are in process and are pending adoption by the European Parliament and the Council, including clarification of approved means of transportation of CO<sub>2</sub> and of responsibilities for physical leakage of CO<sub>2</sub> during transport. However, the current regulatory regime still contains gaps and raises barriers that must be addressed to facilitate BECCS deployment. The significance of regulation is only going to increase as economic policy instruments, designed to incentivize negative emissions, are likely to develop in the coming years, both on the EU level and in Member States. The most urgent regulatory reforms are to:

1. Allow EU Member States to use negative emissions from BECCS to comply with their obligations under one or several of the legislative pillars that underpin the EU's climate objectives, i.e., the Effort Sharing Regulation, the EU Emissions Trading System, and the Land-Use, Land Use Change and Forestry Regulation.

2. Amend the CCS Directive to distinguish between physical leakage of fossil and biogenic CO<sub>2</sub> from geological storage sites. The share of leakage that could be attributed to sustainably sourced biomass, in accordance with sustainability criteria in the Renewable Energy Directive, ought to be exempted from the requirement to surrender emission allowances in the EU ETS or, if BECCS has been economically rewarded, the penalty for leakage should correspond to the level of the reward to avoid creating perverse incentives and/or investment risks.
3. Erase the existing barriers provided by multilateral regimes, such as clarifying whether BECCS is covered by the geoengineering moratorium maintained by the UN Convention on Biological Diversity.

Alongside the key issue of agreeing on economic policy incentives for BECCS, addressing the proposed regulatory barriers is crucial to further encourage BECCS deployment.

## DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. The datasets analyzed for this study can be found in online legal

repositories of the UN, regional multilateral organizations, and the EU. The names of the repositories and accession numbers can be found at: <https://treaties.un.org>; <https://unfccc.int/documents>; <https://docs.imo.org>; <https://www.cbd.int/cooperation/about/documents.shtml>; <https://www.ipcc.ch/documentation>; <https://www.ospar.org/meetings/archive>; <https://helcom.fi>; and <https://eur-lex.europa.eu>.

## AUTHOR CONTRIBUTIONS

The article was planned by MF. TN made the first draft of the article. Thereafter, in collaboration between MF and TN, the article was discussed and revised in all parts including new text and references. Both authors approved the submitted version.

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# CO<sub>2</sub> Utilization and Market Size Projection for CO<sub>2</sub>-treated Construction Materials

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Achieving a net climate benefit requires that captured CO<sub>2</sub> (from stack emissions, directly from air or any other source) does not return to the atmosphere. There are several negative-emissions technologies that will meet this criterion. First, underground storage, possibly coupled with mineralization underground, creates essentially permanent removal. Second, CO<sub>2</sub> can be mineralized above ground into benign solid materials that can be left in the environment. Third, construction materials such as concrete, aggregates, and wall boards can be made with the use of CO<sub>2</sub>. From a climate point of view the lifetime of those materials can be considered permanent. While structures (buildings, roads, etc.) will not last permanently, the mineralized CO<sub>2</sub> in the underlying raw materials will. The latter negative emissions technology category has the added benefit of creating revenue from the sale of the products and thus providing financial incentives for deployment. The potential CO<sub>2</sub> utilization amount and market size for mineral-based construction materials is projected from today to 2050 and is discussed in context of the total addressable market for respective products. A range of scenarios for growth are considered and discussed. CO<sub>2</sub> utilization between 1.0 and 10.8 gigatons per year is projected with a market valuation reaching 0.8–1 trillion USD/year by 2050.

**Keywords:** construction materials, carbon capture and utilization, concrete, aggregates, CO<sub>2</sub> removal, market size and growth

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## INTRODUCTION

Construction, or more broadly, construction materials offer a unique opportunity for innovation in the context of necessary action to address climate change as well as productivity and quality of workplace in the construction industry. The combined impact of construction (11%) and operation (28%) of the built infrastructure is responsible for nearly 40% of total greenhouse gas activity (World Green Building Council, 2019) and therefore substantial progress must be made in this sector to achieve a net-zero carbon economy (Hansen et al., 2022). Concrete, as one of the dominating construction materials contributes 5–8% of the global CO<sub>2</sub> emissions from the production of cement alone (International Energy Agency, 2018). Changes to cement and concrete production and use can therefore have a profound impact on emissions and society (Strunge et al., 2022a). In addition, the construction industry, worldwide, suffers from a substantial shortage of available labor forces. Therefore, the development and instruction of new construction materials offers the opportunity to implement new construction methods to address this shortage. This includes the use of automation, such as 3D-printing of concrete to speed up construction that

can be enhanced with tailor-made CO<sub>2</sub>-treated materials. The urgency to address this problem is exacerbated by the projected growth rate for new housing estimating that the equivalent of one New York City will be built every month until 2060 (Gates, 2021).

Achieving a net-zero scenario requires reducing CO<sub>2</sub> emissions to the minimum possible, and offsetting the remaining emissions with capture and storage, and targeted use of CO<sub>2</sub> to manufacture materials. Broadly, carbon dioxide capture, utilization, and storage (CCUS) are actions that are deemed necessary by the Intergovernmental Panel on Climate Change (Masson-Delmotte et al., 2018) and the International Energy Agency (International Energy Agency, 2020). How to integrate these efforts with overall emission reducing progress is a large-scale challenge (Fankhauser et al., 2021). Accepting the premise that CO<sub>2</sub> must be captured, the opportunity to use that CO<sub>2</sub> as a non-fossil carbon-source to make products creates market-driven approaches that will reduce the cost of achieving a net-zero future. A 2016 study projected the annual use potential for CO<sub>2</sub> of about 4 gigatons/year with a revenue volume of \$1 trillion in 2030, if suitable policy support would be put into action (CO<sub>2</sub> Sciences, 2016). Preference should thus be given to technologies that can utilize CO<sub>2</sub> for useful, revenue-generating carbon dioxide capture and utilization (CCU) products. However, given the volume of negative emissions needed, use in products alone will not be sufficient to supplant the need for underground storage of CO<sub>2</sub>.

Potential products can be categorized into two groups. The first category includes products that cannot be made without carbon and for which no non-carbon alternatives exist, e.g., many chemicals, solvents, fuels, detergents. The second category includes those products that currently are not made with carbon, e.g., CO<sub>2</sub>-cured concrete and carbonated aggregates. Especially the latter offers unique options to incorporate large quantities of CO<sub>2</sub> (Woodall et al., 2019) in uses that are entirely new, and the duration of CO<sub>2</sub> removal could be considered as permanent. It is useful in this context to define two tracks for CO<sub>2</sub>-treated materials.

TRACK 1 materials remove CO<sub>2</sub> for more than 100 years and include construction materials (concrete, aggregates) and additionally, mineralization efforts that are frequently discussed in the context of carbon dioxide capture and storage (CCS) should also be considered here as a valuable resource for building materials. None of these materials have traditionally been made with CO<sub>2</sub> and therefore they provide an attractive opportunity for CO<sub>2</sub> removal with lifetimes in excess of 100 years and the potential to constitute negative emissions technologies. Additionally, new materials, such as carbon fibers and carbon-ceramics could reach such lifetimes.

TRACK 2 materials remove CO<sub>2</sub> for less than 100 years but produce products that require carbon contents and in general will have a shorter timescale than Track 1 technologies before releasing CO<sub>2</sub> back into the atmosphere. Their climate benefit stems from avoided CO<sub>2</sub> emissions by replacing fossil carbon with captured CO<sub>2</sub> in a circular fashion.

Track 1 products are an attractive opportunity with substantial deployment capabilities that can achieve an important positive climate impact (Ravikumar et al., 2021a,b). Recent data

on market size and CO<sub>2</sub> utilization potential are discussed in this perspectives paper.

## MARKET SIZE AND PENETRATION FOR CO<sub>2</sub>-TREATED AGGREGATES AND CONCRETE

Aggregate use in construction is abundant in the form of components in concrete, fillers for roads, asphalt, and much more. In 2020, the global market size for aggregates was estimated as 45 billion metric tons using several sources (Carey, 2018; Aggregates Business, 2019; LafargeHolcim, 2020) and, with an assumed compound annual growth rate (CAGR) of 3.3% by 2050, the annual needs for aggregates would rise to 119 billion metric tons. The CAGR value was determined as an industry average of historical data from CRH, Cemex, Holcim, and Vulcan.

Concrete use is typically estimated based on cement production volume (5.2 billion metric tons in 2020) and in 2020 amounted to about 25 billion metric tons. For the purpose of this perspective, only precast concrete is considered further due to its much higher level of CO<sub>2</sub> use for curing compared to ready-mix concrete, even though it currently only accounts for approximately 30% of the total concrete market (Henrion et al., 2021a). The global precast concrete market in 2020 was 7 billion metric tons and is expected to rise at a CAGR of 5.2% to 32 billion metric tons in 2050. This CAGR is an average of published industry data from CRH, Forterra, Holcim, the National Precast Concrete Association, and reports (Grandview Research, 2021).

A key question will be what fraction of these total addressable markets can be covered with the CO<sub>2</sub>-treated alternative materials? Addressing this question in detail is beyond the scope of this perspective and would have to include factors that concern the resulting material properties (Vance et al., 2013; Mehdipour et al., 2019; Rahmouni et al., 2019; Zhang et al., 2019), necessary certifications (American Concrete Institute, 2020), the local (!) availability of suitable raw materials including CO<sub>2</sub> (National Academies of Sciences and Medicine, 2018, 2019; Woodall et al., 2019; Strunge et al., 2022b), public acceptance (Arning et al., 2021), supporting policy (Di Filippo et al., 2019; Henrion et al., 2021b), and of course cost (CO<sub>2</sub> Sciences, 2016).

Consideration of the above-mentioned factors will be necessary to determine the potential deployment rates for a specific application in a particular region. This perspective examines the potential from a macro- or global level to determine upper bounds for CO<sub>2</sub>-treated aggregates and concrete use.

Carbonation of minerals or waste materials can consume between 0.087 and 0.440 tons of CO<sub>2</sub> per ton of aggregate depending on the starting raw material, conversion process, and resulting product material (Woodall et al., 2019). As a starting point, for 2020, the global amount of suitable waste materials was estimated to about 6.6 gigatons per year (Carey, 2018; Hepburn et al., 2019) while the global annual production of alkaline minerals (wollastonite, olivine, serpentinite) was at 16.2 million metric tons (U.S. Geological Survey, 2022). That then determines the range of potential CO<sub>2</sub> use when multiplied by an assumed market penetration for carbonated aggregates. For

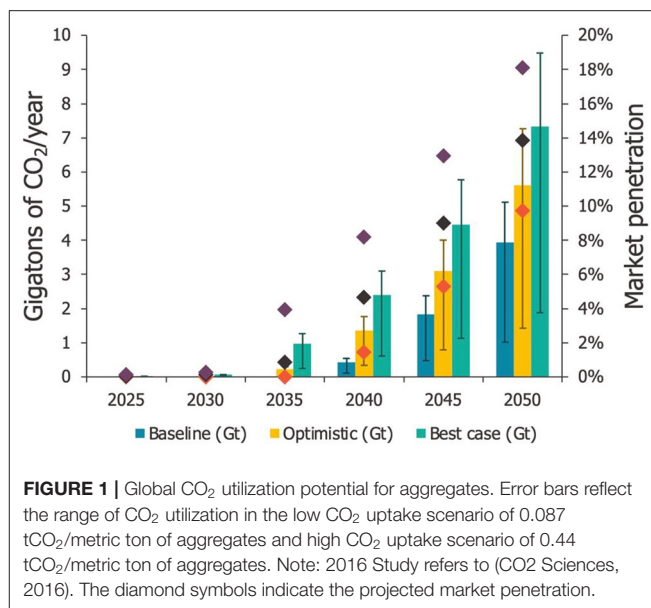
precast concrete, CO<sub>2</sub> utilization estimates range from 0.001 to 0.085 tons per ton of concrete (Zhang and Shao, 2016; Henrion et al., 2021b) but we use a more conservative upper bound of 0.05 tons for the projections. It is noted that while there are indications that CO<sub>2</sub> curing might not affect the natural ability of concrete to further take up CO<sub>2</sub> over its lifetime, this is an ongoing debate for research (Zhang and Shao, 2016).

It will be important, though, to assess the increase in market penetration over time. The market share will be critically dependent on the cost of the CO<sub>2</sub>-treated product. The production cost of the CO<sub>2</sub>-treated product will depend, in part, on the cost of CO<sub>2</sub>. High capture cost, especially when CO<sub>2</sub> is captured from air, will make market competitiveness sensitively linked to a (potential) price on carbon. A price on carbon is a tax or financial penalty for emitting CO<sub>2</sub>. It must be noted, though, that such a price could also be deployed in the form of incentives, such as tax rebates. However, it has been demonstrated that the use of solid waste materials and CO<sub>2</sub> emissions to manufacture building materials can be cost competitive in some regions today due to the cost of waste disposal (Carey, 2018). In general, though, establishing a CO<sub>2</sub>-utilization industry will require policy support in some form and overall, a comprehensive assessment of policy instruments should be researched.

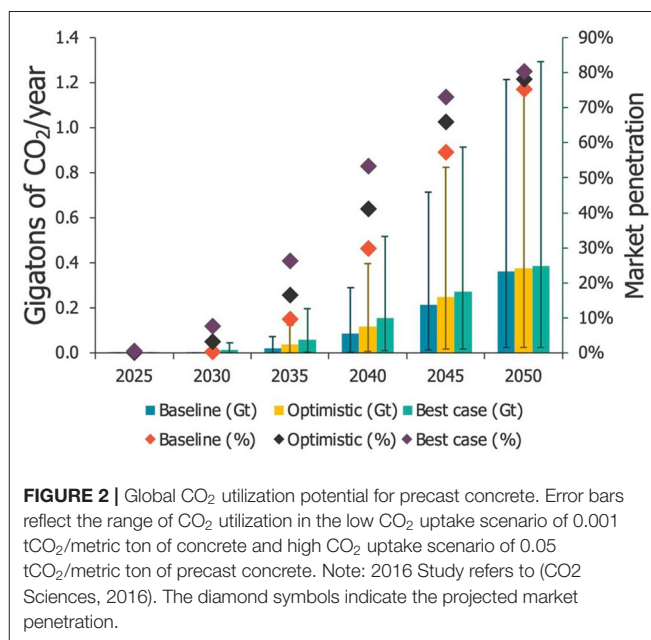
An analysis of the impact of a carbon price resulted in three scenario solutions. For each scenario, the price of carbon is subtracted from the cost of carbonate product and that will result in cost tipping points in the future, enabling increased market penetration. All three scenarios start with a carbon price of USD10/metric ton in 2020 and that is projected to rise to USD100/metric ton as projected by the Carbon Disclosure Project (Carbon Disclosure Project, 2022). The analysis is based on World Bank (World Bank, 2022) data showing that 75% of existing carbon pricing is less than USD10/metric ton. An annual increase of USD2/metric ton was used for a “Baseline” scenario. It is noted that increases in carbon pricing will of course be regionally specific but are assumed to reach USD50/metric ton by 2040. A USD2.5/metric ton annual increase for an “Optimistic” scenario will reach the lower threshold of Carbon Disclosure Project’s analysis of required carbon pricing to meet the targets set by the Paris Agreement in 2015 (Carbon Disclosure Project, 2022). Fastest growth would be achieved in a “Best case” scenario where the carbon price would increase by USD4/metric ton annually, reflecting the higher threshold of the Carbon Disclosure Project analysis.

In 2020, the global average price for aggregates was 10 USD per metric ton, triangulated from various sources (CRH, 2020; LafargeHolcim, 2020; Chinese Aggregates Association, 2022), and is assumed to grow at an annual rate of 1.5% while CO<sub>2</sub>-treated aggregates were priced at 50 USD, based on interviews of producers and in line with Hepburn et al. (Hepburn et al., 2019). Prices for precast concrete were 19 USD per metric ton in 2020 and are assumed to increase at 1.1% annually (based on historical price changes), while the price of CO<sub>2</sub>-cured precast concrete was 26 USD per metric ton.

Market penetration projections by Lux Research Inc. (Lux Research, 2022) were based on a qualitative assessment of



**FIGURE 1** | Global CO<sub>2</sub> utilization potential for aggregates. Error bars reflect the range of CO<sub>2</sub> utilization in the low CO<sub>2</sub> uptake scenario of 0.087 tCO<sub>2</sub>/metric ton of aggregates and high CO<sub>2</sub> uptake scenario of 0.44 tCO<sub>2</sub>/metric ton of aggregates. Note: 2016 Study refers to (CO<sub>2</sub> Sciences, 2016). The diamond symbols indicate the projected market penetration.



**FIGURE 2** | Global CO<sub>2</sub> utilization potential for precast concrete. Error bars reflect the range of CO<sub>2</sub> utilization in the low CO<sub>2</sub> uptake scenario of 0.001 tCO<sub>2</sub>/metric ton of concrete and high CO<sub>2</sub> uptake scenario of 0.05 tCO<sub>2</sub>/metric ton of precast concrete. Note: 2016 Study refers to (CO<sub>2</sub> Sciences, 2016). The diamond symbols indicate the projected market penetration.

competing technologies and industry willingness to adopt. Competing technologies included the possible availability of non-CCU, but low-carbon or zero-carbon technologies for the same or a similar product. Willingness to adopt was based on consideration of the CCU product’s sustainability, performance advantages, and other benefits potentially offsetting a higher cost.

CO<sub>2</sub> use for aggregates could reach between 1.0 and 9.5 gigatons per year in 2050 as shown in **Figure 1** while capturing 18% of the total aggregates market with a revenue generation of USD 182 USD 337 billion.



For precast concrete curing, the projected CO<sub>2</sub> use is <0.1 gigaton per year by 2030 in the best case but will rise to 0.05–1.3 gigatons/year in 2050 as shown in **Figure 2** with a market size that could reach USD 623–USD 666 billion by 2050.

## DISCUSSION

Six years after the publication of CO<sub>2</sub> Sciences' "Global Roadmap for Implementing CO<sub>2</sub> Utilization" (CO<sub>2</sub> Sciences, 2016), the awareness of and the interest in the climate and economic potential of using CO<sub>2</sub> to make products has increased substantially. Especially carbonation of minerals and alkaline waste materials is now an attractive option for permanent carbon dioxide removal from the atmosphere. When coupled with the use of the resulting carbonates as construction materials a large-scale market can be served, and the cost of removal is more easily offset with the revenue from the sale of the materials. The global construction market is growing fast and therefore it is now important to deploy as much CO<sub>2</sub>-treated aggregates and CO<sub>2</sub>-cured concrete as possible to reduce the amount of "embodied carbon" in buildings and other infrastructure (the amount of CO<sub>2</sub> generated in the construction of buildings). Permanently incorporating CO<sub>2</sub> into building materials could help to offset CO<sub>2</sub> emissions occurring elsewhere in the construction process.

It must be emphasized strongly that the data presented above are global data that do not include the granularity of a localized analysis for CO<sub>2</sub> use potential. Preparation and transport of large volumes of heavy raw materials and products in general should discourage transport beyond local distances for environmental and cost reasons. This is particularly true of products such as aggregates and concrete that have low profit margins. The projections for CO<sub>2</sub> utilization in construction materials do highlight that efforts on technology development and policies to support their deployment are meaningful in the context of climate stabilization, even if only a fraction of the global potential comes to market. For example, if only 10% of the 2050 aggregates market is served with CO<sub>2</sub>-treated aggregates, the amount of CO<sub>2</sub> utilization can reach 1.1–5.3 gigatons per year. The IEA projects that in 2050, 5.3 gigatons of CO<sub>2</sub> would have to be

captured and stored while only 0.37 gigatons would be utilized (Table 2.1 in International Energy Agency, 2020). CCU therefore can take on a bigger, and more cost effective, role to reach a net-zero carbon economy. To materialize this amount, it will be important to identify local opportunities to find optimum deployment options in terms of maximum amount of CO<sub>2</sub> removed and positive revenue. Related analyses and plans must factor in local communities needs and constraints in land use and job creation. Lastly, it also must be ensured that the technologies that are intended to use CO<sub>2</sub> to produce construction materials have been designed and analyzed appropriately to ensure a true net-benefit of the CO<sub>2</sub> use (Ravikumar et al., 2021b).

## DATA AVAILABILITY STATEMENT

The data analyzed in this study is subject to the following licenses/restrictions: The data is part of a more comprehensive project that will be made available once it is completed. Requests to access these datasets should be directed to vsick@umich.edu.

## AUTHOR CONTRIBUTIONS

VS led the concept and writing of the paper and integrated contributions from the co-authors. GS and FM equally contributed to the data generation, analysis, and interpretation, and co-edited the manuscript. All authors contributed to the article and approved the submitted version.

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# Maximizing Global Cooling Potential in Carbon Dioxide Removal (CDR) Procurements: A Proposal for Tonne-Year Pricing

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Recently, a consortium of companies including Stripe, Alphabet, Shopify, Meta, and McKinsey allocated US\$925 million for advanced market commitments to kickstart the early-stage Carbon Dioxide Removal (CDR) market. We argue that it is now more important than ever to consider a Global Cooling Potential (GCP) perspective in corporate CDR procurements. Currently, CDR projects are evaluated and priced on a simple cost-per-tonne basis, which fails to monetize storage duration and can ultimately incentivize the large-scale procurement of short-duration CDR. However, the relative duration of carbon storage is a critical aspect of any CDR project given the implications for climate warming from growing atmospheric concentrations of carbon dioxide. In this perspective article, we apply tonne-year carbon pricing to Microsoft and Stripe's initial CDR procurements to demonstrate that a combination of tonne-year pricing and conventional pricing could produce a CDR portfolio that simultaneously prioritizes storage duration, volume, and temporal urgency, which are all important considerations for maximizing GCP.

**Keywords:** Carbon Dioxide Removal (CDR), Negative Emissions Technologies (NETs), carbon market, carbon storage, negative emissions credit

## INTRODUCTION

The Intergovernmental Panel on Climate Change (IPCC)'s 2018 Special Report on Global Warming concluded that, "all pathways that limit global warming to 1.5°C with limited or no overshoot project the use of carbon dioxide removal (CDR) on the order of 100–1000 GtCO<sub>2</sub> over the 21st century" (IPCC, 2018). There are several ways to remove CO<sub>2</sub> from the atmosphere, and each method varies in its ability to scale, the cost of CO<sub>2</sub> removal, and its compatibility with long-duration CO<sub>2</sub> storage. Most commonly, CO<sub>2</sub> can be sequestered in the biosphere, which involves planting trees (Afforestation/Reforestation) and modifying agricultural practices (Soil Carbon) to capture and store CO<sub>2</sub> (Orbuch, 2020; IPCC, 2022). Geosphere-based CDR, involves storing CO<sub>2</sub> outside of the biosphere and typically refers to solutions such as Direct Air Capture (DAC), Bio-Energy Carbon Capture and Storage (BECCS), and Enhanced Mineral Weathering (EMW) (Orbuch, 2020; IPCC, 2022).

Despite the IPCC's conclusion about the enormous scale of required CO<sub>2</sub> removal (IPCC, 2018, 2022), CDR procurement is currently done on a voluntary basis and heavily relies on the goodwill of a select few companies (McLaren et al., 2019; Battersby et al., 2022). In the status quo, global corporations must voluntarily pledge to reduce their carbon footprint, and they may take a number of actions to do so. The lowest cost actions can involve Power Purchasing Agreements (PPAs) with renewable energy suppliers, energy efficiency measures, and Electric Vehicle (EV) procurements, among several other tactics that are frequently used to mitigate CO<sub>2</sub> emissions (IPCC, 2022). After the straightforward, low-cost solutions have been actioned, companies will oftentimes purchase carbon credits to offset their remaining emissions. The problem with the current regime is that it places the burden for high-quality credit procurement on the buyer—many of which are making their first serious attempts to lower their emissions (Wright and Nyberg, 2015). Consequently, the vast majority of companies lack a sufficient framework to make informed decisions that maximize GCP. Therefore, it is unreasonable to expect that the current system will lead to ideal outcomes when history has repeatedly demonstrated that most companies are incapable of independently making the most environmentally efficacious decision (Wright and Nyberg, 2015; McLaren et al., 2019).

Currently, credits for CDR are evaluated and sold using a cost-per-tonne pricing system in which the cost to remove a certain volume of CO<sub>2</sub> is the primary focus. Critically, this pricing framework means that each company will place a different weight on the value of CO<sub>2</sub> storage duration, which results in uneven procurement. Certain companies will prioritize long-duration CDR, while others will not, and ultimately, both types of companies will claim emissions reductions using very different underlying carbon credits. This is, perhaps, an inherent flaw with any voluntary carbon market because positive climate outcomes are reliant on self-policing, discerning buyers (Battersby et al., 2022).

Additionally, assessing the value of CDR credits is an incredibly complex task. In their review of Negative Emissions Technologies (NETs), Fridahl et al. (2020) identified 21 indicators to differentiate the values of various NETs. While the article explains that a summary index comprised of different indicators could aid decision making about CDR deployment, the authors do not propose an all-encompassing index. As it would be challenging to optimize all 21 parameters under one index, corporations might find it more straightforward to focus on a select few values.

For CO<sub>2</sub> mitigation, there is precedent for using summary indices to assess the value of different solutions; In 2007, McKinsey released its CO<sub>2</sub> abatement cost curve, which visualized the cost and abatement potential for a variety of measures (McKinsey, 2007). However, the most abundant forms of CO<sub>2</sub> mitigation effectively avoid emissions permanently. Consequently, it is intuitive to monetize emissions reductions in terms of the cost-per-tonne because the durability for each form of mitigation is all theoretically equal. In contrast, the durability of a tonne removed through CDR varies widely, which is why monetizing storage duration can be quite revealing.

It is therefore prudent to introduce a more comprehensive pricing framework that accounts for the value of long-duration storage. From a climate perspective, the primary goal of CDR should be to maximize cooling temperature effectiveness—more simply described as Global Cooling Potential (Fridahl et al., 2020). With a focus on GCP, the most important parameters for CDR become the timing of removal, the volume of CO<sub>2</sub> removal, and the durability of CO<sub>2</sub> storage. Accordingly, Joppa et al. (2021) note that today's cost-per-tonne pricing does not monetize the duration of storage, which encourages the procurement of low-cost CDR that often has the shortest sequestration periods. However, a new pricing index is not fully comprehensive, as Förster et al. (2022) note that factors like co-benefits are crucial decision variables for deploying CDR.

In the early-stage CDR market, several companies such as Microsoft, Stripe, Alphabet, Shopify, Meta, and McKinsey have emerged as leaders for their efforts to kickstart corporate CDR procurement despite high initial costs (Frontier, 2022). It could be argued that without formal interventions, several companies have been able to develop individualized multi-attribute CDR procurement strategies, and thus, it is not necessary to consider alternative CDR frameworks. However, the aforementioned companies should be seen as the exception—not the rule—when it comes to CDR procurement. The vast majority of companies have not yet engaged with the CDR market, and many companies are still purchasing avoided emissions credits, which have been plagued with issues of additionality, monitoring, reporting, and verification (MRV) (Gillenwater et al., 2007; Miltenberger et al., 2021). That said, significant work must be done on MRV in the CDR market to ensure that these credits are verifiably removing CO<sub>2</sub> from the atmosphere.

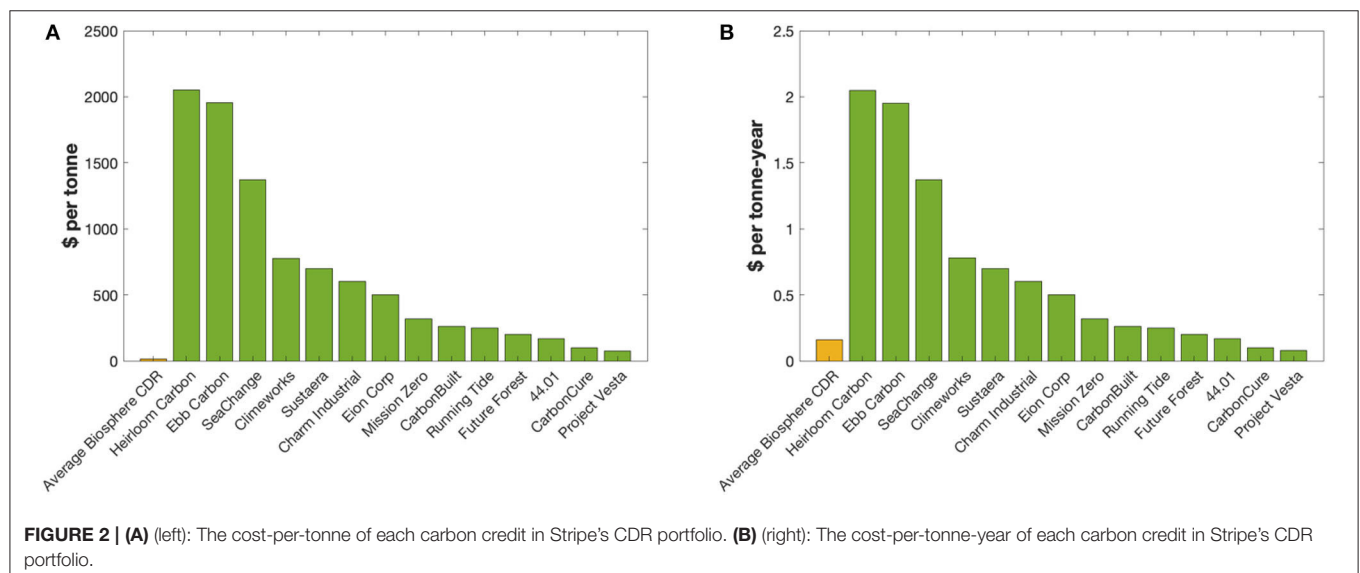
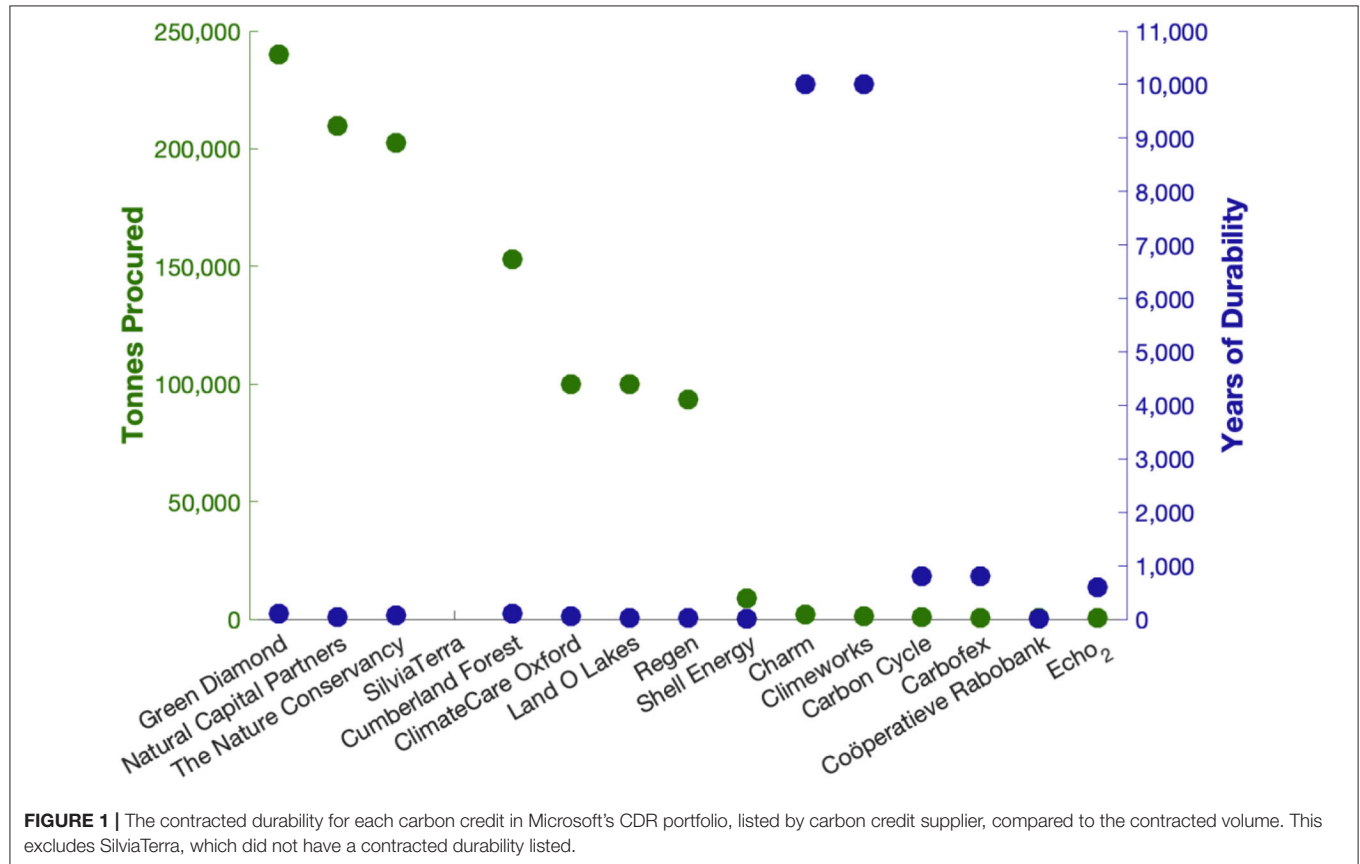
While Microsoft and Stripe have recognized the importance of purchasing CDR, they each have distinct procurement strategies. As expected, every company in this market has its own preferences and will develop a unique multi-attribute CDR purchasing strategy, which results in different portfolios. For instance, Microsoft has largely prioritized purchasing large volumes of CDR, and for the 2021 fiscal year, it signed carbon removal agreements with 15 different organizations to collectively remove 1.3 megatons of CO<sub>2</sub> (Microsoft, 2021). Among the 1.3 megatons, ≈99% of credits in Microsoft's 2021 CDR portfolio have a contracted durability of fewer than 100 years (Microsoft, 2021). In contrast, Stripe has a relatively smaller CDR portfolio because it has prioritized long-term durability over volume. In fact, Stripe only purchases CDR credits from projects that can sequester CO<sub>2</sub> for longer than 1,000 years (Orbuch, 2020). However, the supply of long-duration CDR remains limited and typically costs significantly more than short-duration credits; as a result, Stripe has only purchased around 15,000 tonnes after three rounds of CDR procurement (Stripe, 2022).

In this work, we introduce a novel pricing metric called the cost-per-tonne-year, which can be useful for advancing CDR procurement that maximizes cooling temperature effectiveness. In this paper, we refer to the tonne-year as the volume of CO<sub>2</sub> that is removed from the atmosphere for a certain period of time. Using a function  $f(t)$ , we can model the net-volume of CO<sub>2</sub> that



has been removed from atmospheric CO<sub>2</sub> stocks in year  $t$ , and by taking the area under the curve, we can calculate the tonne-years for a certain volume of removal. Critically, the tonne-year calculation accounts for CO<sub>2</sub> leakage over time because  $f(t)$  will decrease when leakage occurs, which also reduces the area under the curve.

This pricing metric can ideally be used in combination with the standard cost-per-tonne metric to ensure that timing, volume, costs, and storage duration are considered. Decisions rooted in the cost-per-tonne metric can result in more immediate high-volume, low-cost procurement with short-term durability, while decisions rooted in tonne-year pricing



can result in low-volume, high-cost procurement with long-term durability. When these two pricing frameworks are used in combination, they could produce a portfolio that balances the immediate need for both large-scale CO<sub>2</sub> removal and strong durability in order to maximize long-term GCP. It should be stressed, that corporate CDR procurements cannot act as a substitute for large-scale emissions reductions, but rather as a complementary strategy to reduce near-term emissions, offset emissions from hard-to-abate sectors, and achieve long-term net carbon negativity (IPCC, 2001, 2022).

## PRIOR USE OF TONNE-YEAR ACCOUNTING

Decades ago, tonne-year carbon accounting was initially proposed because scholars wanted to create an equivalence factor to directly compare the impact of a certain volume of avoided emissions with a certain volume of temporarily sequestered emissions (Chomitz, 2000; Costa and Wilson, 2000; Fearnside et al., 2000; Cacho et al., 2003). However, in the wake of the Kyoto Protocol, which first established a robust international carbon market, there have been a range of concerns raised about the accuracy, additionality, and environmental impact of credits sold on international exchanges—especially for avoided emissions credits (Gillenwater et al., 2007; Richards and Huebner, 2012; Rosen, 2015; Miltenberger et al., 2021). These concerns, in combination with a growing scientific consensus that CDR is arithmetically required for meeting temperature targets and addressing historical emissions (IPCC, 2018, 2022), means that companies such as Microsoft, Stripe, Alphabet, Shopify, Meta, and McKinsey are integrating CDR credits—not avoided emissions credits—into their new climate strategies (Joppa et al., 2021; Frontier, 2022).

Initial discussions on tonne-year accounting took place when virtually all of the sequestration credits on the market were biosphere-based methods such as afforestation/reforestation and soil carbon sequestration, which have similar storage durations (IPCC, 2000); consequently, durability was not distinct from credit to credit, and it was a less meaningful factor in carbon credit purchases. At the time, CDR was considered a permanent removal if it kept CO<sub>2</sub> out of the atmosphere for 100 years (Cacho et al., 2003). Today, however, there are a wider number of CDR projects being deployed, and as seen in Microsoft and Stripe's portfolio, storage durability can range from decades to a thousand years. As a result, storage duration is more of a concern than ever and warrants consideration in efforts to maximize GCP.

## MICROSOFT AND STRIPE'S CDR PORTFOLIO

In its 2021 procurement round, Microsoft purchased a range of CDR credits, which reflects the diversity of available credits on the voluntary carbon market (Joppa et al., 2021; Microsoft, 2021). Using Microsoft's publicly available data, we can visualize the contracted durability of its CDR portfolio in **Figure 1** which

shows that Microsoft's CDR credits have a variety of contracted durability ranging from 13 years with Coöperatieve Rabobank to 10,000 years with Charm Industrial and Climeworks (Microsoft, 2021). However, **Figure 1** shows the volume of credits purchased from each project, and it is clear that the CDR solutions with the greatest contracted volumes are all short-term credits, which signals a potential misallocation of resources (Microsoft, 2021).

Over 99% of the procured CDR volume in Microsoft's portfolio is derived from projects with <100 years of contracted durability. While there is a benefit to purchasing short-duration credits (Matthews et al., 2022) when it is not yet possible to purchase large volumes of long-duration credits, it is unlikely that the optimal balance between short-term and long-term procurement has been achieved in this portfolio. In the current voluntary system, Microsoft has recognized the temporal urgency for purchasing CDR, but its individualized preferences have resulted in a portfolio that strongly prioritizes volume over durability.

Among early-stage CDR purchases, Stripe is the only company that has published the durability, procurement volume, and cost for each of its CDR purchases, which means that our dataset for tonne-year pricing is limited to Stripe's portfolio (Orbuch, 2020; Stripe, 2022). However, when we apply our conceptual framework to their portfolio, it's still evident that tonne-year pricing could make a valuable addition to current CDR purchasing decisions.

Geosphere-based credits are often several times more expensive than biosphere-based credits, which makes these credits uncompetitive if costs are the only consideration. In funding applications to Stripe Climate, biosphere-based credits were offered for an average of US\$16 per tonne of CO<sub>2</sub>, whereas geosphere-based solutions were purchased by Stripe for an average of US\$304.06 per tonne of CO<sub>2</sub> (Orbuch, 2020; Joppa et al., 2021; Stripe, 2022). Despite the abundance of shorter duration credits, only 0.15% of applications to Stripe met its criteria for long-duration storage (Joppa et al., 2021).

Unlike Microsoft, which purchases credits with a range of durability, Stripe has established a minimum durability criteria of 1,000 years for its credits (Orbuch, 2020). Therefore, Stripe's portfolio comprises geosphere-based CDR credits from DAC, BECCS, and EMW projects, which can offer long-term durability, but its procurement volume is substantially lower than Microsoft's. Because Stripe has prioritized durability over volume, the temporal urgency for large-scale CDR has been sacrificed, which suggests that Stripe also has not achieved an optimal procurement strategy from a GCP perspective.

Using open-sourced information from Stripe, we can visualize the cost-per-tonne and cost-per-tonne-year of Stripe's 2020 and 2021 CDR procurements in **Figures 2A,B**, respectively (Orbuch, 2020; Carbon Plan, 2021; Stripe, 2022). This allows us to compare both methodologies to understand the effect of monetizing storage duration. In **Figure 2A**, it is clear that the average biosphere-based solution is typically a far less expensive mechanism for removing CO<sub>2</sub> from the air. However, when we monetize the duration of storage, shown in **Figure 2B**,

and apply a 100-year durability to the average biosphere-based credit—even though this is an optimistic durability given that the average similar credit in Microsoft's portfolio has a far lower contracted durability—several geosphere-based solutions with a low cost-per-tonne-year become cost competitive with the average biosphere-based solution. If companies utilized both cost methodologies in their CDR purchasing decisions, their strategies would be more effective from a GCP perspective because they would focus less on one dimension of CDR and would instead simultaneously optimize several parameters such as timing, volume, durability, and cost.

Of course, it is very difficult to fully calculate the optimal balance of volume, timing, cost, and durability, and this could certainly warrant a more substantial piece of future research. However, in this article, we are merely signaling that the use of both pricing methodologies would likely produce a more optimal balance from a GCP perspective than a counterfactual scenario where companies maximize individual parts of the equation.

## CONCLUSION

It is important to recognize that an additional pricing framework is not a panacea for the issues that have plagued carbon markets over the last several decades. These issues will likely persist in any voluntary carbon market as long as

companies are not legally constrained by the types of credits that qualify for making claims about emissions reductions. However, a pricing framework that accounts for storage durability could be a meaningful tool when used in combination with the existing conceptual frameworks for corporate CDR procurements. Along with large-scale emissions reductions, a nuanced framework could help produce more holistic corporate responses to the climate crisis that are consistent with maximizing GCP.

## DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. This data can be found here: “<https://github.com/stripe/carbon-removal-source-materials>” and “<https://carbonplan.org>.”

## AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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# Geochemical Negative Emissions Technologies: Part I. Review

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Over the previous two decades, a diverse array of geochemical negative emissions technologies (NETs) have been proposed, which use alkaline minerals for removing and permanently storing atmospheric carbon dioxide (CO<sub>2</sub>). Geochemical NETs include CO<sub>2</sub> mineralization (methods which react alkaline minerals with CO<sub>2</sub>, producing solid carbonate minerals), enhanced weathering (dispersing alkaline minerals in the environment for CO<sub>2</sub> drawdown) and ocean alkalinity enhancement (manipulation of ocean chemistry to remove CO<sub>2</sub> from air as dissolved inorganic carbon). CO<sub>2</sub> mineralization approaches include *in situ* (CO<sub>2</sub> reacts with alkaline minerals in the Earth's subsurface), surficial (high surface area alkaline minerals found at the Earth's surface are reacted with air or CO<sub>2</sub>-bearing fluids), and *ex situ* (high surface area alkaline minerals are transported to sites of concentrated CO<sub>2</sub> production). Geochemical NETS may also include an approach to direct air capture (DAC) that harnesses surficial mineralization reactions to remove CO<sub>2</sub> from air, and produce concentrated CO<sub>2</sub>. Overall, these technologies are at an early stage of development with just a few subjected to field trials. In Part I of this work we have reviewed the current state of geochemical NETs, highlighting key features (mineral resources; processes; kinetics; storage durability; synergies with other NETs such as DAC, risks; limitations; co-benefits, environmental impacts and life-cycle assessment). The role of organisms and biological mechanisms in enhancing geochemical NETs is also explored. In Part II, a roadmap is presented to help catalyze the research, development, and deployment of geochemical NETs at the gigaton scale over the coming decades.

**Keywords:** carbon dioxide removal (CDR), mineral carbonation, enhanced weathering in soils, coastal enhanced weathering, biomineralization, ocean liming, climate change

## HIGHLIGHTS

- Geochemical NETs (*in situ*, *ex situ* and surficial carbon mineralization, enhanced weathering, ocean alkalinity enhancement, etc.) are extensively reviewed.
- The potential role of biotechnology in geochemical NETs is given special focus.
- The Review (Part I) is accompanied by a Roadmap (Part II) to help catalyze development of geochemical NETs at scale in the coming decades.

## INTRODUCTION

To meet the 2°C climate target set out in the Paris Agreement, the atmospheric concentration of CO<sub>2</sub>-equivalent (CO<sub>2</sub>eq) should not exceed 450 ppm (or 430 ppm for the 1.5°C target) (Spier, 2020). To achieve this, a drastic decrease in anthropogenic emissions is required, which can be achieved through the expansion of renewable energy generation and lower emissions from land-use and land-use-change. The International Energy Agency (IEA) estimates that carbon capture and storage (CCS), i.e., capturing and storing CO<sub>2</sub> before it is released to the atmosphere, may prevent upwards of 6 Gt CO<sub>2</sub> yr.<sup>-1</sup> by 2050 (Haszeldine et al., 2018). In addition, negative emissions technologies (NETs) may also need to remove 10 Gt CO<sub>2</sub> yr.<sup>-1</sup> by 2050, and 20 Gt CO<sub>2</sub> yr.<sup>-1</sup> by the end of the century (National Academies of Sciences, Engineering, and Medicine, 2019). These targets necessitate technologies capable of capturing, removing, and storing CO<sub>2</sub> at a large scale.

Carbon can be stored as organic materials, e.g., terrestrial vegetation, ocean biomass, and biochar, or as pure CO<sub>2</sub> deep underground in sedimentary rocks. However, the permanence of these storage media vary greatly, creating uncertainty and legacy issues for industry, policymakers, and regulators (Lackner, 2003). On the other hand, carbon can be permanently stored in the form of the carbonate anion (CO<sub>3</sub><sup>2-</sup>) in solid minerals, e.g., calcium carbonate (CaCO<sub>3</sub>) and magnesium carbonate (MgCO<sub>3</sub>), or in the form of dissolved bicarbonate (HCO<sub>3</sub><sup>-</sup>) in ocean water. These forms of storage can be achieved by three main groups of technologies, commonly referred to as CO<sub>2</sub> mineralization, enhanced weathering, and ocean alkalinity enhancement, collectively referred to here as “geochemical NETs.”

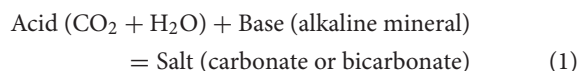
Most geochemical NETs involve enhancing the reactions of alkaline minerals with CO<sub>2</sub> (and H<sub>2</sub>O), mimicking natural chemical weathering reactions of silicate rocks at the Earth's surface, which removes ~1.1 Gt CO<sub>2</sub> yr.<sup>-1</sup> from the atmosphere, primarily stored as ocean bicarbonate (Strefler et al., 2018). The goal of geochemical NETs is to add considerably to this natural removal rate as a tool to combat climate change. In the last two decades, and particularly during the past few years, research on geochemical NETs has grown considerably, with many novel approaches being explored. Several companies and projects have been recently established. Though some are already operating at the kiloton (kt) scale, as a group they are, by and large,

at an early stage of their development, with just a few at the pilot scale.

In Part I of this work, geochemical NETs are reviewed and their potential impacts and limitations discussed. In Part II, a set of projects and interventions that warrant prioritization are presented in the form of a roadmap with the aim of catalyzing the development and deployment of geochemical NETs at the scale necessary to achieve significant carbon removal.

## OVERVIEW OF GEOCHEMICAL NETs

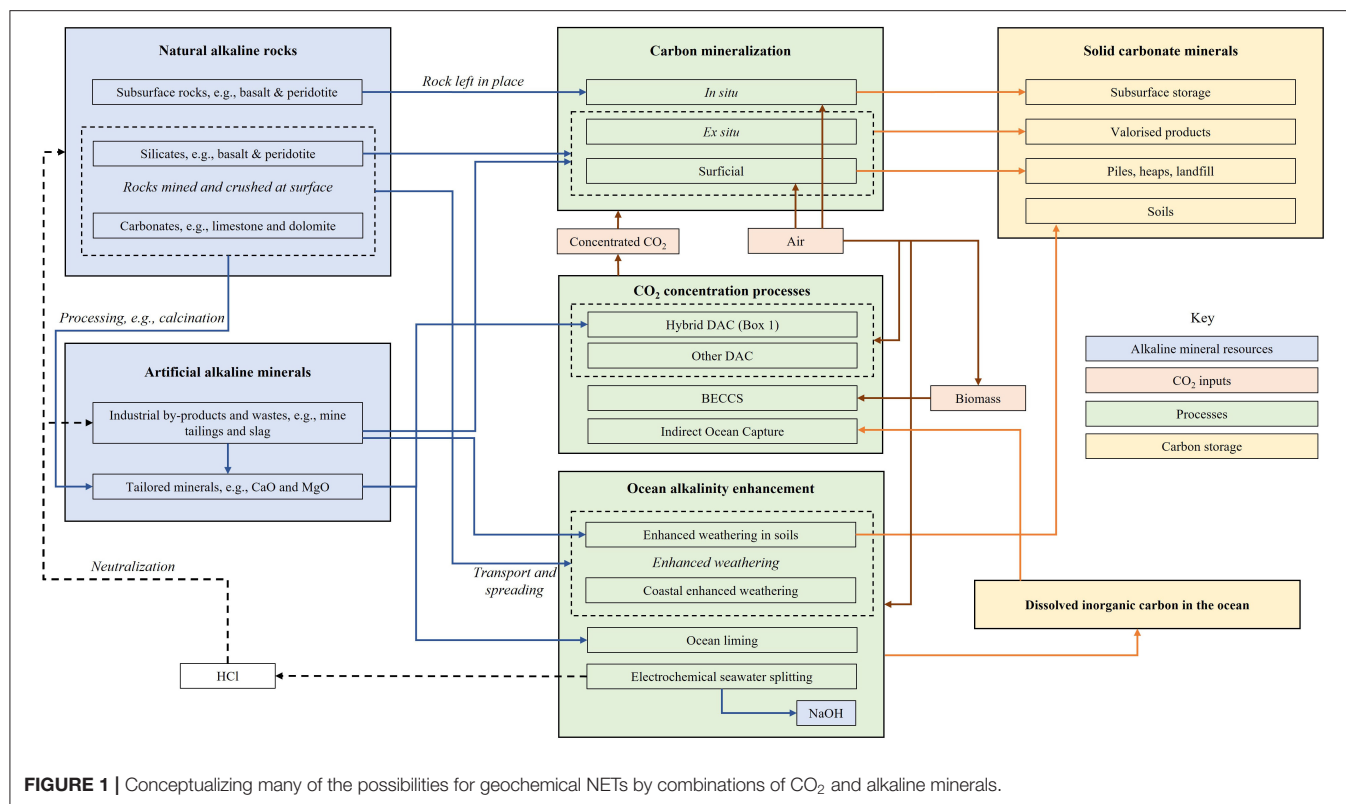
A geochemical NET is any technology which involves the use of substantial amounts of alkaline minerals in its flowsheet and involves enhancing the reaction of CO<sub>2</sub> and mineral alkalinity for the purpose of safely removing and storing CO<sub>2</sub> from the atmosphere as stable carbonate minerals, or dissolved ocean bicarbonate. At a fundamental level, most geochemical NETs are simply an acid-base neutralization of the form given in Equation (1).



**Figure 1** conceptualizes how sources of CO<sub>2</sub> and mineral alkalinity can be combined, giving rise to various geochemical NETs. In order for such a technology to be carbon negative, it must remove significantly more CO<sub>2</sub> than it emits from its life-cycle (Fajardy and Mac Dowell, 2017). Therefore, CO<sub>2</sub> must be removed directly or indirectly from the atmosphere, typically using renewable energy or bioenergy, rather than fossil fuel energy.

In order to create solid carbonate minerals, CO<sub>2</sub> (and H<sub>2</sub>O) must react with alkaline minerals (**Table 1**). Alkaline minerals are simply any natural or artificial mineral that is rich in alkaline earth metals (second column of the periodic table), particularly magnesium (Mg) or calcium (Ca), since these are far more abundant than strontium or barium, etc. (which also form stable carbonate minerals). Common Ca- and Mg-rich minerals are given in section the Common Alkaline Minerals. These minerals are found in natural igneous, metamorphic, and sedimentary rocks (section Naturally Occurring Alkaline Rocks) as well as in industrial by-products and wastes such as mine tailings, cement kiln dusts, fly ash, slag, desalination brines, etc., or minerals tailored for purpose (section Artificial Alkaline Minerals—Industrial By-Products and Wastes, and Tailored Minerals and **Table 2**). Abundant silicate minerals rich in alkali metals (first column of the periodic table), in particular sodium and potassium, may be able to contribute in some geochemical NETs, but their carbonates are too soluble for long-term carbon sequestration. While other elements may also form carbonate minerals (e.g., cadmium, cobalt, copper, iron, lead, manganese, nickel, uranium, zinc) their abundance, stability, or toxicity limit their large-scale reaction with CO<sub>2</sub>.

Most of the reactions between CO<sub>2</sub>, H<sub>2</sub>O and mineral alkalinity (section Reaction Chemistry) are thermodynamically



**FIGURE 1** | Conceptualizing many of the possibilities for geochemical NETs by combinations of CO<sub>2</sub> and alkaline minerals.

favorable, as indicated by their negative Gibbs free energies. The result of these reactions is either a solid carbonate mineral (section Carbonate Products and Other Secondary Minerals), or dissolved ocean bicarbonate. However, owing to kinetic limitations, the reaction between rock outcrops containing natural alkaline minerals and CO<sub>2</sub> at ambient conditions occurs on geological timescales. Therefore, the main goal of geochemical NETs is to considerably enhance the rate of these reactions to a timescale relevant to climate change mitigation by manipulating the kinetics (section Kinetics).

Those geochemical NETs which predominantly produce solid carbonate minerals (section CO<sub>2</sub> Mineralization), can be divided conceptually into *in situ*, *ex situ* and surficial CO<sub>2</sub> mineralization. *In situ* approaches typically involve circulation of CO<sub>2</sub>-rich fluids through alkaline rocks, e.g., basalt or peridotite, in the Earth's subsurface (Matter and Kelemen, 2009). *Ex situ* approaches typically involve reacting high concentration CO<sub>2</sub> with finely ground natural alkaline minerals or artificial alkaline by-products/wastes in engineered reactors. These reactions typically go to completion within minutes using high temperatures, pressures, concentrated CO<sub>2</sub> and/or other reagents such as acids (Sanna and Maroto-valer, 2016). On the other hand, surficial approaches typically involve reaction of air, or CO<sub>2</sub>-bearing fluids/gases, with ground minerals at the Earth's surface, occurring more slowly than *ex situ* reactions. Examples include reactions of natural minerals in controlled environments like greenhouses (Myers and Nakagaki, 2020), or in heaps or piles

of artificial wastes such as slags (Stolaroff et al., 2005) or mine tailings (Wilson et al., 2006, 2009). In both *ex situ* and surficial CO<sub>2</sub> mineralization, the carbonate products may be valorized (sold or utilized), whereas in *in situ* approaches the mineralized CO<sub>2</sub> is safely and permanently stored underground. Surficial approaches may also be harnessed for the purpose of cost-effective direct air capture (DAC) (**Box 1**).

Other geochemical NETs involve the dispersing of alkaline minerals for the purpose of enhanced weathering in large open spaces, exploiting certain environmental conditions. Where the reactive medium is soil, this is referred to as enhanced weathering in soil, or terrestrial enhanced weathering (section Enhanced Weathering in Soils) (Schuiling and Krijgsman, 2006). Where the weathering takes place at beaches and coastal shelves, the method is referred to as coastal enhanced weathering (Montserrat et al., 2017). Coastal enhanced weathering is one approach for ocean alkalinity enhancement (OAE), which is any process that involves increasing alkalinity in the oceans, resulting in atmospheric CO<sub>2</sub> removal. Other methods for OAE include ocean liming (Caserini et al., 2021), and a range of electrochemical processes (House et al., 2007; Davies, 2015; Mustafa et al., 2020). The role of alkaline minerals in ocean-based NETs is discussed in the section Ocean Alkalinity Enhancement. Key features of the different geochemical NETs are summarized in **Table 3**. Finally, biological mechanisms that influence geochemical reactions or transport ions may potentially be integrated into many of the above-mentioned

**TABLE 1 |** Mg-, Ca-, Na- silicate, oxide, hydroxide, aluminate, carbonate, etc., minerals typically encountered in geochemical NETs, with their Gibbs free energies ( $\Delta G_f$ ) and enthalpies ( $\Delta H_f$ ) of formation (Robie and Hemingway, 1995), and in some cases their Gibbs free energies ( $\Delta G_r$ ) of reaction with  $H_2O$  and  $CO_2$ , at 1 atm and 25°C [calculated using data from Robie and Hemingway (1995)].

Mineral	Formula	Molar mass (g mol <sup>-1</sup> )	$\Delta G_f$ (kJ mol <sup>-1</sup> )	$\Delta H_f$ (kJ mol <sup>-1</sup> )	Reaction	$\Delta G_r$ (kJ mol <sup>-1</sup> )	$\Delta H_r$ (kJ mol <sup>-1</sup> )
<b>Carbonation—aqueous</b>							
Calcite	CaCO <sub>3</sub>	100.09	-1128.5	-1207.4	CaCO <sub>3</sub> + CO <sub>2</sub> + H <sub>2</sub> O → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	22.8	-35.9
Chrysotile	Mg <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	277.11	-4034.0	-4361.7	Mg <sub>3</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> + 6CO <sub>2</sub> + 5H <sub>2</sub> O → 3Mg <sup>2+</sup> + 6HCO <sub>3</sub> <sup>-</sup> + 2H <sub>4</sub> SiO <sub>4</sub>	24.7	-308.4
Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>	184.41	-2161.3	-2324.5	CaMg(CO <sub>3</sub> ) <sub>2</sub> + 2CO <sub>2</sub> + 2H <sub>2</sub> O → Mg <sup>2+</sup> + Ca <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup>	48.6	-86.2
Hydromagnesite	Mg <sub>5</sub> (CO <sub>3</sub> ) <sub>4</sub> (OH) <sub>2</sub> ·4H <sub>2</sub> O	546.54	-5864.16	-6514.9	Mg <sub>5</sub> (CO <sub>3</sub> ) <sub>4</sub> (OH) <sub>2</sub> ·4H <sub>2</sub> O + 6CO <sub>2</sub> → 5Mg <sup>2+</sup> + 10HCO <sub>3</sub> <sup>-</sup>	27.9	-357.6
Ikaite	CaCO <sub>3</sub> ·6H <sub>2</sub> O	208.21	-2540.9	-2954.1	CaCO <sub>3</sub> ·6H <sub>2</sub> O + CO <sub>2</sub> → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> + 5H <sub>2</sub> O	12.6	-4.0
Magnesite	MgCO <sub>3</sub>	84.32	-1029.5	-1113.3	MgCO <sub>3</sub> + CO <sub>2</sub> + H <sub>2</sub> O → Mg <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	22.5	-54.1
Monohydrocalcite	CaCO <sub>3</sub> ·H <sub>2</sub> O	118.11	-1361.6	-1498.3	CaCO <sub>3</sub> ·H <sub>2</sub> O + CO <sub>2</sub> → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	18.8	-30.8
Nesquehonite	MgCO <sub>3</sub> ·3H <sub>2</sub> O	138.38	-1723.8	-1977.26	MgCO <sub>3</sub> ·3H <sub>2</sub> O + CO <sub>2</sub> → Mg <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> + 2H <sub>2</sub> O	5.4	-47.5
Sodium carbonate	Na <sub>2</sub> CO <sub>3</sub>	105.99	-1045.3	-1129.2	Na <sub>2</sub> CO <sub>3</sub> + CO <sub>2</sub> + H <sub>2</sub> O → 2Na <sup>+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-29.9	-51.9
Thermonatrite	NaHCO <sub>3</sub>	124.01	-851.2	-949.0	Na <sub>2</sub> CO <sub>3</sub> ·H <sub>2</sub> O + CO <sub>2</sub> → 2Na <sup>+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-26.2	-37.2
Anorthite	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	278.22	-4007.9	-4229.1	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> + 2CO <sub>2</sub> + 3H <sub>2</sub> O → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> + Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	-36.8	-169.2
Diopside	MgCaSi <sub>2</sub> O <sub>6</sub>	216.57	-3036.6	-3210.7	MgCaSi <sub>2</sub> O <sub>6</sub> + 4CO <sub>2</sub> + 6H <sub>2</sub> O → Ca <sup>2+</sup> + Mg <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup> + 2H <sub>4</sub> SiO <sub>4</sub>	25.4	-189.8
Forsterite	Mg <sub>2</sub> SiO <sub>4</sub>	140.71	-2053.6	-2173.0	Mg <sub>2</sub> SiO <sub>4</sub> + 4CO <sub>2</sub> + 4H <sub>2</sub> O → 2Mg <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup> + H <sub>4</sub> SiO <sub>4</sub>	-25.3	-263.2
Jennite	Ca <sub>9</sub> Si <sub>6</sub> O <sub>18</sub> (OH) <sub>6</sub> ·8H <sub>2</sub> O	927.32	-13644.4	-272.0	Ca <sub>9</sub> Si <sub>6</sub> O <sub>18</sub> (OH) <sub>6</sub> ·8H <sub>2</sub> O + 18CO <sub>2</sub> + 10H <sub>2</sub> O → 9Ca <sup>2+</sup> + 18HCO <sub>3</sub> <sup>-</sup> + 6H <sub>4</sub> SiO <sub>4</sub>	-457.5	-1145.9
Larnite	Ca <sub>2</sub> SiO <sub>4</sub>	172.25	-2191.2	-823.0	Ca <sub>2</sub> SiO <sub>4</sub> + 4CO <sub>2</sub> + 4H <sub>2</sub> O → 2Ca <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup> + H <sub>4</sub> SiO <sub>4</sub>	-85.1	-282.0
Rankinite	Ca <sub>3</sub> Si <sub>2</sub> O <sub>7</sub>	288.42	-3748.1	-1293.1	Ca <sub>3</sub> Si <sub>2</sub> O <sub>7</sub> + 6CO <sub>2</sub> + 7H <sub>2</sub> O → 3Ca <sup>2+</sup> + 6HCO <sub>3</sub> <sup>-</sup> + 2H <sub>4</sub> SiO <sub>4</sub>	-83.2	-386.2
Tobermorite	Ca <sub>5</sub> Si <sub>6</sub> O <sub>12</sub> (OH) <sub>10</sub> ·5H <sub>2</sub> O	588.94	-10466.4	-824.6	Ca <sub>5</sub> Si <sub>6</sub> O <sub>12</sub> (OH) <sub>10</sub> ·5H <sub>2</sub> O + 10CO <sub>2</sub> + 7H <sub>2</sub> O → 5Ca <sup>2+</sup> + 10HCO <sub>3</sub> <sup>-</sup> + 6H <sub>4</sub> SiO <sub>4</sub>	-513.0	-941.4
Wollastonite	CaSiO <sub>3</sub>	116.17	-1549.9	-1635.2	CaSiO <sub>3</sub> + 2CO <sub>2</sub> + 3H <sub>2</sub> O → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> + H <sub>4</sub> SiO <sub>4</sub>	-5.1	-103.0
Brucite	Mg(OH) <sub>2</sub>	58.33	-833.5	-924.5	Mg(OH) <sub>2</sub> + 2CO <sub>2</sub> → Mg <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-26.2	-135.2
Lime	CaO	56.08	-603.1	-635.1	CaO + 2CO <sub>2</sub> + H <sub>2</sub> O → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-118.3	-214.7
Periclase	MgO	40.31	-569.2	-601.5	MgO + 2CO <sub>2</sub> + H <sub>2</sub> O → Mg <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-53.5	-172.4
Portlandite	Ca(OH) <sub>2</sub>	74.1	-898.4	-986.1	Ca(OH) <sub>2</sub> + 2CO <sub>2</sub> → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup>	-60.1	-149.5
CAH10	CaAl <sub>2</sub> O <sub>4</sub> ·10H <sub>2</sub> O	488.14	-4622.3	-	CaAl <sub>2</sub> O <sub>4</sub> ·10H <sub>2</sub> O + 2CO <sub>2</sub> → Ca <sup>2+</sup> + 2HCO <sub>3</sub> <sup>-</sup> + 2Al(OH) <sub>3</sub> + 6H <sub>2</sub> O	-68.5	-
C2AH8	Ca <sub>2</sub> Al <sub>2</sub> O <sub>5</sub> ·8H <sub>2</sub> O	478.2	-4812.8	-	Ca <sub>2</sub> Al <sub>2</sub> O <sub>5</sub> ·8H <sub>2</sub> O + 4CO <sub>2</sub> → 2Ca <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup> + 2Al(OH) <sub>3</sub> + 3H <sub>2</sub> O	-125.3	-
C3AH6	Ca <sub>3</sub> Al <sub>2</sub> (OH) <sub>12</sub>	378.32	-5019.3	-	Ca <sub>3</sub> Al <sub>2</sub> (OH) <sub>12</sub> + 6CO <sub>2</sub> → 3Ca <sup>2+</sup> + 6HCO <sub>3</sub> <sup>-</sup> + 2Al(OH) <sub>3</sub>	-165.8	-
C4AH13	Ca <sub>4</sub> Al <sub>2</sub> O <sub>7</sub> ·13H <sub>2</sub> O	755.41	-7327.5	-	Ca <sub>4</sub> Al <sub>2</sub> O <sub>7</sub> ·13H <sub>2</sub> O + 8CO <sub>2</sub> → 4Ca <sup>2+</sup> + 8HCO <sub>3</sub> <sup>-</sup> + 2Al(OH) <sub>3</sub> + 6H <sub>2</sub> O	-238.8	-

(Continued)



TABLE 1 | Continued

Mineral	Formula	Molar mass (g mol <sup>-1</sup> )	$\Delta G_f$ (kJ mol <sup>-1</sup> )	$\Delta H_f$ (kJ mol <sup>-1</sup> )	Reaction	$\Delta G_f$ (kJ mol <sup>-1</sup> )	$\Delta H_f$ (kJ mol <sup>-1</sup> )
C2FH8	Ca <sub>2</sub> Fe <sub>2</sub> O <sub>5</sub> .8H <sub>2</sub> O	416.01	-3919.0	-	Ca <sub>2</sub> Fe <sub>2</sub> O <sub>5</sub> .8H <sub>2</sub> O + 4CO <sub>2</sub> → 2Ca <sup>2+</sup> + 4HCO <sub>3</sub> <sup>-</sup> + Fe <sub>2</sub> O <sub>3</sub> + 6H <sub>2</sub> O	-163.2	-
C3FH6	Ca <sub>3</sub> Fe <sub>2</sub> O <sub>6</sub> .6H <sub>2</sub> O	436.05	-4125.5	-	Ca <sub>3</sub> Fe <sub>2</sub> O <sub>6</sub> .6H <sub>2</sub> O + 6CO <sub>2</sub> → 3Ca <sup>2+</sup> + 6HCO <sub>3</sub> <sup>-</sup> + Fe <sub>2</sub> O <sub>3</sub> + 3H <sub>2</sub> O	-203.8	-
C4FH13	Ca <sub>4</sub> Fe <sub>2</sub> O <sub>7</sub> .13H <sub>2</sub> O	618.27	-6433.7	-	Ca <sub>4</sub> Fe <sub>2</sub> O <sub>7</sub> .13H <sub>2</sub> O + 8CO <sub>2</sub> → 4Ca <sup>2+</sup> + 8HCO <sub>3</sub> <sup>-</sup> + Fe <sub>2</sub> O <sub>3</sub> + 9H <sub>2</sub> O	-276.7	-
Tricarboaluminate	Ca <sub>6</sub> Al <sub>2</sub> (CO <sub>3</sub> ) <sub>3</sub> (OH) <sub>12</sub> .26H <sub>2</sub> O	1147.11	-14536.0	-	Ca <sub>6</sub> Al <sub>2</sub> (CO <sub>3</sub> ) <sub>3</sub> (OH) <sub>12</sub> .26H <sub>2</sub> O + 9CO <sub>2</sub> → 6Ca <sup>2+</sup> + 12HCO <sub>3</sub> <sup>-</sup> + 2Al(OH) <sub>3</sub> + 23H <sub>2</sub> O	-131.0	-
<b>Carbonation – solid</b>							
Anorthite	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	278.22	-4007.9	-4229.1	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> + CO <sub>2</sub> + 2H <sub>2</sub> O → CaCO <sub>3</sub> + Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub>	-59.5	-133.3
Diopside	MgCaSi <sub>2</sub> O <sub>6</sub>	216.57	-3036.6	-3210.7	MgCaSi <sub>2</sub> O <sub>6</sub> + 2CO <sub>2</sub> + 4H <sub>2</sub> O → CaCO <sub>3</sub> + MgCO <sub>3</sub> + 2H <sub>4</sub> SiO <sub>4</sub>	-19.9	-99.8
Forsterite	Mg <sub>2</sub> SiO <sub>4</sub>	140.71	-2053.6	-2173.0	Mg <sub>2</sub> SiO <sub>4</sub> + 2CO <sub>2</sub> + 2H <sub>2</sub> O → 2MgCO <sub>3</sub> + H <sub>4</sub> SiO <sub>4</sub>	-70.3	-155
Gehlenite	Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub>	274.21	-3808.7	-4007.6	Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub> + 2CO <sub>2</sub> + 5H <sub>2</sub> O → 2CaCO <sub>3</sub> + 2Al(OH) <sub>3</sub> + H <sub>2</sub> SiO <sub>4</sub>	-111.6	-237.4
Jennite	Ca <sub>9</sub> Si <sub>6</sub> O <sub>18</sub> (OH) <sub>6</sub> .8H <sub>2</sub> O	927.32	-13644.4	-272.0	Ca <sub>9</sub> Si <sub>6</sub> O <sub>18</sub> (OH) <sub>6</sub> .8H <sub>2</sub> O + 9CO <sub>2</sub> + H <sub>2</sub> O → 9CaCO <sub>3</sub> + 6H <sub>4</sub> SiO <sub>4</sub>	-662.4	-822.8
Larnite	Ca <sub>2</sub> SiO <sub>4</sub>	172.25	-2191.2	-823.0	Ca <sub>2</sub> SiO <sub>4</sub> + 2CO <sub>2</sub> + 2H <sub>2</sub> O → 2CaCO <sub>3</sub> + H <sub>4</sub> SiO <sub>4</sub>	-130.7	-210.2
Lime	CaO	56.08	-603.1	-635.1	CaO + CO <sub>2</sub> → CaCO <sub>3</sub>	-141.0	-178.8
Merwinite	MgCa <sub>3</sub> Si <sub>2</sub> O <sub>8</sub>	328.71	-4339.4	-4566.8	MgCa <sub>3</sub> Si <sub>2</sub> O <sub>8</sub> + 4CO <sub>2</sub> + 4H <sub>2</sub> O → 3CaCO <sub>3</sub> + MgCO <sub>3</sub> + 2H <sub>4</sub> SiO <sub>4</sub>	-205.3	-371.5
Rankinite	Ca <sub>3</sub> Si <sub>2</sub> O <sub>7</sub>	288.42	-3748.1	-1293.1	Ca <sub>3</sub> Si <sub>2</sub> O <sub>7</sub> + 3CO <sub>2</sub> + 4H <sub>2</sub> O → 3CaCO <sub>3</sub> + 2H <sub>4</sub> SiO <sub>4</sub>	-151.5	-386.2
Tobermorite	Ca <sub>5</sub> Si <sub>6</sub> O <sub>12</sub> (OH) <sub>10</sub> .5H <sub>2</sub> O	588.94	-10466.4	-824.6	Ca <sub>5</sub> Si <sub>6</sub> O <sub>12</sub> (OH) <sub>10</sub> .5H <sub>2</sub> O + 5CO <sub>2</sub> + 2H <sub>2</sub> O → 5CaCO <sub>3</sub> + 6H <sub>4</sub> SiO <sub>4</sub>	-626.8	-761.9
Wollastonite	CaSiO <sub>3</sub>	116.17	-1549.9	-1635.2	CaSiO <sub>3</sub> + CO <sub>2</sub> + 2H <sub>2</sub> O → CaCO <sub>3</sub> + H <sub>4</sub> SiO <sub>4</sub>	-27.8	-67.1
Brucite	Mg(OH) <sub>2</sub>	58.33	-833.5	-924.5	Mg(OH) <sub>2</sub> + CO <sub>2</sub> → MgCO <sub>3</sub> + H <sub>2</sub> O	-48.7	-81.1
Portlandite	Ca(OH) <sub>2</sub>	74.1	-898.4	-986.1	Ca(OH) <sub>2</sub> + CO <sub>2</sub> → CaCO <sub>3</sub> + H <sub>2</sub> O	-82.8	-113.6
Periclase	MgO	40.31	-569.2	-601.5	MgO + CO <sub>2</sub> → MgCO <sub>3</sub>	-75.9	-118.3
					Amorphous silica precipitation*		
					H <sub>4</sub> SiO <sub>4</sub> → SiO <sub>2</sub> + 2H <sub>2</sub> O	-22.7	-22.3

\*The enthalpy and free energy for precipitation of SiO<sub>2</sub> from H<sub>4</sub>SiO<sub>4</sub> is given (last row) to enable calculation of the free energy and enthalpy of carbonation reactions where SiO<sub>2</sub> is the product.

**TABLE 2** | Current production values for some common artificial minerals used in geochemical NETs, with estimated production values and NET potentials by 2050.

	Current annual production levels (Gt of material)	2050 forecast production (Gt of material)	2050 NET potential (Gt CO <sub>2</sub> yr. <sup>-1</sup> )	References
Ash	0.6–1	17–29	0.07–0.14	Renforth, 2019
Cement—uptake into cement and construction and demolition wastes	1.4–5.8	40–170	1.4–2.0	Renforth, 2019
Mine tailings	9–17		1.1–4.5	Bullock et al., 2021
Red mud	0.12	3.5	<0.1	Renforth, 2019
Slag (BOF, BFS)	0.17–0.5	5–15	0.3–0.5	Renforth, 2019
Desalination brines (dry NaOH)	0.12	0.06	0.2	Gao et al., 2017; Jones et al., 2019; Ihsanullah et al., 2021
Lime (CaO)	0.43	0.9–1.6*	Unknown, but potentially on the order of 0.5–5***	Renforth, 2019
Magnesia (MgO)	0.14	0.5**		José et al., 2020

\*Forecast production lower estimate from Renforth (2019), upper estimate assuming additional 10% yr.<sup>-1</sup> growth.

\*\*Upper estimate assuming additional 10% yr.<sup>-1</sup> growth.

\*\*\*Lower estimates assuming once through capacity (e.g., ocean liming) on production forecast. Higher estimates for proposals that consider looping Ca or Mg (McQueen et al., 2020) with subsequent geological storage.

#### BOX 1 | Harnessing surficial mineralization processes for direct air capture.

Direct Air Capture (DAC) requires the removal of CO<sub>2</sub> from the air to produce a concentrated source of CO<sub>2</sub>. This concentrated CO<sub>2</sub> can then either be utilized or permanently stored. Surficial mineralization processes have been proposed that fit this definition. For example calcium oxide (Hanak et al., 2017; Hanak and Manovic, 2018) or magnesium oxide (McQueen et al., 2020) looping systems. Calcium looping is a pre- or post-combustion CO<sub>2</sub> capture technology which uses high temperatures. Carbonation is usually performed at around 650°C to form calcium carbonate (CaCO<sub>3</sub>). The carbonates, while still hot, are then fed into the next part of the system where they are calcined above 900°C. The calcination step regenerates the lime and produces a more concentrated source of CO<sub>2</sub> which is suitable for CCS (Martínez et al., 2018). Hybrid surficial DAC works similarly, except that CaO or MgO is carbonated under ambient conditions in air, at potentially unlimited scale. The carbonation step in air has slower reaction kinetics than calcium looping processes and is thought to be dependent on a relative humidity above 55% (Erans et al., 2020; Samari et al., 2020). For further details on these and other DAC systems see (Sanz-Pérez et al., 2016; Okesola et al., 2018; McQueen et al., 2021a).

methods to improve efficiency (section Application of Biotechnology to Geochemical NETs).

## ALKALINE MINERAL RESOURCES

### Common Alkaline Minerals

A mineral is an inorganic solid with distinctive chemical and physical properties, composition, and atomic structure, whereas rocks are an assemblage of minerals. In geochemical NETs, the alkalinity for the neutralization reaction (Equation 1) is usually supplied by abundant Ca- and Mg-rich silicate (or aluminosilicate) minerals, and in some cases the oxides, hydroxides or carbonates of calcium and magnesium (see

**Table 1**). Potential material resources for magnesium-bearing minerals are much larger than that of calcium owing to their natural availability, while on the other hand, markets for magnesium-based products are much smaller than that of calcium-based products. Minerals rich in other cations such as Na, K, Fe are also considered in some approaches (Khesghi, 1995; Palandri and Kharaka, 2005; Campbell, 2019).

### Naturally Occurring Alkaline Rocks

Alkaline minerals are found in alkaline rocks, including: (i) igneous rocks, such as basalt and peridotite (McGrail et al., 2006; Kelemen and Matter, 2008; Matter and Kelemen, 2009; Clark, 2019; Kelemen et al., 2020), (ii) metamorphic rocks, such as serpentinites (Okamoto et al., 2006; Power et al., 2013b; Bide et al., 2014; Dichicco et al., 2015), and (iii) sedimentary rocks such as limestone and dolomite (Rau and Caldeira, 1999; Rau et al., 2007; Rau, 2011). There are two main types of igneous and metamorphic alkaline rock considered for geochemical NETs: (i) mafic rocks such as basalt, and (ii) ultramafic rocks such as peridotite and serpentinite. Mafic and ultramafic rocks are chemically and physically distinct. For example, mafic rocks typically contain 15–28% MgO, 1–15% CaO, and 46–54% SiO<sub>2</sub> (among other minor components), whereas ultramafic rocks typically contain 35–46% MgO, 5–15% CaO, and 42–48% SiO<sub>2</sub> (Sen, 2014). Depending on the particular geochemical NET, some rock types might be more suitable than others, e.g., olivine may be more promising than basalt in enhanced weathering approaches. For *in situ* mineralization, both mafic (e.g., basalt) and ultramafic (e.g., peridotite and serpentinite) formations with suitable properties, such as high porosities and permeabilities, will allow for cost-effective storage. Together, mafic and ultramafic rocks represent over 90 teratonnes (Tt) of resources, sufficient to store the equivalent of 700-years worth

**TABLE 3 |** Qualitative comparison of geochemical NET processes.

	CO <sub>2</sub> mineralization			Enhanced weathering and ocean alkalinity enhancement			
	<i>In situ</i>	<i>Ex situ</i>	Surficial	Enhanced weathering in soils	Enhanced weathering at coasts	Ocean liming	Electrochemical seawater splitting
Where?	Subsurface	Reactors	Heaps, piles, ponds, greenhouses	Forest and agricultural soils	Beaches and coastal shelves	Oceans	Coastal zones and oceans
CO <sub>2</sub> source	Suited for industrial flue gases, but also air/DAC	Industry (potentially also DAC)	Air/DAC/industry	Air	Air	Air	Air
If concentrated CO <sub>2</sub> is used, is it retained?	Yes, minor losses	Yes	Potential losses	N/A	N/A	N/A	N/A
CO <sub>2</sub> transport?	Some approaches	No	Some approaches	N/A	N/A	N/A	N/A
Mineral transport?	Rocks in place	Rocks transported to CO <sub>2</sub> sources	Minimal transport, some spreading	Transport and spreading	Transport and spreading	Transport and spreading	Rocks transported to electrochemical reactor
Carbon products	Carbonate minerals	Carbonate minerals	Carbonate minerals	Carbonate minerals, ocean (bi)carbonate	Ocean (bi)carbonate	Ocean (bi)carbonate	Ocean (bi)carbonate
Can carbon products be utilized?	No	Yes	Yes	No	No	No	No
Temperature and pressure	Rock formation dependent (depth)	Likely non-ambient	Ambient	Ambient	Ambient	Non-ambient (calcination)	N/A
Enhancements	CO <sub>2</sub> concentration, reaction driven cracking, H <sub>2</sub> O/CO <sub>2</sub> ratio, additives	Grinding, mixing, agitation, sonication, acids, salts, additives, pre-treatments, CO <sub>2</sub> concentration, solid-liquid ratio, humidity	Grinding, mixing, agitation, sonication, dispersing minerals, sparging, CO <sub>2</sub> concentration, solid-liquid ratio, humidity	Comminution. Physical, chemical, and biological location-dependent weathering	Comminution. Physical, chemical, and biological location-dependent weathering	Calcination to produce highly reactive CaO	Electricity
CO <sub>2</sub> removal rate	Days to years	Minutes	Weeks to months	Years	Years	Weeks	Weeks
Potential removal scale	Gt CO <sub>2</sub> yr. <sup>-1</sup>	Mt CO <sub>2</sub> yr. <sup>-1</sup>	Mt CO <sub>2</sub> yr. <sup>-1</sup>	Mt–Gt CO <sub>2</sub> yr. <sup>-1</sup>	Mt–Gt CO <sub>2</sub> yr. <sup>-1</sup>	Mt–Gt CO <sub>2</sub> yr. <sup>-1</sup>	Mt CO <sub>2</sub> yr. <sup>-1</sup>
Emissions reduction (ER) or NET	ER or NET	ER or NET	ER or NET	NET	NET	NET	NET
Monitoring and verification	Potentially straightforward	Straightforward	Potentially straightforward	Difficult	Difficult	Difficult	Difficult

of global CO<sub>2</sub> emissions (Bide et al., 2014). For *ex situ* and surficial mineralization, as well as for enhanced weathering, near-surface deposits of mafic and ultramafic alkaline rocks could be mined, crushed, and ground to create high surface areas to facilitate a reasonable rate of reaction with CO<sub>2</sub>. In this regard, the available rock resources that could be used for geochemical NETs at the Earth's surface are plentiful, since the estimates for global sand, gravel, and stone reserves amount to more than 190 Tt (Sverdrup et al., 2017). If only a small part of this industry were to be redirected toward production of crushed alkaline rocks for surface geochemical NETs, then many Gt CO<sub>2</sub> yr.<sup>-1</sup> would be achieved in the near future. The reason is that a robust and expanding industry is already in place, i.e., the construction aggregates industry, which annually extracts and processes 50 billion tons (Gt) of rocks (range 47–59 Gt) (Sverdrup et al., 2017). Finally, it may be possible to use natural carbonate rocks for enhanced weathering and remove CO<sub>2</sub> from air in the form of bicarbonate (Kirchner et al., 2020).

## Artificial Alkaline Minerals—Industrial By-Products and Wastes, and Tailored Minerals

In addition to naturally occurring mineral resources, some geochemical NETs can also exploit abundant artificial mineral resources (Table 2). These are typically wastes or by-products of industrial processes, landscaping, or quarrying (Dijkstra et al., 2019). On a global scale, it is estimated that 7 Gt of these alkaline mineral by-products/wastes are produced annually, with a combined potential to capture and store CO<sub>2</sub> away from the atmosphere at 2.9–8.5 Gt yr.<sup>-1</sup> by 2100 (Renforth, 2019). More specifically, these materials include: (i) iron and steelmaking slags (blast furnace, basic oxide, electric arc furnace, ladle furnace, and argon oxygen decarburization slags) (Mayes et al., 2018; Pullin et al., 2019; Reddy et al., 2019; Luo and He, 2021); (ii) cement wastes (cement and concrete wastes, construction and demolition wastes, cement kiln/bypass dust, recycled calcium sulfates, and blended hydraulic slag cement) (Huntzinger et al., 2009a; Medas et al., 2017; Pedraza et al., 2021); (iii) ashes and relevant residues [bottom ash from furnaces and incinerators (municipal solid waste incinerator bottom ash, fly ash, boiler ash, coal slag, oil shale ash), air pollution control residues (cyclone dust, cloth bag dust), and fuel combustion ashes (coal fly ash, lignite fly ash, oil shale, biomass ashes)] (Alba et al., 2001; Baciocchi et al., 2006; Sun et al., 2008; Zhang et al., 2008; Montes-Hernandez et al., 2009; Prigione et al., 2009; Lombardi et al., 2016; Brück et al., 2018; Liu et al., 2018; Ji et al., 2019; Vassilev et al., 2021); (iv) mine and mineral processing wastes (asbestos tailings, nickel tailings, diamond tailings, and red mud) (Wilson et al., 2010, 2014; Power et al., 2014, 2020; Gras et al., 2017; Mervine et al., 2018); (v) alkaline paper mill wastes (lime kiln residues, green liquor dreg, paper sludge) (Pérez-López et al., 2008; Sun et al., 2013; Li and Sun, 2014; Spínola et al., 2021); and (vi) reject brines from desalination (Mustafa et al., 2020). The latter can be employed by electrochemical approaches that aim at removing acidity (HCl) from seawater and return alkalinity (NaOH). Currently, more than 95 million m<sup>3</sup> of desalinated water is produced daily on a

global scale, which is responsible for generating more than 141 million m<sup>3</sup> of brine each day that is typically discharged into the oceans, often negatively affecting the receiving ecosystems (Jones et al., 2019). This number is on the rise, since recent estimates suggest that by 2030 the global desalination capacity will be more than 200 million m<sup>3</sup> day<sup>-1</sup> (Ihsanullah et al., 2021), while this number could be more than tripled by 2050 since the total global desalination population is projected to increase by 3.2-fold in 2050 compared to the present (Gao et al., 2017). These very large volumes of reject brines (waste) present certain advantages for OAE, since their mean salinity is twice that of seawater (Ihsanullah et al., 2021), suggesting that if they were used for OAE, CO<sub>2</sub> removal at the Mt yr.<sup>-1</sup> scale at least could be achieved in the nearterm.

Regarding the solid alkaline waste materials, these are generally low-cost (Huijgen et al., 2005) and often deposited in heaps or buried at the shallow subsurface, implying that these are more accessible and more readily available than natural minerals. Furthermore, most legacy deposits may be only partially weathered, suggesting their great potential for CO<sub>2</sub> removal. For example, 40–140 years after deposition, a slag deposit in Consett, England, which is estimated to be over 30 Mt, has only reached ~3% of its CO<sub>2</sub> sequestration potential (Pullin et al., 2019). Artificial alkaline minerals also tend to have higher reactivities than natural minerals, due to their activation by various industrial pre-treatments (e.g., grinding and heat treatment), which often create high surface areas and higher crystal disorder (La Plante et al., 2021a). However, compared to natural alkaline rocks, they are less abundant and may contain more labile toxic metals, possibly making their use problematic in large-scale geochemical NETs.

Therefore, rather than using wastes and by-products of existing industrial processes, artificial alkaline minerals, tailored for the purpose of negative emissions, could be more promising. For example, the carbonates of calcium and magnesium can be calcined, the CO<sub>2</sub> generated by their decomposition could be captured and stored, while the resulting high-reactivity oxides (CaO and MgO) could be used in different NETs such as power generation using an integrated solid-oxide fuel cell (Hanak et al., 2017), metal oxide looping DAC (see Box 1) (McQueen et al., 2020), or ocean liming applications (Renforth and Kruger, 2013; Renforth et al., 2013). Substances other than CaO and MgO have also been investigated for hybrid DAC systems, such as sodium and potassium oxides, and related compounds (Nikulshina et al., 2008; Campbell, 2019).

## REACTION CHEMISTRY

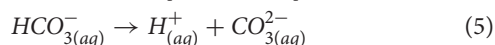
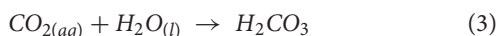
Reactions of alkaline minerals with CO<sub>2</sub> can occur as gas-solid systems (e.g., Equation 2).



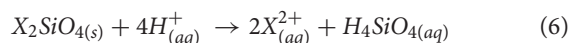
Where X is Mg or Ca. Humidity is usually required to catalyze these reactions. See section Gas-Solid Kinetics for discussion on the kinetics of gas-solid reactions.



Alternatively, and more commonly, reactions of alkaline silicate minerals with CO<sub>2</sub> occur in the aqueous phase. First, CO<sub>2</sub> dissolves in water forming carbonic acid (H<sub>2</sub>CO<sub>3</sub>), which releases acidity, H<sup>+</sup>, into solution:



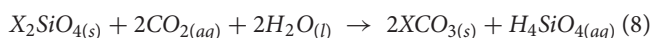
Alkaline mineral surfaces then react with H<sup>+</sup>:



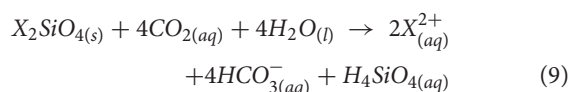
Over time, carbonate minerals may precipitate:



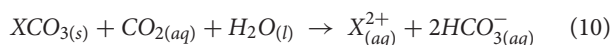
Overall the reaction is:



Theoretically, 1 mol of CO<sub>2</sub> is removed for every 1 mol of alkaline metal. Similar reactions can occur for a wide array of alkaline silicate minerals (**Table 1**). In some geochemical NETs, such as coastal enhanced weathering, the goal is to remove carbon and store it as dissolved ocean bicarbonate, rather than minerals:

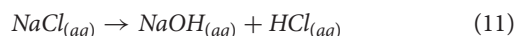


In this case, 2 mol of CO<sub>2</sub> are theoretically removed for every 1 mol of alkaline metal (example reactions are given in **Table 1**). The residence time of bicarbonate is tens to hundreds of thousands of years in the ocean and thus it can be considered a stable store of carbon since abiotic mineral carbonate formation is kinetically inhibited by the ocean's chemistry (Renforth and Henderson, 2017). Alkaline carbonates can also be used to remove CO<sub>2</sub>:

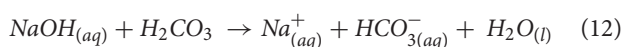


In this instance, 1 mol of CO<sub>2</sub> is theoretically removed for every 1 mol of alkaline metal, assuming the carbon is stored as ocean bicarbonate.

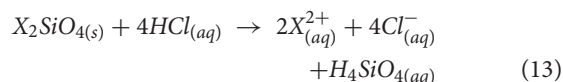
Some geochemical NETs do not react alkaline minerals with CO<sub>2</sub> or H<sub>2</sub>CO<sub>3</sub> directly, but instead react alkaline minerals with other acids, which are by-products or wastes of other NETs. For example, electrochemical seawater dialysis may produce HCl (House et al., 2007; Davies, 2015):



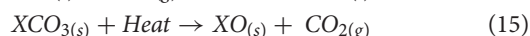
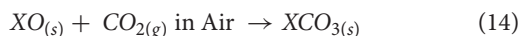
Where NaOH is used for ocean alkalinity enhancement and CO<sub>2</sub> removal:



And where HCl is disposed of by reaction with alkaline minerals.



Furthermore, some geochemical NETs capture CO<sub>2</sub> from air and produce concentrated CO<sub>2</sub> gas in a looping process (McQueen et al., 2020):

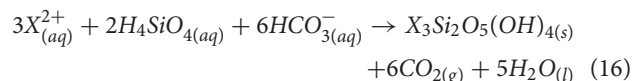


The concentrated CO<sub>2</sub> stream can then be used or safely stored geologically. This process may be possible with minerals other than Ca- and Mg- oxides.

## CARBONATE PRODUCTS AND OTHER SECONDARY MINERALS

Solid products of geochemical NETs primarily include carbonate minerals such as calcite (CaCO<sub>3</sub>), magnesite (MgCO<sub>3</sub>), dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>), and various hydrated magnesium carbonates (Mg<sub>5</sub>(CO<sub>3</sub>)<sub>4</sub>(OH)<sub>2</sub>·nH<sub>2</sub>O). These are stable enough to be stored for long time periods. Other carbonate minerals such as siderite (FeCO<sub>3</sub>), dawsonite (NaAl(CO<sub>3</sub>)(OH)<sub>2</sub>), and ankerite (Ca(Fe,Mg,Mn)(CO<sub>3</sub>)<sub>2</sub>) can act as stores of carbon, but may only be stable in subsurface environments (Hellevang et al., 2005; Snæbjörnsdóttir et al., 2014; Yu et al., 2020).

Besides carbonates, other products of mineral carbonation and weathering include silica, iron oxides, and clays. These secondary minerals, including the carbonate products, can occlude reactive surfaces, halting further reaction (Béarat et al., 2006; Andreani et al., 2009; Maher et al., 2009; Saldi et al., 2013; Sissmann et al., 2014). The role of clay mineral formation *via* “reverse weathering” (Equation 16) is a subject of ongoing debate within several geochemical NETs, as these reactions may inhibit their CO<sub>2</sub> sequestration efficiencies (Montserrat et al., 2017; Oelkers et al., 2018; Renforth and Campbell, 2021).



Where X is a cation such as Mg<sup>2+</sup> or Ca<sup>2+</sup>. Successful geochemical NETs will likely include approaches for avoiding or minimizing the extent and impact of secondary minerals.

## KINETICS

The field of kinetics involves the study of reaction rates, and provides the basis for reactor design and system optimization. Although conversion of alkaline minerals into carbonates is thermodynamically favored in the presence of CO<sub>2</sub>, the reactions are kinetically inhibited. To become an effective tool for climate change mitigation, conversion rates must be enhanced considerably. **Table 3** summarizes some common enhancements. In geochemical NETs, there are many competing effects, and

trade-offs will be required. For example, maintaining a low pH can significantly increase the dissolution rate of silicate minerals but will limit the formation of carbonate minerals, while elevated temperatures favor mineral dissolution and carbonate precipitation, they also lead to lower CO<sub>2</sub> solubility. These competing effects are particularly relevant to direct carbonation. Separating dissolution and precipitation allows each process to be optimized independently (indirect carbonation). See section *Ex situ* for more details on direct vs. indirect approaches. Generally speaking, most geochemical NETs are CO<sub>2</sub>-mineral-water systems that can be divided into gas-solid (section Gas-Solid Kinetics), or aqueous (section Aqueous Phase Kinetics). In the latter, three main processes occur: (i) mineral dissolution (section Mineral Dissolution); (ii) CO<sub>2</sub> dissolution and hydration (section CO<sub>2</sub> Dissolution and Hydration); and (iii) precipitation of carbonate minerals (section Carbonate Precipitation). Biological influences on kinetics are discussed separately in the section Application of Biotechnology to Geochemical NETs.

## Gas-Solid Kinetics

Gas-solid kinetics are relevant to *ex situ* (Baciacchi et al., 2009) and surficial (Myers and Nakagaki, 2020) CO<sub>2</sub> mineralization, as well as DAC (McQueen et al., 2020). These systems operate with gaseous (humidity), rather than liquid water, thus avoiding significant leaching of potentially toxic metals (El-Naas et al., 2015). The reaction of spherical particles of natural and artificial alkaline minerals with CO<sub>2</sub> is usually limited by ion diffusion through a growing product layer, a process often described by a shrinking core model such as in Equation (17) (Yagi and Kunii, 1955).

$$t = \frac{\rho_{\text{solid}} r^2}{6DC_{\text{gas}}} \left[ 1 - 3 \left( \frac{r-d}{r} \right)^2 + 2 \left( \frac{r-d}{r} \right)^3 \right] \quad (17)$$

where  $t$  is time (s),  $\rho_{\text{solid}}$  is the molar density of Ca or Mg in the solid phase (mole m<sup>-3</sup> of mineral),  $r$  is the particle radius (m),  $d$  is the thickness of the product layer (m),  $C_{\text{gas}}$  is the CO<sub>2</sub> concentration in the gas phase (moles m<sup>-3</sup> of gas), and  $D$  is the carbonate ion diffusivity through the product layer (m<sup>2</sup> s<sup>-1</sup>). The ion diffusivity has an Arrhenius temperature dependence, thus increasing temperature increases carbonation rate (Li, 2020). According to Equation (17), using pure CO<sub>2</sub> rather than ambient air increases the reaction rate by 3 orders of magnitude whereas grinding from 10 mm to 10  $\mu$ m increases mineralization rates by 6 orders of magnitude. Values of  $D$  for relevant minerals can vary across 7 orders of magnitude depending on the mineral composition and structure (Myers et al., 2019). Other kinetic enhancements are possible for gas-solid processes. For example, in fluidized bed processes, the use of a nanosilica additive increased the gas-solids contact efficiency and carbonation rates of Ca(OH)<sub>2</sub> (Pontiga et al., 2013), while attrition has been shown to prevent the build-up of passivating product layers, improving CO<sub>2</sub> uptake by CaO (Chen et al., 2012).

Humidity also plays a crucial role in gas-solid approaches. For example, the rate and extent of reaction between portlandite (Ca(OH)<sub>2</sub>) and CO<sub>2(g)</sub> (60–90°C) was found to increase

significantly with increasing humidity, proposed to be due to the rate limiting step of dissolution of Ca(OH)<sub>2</sub> in adsorbed surface water (Shih et al., 1999). For brucite (Mg(OH)<sub>2</sub>), dehydroxylation/rehydroxylation processes have been shown to induce morphological changes, including translamellar cracking and delamination, that can serve to enhance carbonation reactivity *via* disruption of the passivating product layer (McKelvy et al., 2001; Fagerlund et al., 2012). Humidity is found to have similar mechanistic effects on gas-solid carbonation of natural silicate minerals such as wollastonite (CaSiO<sub>3</sub>) (Longo et al., 2015) and chrysotile (Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>) (Larachi et al., 2010, 2012), mine tailings (Veetil and Hitch, 2020), and industrial alkaline by-products/wastes such as air pollution control residue (Baciacchi et al., 2006), fly ash (Liu et al., 2018) and calcium silicate hydrates found in hydrated portland cement (Steiner et al., 2020).

## Aqueous Phase Kinetics Mineral Dissolution

Mineral dissolution is the degradation of a solid mineral in aqueous media, with the subsequent release of soluble species such as Mg<sup>2+</sup>/Ca<sup>2+</sup> and H<sub>4</sub>SiO<sub>4</sub> (e.g., Equation 6). The rate of carbon sequestration in geochemical NETs is often limited by the mineral dissolution rate. Mineral dissolution can be described by the rate law:

$$\text{Rate} = SA \cdot k_0 \cdot e^{-E_A/RT} \cdot \prod_i a_i^{n_i} \cdot f(\Delta G) \quad (18)$$

where  $SA$  is the reactive surface area,  $k_0$  is the standard rate constant,  $E_A/RT$  is apparent activation energy divided by the gas constant,  $R$ , and temperature,  $T$ ,  $a_i$  is the activity of aqueous species  $i$  to the power of  $n_i$ , and  $f(\Delta G)$  is a function of the Gibbs free energy change (see Lasaga, 1998; Black et al., 2015 for more detail).

As implied in Equation (18), the rate of dissolution is directly proportional to the reactive surface area (Brantley and Mellott, 2000). For earth-surface geochemical NETs that make use of rocks, crushing and milling is needed to increase reactive surface areas (Haug et al., 2010; Moosdorf et al., 2014; Rigopoulos et al., 2016). For subsurface mineralization, high vesicularity basalts provide large reactive surface areas (Galeczka et al., 2014; Xiong et al., 2018). Temperature also plays an important role in mineral dissolution, since the rate constant in Equation (18) greatly depends on temperature and even small increases in the temperature will largely increase mineral dissolution rates. As a result, in *ex situ* approaches mineral dissolution rates are often enhanced through temperature increase (Gerdemann et al., 2007). For *in situ* approaches, greater depths are prioritized, since the naturally warmer underground temperatures will greatly enhance carbonation rates, by up to 76 times compared to ambient surface rocks (Paukert et al., 2012). Indeed, fully carbonated peridotites (listevenites) give a good indication of the enormous potential of carbonation of ultramafic rocks at high temperatures (Falk and Kelemen, 2015). Finally, for enhanced weathering at the Earth's surface, warm tropical regions are typically prioritized, since in these areas mineral dissolution rates are greatly accelerated (Kohler et al., 2010).

The composition of the aqueous phase also plays an important role. For example, the dissolution rates of minerals such as forsterite and apatite increase linearly with decreasing pH (Brantley, 2008). However, others may show a non-linear dependence, for instance albite has a parabola-shaped dependence, having a minimum at pH ~5 (Gislason et al., 2014). In some geochemical NETs, carbonic acid provides the acidity needed to enhance dissolution (see Equation 3) (O'Connor et al., 2000; Kanakiya et al., 2017) while in others, organic and inorganic acids will provide acidity (van Hees et al., 2000; Kakizawa et al., 2001; Teir et al., 2007a,b). Organic acids can also catalyze silicate dissolution by acting as chelators, which complex and solubilize cations in the mineral crystal framework (Drever and Stillings, 1997; Lazo et al., 2017; Oelkers et al., 2018). Inorganic ligands, such as sulfate and phosphate (Pokrovsky et al., 2005) may also enhance mineral dissolution.

Dissolution rates are also dependent on the solids' composition. In silicate minerals, the dissolution rate is controlled by breaking of the shortest and strongest (usually the Si–O) bonds. Thus, minerals with a low degree of silica polymerization (e.g., olivine) dissolve at faster rates than minerals with higher degrees (e.g., quartz) (Goldich, 1938). For this reason, artificially tailored minerals such as MgO and CaO exhibit much faster dissolution rates than silicates, making them good candidates for OAE (Kheshgi, 1995; Renforth and Henderson, 2017). Furthermore, calcium-rich minerals dissolve faster than their magnesium-rich counterparts, owing to the comparatively weaker Ca–O bond (Brantley, 2008). The presence of transition metals and their potential for reduction–oxidation (redox) reactions, particularly Fe and Mn, can have a substantial impact on dissolution rates, e.g., Fe(III)–O bond is stronger than Fe(II)–O bond, suggesting that reductive conditions could increase mineral dissolution rates (Brantley, 2008). Silicates tend to dissolve non-stoichiometrically, i.e., the ratio of release rates for the various species is not equal to the stoichiometry of the starting mineral, often because their most soluble elements, e.g., Na, K, Ca, Mg, are released preferentially (Brantley, 2008). Such incongruent dissolution may lead to the formation of a silica-rich outer layer on the particle surfaces, inhibiting further dissolution (Béarat et al., 2006; Andreani et al., 2009; Maher et al., 2009; Saldi et al., 2013; Sissmann et al., 2014). Surfaces can also be passivated by precipitating secondary minerals which limit diffusion of reactants and products (King et al., 2010). Agitation and sonication have been employed to reduce the impact of surface passivation (Santos and Van Gerven, 2011). Organisms can also prevent secondary mineral precipitation by secreting organic chelators (Liermann et al., 2000; Buss et al., 2007; Torres et al., 2019).

**Figure 2** shows the CO<sub>2</sub> removal potential *via* enhanced weathering (mineral dissolution) for the most relevant alkaline minerals contained within various types of mine tailings over a 50-year period (Bullock et al., 2022). The removal potential is determined according to weathering (Equation 6) *via* a shrinking core model under two conditions: “unimproved” (pH of 6–8, and common grain sizes for these materials, e.g., 75 μm for platinum group metal (PGM) tailings) and “improved” (pH of 3–4, and grain sizes of 10 μm for all

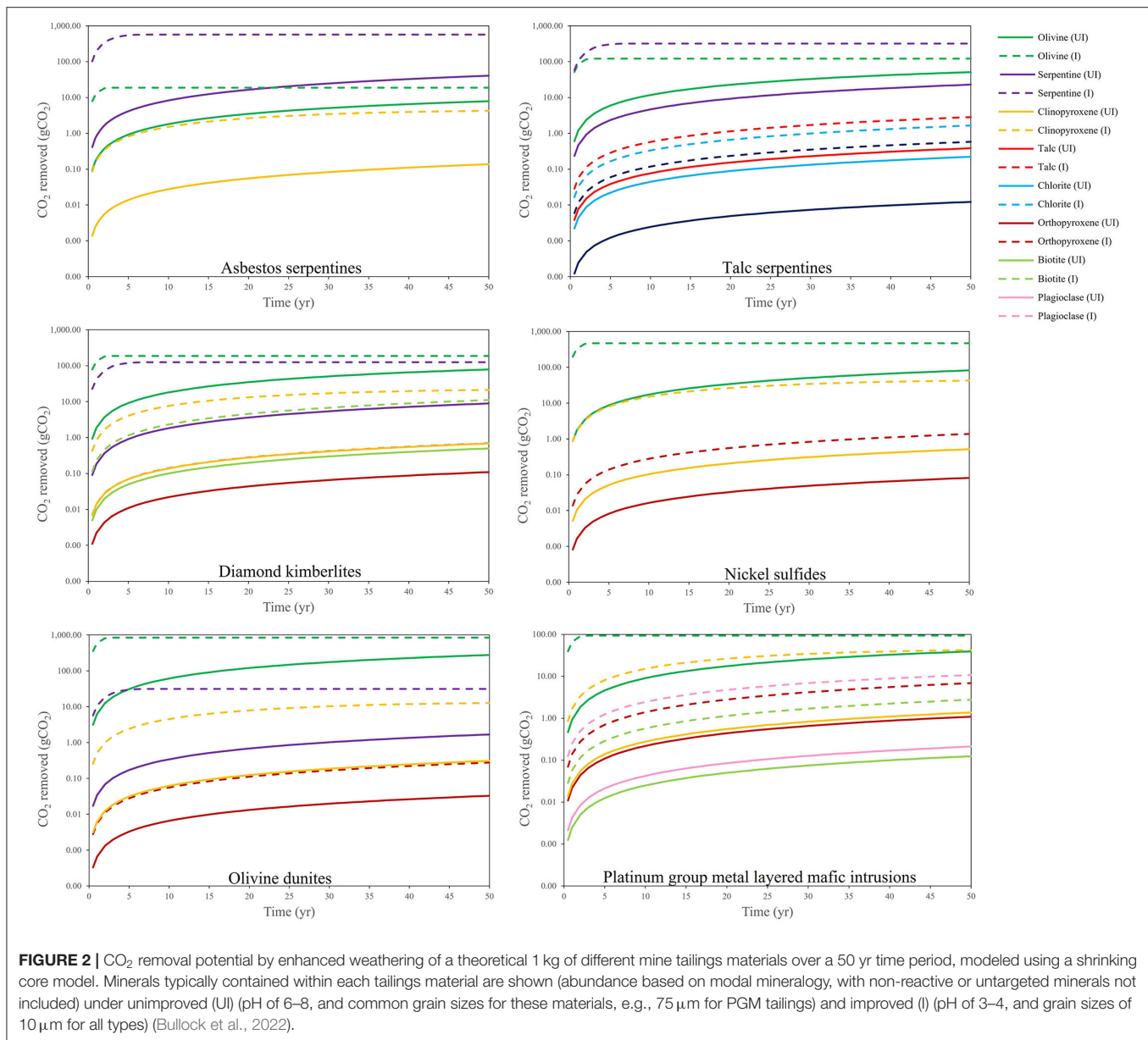
types). Tailings containing high abundances of olivine, serpentine and clinopyroxene show the highest CO<sub>2</sub> removal potential due to their favorable kinetics. The rates of CO<sub>2</sub> removal are estimated to become substantially augmented using improved conditions. Specifically, Bullock et al. (2022) estimate that the average annual global CO<sub>2</sub> removal potential of tailings weathered over 2030–2100 to be ~93 (unimproved conditions) to 465 (improved conditions) Mt CO<sub>2</sub> yr.<sup>−1</sup>. These data clearly demonstrate the enormous impact that enhancing the reaction kinetics can have on the CO<sub>2</sub> removal potential of alkaline materials.

## CO<sub>2</sub> Dissolution and Hydration

In low and neutral pH aqueous solutions CO<sub>2</sub> will react with water and form carbonic acid, H<sub>2</sub>CO<sub>3</sub>, with deprotonation to form bicarbonate, HCO<sub>3</sub><sup>−</sup>, and carbonate, CO<sub>3</sub><sup>2−</sup> (Equations 3–5) (Knoche, 1980). As the pH increases, the equilibrium shifts further to the right, increasing the concentrations of HCO<sub>3</sub><sup>−</sup> and CO<sub>3</sub><sup>2−</sup>. At higher pH values (>8) the CO<sub>2</sub> reaction mechanism changes, with HCO<sub>3</sub><sup>−</sup> forming directly *via* the much faster reaction (Morel and Hering, 1993):



This behavior is the essence behind many geochemical NETs which use dissolved mineral alkalinity as the driving force for CO<sub>2</sub> capture and sequestration. However, in some geochemical NETs, mineral dissolution is not the limiting factor, but rather they are constrained by the CO<sub>2</sub> availability, particularly where atmospheric air is the CO<sub>2</sub>-bearing gas (Power et al., 2013b; Gras et al., 2017). According to Henry's Law, doubling the CO<sub>2</sub> partial pressure approximately doubles the CO<sub>2</sub> solubility (Henry and Banks, 1803). Increasing CO<sub>2</sub> partial pressure can be achieved by increasing the total pressure or by increasing the gas phase CO<sub>2</sub> concentration (O'Connor et al., 2000, 2001). In some geochemical NETs, CO<sub>2</sub> is artificially pre-concentrated by DAC or bioenergy with carbon capture and storage (BECCS) (Kelemen et al., 2020). Others exploit the Earth's natural mechanisms for pre-concentrating CO<sub>2</sub>. For example, in enhanced weathering in soils, the weathering rate of the applied alkaline minerals is accelerated due to elevated concentrations of CO<sub>2</sub> in the soil pores, which traces back to microbial/plant respiration (Robbins, 1986), while some ocean NETs may take advantage of the higher carbon concentration by volume of seawater compared to air (de Lannoy et al., 2018). Furthermore, diffusion of CO<sub>2</sub> into the aqueous phase can be artificially accelerated through bubbling (Legendre and Zevenhoven, 2017; Abe et al., 2021), stirring (Gadikota, 2020), spraying solution in scrubbing towers (Gunnarsson et al., 2018), spraying fluids rich in mineral dissolution products (Stolaroff et al., 2005), sparging (Kelemen et al., 2020), or by using thin films of alkaline solution trickled over high surface area packing materials (Keith et al., 2018). The rate of hydration of CO<sub>2</sub> into carbonic acid can be catalyzed by the enzyme carbonic anhydrase (CA) (Lindskog, 1997) (section The Influence of Organisms and Biological Mechanisms on the Chemical Reactions Underlying Geochemical NETs). In coastal enhanced weathering, the CO<sub>2</sub> concentration from surface to

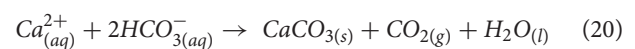


seabed is constantly resupplied since the shallow, high-energy coasts enable rapid sea-air mixing and equilibration. Other factors might also influence CO<sub>2</sub> dissolution rates. For example, CO<sub>2</sub> solubility in aqueous solutions decreases with increasing temperature, as related *via* the temperature dependence of Henry's coefficient (Carroll et al., 1991), and also with increasing salinity due to the “salting out effect” (Setschenow, 1889; Yasunishi and Yoshida, 1979).

### Carbonate Precipitation

Most geochemical NETs require production of dry solid carbonate minerals, and in some of these approaches carbonate precipitation is the rate-limiting step. For example, in OAE, precipitation of carbonate minerals reduces the efficiency of the

overall sequestration by release of CO<sub>2</sub>:



In general, precipitation occurs when the aqueous medium is oversaturated with respect to the mineral that precipitates, i.e., the ionic activity product is higher than the equilibrium constant; whereas dissolution occurs when the aqueous medium is undersaturated with respect to these minerals (Brantley, 2008). For calcium carbonate precipitation (Equation 7), the stoichiometric solubility product,  $K_{sp}^*$ , is defined by:

$$K_{sp}^* = [\text{Ca}^{2+}]_{sat} \times [\text{CO}_3^{2-}]_{sat} \quad (21)$$



where  $[Ca^{2+}]_{sat}$  and  $[CO_3^{2-}]_{sat}$  are the equilibrium concentrations of each species in a solution saturated with  $CaCO_3$  (at a specific temperature, pressure, and salinity). The saturation state,  $\Omega$ , is then defined as:

$$\Omega = [Ca^{2+}][CO_3^{2-}]/K_{sp}^* \quad (22)$$

where  $[Ca^{2+}]$  and  $[CO_3^{2-}]$  are the concentrations of each species in solution. If  $\Omega = 1$ , the solution is at equilibrium with the mineral phase. If  $\Omega < 1$ , the aqueous phase is undersaturated, and calcium carbonate is expected to dissolve, whereas if  $\Omega > 1$  then the aqueous phase is oversaturated, and calcium carbonate is expected to precipitate. However, even when saturated, carbonate minerals may not always precipitate. For example, the presence of  $Mg_{(aq)}^{2+}$  is known to inhibit calcium carbonate formation (Morse et al., 2007). Similarly, organic and inorganic ligands present in the aqueous phase can inhibit calcium carbonate formation *via* complexation and adsorption (Morse et al., 2007), although some enhance the precipitation rates by accelerating desolvation kinetics (Schott et al., 2009). The rate of carbonate precipitation increases with increasing pH, owing to the shift of aqueous equilibrium toward  $CO_3^{2-}$  (Ruiz-Agudo et al., 2011). Higher temperatures and pressures result in greater precipitation of calcium carbonate and (hydrated) magnesium carbonates (Zeebe and Wolf-Gladrow, 2001; Hänchen et al., 2008). In turbulent conditions, eddy formation increases the diffusion rates of species enhancing carbonate precipitation (Dreybrodt et al., 1997). Precipitation of  $CaCO_3$  is  $\sim 4$  orders of magnitude faster than precipitation of  $MgCO_3$ , as  $Ca^{2+}$  is much larger than  $Mg^{2+}$  and the water molecules in its coordination sphere are held more loosely, enabling faster exchange with carbonate (Schott et al., 2009). Notably, microorganisms have been observed to catalyze the nucleation of  $MgCO_3$  (McCutcheon et al., 2019). Microorganisms can catalyze nucleation of carbonate precipitation by concentrating cations near the surfaces of cell walls or extracellular polymeric substances (Dupraz et al., 2009).

## PROCESSES

Processes for geochemical NETs that produce solid carbonate minerals *via*  $CO_2$  mineralization include *in situ*, *ex situ* and surficial approaches. Processes which increase the ocean's storage capacity of dissolved inorganic carbon (DIC) are termed OAE and include coastal enhanced weathering, electrochemical seawater splitting, and ocean liming. Enhanced weathering in soils produces both carbonate minerals (carbonation) and also lead to ocean alkalization since cations from rock dissolution will remain dissolved in water and eventually be transferred to the oceans (Renforth, 2012; Lefebvre et al., 2019). **Table 3** summarizes some of the defining features of each of these processes. Furthermore, geochemical NETs which use alkaline minerals in their flowsheets but which do not necessarily result in carbon storage are introduced in **Box 1**. Note that descriptions of geochemical NETs, e.g.,  $CO_2$  mineralization, enhanced weathering, OAE, etc., in other sources may vary from the ones in this review, and that their defining features may overlap in one or more ways.

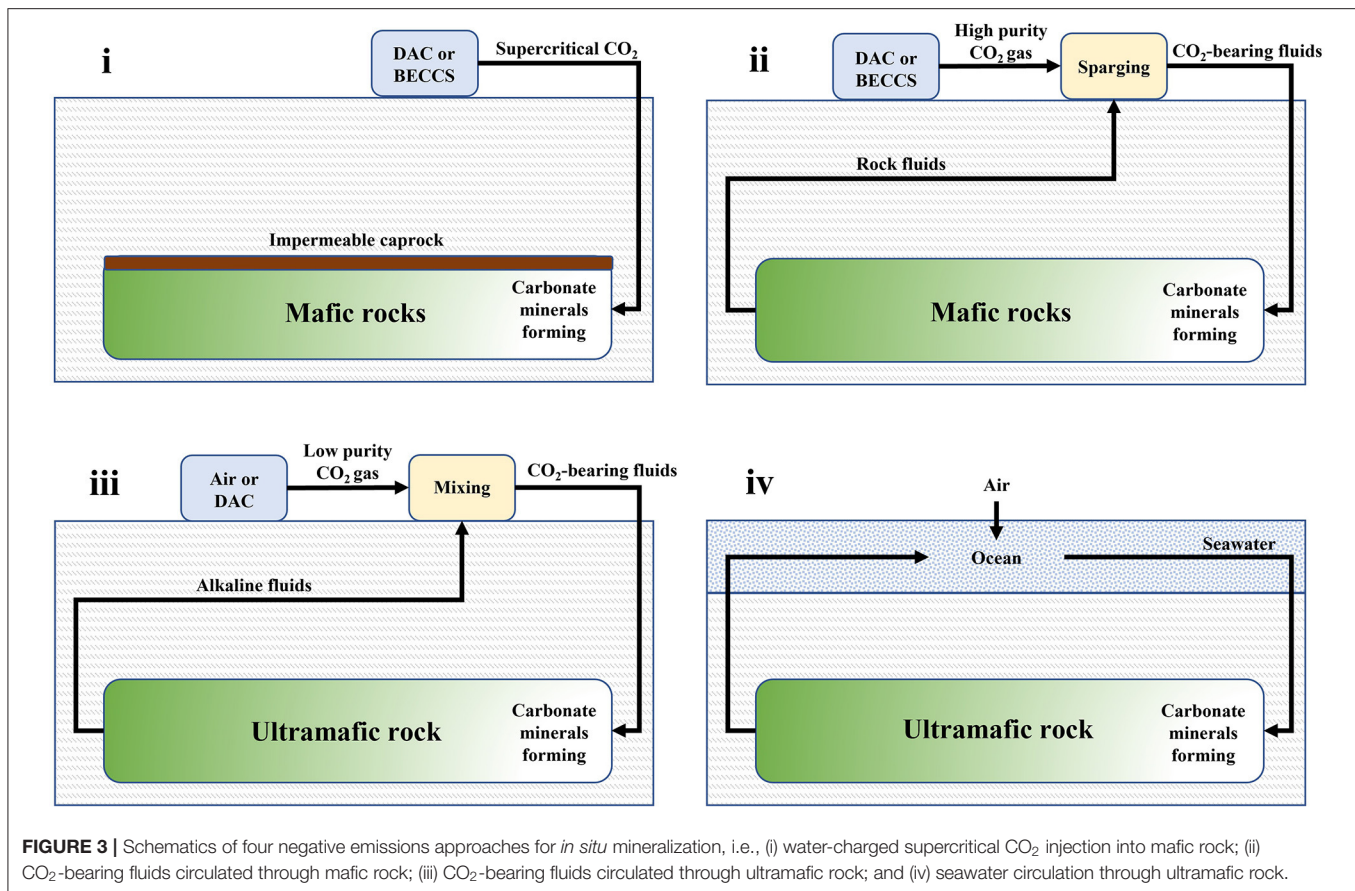
## $CO_2$ Mineralization

### *In situ*

In CCS,  $CO_2$  is typically injected as a pure supercritical fluid into geological formations, such as deep sedimentary formations, salt mines, depleted oil fields, or unmineable coal seams, where it becomes trapped in rock pores and structural spaces, with minimal  $CO_2$  mineralization. An impermeable caprock is needed in order to limit leakage and long-term monitoring is required (Zhang and Song, 2014). In *in situ* mineralization, the focus is on mineral storage, rather than pore and structural storage. This is achieved by injecting supercritical  $CO_2$ , or  $CO_2$ -rich fluids, into alkaline geological rock formations. Once injected, the  $CO_2$  creates a low pH zone within the rock, enhancing dissolution of the surrounding silicate minerals and causing  $Mg^{2+}$  and  $Ca^{2+}$  to be released (Equation 6). As mineral dissolution increases, the pH begins to increase, which in turn induces the precipitation of stable carbonate minerals (Equation 7). With mineralization there is less need for long-term monitoring of the storage efficacy compared to traditional forms of CCS, particularly when  $CO_2$  is pre-dissolved prior to injection. However, pre-dissolution incurs additional cost and complexity compared to injection of pure supercritical  $CO_2$  (Blondes et al., 2019).

As mentioned above, there are two main types of alkaline rock formation suitable for *in situ* mineralization: (i) mafic rocks such as basalt, and (ii) ultramafic rocks such as peridotite and serpentinite. Due to their higher alkalinity, ultramafic rocks have greater potential for  $CO_2$  mineralization per cubic volume of rock, while their exothermic reaction with  $CO_2$  releases larger amounts of heat which is beneficial for the mineralization reaction kinetics (National Academies of Sciences, Engineering, and Medicine, 2019). However, ultramafic rocks are usually found at greater depths, are less porous and permeable, and have a wider range of crystal size than mafic rocks (Kelemen and Matter, 2008). For both, capacity is large (section Naturally Occurring Alkaline Rocks), and these rocks are widely geographically distributed (Pilorgé et al., 2021). For example, extensive reserves of onshore flood basalts exist in the US, India, and Russia. However, most of the potential lies offshore, as the majority of the seafloor is composed of basalt. Although most peridotite is deeply buried, near-surface deposits can be found in locations such as Oman, United Arab Emirates, the Mediterranean, the Pacific Islands and New Zealand.

There are four main approaches to negative emissions *via in situ* mineralization which are based on the rock types and engineering methods employed, with two of them being already demonstrated at the kt  $CO_2$   $yr^{-1}$  removal scale (**Figure 3**). In the first (i) approach,  $CO_2$  is injected into porous mafic rock formations, e.g., basalt, as a supercritical fluid. An impermeable caprock is needed to minimize  $CO_2$  leakages. If the rocks are not already water-saturated, then some water can be co-injected alongside the supercritical  $CO_2$  to facilitate mineralization. This has been successfully demonstrated in 2013 by the Wallula project, whereby 977 tons of water-saturated supercritical  $CO_2$  from the industry were injected into a permeable Columbia River basalt at a depth of 900 m over a 3-week period (McGrail et al., 2011, 2014, 2017a,b; Spane et al., 2012; White et al., 2020; Holliman



et al., 2021). Although CO<sub>2</sub> mineralization was observed in sidewall core samples, evidence suggests that much of the CO<sub>2</sub> remains structurally trapped (White et al., 2020; Holliman et al., 2021).

In the second (ii) approach, an impermeable caprock is not present and thus pre-dissolution of CO<sub>2</sub> in reservoir fluids or seawater is required, prior to injection into porous basalt. Recirculation of the CO<sub>2</sub>-carrier fluids helps maintain a constant rock formation pressure, reducing the chance of seismic activity, as well as enabling monitoring of the extent of mineralization *via* tracers such as <sup>14</sup>C rich CO<sub>2</sub> or Ca isotopes (Matter et al., 2016; Snæbjörnsdóttir et al., 2017; Gíslason et al., 2018; von Strandmann et al., 2019; Clark et al., 2020). This approach was adopted at a geothermal energy plant in Hellisheiði, Iceland (Carbfix project). Specifically, CO<sub>2</sub> (and H<sub>2</sub>S) emitted by the process were pre-dissolved by sparging in water and then co-injected with geothermal brine into highly-fractured basalt at depths of 300–1000 m (Matter et al., 2009; Gíslason et al., 2010; Gutknecht et al., 2018). Each ton of CO<sub>2</sub> required ~25 tons of water. To date, more than 70,000 tons of CO<sub>2</sub> have been injected with an estimated 60% successfully mineralized (Clark et al., 2020). In 2021, the world's first DAC-mineralization plant (Project Orca) went live as a collaboration between CarbFix and the company Climeworks, with the goal to annually remove 4 kt CO<sub>2</sub>. Modeling studies indicate that basalt carbonation may be limited by alkalinity constraints and lead to the existence of

unreacted free-phase CO<sub>2</sub> (Tutolo et al., 2021). However, recent field-scale three-dimensional transport models of the CarbFix injection site indicate mineralization rates remain high even after many years of injection and that 300 Mt CO<sub>2</sub> can be stored using just 10% of the rock pore space (Ratouis et al., 2022). The role that secondary minerals, e.g., clays and zeolites, play in the reactivity of CO<sub>2</sub> with basalt is yet to be fully understood. Furthermore, the evolution of dissolution and precipitation fronts and their effect on rock permeability and fluid flow during the injection period is another phenomenon with great uncertainty (Lisabeth et al., 2017; Peuble et al., 2018).

In the third (iii) approach, alkaline geological fluids are extracted from ultramafic rock formations, such as peridotite, and allowed to absorb CO<sub>2</sub> from air in surface ponds creating DIC. The fluids are then recirculated through the rock where DIC reacts and forms carbonate minerals. This speculative approach is based on natural terrestrial alkaline springs and their associated surface travertine deposits (Kelemen and Matter, 2008; Kelemen et al., 2011; Power et al., 2013b). However, the circulation of fluids with such low concentrations of dissolved carbon could be prohibitively expensive. Therefore, it has been suggested that this approach can be scaled cost effectively by combination with DAC that produces low purity (3–5% wt.) CO<sub>2</sub> (Kelemen et al., 2020). Although the process seems promising, challenges remain. For example, as the fluid pathways become filled with product carbonate minerals the permeability of the rock formation would

reduce, inhibiting CO<sub>2</sub> transport to unreacted rock further from the injection well and causing the system to become self-limiting. On the other hand, volume expansion during carbonation can create enough force to fracture the host rock, maintaining permeability. Such “reaction-driven cracking” could increase permeability, thus enhancing the efficiency of *in situ* mineralization in peridotite (Kelemen and Hirth, 2012; Kelemen et al., 2013; Sohn, 2013; Evans et al., 2020). Overall, the balance between clogging and cracking during *in situ* mineralization in peridotite remains a key uncertainty.

In the fourth (iv) and final approach, seawater is circulated through ultramafic rocks near the oceans (Kelemen and Matter, 2008). Although largely speculative, some natural analogs exist which indicate potential feasibility (Grozeva et al., 2017; Kelemen, 2017; Picazo et al., 2020). This approach simultaneously increases alkalinity of the ocean, while removing DIC in seawater by reaction with peridotite, thus creating a double driving force for CO<sub>2</sub> drawdown from the air into the ocean *via* manipulation of the Revelle factor (Eggleston et al., 2010). Thermal gradients between the rock and seawater could drive natural circulation.

DAC with *in situ* mineralization is energy intensive and can require significant heat for regenerating capture sorbents, in addition to significant power input for CO<sub>2</sub>/air sparging and fluid pumping. In both the CarbFix and Orca projects, the heat and power needs are met by geothermal energy (Marieni et al., 2018; Adams et al., 2020). Geothermal fluids typically have temperatures of 70–250°C (Zarrouk and Moon, 2014) enabling integration with DAC systems whose synthetic sorbents (usually amine-based polymers) are regenerated in a similar range. Tectonically active areas such as the Western United States, Alaska, Hawaii, British Columbia, Indonesia, the Philippines, Italy, Turkey, New Zealand, Japan, Iceland, Kenya, Mexico, El Salvador, and Central America (Zarrouk and Moon, 2014) could provide low-cost opportunities for geothermal powered DAC and mineralization. As new geothermal technologies develop, such opportunities could expand elsewhere (Olasolo et al., 2016). Alternatively, the CO<sub>2</sub> and power requirement for *in situ* mineralization could be simultaneously provided by bioenergy (Turner et al., 2018), which has the advantage of lower leveled cost of CO<sub>2</sub> capture than DAC. However, pipelines would be needed for transportation of CO<sub>2</sub>, and issues with biodiversity, land requirements, sustainability, and scalability could arise (Burns and Nicholson, 2017; Smith et al., 2019).

*In situ* mineralization may have fewer adverse environmental and human health effects than surface-based geochemical NETs, since materials are mostly contained beneath the Earth's surface where they have little direct impact on ecosystems and biodiversity, and typically use less land and fresh water than other NETs (National Academies of Sciences, Engineering, and Medicine, 2019). Wastewater could be co-injected with CO<sub>2</sub> for dual benefit (Phan et al., 2018). *In situ* mineralization could have other potential co-benefits, such as enabling the transition of workers from fossil fuel industries into the clean energy sector where near-identical skills are required. Reaction-driven cracking could be applied to *in situ* mining of metals and uranium (Kelemen et al., 2020). On the other hand, there

are potential risks with the injection of CO<sub>2</sub> and fluids for geological storage, including: (i) production and leakage of methane (CH<sub>4</sub>) and hydrogen sulfide (H<sub>2</sub>S) to the atmosphere by CO<sub>2</sub>-reducing bacteria (Guyot et al., 2011); (ii) groundwater acidification (Li et al., 2018); (iii) heavy metal mobilization, which could contaminate local water supply (de Orte et al., 2014); and (iv) increasing seismicity (Blondes et al., 2019). The latter presented certain key challenges, which were identified during the initial stages of the CarbFix project where several (micro)seismic events were initially observed (Hjörleifsdóttir et al., 2021). Improved engineering methods, i.e., constant recirculation of the fluids, reduced seismic occurrences, while engagement with local residents aided public acceptance of the project. Partnership with Climeworks, as well as the addition of a geothermal lagoon for bathing also aided in improving public acceptance (Aradóttir and Hjálmarsson, 2018).

### Ex situ

*Ex situ* routes were the first approaches for CO<sub>2</sub> mineralization to be investigated for the purpose of climate change mitigation (Lackner et al., 1995), particularly focusing on reducing point source emissions. *Ex situ* mineralization involves reacting high surface area alkaline minerals with CO<sub>2</sub>-rich gases, mainly in engineered reactors (Gerdemann et al., 2007). *Ex situ* approaches using crushed natural rocks rich in minerals such as olivine (Kwon et al., 2011), serpentine (Park and Fan, 2004; Wang and Maroto-Valer, 2011b; Nduagu et al., 2012), and wollastonite (Huijgen et al., 2006; Daval et al., 2009; Xu et al., 2019) have been investigated, but industrial alkaline wastes and by-products, such as mine tailings (Bodénan et al., 2014) or iron and steel slags (Yadav and Mehra, 2017), are likely better suited to *ex situ* processes owing to greater reactivity than their natural counterparts, as discussed in the section Artificial Alkaline Minerals—Industrial by-Products and Wastes, and Tailored Minerals. High temperatures and pressures (Domingo et al., 2006), high CO<sub>2</sub> partial pressures (Li et al., 2019), additives (Krevor and Lackner, 2009), and mechanical (Fabian et al., 2010; Li and Hitch, 2018), or heat activation (Farhang et al., 2019) could be used to capture and store CO<sub>2</sub> within timeframes relevant to industrial processes. Although *ex situ* processes are likely best integrated with readily available sources of concentrated CO<sub>2</sub> from industry, integration with DAC may also be possible. For example, OCO Technology, which makes carbonate construction materials, is now working with London-based Mission Zero Technologies, to use CO<sub>2</sub> sourced by air capture (OCO Technology, 2021).

*Ex situ* processes can be broadly categorized as either “direct” or “indirect.” Direct CO<sub>2</sub> mineralization occurs in one step, as a gas-solid (Kwon et al., 2011; Liu et al., 2018) or as a gas-liquid-solid process (Benhelal et al., 2019; Li et al., 2019). Indirect CO<sub>2</sub> mineralization methods use multiple steps which overall result in the dissolution of a silicate mineral and the creation of a carbonate mineral. First Mg and/or Ca is extracted from the mineral feedstocks, followed by reaction with CO<sub>2</sub>. This is usually achieved by a pH swing approach using reagents, e.g., hydrochloric acid (Lackner et al., 1995; Ferrufino et al., 2018),



acetic acid (Kakizawa et al., 2001), ammonium salts (Wang and Maroto-Valer, 2011a; Highfield et al., 2012), ammonia and brine (based on solvay process) (Huang et al., 2001) or molten salt ( $\text{MgCl}_2 \cdot n\text{H}_2\text{O}$ ) (Wendt et al., 1998), where the acidic reagents aid in mineral dissolution and the alkali reagents aid in carbonate precipitation. Reagents should be recycled as part of the process. Direct processes have the advantage of greater simplicity, whereas indirect approaches have the advantage of faster throughput and the production of high purity carbonate minerals (Zevenhoven et al., 2011). Direct mineralization requires pure  $\text{CO}_2$  for reaction (necessitating integration with either DAC or BECCS), whereas some indirect processes produce reactive alkaline hydroxides that may be suitable for direct reaction with atmospheric  $\text{CO}_2$ .

*Ex situ* approaches can also produce useful carbonated products (Fernández Bertos et al., 2004; Hills et al., 2020; Qiu, 2020). Other valuable side products such as hydrogen could enable *ex situ* processes to become more economical (Kularatne et al., 2018). However, life cycle assessments (LCAs) frequently show that not all *ex situ* approaches result in negative emissions (Ncongwane et al., 2018; Thonemann et al., 2022). Further information on different *ex situ* processes can be found elsewhere (Sanna et al., 2014; Veetil and Hitch, 2020; Yadav and Mehra, 2021).

## Surficial

Surficial  $\text{CO}_2$  mineralization is any process by which low purity  $\text{CO}_2$  (either from the air, or low  $\text{CO}_2$  concentration gases and liquids) is reacted with alkaline materials in piles, fields, pools, or large indoor spaces such as greenhouses. Surficial processes generally require less intensive reaction conditions than *ex situ* processes, with carbon removal occurring over weeks to months, rather than minutes. Surficial approaches allow minerals to be carbonated near to their site of production, thus reducing mineral transportation costs. Like *ex situ* approaches, surficial approaches enable the sale of the carbonated minerals, for example, as aggregates for the building and construction sector (Huntzinger et al., 2009a,b; Liu et al., 2021).

Surficial mineralization of crushed alkaline materials was investigated by Myers and Nakagaki (2020) who proposed a gas-solid method whereby finely crushed materials are spread thinly in vertical tiers in a greenhouse. Solar panels drive fans which continuously supply fresh air over the layers of material and trays of water provide the necessary humidity. This approach suggested the use of a variety of alkaline materials from natural mafic and ultramafic rocks to anthropogenic materials such as slag and lime. Other surficial approaches have investigated carbonation of existing mafic and ultramafic mine tailings (Wilson et al., 2006; Power et al., 2010, 2014, 2020; Mervine et al., 2018; Kelemen et al., 2020) and industrial wastes such as slag (Stolaroff et al., 2005). Artificial materials are usually favored due to their greater reactivity than natural minerals. In general, most industrial wastes and by-products (Table 2) present promising opportunities for surficial mineralization due to their wide availability and relatively low cost (Renforth, 2019).

$\text{CO}_2$  availability is often the limiting factor in ambient weathering of mine tailings and other industrial alkaline wastes

(Wilson et al., 2009; Pullin et al., 2019). Therefore, increasing the  $\text{CO}_2$  supply in surficial processes using DAC to provide low purity  $\text{CO}_2$  could lower overall costs compared to air (Kelemen et al., 2020). While higher purity  $\text{CO}_2$  could theoretically be used, significant losses would occur for systems which are not closed. The availability of humidity in the air could also be a limiting in some cases, with some studies quoting a minimum requirement of 55–60% relative humidity required for the reaction to take place (Erans et al., 2020; Samari et al., 2020).

Surficial mineralization of anthropogenic waste materials may serve a dual purpose of waste management (*via* a reduction in liability associated with hazardous materials) in addition to  $\text{CO}_2$  removal. This may be achievable at greater scale and lower cost than *ex situ* approaches. For example, the building and construction industry is thought to be accountable for 40% of solid waste worldwide (Shan et al., 2017). Stockpiles of steel slag produce highly alkaline leachates ( $\text{pH} > 10$ ) (Yi et al., 2012) that can lead to environmental issues surrounding potential heavy metal mobilization and local pollution (Mayes et al., 2008). Carbonation reduces the pH of these wastes, and reduces the mobility of toxic metals. Similarly, carbonation destroys the hazardous asbestiform aspect of some mine tailings (Bobicki et al., 2012). Likewise, 70 million tons of highly alkaline red mud, a waste product of alumina production, are generated annually. Disposal of this waste is challenging due to aluminum toxicity and leaching of alkalinity into groundwater supplies (Bobicki et al., 2012). Carbonation mitigates these effects (Renforth, 2012) and enables the products to be used as a soil amendment, a reagent for removal of nitrogen and phosphorus from wastewater, a fertilizer additive, brick manufacture, plastic filler, and cement production (Bonenfant et al., 2008).

Most of the work conducted on mineralization processes has focused on *ex situ* approaches where high conversion can be reached rapidly (Sanna et al., 2014). Alternative surficial approaches are emerging which can potentially combine the scalability of enhanced weathering with the ability to produce useful carbonate products, or to remediate hazardous industrial mineral wastes. However, the kinetics of ambient mineralization are poorly understood, and more work is needed.

## Enhanced Weathering in Soils

Enhanced weathering in soils aims to accelerate the natural process of weathering through the spreading of crushed Mg- and Ca-rich silicate rocks in agricultural, urban, and forest soils (Renforth, 2012; Hartmann et al., 2013; Beerling et al., 2020; Haque et al., 2020b). Carbonate rocks, such as limestone or dolomite, could also be used for enhanced weathering in soils, however, (i) they are unlikely to achieve the same spatial flux of alkalinity (Renforth and Campbell, 2021); (ii) they have a lower  $\text{CO}_2$  sequestration potential; and (iii) they deliver fewer co-benefits than silicate rocks (Beerling et al., 2018). Through enhanced weathering in soils, atmospheric  $\text{CO}_2$  is drawn down into the soil, dissolved into porewaters and transformed into bicarbonate ( $\text{HCO}_3^-$ ) and carbonate ( $\text{CO}_3^{2-}$ ) anions. The result of this process includes carbon stored as carbonate minerals in soils, or dissolved bicarbonates and carbonates draining into



surface waters and eventually transported to the ocean where they contribute to ocean alkalinity (Renforth and Campbell, 2021). Depending on the type of mineral used, enhanced weathering in soils has the potential to remove between 0.3 and 1.25 tons of atmospheric CO<sub>2</sub> per ton of mineral dissolved (Renforth, 2012; Moosdorf et al., 2014; Haque et al., 2019), although the maximum scalable potential when using rocks is <1. Cost estimates for enhanced weathering in soils vary by country ranging from US\$55–190 per ton of CO<sub>2</sub> removed, with estimates for China, India, Indonesia or Brazil at the lower end, and USA, Canada and European countries at the higher end of the cost range (Beerling et al., 2020).

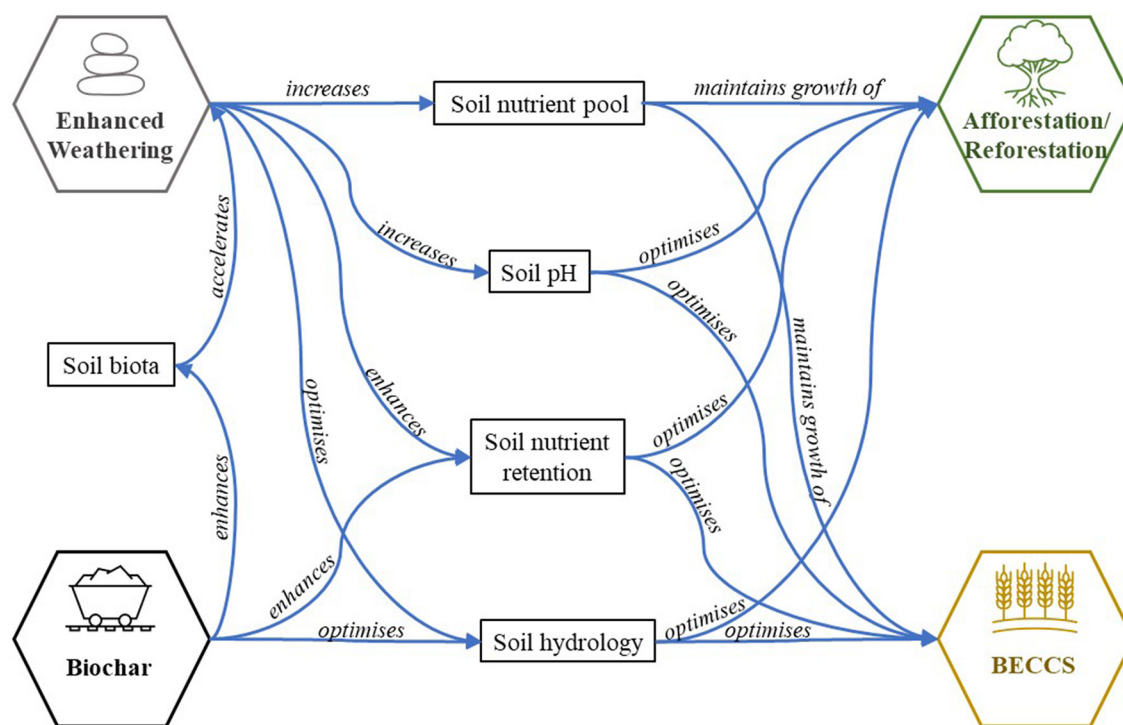
The weathering rate of silicate minerals depends on several abiotic and biotic factors. Specifically, it increases with increasing surface area (Steffler et al., 2018), while higher pH, lower temperature, and precipitation rates, along with varying soil CO<sub>2</sub> partial pressure can negatively affect the weathering rates (Verbruggen et al., 2021). Biogeochemical and biomechanical activity can also affect weathering rates in soils (Vicca et al., 2022). Plants may enhance silicate mineral weathering in soils through their roots and associated mycorrhizal fungi, *via* diverse mechanisms such as the release of organic acids (Taylor et al., 2009; Thorley et al., 2015; Verbruggen et al., 2021) and secretion of acids or stimulation of acid-generating nitrification by nitrogen-fixing plants (Bolan et al., 1991; Epihov et al., 2017; Perakis and Pett-Ridge, 2019). Invertebrates in soil also contribute to weathering, both chemically, through the action of gut microbiota, and mechanically by biopedturbation (Van Groenigen et al., 2019; Vicca et al., 2022).

Besides capturing CO<sub>2</sub>, enhanced weathering in soils also presents potential associated benefits. For example, soil pH is increased by alkalinity fluxes, eroded soils are replenished in the long-term with macro (e.g., Mg, Ca, K, P, and S) and micronutrients (e.g., B, Mo, Cu, Fe, Mn, Zn, and Ni) (Leonardos et al., 1987; Hartmann et al., 2013; Anda et al., 2015), while plant resilience to biotic and abiotic stress improves (Beerling et al., 2018). Particularly in agricultural land, enhanced weathering in soils could help revert agricultural soil erosion, act as a liming agent, and help reduce the use of fertilizers and pesticides (Kantola et al., 2017; Beerling et al., 2018, 2020; Haque et al., 2020b). As croplands cover 10% of the Earth's land surface (Monfreda et al., 2008), there is potential for large-scale application. Additionally, the equipment currently used in farming (e.g., lime spreaders) can be easily adapted to spread ground rocks on agricultural lands. Forested areas also provide opportunity for enhanced weathering in soils. The associated benefits of applying silicate rock dust on soils could promote plant growth and survival rate, through replenishing soil nutrients (Leonardos et al., 1987; Hartmann et al., 2013; Anda et al., 2015) and through increasing plant resilience to external stresses (Beerling et al., 2018). Moreover, mycorrhizal fungi associated with plant roots are one of the main drivers of silicate rock weathering in soils (Taylor et al., 2009; Thorley et al., 2015; Verbruggen et al., 2021). Urban and brownfield soils also present certain advantages for soil enhanced weathering (Manning and Renforth, 2013). In urban soils, enhanced weathering can be integrated in the landscape design to create carbon sinks, such

as “carbon capture gardens,” while brownfields have the potential to develop value by removing CO<sub>2</sub> as part of their remediation process (Manning and Renforth, 2013).

Alongside its many potential co-benefits, enhanced weathering in soils also has several drawbacks and risks, associated with comminution (which is the most energy demanding step of the process) (Renforth, 2012), transportation (as the distance from quarry to field increases, the CO<sub>2</sub>eq emissions increase and by extension the CO<sub>2</sub> sequestration potential declines) (Lefebvre et al., 2019), and pollutants embedded into the mineral matrix being released in the environment through weathering (Haque et al., 2020a). For example, depending on the chemical composition of the parent material, the weathering of silicate minerals such as olivine might release heavy metals including chromium (Cr), nickel (Ni), or other elements [e.g., silicon (Si)], affecting the receiving ecosystems. Particularly in croplands, if these elements are present in concentrations higher than recommended by soil quality guidelines, they could be incorporated in the food chain, acting as pollutants and also affecting human and environmental health (ten Berge et al., 2012; Haque et al., 2020a). Plants with metal-accumulating mechanisms have been proposed as a strategy for preventing contamination of soils and water with toxic metals during enhanced weathering (Suhrhoff, 2022). There are also concerns about the effect of alkalinity addition to freshwater ecosystems, which have been shown to be sensitive to pH changes (Morgan, 1987; Wyatt and Stevenson, 2010; Pulido et al., 2012). Furthermore, the mining of natural rocks can have an ecological impact on wildlife, and/or require deforestation (Edwards et al., 2017). To reduce the need for mining and lower the overall cost, silicate-rich non-hazardous by-products or wastes from industrial processes, such as iron and steel slag (Das et al., 2019) or cement kiln dust (Beerling et al., 2020), can be used. Nevertheless, long-term comprehensive field studies across different climates and soil types are required to assess the suitability of spreading industrial alkaline wastes or by-products in soils (Beerling et al., 2020).

Finally, enhanced weathering in soils could be combined with other NETs, such as afforestation or reforestation, or with the feedstock crops used in BECCS and biochar. A combination of different approaches implemented together has a greater potential of achieving the CO<sub>2</sub> removal capacity that is needed (Minx et al., 2018) and reducing operational costs (Beerling et al., 2018). Additionally, combining enhanced weathering in soils with afforestation or reforestation and biochar could also lead to synergies, both through negating one another's potential negative impacts, and through increasing the carbon capture uptake (Amann and Hartmann, 2019). **Figure 4** shows the synergistic effects of combining enhanced weathering in soils, BECCS, afforestation/reforestation, and biochar. One proposed co-deployment scenario is the combination of enhanced weathering and biochar onto land used to grow crops for BECCS. In this scenario, the crushed rocks would act as a source of micro- and macronutrients (Leonardos et al., 1987; Hartmann et al., 2013; Anda et al., 2015), while biochar would increase nutrient release (Atkinson et al., 2010) and crop productivity (Jeffery et al., 2011; Kantola et al., 2017).



**FIGURE 4 |** Summary of effects from combining terrestrial NETs: enhanced weathering, biochar, afforestation/reforestation and bioenergy with carbon capture and storage (BECCS). Adapted from Amann and Hartmann (2019).

## Ocean Alkalinity Enhancement

Seawater covers the majority of the Earth's surface (Shiklomanov, 1993) and is a large natural reservoir of carbon. Specifically, DIC in ocean water, which is dissolved  $\text{CO}_2$  gas and bicarbonate and carbonate ions, is 140 times higher, by volume, than gaseous carbon in the atmosphere (de Lannoy et al., 2018). An equilibrium between atmospheric  $\text{CO}_2$  and surface ocean waters exists, described by the Revelle factor, which accounts for  $\sim 25\%$  of anthropogenic (surplus)  $\text{CO}_2$  emissions already absorbed (Watson et al., 2020). The main mechanism for  $\text{CO}_2$  mineralization in the oceans is its dissolution in seawater, since when  $\text{CO}_2$  reacts with water ( $\text{H}_2\text{O}$ ) it forms carbonic acid ( $\text{H}_2\text{CO}_3$ ), bicarbonate ( $\text{HCO}_3^-$ ) and acidity ( $\text{H}_3\text{O}^+$ ), the latter mainly neutralized by carbonate ions ( $\text{CO}_3^{2-}$ ) forming again bicarbonate, but also contributing to acidification (Egleston et al., 2010). As a result, the oceans are annually sequestering  $\sim 0.5$  Gt  $\text{CO}_2$  from the atmosphere (Renforth and Henderson, 2017). However, the capacity of the oceans for  $\text{CO}_2$  uptake is not infinite and cannot cope with the increasing anthropogenic  $\text{CO}_2$  emissions. Furthermore,  $\text{CO}_2$  is acidic and therefore its uptake by surface ocean waters comes at the expense of their natural alkalinity, i.e., their mean pH has dropped by  $\sim 0.1$  units over the past two centuries from an initial average of  $\sim 8.2$  (Caldeira et al., 2005), and is projected to further decline by  $\sim 0.3$  by 2100 (Figuerola et al., 2021). Note that pH scale is logarithmic and therefore the  $\sim 0.1$  drop corresponds to around a 26% reduction of the surface ocean waters' mean alkalinity.

Research has previously explored injection of  $\text{CO}_2$  into the deep ocean (Caldeira et al., 2005), however, the possibility of storing neutralized  $\text{CO}_2$  as increased alkalinity is gaining increasing attention (Renforth and Henderson, 2017), along with the removal of DIC from seawater. Specifically, two main ocean-based NETs include: (i) removing carbon (DIC) from seawater, similar to DAC, using electrochemical processes (de Lannoy et al., 2018; Eisaman et al., 2018; La Plante et al., 2021a); and (ii) artificially increasing ocean alkalinity for enhancing seawater's carbon sequestration capacity (Renforth and Henderson, 2017), a process popularly known as ocean alkalinity enhancement (OAE) (Bach et al., 2019; Gagern et al., 2019). For the first pathway, given that the majority of carbon in seawater is in the form of carbonate and bicarbonate ions,  $\text{CO}_2$  extraction processes must "swing" alkalinity to extract as much DIC as possible. DIC-depleted seawater will then return to the oceans, where atmospheric  $\text{CO}_2$  will be taken up to maintain the air-ocean equilibrium. Therefore, ocean water can be used for both carbon capture and/or storage. Other  $\text{CO}_2$  storage pathways exist, however these are not long-term DIC storage, but rather "ephemeral" storage up to 1,000 years of pure phase  $\text{CO}_2$ , such as dry ice torpedoes where  $\text{CO}_2$  is released from the sea surface with the goal being to penetrate the sea floor (Herzog and Drake, 1996), and intentional storage of  $\text{CO}_2$  in deep ocean, as solid  $\text{CO}_2$  hydrate, possibly in depths higher than 3,000 m, where  $\text{CO}_2$  is denser than seawater and long-term storage is promoted (Caldeira et al., 2005). However, these storage pathways have been associated with environmental impacts on deep ocean ecosystems (Caldeira et al., 2005), while,

even in the deep ocean solid CO<sub>2</sub> hydrate will slowly decompose (GHG, 2004).

Electrochemical processes aim at controlling seawater's pH and trigger the removal of DIC (Sharifian et al., 2021). In the context of geochemical NETs, an acid (HCl) and an alkali (NaOH) are first produced, by seawater/brine electrolysis or electrodialysis, which can be used for two different distinct processes, i.e., the “acid” and the “base” process, respectively. In the acid process, HCl is used to acidify (to pH ~4) degassed (O<sub>2</sub> and N<sub>2</sub> have been removed) seawater, thus converting DIC to CO<sub>2</sub> gas, which is then captured (e.g., using hollow fiber membrane contactors). In the base process NaOH is directly added to seawater to raise the pH (>9) and precipitate carbonates in the form of CaCO<sub>3</sub> (de Lannoy et al., 2018). Subsequently, the natural alkalinity is restored in the decarbonized seawater of both processes, which is then returned to the ocean to recapture CO<sub>2</sub> and store it as bicarbonate (Eisaman et al., 2018). It could be possible to directly release the elevated-pH seawater from the base process to the ocean, since this will lead to additional atmospheric CO<sub>2</sub> uptake due to ocean alkalinity enhancement. Even though only the base process comprises a direct mineralization pathway (CaCO<sub>3</sub> is the final product), the gaseous CO<sub>2</sub> acid process can also be used to produce carbonate minerals. Single-step carbon sequestration and storage is similar to the base process since carbonate minerals are electrochemically (flow-through membraneless electrolysis) formed from DIC and divalent cations (Ca and Mg) that are naturally contained in seawater, however, in this case, without the need of fine-pore membranes but rather coarse-mesh electrodes (La Plante et al., 2021b). The main challenge in electrochemical processes pertains to high electricity and raw material (mainly electrodes or membranes) inputs (Sharifian et al., 2021), which raise costs and possibly environmental impacts.

In OAE alkaline materials, such as natural or artificial minerals and industrial waste/by-products (Renforth, 2019), are used to increase the oceans' alkalinity, thereby adding to its natural capacity as a carbon sink, i.e., CO<sub>2</sub> is permanently (>10,000 years) stored as aqueous bicarbonate ions (Renforth and Henderson, 2017). Three main methods for addition of alkaline materials to the ocean have been proposed: (i) coastal enhanced weathering (Meysman and Montserrat, 2017; Renforth and Campbell, 2021), whereby crushed or pulverized silicate minerals are spread onto beaches and coastal shelves and waves and currents promote their dissolution and the release of alkalinity into the ocean; (ii) ocean liming, whereby lime is spread on the open ocean for artificial alkalinity enhancement (Paquay and Zeebe, 2013; Renforth et al., 2013); and (iii) electrochemical approaches, which do not aim to remove DIC from seawater, but produce alkaline agents that can be used for OAE (House et al., 2009). Wastes from different industrial processes could also be used for OAE at scale (Renforth, 2019). However, alkaline wastes can be enriched with harmful pollutants, such as trace (heavy) metals (Gomes et al., 2016), and therefore, more likely, might find limited application for OAE, at least in the near future.

For silicate minerals to be quickly dissolved in the upper ocean, pulverization (<1 μm) is required (Meysman and Montserrat, 2017), which is energy intensive and not practical

(Hangx and Spiers, 2009). On the other hand, it can take up to thousands of years for olivine sand to fully dissolve in coastal environments (Hangx and Spiers, 2009), but high-energy shallow marine environments (coastal shelves and beaches) can greatly accelerate olivine dissolution (Schuiling and de Boer, 2011). Specifically, natural forces acting on the coastal environments, such as waves and currents, can be used to enhance the dissolution rates of silicate minerals (Montserrat et al., 2017). Among the industrially mined silicate minerals, olivine has been found promising for coastal enhanced weathering, since it is abundant and readily available (olivine mines can be found across the world), while its dissolution rate is significantly higher (three orders of magnitude) compared to ordinary quartz (Meysman and Montserrat, 2017). Similar to enhanced weathering in soils, coastal enhanced weathering can capture 0.28 g C per g olivine dissolved in ocean water (Köhler et al., 2013). On the other hand, olivine also contains various nutrients (mainly silicic acid and iron) that improve phytoplankton growth (fertilization), leading to a maximum carbon capture capacity of up to 0.57 g C per g of olivine spread (Hauck et al., 2016). Nonetheless, among others, silicic acid can alter marine biology, shifting phytoplankton species composition toward silicifiers such as diatoms (Köhler et al., 2013), while phytoplankton fertilization cannot be considered as a direct mineralization pathway. Silicifier growth can also turn the color of seawater to greener shades (Bach et al., 2019). Overall, coastal enhanced weathering appears promising, however, many open questions exist, including non-stoichiometric dissolution, pore-water saturation in the seabed, possible secondary reactions, and the exact ecosystem and feedback effects of large-scale olivine dissolution (Montserrat et al., 2017).

The surface ocean waters are supersaturated with respect to calcite (CaCO<sub>3</sub>), i.e., limestone, and therefore its “activation,” typically through calcination prior to its spreading for OAE, has been proposed (Kheshgi, 1995). As such, ocean liming is typically achieved using artificial alkaline minerals such as calcium oxide (CaO) or more likely calcium hydroxide (Ca(OH)<sub>2</sub>) (Caserini et al., 2021), which will rapidly dissolve and release alkalinity in seawater (Justnes et al., 2020). Specifically, limestone is mined, crushed, washed, milled, and then calcined, before being transported and dispersed into surface ocean waters for alkalinity enhancement. It has been reported that 1.4–1.7 tons of limestone can uptake 1 ton of CO<sub>2</sub> from the atmosphere (Renforth et al., 2013). Since (bi)carbonate ions are added to seawater, ocean liming can benefit calcifiers such as corals (Comeau et al., 2013; Feng et al., 2016), coccolithophores, foraminiferans, and pteropods (Figueroa et al., 2021) which are threatened by ocean acidification (Doney et al., 2020). The rapid growth of calcifiers such as coccolithophores can give a bluer or whiter shade to the ocean color (Bach et al., 2019). Ocean liming faces several engineering challenges (Renforth et al., 2013), particularly in capturing and storing the carbon emissions from limestone decomposition (Paquay and Zeebe, 2013) and ocean spreading (Caserini et al., 2021), along with uncertainties pertaining to the responses of marine organisms to large-scale lime addition (Paquay and Zeebe, 2013; Das and Mangwani, 2015). Furthermore, ocean liming appears to



allow calcifiers to outcompete silicifiers, while coastal enhanced weathering allows for the opposite (Moore, 2021). This implies that these NETs can be used in tandem to maximize the uptake of atmospheric CO<sub>2</sub> and maintain the balance between different marine (micro)organisms.

Finally, it is possible to use electrochemistry to remove acidity, in the form of HCl, from seawater or from brines [e.g., from the desalination industry (Mustafa et al., 2020)] and store or neutralize this acidity (e.g., using silicate minerals) and return alkalinity (NaOH is produced along with HCl while excess alkalinity can also be produced from the dissolution of silicate minerals) to the oceans (House et al., 2007; Davies, 2015). Electrolysis or electrodialysis can be used to split seawater/brines and generate acidity and alkalinity; however, the proposed processes are typically energy intensive, with values in the range of 3–18 GJ per t of CO<sub>2</sub> removed from the atmosphere (Renforth and Henderson, 2017). Brines with high salt concentrations can be used to improve the process efficiency in acid (HCl) and alkali (NaOH) production; however, it is unlikely that the existing brines, primarily from the desalination industry, are sufficient for scaling up this process to the gigaton range. Therefore, it is more likely that electrochemical processes for OAE will be used complementary to coastal enhanced weathering and ocean liming, at least until the technology matures and becomes more energy efficient.

## Application of Biotechnology to Geochemical NETs

Organisms are known to control many of the key reactions underlying geochemical NETs (Table 4), and microbial weathering strongly influences geologic cycling (Finlay et al., 2020; Samuels et al., 2020). The organisms, biomolecules and metabolisms underlying biogeochemical activity provide a diversity of mechanisms that can be integrated into geochemical NETs. Further, they can potentially be enhanced through protein engineering, directed evolution, metabolic engineering or other synthetic biology methods. Examples of naturally-existing mechanisms, applications of biotechnology to geochemical NETs to date, and future perspectives are discussed in the following three subsections.

### The Influence of Organisms and Biological Mechanisms on the Chemical Reactions Underlying Geochemical NETs

Many bacteria and fungi promote dissolution of alkaline minerals by altering the chemical microenvironment at the mineral surface, in some cases for liberation of essential nutrients (Rogers and Bennett, 2004). Mechanisms include acidification *via* secretion of protons and weak organic acids, generation of carbonic acid as a byproduct of respiration, production of strong acids by chemolithotrophs, and production of polymeric and small molecule organic acids that act as chelators that catalyze mineral dissolution (Barker et al., 1997; Drever and Stillings, 1997; Nordstrom and Southam, 1997; Bennett et al., 2001; Lazo et al., 2017; Pokharel et al., 2019; Gerrits et al., 2021). Fungi can also increase the surface area of rocks by exerting mechanical forces that induce cracking (Bechinger

et al., 1999). Lichen (a mutualism between a fungus and a cyanobacterium) are particularly active in silicate mineral weathering, through penetration of hyphae and thalli, secretion of organic acids, and provision of dissolved carbonate and acidification through respiration (Chen et al., 2000). Rates of secretion of weathering agents by fungi and bacteria are reported to vary by mineral substrate and nutrient availability (Bennett et al., 2001; Schmalenberger et al., 2015), indicating that weathering can occur by mechanisms that are actively regulated by organisms. Organisms can also facilitate alkaline mineral dissolution by inhibiting the formation of passivating iron oxide layers; one such mechanism is secretion of small-molecule chelators known as siderophores (Liermann et al., 2000; Buss et al., 2007; Ahmed and Holmström, 2014; Torres et al., 2019). There has been some disagreement over the extent to which microbes and organic acid chelators catalyze mineral dissolution, and the range of minerals on which they act (Pokrovsky et al., 2009, 2021; Oelkers et al., 2015), indicating the need for further discourse and research to obtain clarity on the contexts in which microbial weathering occurs. Microbial biofilms have also been observed to reduce mineral dissolution rates in some contexts by inhibiting the exchange of ions with the bulk solution (Ullman et al., 1996; Lüttge and Conrad, 2004). Plant roots can also secrete organic acids that act as catalytic chelators (Ryan et al., 2001), and also support fungal and bacterial communities with weathering activity (Kang et al., 2017; Ribeiro et al., 2020; Verbruggen et al., 2021), which together contribute to weathering within the rhizosphere.

Microbially-mediated precipitation of carbonates is also widespread (Zhu and Ditttrich, 2016; Görgen et al., 2020), and its mechanisms are more thoroughly characterized than those of silicate dissolution. Many organisms promote carbonate precipitation by generating alkalinity or increasing ionic saturation state through metabolic activity. Photosynthetic organisms, which are thought to have influenced the majority of calcium carbonate formation through Earth's history (Altermann et al., 2006; Riding, 2006), promote carbonate precipitation by increasing pH through conversion of HCO<sub>3</sub><sup>-</sup> to CO<sub>2</sub> and OH<sup>-</sup>, after which CO<sub>2</sub> is assimilated into biomass and OH<sup>-</sup> is released (Power et al., 2007; Kamennaya et al., 2012). Ureolytic bacteria increase pH by producing NH<sub>4</sub><sup>+</sup>, OH<sup>-</sup> and CO<sub>3</sub><sup>2-</sup> through hydrolysis of urea (Zhu and Ditttrich, 2016). Microbial degradation of amino acids as an energy source, called ammonification, also produces NH<sub>4</sub><sup>+</sup>, OH<sup>-</sup> and CO<sub>3</sub><sup>2-</sup> (González-Muñoz et al., 2010). Anaerobic oxidation of organic matter *via* reduction of nitrates by denitrifying bacteria, or reduction of sulfates by sulfate-reducing bacteria, produces alkalinity *via* consumption of protons and thereby also favors carbonate precipitation (Baumgartner et al., 2006; Martin et al., 2013). Oxalotrophic bacteria also promote carbonate precipitation by liberating both CO<sub>3</sub><sup>2-</sup> and Ca<sup>2+</sup>, along with CO<sub>2</sub>, through metabolism of solid calcium oxalate, which is abundant in many soils (Cromack et al., 1977; Braissant et al., 2002). Aerobic oxidation of organic matter by heterotrophs can also promote carbonate precipitation by producing CO<sub>2</sub>, when CO<sub>2</sub> is limiting (Dupraz et al., 2009; Sánchez-Román et al., 2011).



**TABLE 4 |** Influence of biological agents on reactions or ions underlying geochemical NETs.

Type	Biological agent	Effects	Process
Organisms	Silicate-weathering microbes (e.g., <i>Knufia petricola</i> )	Acceleration of silicate dissolution, e.g., 7x increase in olivine dissolution by <i>Knufia petricola</i> in benchtop experiment (Pokharel et al., 2019).	Microbes can acidify the mineral surface microenvironment by secreting organic acids or by respiration; secrete small-molecule or polymeric organic acids/chelators that catalyze silicate dissolution; or prevent surface passivation by chelation of iron (Lazo et al., 2017; Torres et al., 2019).
	Strong acid-generating microbes (e.g., <i>Acidithiobacillus</i> )	Acceleration of silicate dissolution, e.g., 39 and 84% of Mg liberated from chrysotile mine tailings in stoichiometric and excess sulphuric acid conditions, in benchtop experiment (McCutcheon et al., 2015)	Chemolithotrophs can produce strong acids through metabolism of minerals, such as production of sulphuric acid by <i>Acidithiobacillus</i> sp. through metabolism of metal sulfides, which readily dissolve alkaline minerals (Nordstrom and Southam, 1997; Power et al., 2010; Schippers et al., 2014).
	Carbonate-precipitating microbes (e.g., cyanobacteria)	Promotion of carbonate precipitation, e.g., 18x increase in carbonate formation from serpentine mine tailings, with cyanobacteria-dominated microbial consortium (McCutcheon et al., 2016).	Phototrophs, ureolytic bacteria, and other microbes can increase pH and generate (bi)carbonate ions through metabolic activity; various microbes, and particularly cyanobacteria, secrete EPS that nucleate carbonate precipitation; and heterotrophs can supply CO <sub>2</sub> by oxidizing organic carbon or metabolizing cation-saturated EPS (Dupraz et al., 2009; Zhu and Ditttrich, 2016).
	Silica-forming microbes (e.g., diatoms)	Sequestration of waste silicic acid, the buildup of which may inhibit mineral carbonation, e.g., diatoms sequestered 87% of silicon in microbial carbonation experiments with synthetic alkaline wastewater (McCutcheon et al., 2019).	Diatoms and siliceous sponges form silica shells or skeletons (Sumper and Kröger, 2004; Müller et al., 2013a). Photosynthetic diatoms are found naturally growing in alkaline mine wastewater.
Enzymes and proteins	Carbonic anhydrase (CA)	Alleviates CO <sub>2</sub> hydration kinetics as a rate-limiting factor	CAs are metalloenzymes that catalyze equilibration between dissolved CO <sub>2</sub> and carbonic acid (Mesbahuddin et al., 2021). Some CAs can also inhibit or nucleate carbonate precipitation depending on ionic conditions (Miyamoto et al., 2005; Rodriguez-Navarro et al., 2019).
	Silicateins and silaffins	Sequestration of waste silicic acid, the buildup of which may inhibit mineral carbonation.	Catalysis of polymerization of silica from silicic acid (Sumper and Kröger, 2004; Müller et al., 2013a).
	Calcareous shell matrix proteins	Various functions, including nucleation and inhibition of carbonate precipitation, mineralization templating, CA activity, concentrating ions.	A complex matrix of proteins and other biomolecules coordinates the formation, morphology and nanoscale structure of shells in mollusks and corals (Falini et al., 2015; Marin, 2020).
	Ion transporters	Manipulation of ion concentrations in microenvironments, including Ca <sup>2+</sup> , Mg <sup>2+</sup> , H <sup>+</sup> , HCO <sub>3</sub> <sup>-</sup> , and H <sub>4</sub> SiO <sub>4</sub>	Passive or active (ATP-dependent) transport of ions to manipulate intra- or extracellular concentrations, to provide input ions for photosynthesis, calcareous shell formation, siliceous shell formation, or other purposes (Martin-Jezequel et al., 2000; Dominguez, 2004; Maguire, 2006; Buch-Pedersen et al., 2009; Reinfelder, 2011; Knight et al., 2016).
Non-enzymatic biocatalysts	Extracellular polymeric substances (EPS)	Promotion of carbonate precipitation; catalysis or inhibition of silicate dissolution	Negatively charged EPS adsorbs and concentrates cations, which nucleate carbonate precipitation, and digestion of EPS by heterotrophs liberates bound cations and produces CO <sub>2</sub> to raise the saturation state (Dupraz et al., 2009). Some EPS can catalyze silicate dissolution by concentrating protons or chelating cations, and others may inhibit silicate dissolution by adsorbing to mineral surfaces (Welch et al., 1999).
	Organic acids and chelators	Catalysis of silicate dissolution, e.g., 10–100-fold acceleration of forsterite dissolution by oxalic and phthalic acids (Oelkers et al., 2018).	Organic acids, which frequently also act as chelators, provide acidity and/or complex and solubilize cations within the mineral crystal framework in a pH-dependent manner (Lazo et al., 2017; Oelkers et al., 2018).
	Siderophores	Inhibition of the formation of passivating iron oxide.	Siderophores are a large class of microbial and plant chelators that bind diverse cations, particularly Fe ions, at high affinity and promote their uptake from the environment (Ahmed and Holmström, 2014).

Carbonate precipitation can also be influenced through biophysical mechanisms. Some extracellular materials secreted by microbes, broadly termed extracellular polymeric substances

(EPS), contain a diversity of negatively charged or chelating chemical moieties that bind cations, and are observed to either inhibit or promote carbonate precipitation in different

contexts. Under conditions of low saturation of bound cations, EPS can inhibit carbonate precipitation by depleting the microenvironment of cations, while under conditions of high saturation, EPS can promote carbonate precipitation by enriching the microenvironment with cations (Dupraz et al., 2009). Degradation of EPS by heterotrophs, which liberates bound cations and (bi)carbonates, can also induce carbonate precipitation (Dupraz et al., 2009). The surfaces of microbial cells themselves, which often bear negatively charged functional groups, can also promote carbonate precipitation by attracting cations (Dupraz et al., 2009). High EPS production in combination with alkalinity generation make cyanobacteria particularly effective at nucleating and precipitating carbonates (De Philippis et al., 2001; Decho et al., 2005; Braissant et al., 2009).

Notably, biological precipitation of magnesite ( $\text{MgCO}_3$ ) has been observed at room temperature (McCutcheon et al., 2019; Zhang et al., 2020), despite being inhibited at temperatures below  $80^\circ\text{C}$  in abiotic conditions due to the high energy of dehydration of aqueous  $\text{Mg}^{2+}$  (Saldi et al., 2009). The proposed mechanism for magnesite precipitation at ambient temperature is dehydration by the negatively charged EPS and cell surfaces (Power et al., 2017). Magnesite precipitation is preferable compared to hydrated magnesium carbonates, since it contains a higher stoichiometric fraction of carbon.

Enzymes and peptides are also known to catalyze reactions relevant to geochemical NETs, including hydration of  $\text{CO}_2$  and carbonate precipitation. Of these,  $\text{CO}_2$  hydration is the best studied and presently the most tractable to engineering. Carbonic anhydrases (CAs) are metalloenzymes that catalyze equilibration between dissolved  $\text{CO}_2$  and carbonic acid (Lindskog, 1997). They are among the most efficient enzymes known in nature, with their rates often being diffusion-limited. They exist as  $\geq 8$  evolutionarily distinct families of CA genes across all domains of life (Mesbahuddin et al., 2021), with diverse enzymatic parameters. CAs are involved in diverse processes, including biomineralization of calcium carbonates (Bertucci et al., 2013; Müller et al., 2013b; Karakostis et al., 2016; Sharker et al., 2021). CAs commonly have additional activity, including inhibition or promotion of carbonate precipitation (Miyamoto et al., 2005). A recent study found that bovine CA directly influences carbonate precipitation mechanisms in multiple ways: it interacts with growing calcium carbonate crystals, thereby modifying their growth and morphology; it can also inhibit precipitation under conditions of low  $\text{CO}_2$ , possibly by stabilizing pre-nucleation ion complexes; and it can undergo conformational changes and oligomerization under high pH or high  $\text{CO}_3^{2-}$  and  $\text{Ca}^{2+}$  concentrations, abrogating anhydrase activity and instead templating nucleation of calcium carbonate (Rodríguez-Navarro et al., 2019). It has been suggested that the above mechanisms of CA are involved in the controlled growth of calcareous structures like shells (Rodríguez-Navarro et al., 2019). Carbonic anhydrase has also been demonstrated to catalyze the dissolution of calcite, possibly by catalyzing the transfer of protons into the mineral lattice during protonation of  $\text{CO}_3^{2-}$  (Dong et al., 2020).

Multicellular and unicellular organisms also demonstrate exquisite control over carbonate precipitation using combinations of the aforementioned mechanisms during

the formation of shells and intracellular carbonate inclusions. Intracellular formation of carbonates in bacteria has been observed, including in environments that are undersaturated with respect to carbonate mineral phases, suggesting that active mechanisms control the ion transport and mineral precipitation, though the precise mechanisms of control are as-yet unclear (Görger et al., 2020). Shells in coccolithophores, mollusks, corals and reptilian and avian eggs are typically  $>95\%$   $\text{CaCO}_3$  by mass, with the remaining organic matter consisting of diverse proteins and polysaccharides functioning to scaffold, nucleate, or remodel calcium carbonate, including intracrystalline proteins (Falini et al., 2015; Taylor et al., 2017; Marin, 2020; Gautron et al., 2021). While the specific biomolecular functions of most shell organic components are unclear, many functions have been identified, including proteins that promote or inhibit calcium carbonate nucleation, as well as CAs (Evans, 2019; Marin, 2020). Interestingly, coccolithophores produce individual calcium carbonate particles known as coccoliths intracellularly within specialized vesicles, after which the particles are secreted onto the cell surface to form a shell (Taylor et al., 2017).

Other enzyme families, especially enriched in ocean-dwelling siliceous organisms, facilitate silica polymerization. Silicateins are enzymes found in sponges that catalyze polymerization of amorphous silica (Müller et al., 2013a), and silaffins are highly post-translationally modified peptides found in diatoms that do the same (Sumper and Kröger, 2004; Lechner and Becker, 2015). Silicase is an enzyme reported to catalyze the dissolution of amorphous silica (Schröer et al., 2003), though we are unaware of other published work in which silicase activity is documented. Reproduction of the original results should be pursued to inspire confidence in their robustness.

Further, active biological transport mechanisms exist for concentrating or depleting  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{H}^+$ ,  $\text{HCO}_3^-$ , and  $\text{H}_4\text{SiO}_4$  within microenvironments (Martin-Jezequel et al., 2000; Dominguez, 2004; Maguire, 2006; Buch-Pedersen et al., 2009; Reinfelder, 2011; Knight et al., 2016). For example, carboxysomes are cyanobacterial organelles within which bicarbonate is actively concentrated and then converted to  $\text{CO}_2$  by CA, thereby providing high levels of  $\text{CO}_2$  to the photosynthetic enzyme RuBisCO (Price et al., 2008).

Some research has investigated the influence of supercritical  $\text{CO}_2$  on microbes, with potential relevance to the use, or passive influence, of microbes on subsurface  $\text{CO}_2$  mineralization for storage. While supercritical  $\text{CO}_2$  generally inhibits microbial growth (Yu and Chen, 2019), multiple studies have found organisms with only mildly inhibited growth under supercritical  $\text{CO}_2$  and have investigated its effects on community structure and geochemistry (Peet et al., 2015; Santillan et al., 2015; Jin and Kirk, 2016; Freedman et al., 2017, 2018; Ham et al., 2017; Li et al., 2017; Boock et al., 2019).

Collectively, the organisms and biological mechanisms discussed in this section constitute a starting point for the integration of biology into alkaline mineral NETs.

### Biologically-Enhanced Geochemical NETs

Biotechnology research has explored applied methods for geochemical NETs to a limited degree. Pioneering examples

to date have focused primarily on enhanced dissolution and carbonation of mine tailings (Power et al., 2010; McCutcheon et al., 2014) at laboratory scale.

Sulfur-oxidizing bacteria have been proposed for use in dissolution of alkaline minerals by catalyzing the production of sulphuric acid from sulfides, e.g., sulphidic mine tailings (Power et al., 2010). Similar approaches are used for biomining of copper and other metals (Schipper et al., 2014). Laboratory optimization experiments found that such an approach liberated 39% of Mg from chrysotile mine tailings while maintaining leachate pH suitable for carbonate precipitation, and up to 84% of Mg in acidic leachate (McCutcheon et al., 2015). The cost of transporting acid-generating feedstocks, which are consumed stoichiometrically as silicates dissolve, may be strongly influenced by relative geographic location of tailings and feedstocks (Power et al., 2014). Combination of this approach with microbially-mediated carbonate precipitation has been proposed for carbon sequestration in ultramafic mine tailings or other *ex situ* applications (Power et al., 2014).

Cyanobacteria have been proposed as catalysts for precipitation of carbonates in alkaline mineral NETs (Jansson and Northen, 2010). In laboratory experiments, a cyanobacteria-dominated consortium sourced from an alkaline tailings lake was inoculated into columns of acid-leached chrysotile tailings and caused precipitation of a crust of magnesium carbonates containing ~18-fold more carbon than abiotic controls (McCutcheon et al., 2016). Large-scale bioreactor experiments simulating an artificial alkaline wetland with inoculation of a microbial consortium confirmed cyanobacteria-mediated formation of a magnesium carbonate crust, but found that depletion of essential nutrients, including phosphate, in the reaction medium limited microbial growth and EPS production in the majority of the bioreactor, drastically reducing mineral precipitation (McCutcheon et al., 2014). Increased nutrient delivery in a follow-up experiment resulted in biofilm growth that outpaced carbonate precipitation (McCutcheon et al., 2019); these studies indicate that nutrient distribution is an important consideration in scaled deployment. The latter study also found that diatom growth successfully sequestered ~87% of the silicon in the bioreactor, providing a sink for silicon waste produced during alkaline mineral dissolution. A microbial carbonation experiment on site at a tailings pile failed to produce a cemented carbonate-chrysotile crust; the authors hypothesized that accessibility of water and CO<sub>2</sub>, as well as effects of weather, limited microbial growth and carbonate precipitation (McCutcheon et al., 2017). Failure of the latter experiment illustrates the need for increased application-focused research and lends support to the notion that microbial carbonation of tailings may benefit from more controlled environments like bioreactors as compared with open tailings piles.

Oxidation of waste organics to CO<sub>2</sub> in the presence of alkaline minerals has been proposed as a mechanism for promoting carbonation in contexts where CO<sub>2</sub> accessibility is limiting; such an approach sequesters carbon that was fixed into organics, preventing its decay and the subsequent return to the atmosphere (Mitchell et al., 2010; Power et al., 2011, 2014). Heterotrophic bacteria have been shown to promote carbonate precipitation

*via* oxidation of organic matter in laboratory experiments with phototrophic and heterotrophic consortia in mine tailings (Power et al., 2011) and ureolytic microbes in synthetic brine (Mitchell et al., 2010).

These early investigations into microbially-enhanced carbon sequestration in mine tailings illustrate promise for more scaled deployment. Carbon sequestration potential using microbes in mine tailings has been variously estimated at 123 t ha<sup>-1</sup> yr<sup>-1</sup> (McCutcheon et al., 2016) to 222–238 t ha<sup>-1</sup> yr<sup>-1</sup> (McCutcheon et al., 2019) based on rates found in laboratory experiments, or 175 Mt yr<sup>-1</sup> globally if carbonation is taken to completion, which could also be economically viable (Power et al., 2014). Applications using minerals mined for the purpose of weathering would increase this potential drastically. However, field trials and pilot tests, as well as integration and optimization of the combined use of phototrophs, heterotrophs, enzymes and biostimulants will be required to determine the true benefits and costs these approaches can bring to carbon removal and sequestration.

Some authors have proposed the biomass produced during phototroph-mediated carbonate precipitation could be used for production of fuels or other commodity chemicals (Ramanan et al., 2010; Power et al., 2011), providing a second source of economic utility. The biomass could also be fed to heterotrophs, producing carbonic acid and other organic acids that would dissolve alkaline minerals and further precipitate carbonate.

CA has been investigated for applications in carbon capture, utilization and storage (CCUS), primarily in point source CO<sub>2</sub> scrubbers (Alvizo et al., 2014; Bose and Satyanarayana, 2017; Giri et al., 2020; Mesbahuddin et al., 2021). Some research has investigated the use of CA in geochemical NETs. Bovine CA has been shown to efficiently promote carbonation of alkaline minerals by alleviating the hydration of CO<sub>2</sub> as a kinetic barrier, with 240 and 360% increased sequestration rates of carbon in brucite under non-optimized conditions with sparging of 10% CO<sub>2</sub> (Power et al., 2016) or ambient air (Power et al., 2013a), respectively. *E. coli* expressing recombinant CA were also shown to facilitate carbonate precipitation (Kim et al., 2012; Tan et al., 2018). The native CA activity in unicellular algae was found to facilitate carbonate precipitation during sparging of 10% CO<sub>2</sub> in alkaline media while producing algal biomass. These experiments warrant further research into deployment of CA in geochemical NETs.

Ureolytic (Mitchell et al., 2010) and denitrifying (Martin et al., 2013) bacteria have also been explored for their potential to mediate carbonate precipitation for carbon sequestration. However, the utility of these alkalinity-generating heterotrophs is limited by the requirement that the carbon should be delivered as a reduced carbon feedstock (urea in the case of ureolytic), since they only produce enough alkalinity to mineralize the carbonate their metabolism produces (Mitchell et al., 2010). However, they may be useful for sealing pores *via* carbonate precipitation to form caprock for subsurface storage of supercritical CO<sub>2</sub> (Cunningham et al., 2009).

While most engineered biomineralization research has been conducted under ambient pressures, a few studies have been performed at higher pressures more relevant to subsurface

mineralization; however these studies were performed using ureolytic and denitrifying bacteria for the purpose of caprock formation (Martin et al., 2013; Mitchell et al., 2013). Organisms have been found growing as deep as 2.8 km in the subsurface, so the challenge of growth under high pressure is not insurmountable (Chivian et al., 2008; Glombitza et al., 2016).

In addition to sequestering carbon, biomineralization of alkaline minerals can provide a means for bioremediation of toxic metals or other pollutants by locking them within solid carbonate (Gadd, 2010; Zhu and Dittrich, 2016; Krajewska, 2018; Jain and Arnepalli, 2019; Ehrlich et al., 2021). Microbially-mediated carbonation of mine tailings may serve the dual purpose of sequestering carbon and remediating mine sites (McCutcheon et al., 2016).

It has also been proposed that alkaline minerals could be integrated into industrial microbial wastewater treatment and biogas production systems, where they would both neutralize acidity and sequester CO<sub>2</sub> (Lu et al., 2018), with laboratory work showing promising results. In the methods proposed, microbes are not used for direct interaction with the minerals, but rather the minerals are integrated into existing microbial digestion or microbial electrosynthesis processes.

### Toward Further Integration of Biology Into Geochemical NETs

Despite these exciting examples, we posit that the possibility-space of biologically-enhanced geochemical NETs is drastically under-explored given the broad ability of biological mechanisms to influence the key reactions. Organisms or biomolecules could be incorporated into the abiotic geochemical NETs described in the previous sections to improve reaction kinetics or prevent inhibitory passivation.

In environments where the dissolution rate of CO<sub>2</sub> is limiting, CAs could be deployed in solution, immobilized on surfaces or within water-permeable beads (Xu et al., 2021), or produced by organisms engineered to secrete or display it (Zhu et al., 2022). CAs could also be used to catalyze the dissolution of calcite, generating alkalinity (Subhas et al., 2017). Silicase enzymes, if developed, could be useful for promoting the dissolution of alkaline minerals that contain polymerized silica, extending fast dissolution beyond the typically-favored orthosilicates (i.e., extend enhanced weathering beyond olivines to include pyroxenes, feldspars, etc.). Silicasases could also help to dissolve passivation layers of amorphous silica.

Microbes could also be co-injected alongside aqueous or supercritical carbon dioxide to facilitate *in situ* mineralization. Microbes and enzymes could be particularly useful in reactor-based mineral weathering environments, where conditions can be more precisely controlled.

Microbes could also be applied alongside minerals that are distributed for terrestrial or coastal enhanced weathering efforts (Ribeiro et al., 2020). Given that environmental microbes generally function in communities, emerging methods for modification of microbial communities (Lawson et al., 2019) could be developed for use in many of the applications proposed above (e.g., in soil). If non-native or engineered microbes were used, the impact of uncontained release on indigenous ecosystems would need to be carefully considered, though

challenges to stable establishment of non-native microbial communities is more likely to be encountered than unwanted proliferation (Albright et al., 2022).

Given the ability of organisms to alter chemical microenvironments, they may be useful in contexts where different conditions are desired at the mineral surface compared to the bulk solution; for example, acidic conditions could be generated at a mineral surface to enhance dissolution kinetics while more basic conditions are maintained in the bulk to favor carbonate precipitation, or catalysts could be secreted at the mineral surface to avoid costs of chemically altering the bulk. In general, the microbes or biomolecules used could be native or non-native, and unmodified or engineered. Engineered organisms may provide superior performance, as they could be optimized both for the unnatural chemical and physical environments created by geochemical NETs and for their specific roles in technologies; for example, catalysts discovered using molecular engineering methods could be produced metabolically by engineered organisms.

Biological approaches may also enable tuning of the characteristics of the output materials of geochemical NETs for improved utility; for example, carbonate or silica particles could be produced biologically for use as cement additives that may reduce emissions from cement production, or sequester carbon in concrete, while improving concrete performance (Müller et al., 2013a; Gadikota et al., 2015; Show et al., 2015; Singh et al., 2015; Iravani and Varma, 2019). Diatoms could also be used for low-cost production of silica particles, potentially with surface functionalization or enzymatic display, for use in concrete or other applications (Sardo et al., 2021).

Deployment of microbes or enzymes in the context of geochemical NETs may require tolerance of extreme conditions of pH, ionic strength, temperature, and/or pressure. Enzymes may need to be engineered for stability and/or isolated from extremophilic organisms (Packer and Liu, 2015; Mamo and Mattiasson, 2016; Ma et al., 2019; Ren et al., 2019; Yin et al., 2019); such work is already underway for carbonic anhydrase (Mesbahuddin et al., 2021). Deployed organisms, whether engineered or unmodified, will also need to tolerate those conditions; in the case of engineered organisms, this will require the development of synthetic biology methods for organisms that are presently non-standard (Wannier et al., 2020; Filsinger et al., 2021).

Many of the biogeochemical mechanisms discussed in this section have not received much attention from molecular biotechnology. Development of high-throughput platforms for their study and engineering will benefit both their application to NETs and the study of their underlying biology. For example, platforms for highly multiplexed screening, selection, or directed evolution of variants of enzymes or organisms that dissolve silicates or precipitate carbonates would be useful both for discovering natural mechanisms and for engineering. Such platforms are ubiquitous in well-developed medical domains of molecular biotechnology (Packer and Liu, 2015; Zeymer and Hilvert, 2018; Zeng et al., 2020; Madhavan et al., 2021).

Risks of environmental biocontamination should be considered before deployment of organisms in uncontained environments, such as mine tailings ponds or the subsurface.



While engineering organisms for environmental deployment is a new frontier, we note that it is not without precedent, as there are many such research efforts underway (Coleman and Goold, 2019; Jaiswal and Shukla, 2020; Janssen and Stucki, 2020; Rylott and Bruce, 2020; Zhou et al., 2022), including field trials (Carvalho et al., 2015). Advances in biocontainment may provide options for restricting the spread of engineered microbes outside the intended environment (Lee et al., 2018).

Given the apparent potential for biotechnology to contribute to geochemical NETs, we encourage the research and funding communities to more thoroughly investigate possibilities, including designs for possible NETs incorporating biological mechanisms and applied engineering of specific mechanisms, as well as the underlying biogeochemical science. Design and techno-economic considerations should be examined, accounting for the constraints biotechnology could alleviate and introduce, for example, reductions in capital and operating expenditures resulting from improved kinetics and extent of reaction, as well as methods and costs for delivering feedstocks to stimulate organism growth. The diversity of organisms and biomolecules that manipulate key reactions, as well their chemical mechanisms and metabolic and genomic underpinnings, should be more comprehensively examined. New research tools should be developed that will benefit both the fundamental biology and engineering of relevant mechanisms. Although biotechnology applications for geochemical NETs are currently under-developed, they may prove to play an important role with continued dedicated efforts. Possible applications range in complexity, with some accessible today while others will require years or decades of concomitant advances in basic biology, biotechnology, and NETs engineering. The latter group may assist in driving down costs to enable the crucial scaling of NETs by mid-century.

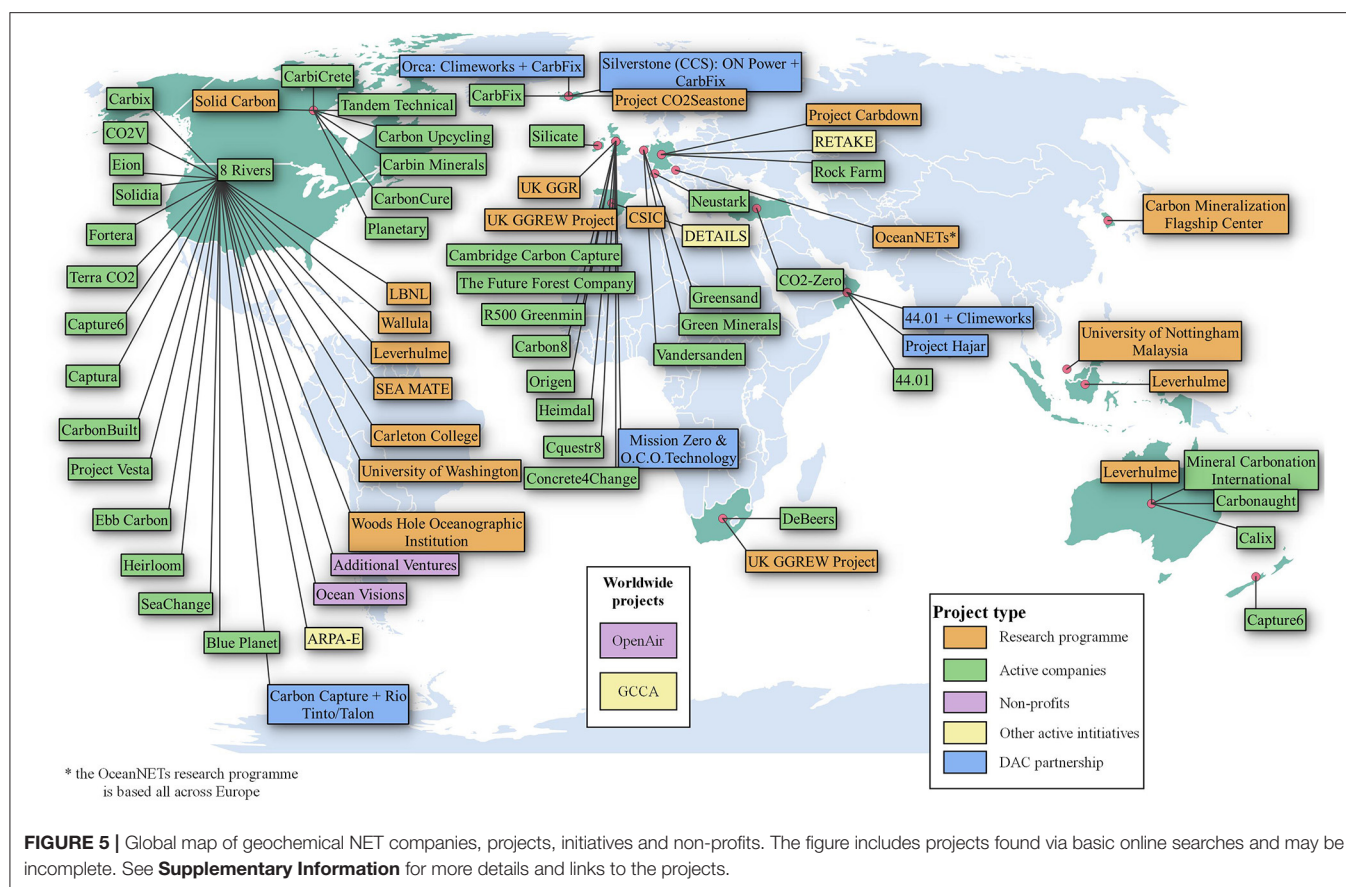
## LIFE CYCLE ASSESSMENT ON GEOCHEMICAL NETs

Research has mainly focused on techno-economic aspects of different geochemical NETs, and NETs in general. However, this is not the case for other important aspects such as their social perspective, including social acceptance, and their environmental perspective, the latter typically examined using the life cycle assessment (LCA) methodology. Specifically, even though (geochemical) NETs aim at removing CO<sub>2</sub>, they could also be responsible for emitting GHGs and other pollutants throughout their life cycle (Cooper et al., 2022). Therefore, LCA can play an important role in both identifying such emissions and minimizing them, and also in comparing, from the environmental perspective, different NETs to identify the most promising solution under the local conditions. The reason is that the environmental performance of NETs depends not only on the technology but also on local conditions and spatial restrictions. For example, Lefebvre et al. (2019) found that transportation (distance from quarry to field) was a key limitation to enhanced weathering in soils, whereas Deutz and Bardow (2021) studied the environmental sustainability of the DAC plants in Hinwil and Hellisheiði, operated by Climeworks, and noted that the LCA

results are very sensitive to the energy sources. Furthermore, when comparing different NETs, a wide range of environmental impacts should be considered (McQueen et al., 2021b) as well as other aspects of each technology. For example, Cooper et al. (2022) compared the effectiveness in carbon sequestration of afforestation/reforestation, enhanced weathering, DAC, and BECCS and noted that even though the first had the lowest environmental impact it exhibited very low carbon removal rates, whereas BECCS had a lower impact on climate change and toxicity compared to enhanced weathering and DAC, but a much higher impact on land use.

However, more research is required on LCA, since many challenges pertaining to different functional units, system boundaries, the climate change-energy nexus, and the timing of GHG emissions and removals make the direct comparison of different NETs difficult (Goglio et al., 2020). Furthermore, when system expansion has been used in LCA, avoided emissions, due to substitution of certain processes, have been misinterpreted as negative emissions, i.e., as carbon removal from the atmosphere, and therefore there is a need to distinguish between avoided and negative emissions, along with consistency in system boundaries and functional units (Terlouw et al., 2021). For example, when 27 LCA studies on NETs were reviewed by Tanzer and Ramírez (2019), 41% (11 studies) labeled avoided emissions as negative emissions, while it was also noted that system boundary choices also play an important role on the perceived emission balance of a NET. For this reason, guidelines for LCA studies on NETs have been proposed (Müller et al., 2020), while detailed life cycle inventory (LCI) data for different spatial extents, power systems, and chemical processes pertaining to NETs are also required (Cruz et al., 2021). The importance of the system boundary, allocation/system expansion, data availability and accuracy, parameter uncertainty, permanence of CO<sub>2</sub> removal, and the need for common guidelines in LCAs for NETs has been also highlighted (McQueen et al., 2021b). In this regard, the integration of LCA with techno-economic analysis (TEA) can decrease NETs uncertainty and improve technology readiness levels (TRL) (Li and Wright, 2020).

Finally, apart from capturing and permanently storing CO<sub>2</sub>, (geochemical) NETs can also be associated with a wide range of positive but also negative environmental impacts, which have yet to be fully identified and therefore are not typically included in the system boundary of LCA studies. For example, ocean acidification greatly impacts marine ecosystems and reliant human communities (Doney et al., 2020), particularly affecting tropical coral ecosystems (Comeau et al., 2013; Feng et al., 2016). However, these positive effects of alkalinity addition, along with possible negative ones such as changes in the primary productivity, respiration, and photophysiology of living organisms in alkalinized seawater (Gore et al., 2019) or freshwater (Mant et al., 2013) have yet to be fully identified and quantified and therefore cannot be captured in LCA studies that examined ocean alkalinity enhancement NETs. This is also the case for terrestrial NETs. For example, although the potential co-benefits of dispersive enhanced weathering approaches are numerous, these are difficult to quantify and use as inputs in LCA studies, since their potentially irreversible effects may not become apparent until years after application, at which



**FIGURE 5 |** Global map of geochemical NET companies, projects, initiatives and non-profits. The figure includes projects found via basic online searches and may be incomplete. See **Supplementary Information** for more details and links to the projects.

point the socio-technical systems may already be entrenched [see Collingridge dilemma (Collingridge, 1979)]. Therefore, there is a need for more research on the biological responses of different NETs, as to identify and quantify both the negative and positive effects and then use this data as inputs in LCA studies dealing with NETs. By doing so, more robust and reliable LCA results will be obtained which could also play an instrumental role in improving the social acceptability of NETs. Specifically, public acceptability can be a potential constraint on the research and deployment of NETs (Bertram and Merk, 2020) and particularly of the ocean-based NETs, which might face a greater public acceptability challenge than their terrestrial counterparts (Cox et al., 2021). To this end, apart from the environmental perspective, the social and economic perspectives should also be considered, thus examining the overall life cycle sustainability of each NET and effectively communicating the results to improve public perceptions and acceptability.

## THE CURRENT STATE OF GEOCHEMICAL NETs

The recent Innovation for Cool Earth Forum (ICEF) Roadmap underlined that geochemical NETs have the lowest ratio of policy coverage to potential impact compared to other NET approaches. Most policy frameworks [e.g., 45Q, CA LCFS, EU CTS (Low Carbon Fuel Standard; EU Emissions Trading

System (EU ETS); Internal Revenue Service, 2021)] have criteria that unintentionally exclude geochemical NETs. International agreements prohibit commercial NETs that involve the release of alkaline feedstock into the ocean. However, some government agencies such as the Advanced Research Projects Agency-Energy (ARPA-E) in the US are exploring the potential contribution of alkaline mineral-based technologies to negative emissions, enhanced metal recovery, and the decarbonization of industry and mining (ARPA-E, 2021).

While geochemical NETs remain a niche industry, the emergence of recent projects, such as those that offer carbon removal credits or sequester carbon within concrete, suggest that the industry is expanding. New companies are involved in piloting projects, some partnering with DAC companies that provide input CO<sub>2</sub>, e.g., the *in situ* mineralization projects underway in Iceland and Oman. Early but increasing exploration by large organizations in agriculture, mining, and concrete includes diamond miner DeBeers, who is experimenting in-house with mineralizing kimberlite (DeBeers group, 2019), Rio Tinto, who recently invested in a CarbonCapture DAC+mineralization project (Hiar, 2021), and The Global Cement and Concrete Association (80% of global production), whose 2021 roadmap calls for industrial-scale carbon capture at calcination sites (GCCA, 2021).

A list of companies and projects currently known to the authors is shown geographically in the map given in **Figure 5**. This map illustrates the range of projects underway today,

but underlines that we are far from having a large-scale, high-functioning geochemical NETs industry, as many of these projects are in their infancy and have yet to deploy geochemical NETs or remove substantial amounts of CO<sub>2</sub>.

## FINAL REMARKS

In this review, the body of knowledge on geochemical NETs was distilled and comprehensively discussed, focusing on terrestrial, subterranean, and ocean-based geochemical NETs. Over the last two decades, an enormous amount of work has been published relating to geochemical NETs, suggesting the large strides that have been made in this regard. It appears that these technologies have great potential for carbon removal and storage, owing to the vast resource of low cost natural and artificial alkaline minerals, their thermodynamically favorable reactions with CO<sub>2</sub>, and the long-term stability of the carbon-bearing products they form. However, the field of geochemical NETs remains, by and large, nascent with few fully-fledged technologies being available. To avoid the worst effects of climate change the introduction of geochemical NETs at scale (Gt CO<sub>2</sub> yr.<sup>-1</sup>) is required as soon as possible. To achieve this, proactive, deliberate, and strategic initiatives are necessary for accelerating technology maturity along with substantially improving the public acceptance of these technologies, which is required for their large-scale introduction. For this reason, in Part II of this work, a roadmap, which is accessible and actionable to both specialist and non-specialist actors, is put forth with the goal of catalyzing the implementation of gigaton-scale removal on the timeframes that the Intergovernmental Panel on Climate Change (IPCC) and other key organizations suggest that is needed from as soon as 2025.

## AUTHOR CONTRIBUTIONS

All authors contributed to the drafting and revision of the manuscript, and read and approved the final submitted version.

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## SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fclim.2022.879133/full#supplementary-material>

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## NOMENCLATURE

Carbonic anhydrase (CA): Enzyme that catalyzes equilibration between dissolved CO<sub>2</sub> and carbonic acid.

CO<sub>2</sub> mineralization: also referred to as mineral carbonation, a process by which CO<sub>2</sub> becomes a solid mineral, namely carbonate. The term carbon mineralization is also widely used in the field and should not be confused with conversion of organic carbon into CO<sub>2</sub>.

Direct air capture (DAC): an engineered process of capturing carbon dioxide directly from ambient air and generating a concentrated stream of CO<sub>2</sub> for sequestration or utilization.

Dissolved inorganic carbon (DIC):  $\text{HCO}_3^-_{(\text{aq})} + \text{CO}_3^{2-}_{(\text{aq})} + \text{CO}_{2(\text{aq})}$

Enhanced weathering: a method whereby crushed alkaline minerals, typically Mg- and Ca-rich silicates, are spread in the environment where they undergo physical, chemical and biological weathering, thus removing CO<sub>2</sub> from the atmosphere and storing it as carbonate minerals and ocean bicarbonate. Two main types of enhanced weathering exist—coastal enhanced weathering and enhanced weathering in soils. Over longer time frames, these are also methods of ocean alkalinity enhancement.

Extracellular polymeric substances (EPS): A broad term for biological polymers secreted by microbes. Depending on specific properties, EPS can influence ion concentrations in microenvironments, nucleate carbonate precipitation, and catalyze mineral dissolution.

*Ex situ* mineralization: High surface area alkaline minerals are reacted with CO<sub>2</sub>-rich gases in engineered reactors, enhanced by elevated temperature and pressure, or using reagents. Mineral

reactants are usually transported to a site of CO<sub>2</sub> production. Carbonate mineral products may be utilized.

Geochemical NETs: Any negative emissions technology which involves substantial amounts of alkali or alkaline minerals in its flowsheet.

Indirect ocean capture (IOC): Technologies which remove dissolved inorganic carbon from the ocean, and thus CO<sub>2</sub> from the atmosphere, *via* air-ocean gas exchange due to the pH sensitivity of the ocean's carbonate buffer system.

*In situ* mineralization: CO<sub>2</sub>-bearing fluids or wet supercritical CO<sub>2</sub> are injected into suitable rock formations beneath the Earth's surface where the CO<sub>2</sub> is mineralized.

Negative emissions technologies (NETs): Any technology which removes CO<sub>2</sub> from the air, directly or indirectly, for the purpose of climate change mitigation. Also known as carbon dioxide removal (CDR) technologies.

Ocean alkalinity enhancement (OAE): Technologies that increase the alkalinity of seawater to enhance the ocean's natural carbon sink.

Ocean liming: Spreading soluble alkaline minerals in the ocean to increase ocean alkalinity.

Surficial mineralization: Air, or low purity CO<sub>2</sub>-bearing gases or fluids, are reacted with high surface area natural alkaline rocks, mine tailings or other alkaline industrial wastes, in large piles, heaps, or in controlled spaces such as greenhouses, forming carbonate minerals. Surficial processes, like enhanced weathering, occur more slowly than *ex situ* processes but unlike enhanced weathering, minerals are not transported or spread in the environment.





# The ABC of Governance Principles for Carbon Dioxide Removal Policy

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Climate change mitigation actions, including those aimed at developing and scaling carbon dioxide removal (CDR) activities spanning the industrial, energy, and agroforestry sector, emerge in a context of internationally shared norms that include governance objectives, legal provisions and informal expectations, and societal expectations. Established governance principles provide normative orientation for policy including when targeting the development and scaling of CDR. Knowledge of these principles can guide effective discussion and evaluation of policy options. To facilitate discussion of mitigation options among experts and CDR practitioners, this study excerpts governance principles from legislative texts, the climate governance literature, and the CDR literature with relevance to CDR policy considerations. To illustrate the relevance of the governance principles found for evaluating policy options, we apply them to three technology groups of CDR: Bioenergy with Carbon Capture and Storage (BECCS), Direct Air Carbon Capture and Storage (DACCS), and forestry. This exercise indicates the importance of more intensive attention to the normative dimension of mitigation policies in ongoing deliberative and planning processes. Such efforts can help disentangle normative and factual dimensions and sources of (dis)agreement on the role of CDR in specific climate policy contexts.

**Keywords:** policy instruments, climate change mitigation, norms, principles, public acceptance, negative emissions, carbon dioxide removal, governance

## INTRODUCTION

Carbon dioxide removal (CDR) methods<sup>1</sup> remove CO<sub>2</sub> from the atmosphere into durable storage (IPCC, 2022)<sup>2</sup>. CDR represents a rapidly growing, albeit contentious, topic in climate governance. Research on the techno-economic feasibility and mitigation potential of various CDR approaches is substantial, and a growing body of literature explores the ethics of CDR. However, emerging deliberations on policy design appear largely detached from established governance principles that have guided mitigation policy for decades.

<sup>1</sup> We use terminology from the IPCC's 6th Assessment Report. We would like to note that "methods" is often understood in the climate policy community as meaning methodologies for assessing the baseline emissions and monitoring activity emissions. Here "methods" means concrete approaches leading to CDR.

<sup>2</sup> We refer to CDR as Carbon Dioxide Removal, but acknowledge, that in principle also other GHG could be removed and durably stored away from the atmosphere (GHG removal) via "negative emissions technologies".

Policy action on CDR has been slow – in part due to a lack of orientation for policy makers (Geden and Schenuit, 2020). Against this backdrop, this article identifies possible governance principles applicable to CDR policy design. The article also points to possible interpretations toward decision-making guidance and legitimacy. Engaging with the normative dimension of CDR policy may help identify genuine differences in opinion regarding CDR implementation and finding common ground.

The article seeks to offer insights for two types of readers: 1. Seasoned climate policy actors to whom *CDR* represents an emerging policy field toward climate mitigation; and 2. CDR experts and practitioners to whom *policy* represents the next frontier toward CDR implementation. To bring both on board, we start with a brief history of CDR, followed by an outline of the concept and relevance of governance principles.

## A Brief History of CDR

Considering the notion of CDR solely based on the integrated assessment model (IAM) literature, one might easily think that CDR methods emerged on a blank normative slate (Low and Honegger, 2020), despite the rich history of removals recognized in social science literatures (Carton et al., 2020). Within IAM frameworks, CDR represents a variable in a modeled pathway designed to achieve a given target (for instance, a predetermined level of end-of-century warming). The one-dimensionality of this approach has been criticized in the past (Rogelj et al., 2019), but remains the frame of reference in climate policy. Although over a decade has passed since the first conception of combining bioenergy generation with carbon capture and storage (BECCS; Obersteiner et al., 2001), CDR has largely remained an abstract notion to climate policy makers in many countries. The degree to which well-below 2°C compatible IAM scenarios rely on dramatic scale up of CDR – alongside an unprecedented pace of decarbonization – arguably remains understated in public debates (Michaelowa et al., 2018).

With the emergence of serious public pressure to increase GHG mitigation, net-zero pledges have lately been communicated by many state- and non-state actors. CDR is now also unambiguously understood as legally representing a form of the mitigation of climate change for all intents and purposes of international (and thus also domestic) climate change governance (Honegger et al., 2021a). Consequently, CDR has – in principle – been firmly situated in the realm of climate change mitigation policy. Yet – in practice – CDR is hardly mentioned in the nationally determined contributions (NDCs; see e.g., Borth and Nicholson, 2021), which are the backbone of mitigation action under the Paris Agreement (PA). This will have to change, if the proposed net-zero pledges and eventually the 2°C – let alone the 1.5°C – goals are to be achieved.

## Governance Principles – Concept and Relevance

Climate policy experience suggests decisionmakers respond to political demands rather than to modeling results. Political demands are embedded in both a web of societal expectations and defined governance goals, each with associated norms that

political action is supposed to meet. While the relevance of CDR policies is self-evident for CDR actors, the same is not necessarily true for other societal actors. Indeed, it appears as though for several years, policymakers lacking (normative) orientation have avoided CDR (Geden et al., 2019) and that presently they still struggle situating policy within societal expectations. It is thus important to reflect on the normative dimension of governance goals (such as those regarding climate action or sustainable development) as well as societal expectations in order to define principles that offer well-reasoned guidance for CDR policy implementation. Both stakeholders pushing for CDR use and actors concerned over their discursive relevance may benefit from examining the normative basis of their demands within norms including governance goals and societal expectations. This will help shaping policy instrument design on a sound normative basis.

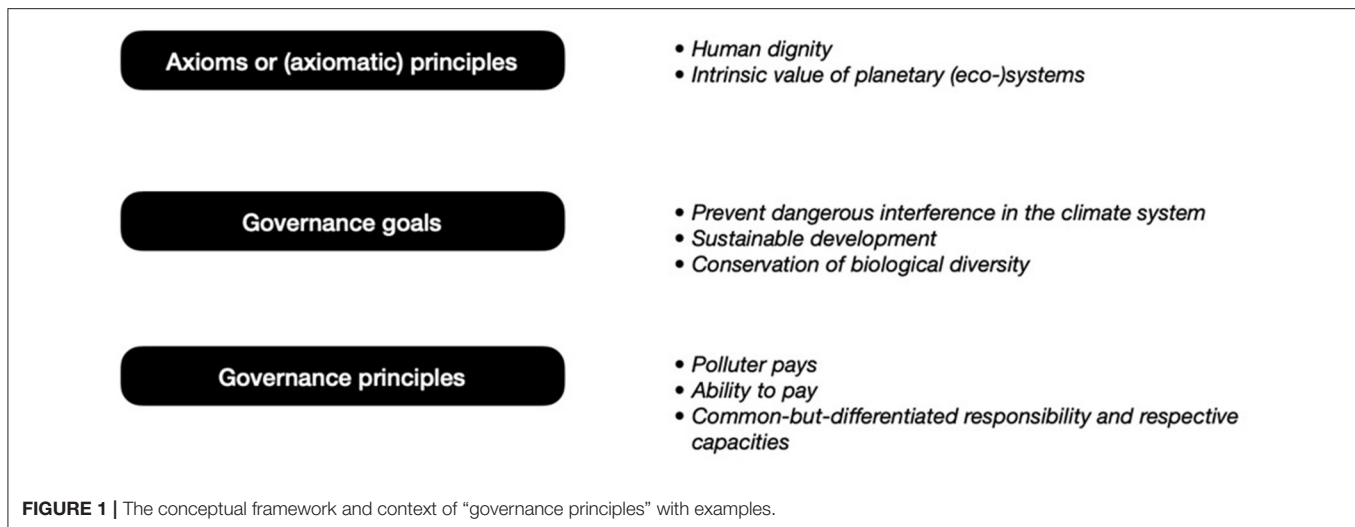
## Review and Tentative Interpretation

In this article, we review key governance literature and legal texts to offer a conceptual framework regarding *norms* and *principles* relevant to CDR, including governance goals and societal expectations. The starting point for our literature selection was the PA and related UNFCCC documents, as they embody key norms for international climate governance. Our second step was to identify norms expressed in the emerging CDR policy literature *via* searches on Google Scholar based on keywords such as “CDR”, “governance”, “principles” and closely related concepts in different combinations. We selected articles based on recency and impact (number of citations) and complemented them with papers deemed particularly impactful in regards to their normative claims or demands. To reduce normative blind spots we complemented the selection through a project-workshop (with the CDR-PoEt consortium) in which we sought to map relevant norms and principles as well as their interlinkages and trade-offs.

After giving an overview on the key principles identified from our literature search, we outline how CDR policy can be situated within them. We focus on climate change mitigation governance and broader environmental governance on the international and national level, but also examine normative claims developed in the CDR-specific policy and governance literature. We then offer possible interpretations and applications of these presented norms and principles to policies targeting three groups of CDR methods, namely BECCS, direct air carbon capture and storage (DACCS), and forestry.

## ESTABLISHED AND EMERGING NORMS AND PRINCIPLES RELEVANT FOR CDR

Most policy decisions – including on climate change mitigation – take place at national or sub-national levels. However, we believe that they are normatively influenced by international governance norms, goals and principles. Our conceptual framework, review and examples focus on the international level, yet it points also to a need for localized (regional, national, and sub-national) mapping of norms and principles.



## Conceptual Framework: Governance Principles

In order to develop, select or evaluate policies for regulating CDR, it is necessary to know what policies should (not) do or be. This issue is the subject of the normative dimension. Normative discourses in governance and policy as well as in related scientific disciplines (political science, philosophy, economics, and others) are closely linked to the concepts of “norms” and “principles.” However, these concepts hold somewhat different meanings within disciplinary and governance contexts. In what follows, we propose to use a possible cross-disciplinary, broad understanding of norms and to view principles as norms that stand at a particular point in the development of a theory or a structure of justification. Since the concepts of governance goals and societal expectations are frequently used in the discussion of governance principles, their location in the framework presented will be briefly outlined here.

Norms provide shared benchmarks to assess courses of action as right or wrong. Such benchmarks take various forms (cf. Mittelstraß, 2004). Understood in regulatory terms, norms comprise benchmarks in the form of rules for action, objectives, and rules constituting institutions. Within a descriptive understanding of norms, these include laws as well as customs. Finally, norms, as understood in moral theories, comprise benchmarks in the form of moral value judgments. We propose to use an overarching notion of norms for the development of policy instruments, encompassing all the aforementioned forms of benchmarks. With such a broad understanding of the concept, governance goals and societal expectations can also be interpreted as norms. Governance goals can be understood as norms within a regulatory understanding. Societal expectations usually represent norms in a regulative sense that are not (yet) institutionalized or established as a rule. These normative expectations that something should (not) be done are expressed, for example, in stakeholder or civil society surveys and are an important component of democratic policy development.

Across disciplines, *principles* are often used to mean *norms* that stand at the beginning of a (thought-) process. In the political context, for example, objectives such as equal pay or subsidiarity are referred to as principles that form the basis for political action (cf. European Union). In the context of international environmental law, principles are defined as “bedrock of this field” (Rajamani et al., 2021). These include, for example, the principles of precaution or of common but differentiated responsibilities (cf. Rajamani et al., 2021). In a philosophical context, principles originally denote axioms, i.e., insights that cannot and need not be proven, from which norms are usually derived. Axioms include, for example, the principle of causality or the principle of utility (cf. Rescher, 2012).

What the various norms and axioms referred to as principles have in common is that they stand at the beginning of the development of a theory or systems or a structure of justification, for example, at the beginning of the development of an ensemble of policy instruments.

We can distinguish principles of different order. A norm may constitute a principle in a certain discourse, while it does not do so in another. For example, *human rights* represent a principle if they form the axiomatic basis for climate action. However, they can also “merely” represent a norm when derived from the principle of human dignity as the subject of a moral theory. Since *human rights* are not the basis of theory development in the latter discourse, they do not constitute an (axiomatic) principle in this context. Axioms, therefore, represent the original principles, since they are generally not further substantiated or derived from any other principles (Figure 1).

We thus propose the term *governance principle* as follows: norms that are at the beginning of the development and justification of an ensemble of policy instruments and their evaluation. Based on this conceptual framework, we map norms that can serve as governance principles for the development of policies targeting CDR.

## International Governance Principles in Context of Climate Change Mitigation

Norms that can act as governance principles for mitigation under the international climate policy regime of the UN Framework Convention on Climate Change (UNFCCC) can be found in the legal texts directly – including as explicit provisions as well as by way of governance goals – and in the PA's governance architecture (Bodansky and Rajamani, 2015).

Provisions also point to governance principles embedded in governance contexts other than directly regarding climate change: (a) the consideration of various rights (including human rights and indigenous peoples' rights); (b) efforts toward the eradication of poverty; and (c) contributions to sustainable development. These are further examined in Section *Governance Principles Situated in Other Fields of Global Environmental Governance* and following.

We map potential climate-related governance principles embodied in the PA architecture in the following sub-sections and address the four main pillars in turn: (i) ambitious global long-term temperature goals translated into net zero targets, (ii) bottom-up national mitigation contributions, (iii) an ambition mechanism “ratcheting up” the contributions over time, bolstered by reporting of all countries on action and progress, and (iv) international cooperation and support. We discuss these elements below.

### Potential Governance Principles in the Context of Mitigation Ambition

There are numerous provisions in the PA on the aggregate pace and volume of mitigation efforts and regarding Parties' obligations of conduct. Art. 2.1a defines the long-term temperature goal of holding warming to “well below” 2°C, and to pursue efforts to limit warming to 1.5°C, inducing a collective obligation of result (Rajamani and Werksman, 2018). Article 4 induces an obligation of conduct including regarding CDR: Art. 4.1 specifies the global goal to reach “a balance between anthropogenic emissions by sources and removals by sinks of GHG in the second half of this century.” CDR is thus observed to be an important part of mitigation (Honegger et al., 2021a). Each new iteration of NDCs (at least every 5 years; Art. 4.9) is to represent a progression over time in its ambition and reflects the parties' highest possible ambition (Art. 4.3). NDCs are to become increasingly comprehensive regarding economic sectors (Art. 4.4), and GHG covered across sources and sinks (UNFCCC, 1992; Art. 3).

Together, these provisions suggest possible governance principles to include (as listed in **Table 1**) (a) that CDR should be considered in NDCs, (b) that the efforts should not weaken other mitigation efforts, and (c) that they should be commensurate with the collective ambition of achieving net-zero GHG emissions (balance) and perhaps net-negative global emissions.

### Potential Governance Principles Regarding International Support and Cooperation on Mitigation and Adaptation

The UNFCCC and the PA include provisions regarding technology-transfer for accelerating the adoption of innovative

mitigation and adaptation technologies globally (Art. 10, PA). Institutions such as the Technology Mechanism [including the Technology Executive Committee (TEC) and the Climate Technology Centre & Network (CTCN)] or the Green Climate Fund (GCF) have been set up to contribute *via* financial, capacity and other types of international support (Azam, 2021). The Paris Agreement provides for international cooperation to “allow for higher ambition” (Art. 6), which is widely regarded to fulfill a norm of mitigation efficiency (relating to *ambition*; Edmonds et al., 2021). Relevance of these provisions and institutions could increase for CDR due to large capacity- and cost- differentials as well as co-benefit potentials. Significant differences in respective responsibility (see below regarding fair-share norms) could further add to expectations of international climate finance for mobilization of CDR as international transfers can significantly improve the distributive implications of climate action for world regions with limited resources (Lenzi et al., 2021). In this context, the term “climate justice” has been widely employed by civil society stakeholders; “climate justice” considerations can have relevant repercussions on the ranking of alternative types of CDR. CDR technology could be relevant beyond mitigation: There are significant expectations for adaptation support, where certain CDR methods could potentially play a role also due to co-benefits (Buck et al., 2020).

These observations suggest governance principles to include also: (d) technology-transfer may need to help strengthen capacities for CDR implementation globally, (e) international cooperation may improve CDR efficiency (f) climate finance may mobilize CDR.

### Potential Governance Principles Regarding Environmental Integrity

Environmental integrity has emerged as a key – albeit somewhat complex – norm particularly relevant to international cooperation (PA Art. 6) but also mitigation more generally (PA Art. 4.13). A simplified definition by Schneider and La Hoz Theuer (2019)– applied to international carbon markets – stipulates that a policy or mitigation action leads to the same or lower aggregated global emissions. Generalizing their framework suggests that to achieve environmental integrity at least four conditions need to be fulfilled jointly: proper accounting; robust tracking (MRV) of mitigation results; ambition of the mitigation target; and spillover incentives for future action. These are intertwined and also relate to ambition, insofar that correct MRV is a precondition for tracking effective progress, and transparency is a condition for judging *ex-ante* the ambition level as well as *ex-post* the progress made. To achieve environmental integrity for CDR action, the durability (or “permanence”) of carbon storage needs to be consistently addressed in accounting and MRV. This may require clarifying monitoring and reporting processes over defined periods of time, as well as liability associated with storage reversal or “leakage.”

The above observations suggest that governance principles may also include an umbrella principle that CDR policies should be environmentally integer. Sub-principles operationalizing this umbrella principle (beyond previous ones regarding ambition) could include (g) consistent accounting for CDR results applying



conservative baselines and including leakage, (h) application of robust MRV methodologies including regarding leakage.

### Potential Governance Principles Regarding Fair-Share Efforts

Given that mitigating climate change is a globally shared challenge, norms regarding regions' countries', or companies' mitigation efforts involve considerations of distributive justice and equity. The same is true for any comparison of present and future efforts (intergenerational justice) inevitably included in the notion of mitigation pathways and policy planning. The norm of *common-but-differentiated responsibilities and respective capacities* (CBDR-RC) referred to explicitly as a principle under the UNFCCC and further qualified as "in the light of different national circumstances" under the PA (Art. 2.2) is central to fair-share considerations of mitigation efforts within individual national contributions and on aggregate for regional or global efforts. Fair-share considerations are increasingly being applied to CDR, where both the intragenerational (e.g., which economies should install and/or finance CDR activities) and the intergenerational (e.g., long-term permanence) dimensions exist (Lenzi et al., 2018; Fyson et al., 2020). Consideration of responsibility or proportionality based on historic emissions, capability and ability-to-pay, or 'per-capita' equality are central to this discussion. There is also a growing realization that biophysical limits would affect the respective capacities and thus may also influence fair-share judgments (but not of financial responsibility; Pozo et al., 2020). In many cases, especially for land-based CDR, potentials are high in low- and middle-income countries with relatively low historic emissions, such that inverse trends between responsibility and biophysical limits need to be navigated (Honegger and Reiner, 2018). The Polluter Pays Principle (PPP) represents one particular expression of intra- or intergenerational justice invoked for mitigation overall whereby revenues collected from emitters would flow to CDR implementation to clean-up previous pollution (Stainforth, 2021). The notion of forcing emitters to completely balance their emissions with permanent carbon removal has led to the idea of imposing a Carbon Removal Obligation on emitters (Bednar et al., 2021), which might meet both inter- and intragenerational justice.

The above observations suggest governance principles to include an umbrella principle for the (i) consideration of inter- and intragenerational equity which can be operationalized to include (j) common-but-differentiated responsibilities (k) differentiation by capacities and (national) circumstances.

### Potential Governance Principles on the National Appropriateness of Policy and Metrics

There are arguments based on the governance architecture of the PA (see Section *Potential Governance Principles in the Context of Mitigation Ambition*) as well as theoretical arguments that policies and their metrics of effectiveness need to be nationally determined and appropriate. Grubb (2014) makes the theoretical case for a broad policy mix that allows rapid development and diffusion of mitigation technologies. The theoretical economists' view that a single international emissions

trading scheme would be singularly effective and efficient is increasingly being questioned (Haïtes, 2020). CDR policies thus ought to be part of a policy ensemble mobilizing a nationally determined range of mitigation technologies, including by funding of research and development, providing incentives for roll-out of maturing technologies, and overcoming non-monetary barriers. Targets for each policy instrument need to be specific enough to judge the respective instrument's ability to achieve them. Metrics of judgment against clearly formulated objectives also need to be nationally determined and span both the near- and the long-term: An intervention can for example be *effective* in increasing the number of CDR installations in the mid-term but remain *inefficient* at delivering meaningful mitigation results in the long-term (or vice-versa), if the costs of the technologies are not declining over time (Honegger et al., 2021c). Clear policy objectives, metrics and aligned policy mixes thus appear to be requirements following from norms regarding ambition, "environmental integrity" and particularly effectiveness. Efficiency – albeit highly relevant for achieving the highest-possible ambition – cannot be viewed in isolation, but within a long-term perspective to avoid solely focussing on temporary low-cost solutions that do not solve the problem, such as non-sustainable biomass-based CDR in settings with high risk of reversals.

The above observations point to further governance principles including on the (l) national determination of clear objectives, policies and metrics for CDR and (m) consideration of both short- and long-term effectiveness and efficiency.

### Potential Governance Principles Regarding Public Deliberation and Participation

Addressing a public good like climate change mitigation requires the public's support for policy instruments at national and local levels. The former may be particularly relevant in case significant costs be associated with the CDR measure. The latter is required to avoid rejection of necessary local infrastructure – often seen in the context of NIMBY (not in my backyard) movements. The scale-up of CDR comes with (perceived) risks and concerns associated both with the measures themselves, and with the broader societal implications and costs. Many of the expectations of outcome identified in this paper are potentially interwoven with public opinion of and support for CDR policies and projects. At the same time, there are long-established expectations and rules for public deliberation and participatory decision-making in environmental governance (Okoro and France, 2019), which may serve as governance principles. Early and consistent public deliberation is thus both a means and an end for climate change mitigation – particularly for measures that do not (appear to) have a self-interested and influential proponent, as may be the case for several types of CDR (Buck, 2019). Transparent and public deliberation processes can help address and alleviate concerns that could otherwise result in opposition at the local (NIMBY) (e.g., Pind Aradóttir and Hjálmarsson, 2018), or national level (Klinke and Renn, 2021). Integrative risk governance seems a prerequisite to legitimate policymaking; the procedural rule or objective regarding the development of policies thus includes deliberation among

experts and epistemic communities (epistemic deliberation), societal stakeholder groups (associational deliberation), and the general public (public deliberation) (Klinke and Renn, 2021). Perception of risks and fairness (both intragenerational and intergenerational) of policies are highly interlinked with trust in the implementing actors and communicators (experts, planners, and decisionmakers; Honegger and Reiner, 2018) and with intuitive public narratives, as found by studies of the public perception of CCS in Norway and Germany (Dütschke et al., 2016; Merk et al., 2022).

The above suggests an additional governance principle to ensure (n) procedural justice and (o) public participation and stakeholder involvement including through a transparent policy deliberation and design process.

## **Governance Principles Situated in Other Fields of Global Environmental Governance**

### **Potential Governance Principle Regarding the Sustainable Development Goals**

The perhaps broadest and most encompassing formulation of a common normative vision for the future can be found in the 17 sustainable development goals (SDGs) adopted by the United Nations – alongside the PA – in 2015. A particularity of the SDGs as a normative backdrop for technology and policy assessment is their integration of biophysical, socio-economic, political, and institutional dimensions and the corresponding recognition of their respective interrelations. One attempt at assessing CDR approaches against the backdrop of the SDGs found the broad range of the objectives to helpfully force the integration of hard-to-integrate factors into assessments (Honegger et al., 2021a). This wide lens could serve as a normative backdrop also for future regional and local evaluations of CDR proposals, where localized assessment of specific approaches is needed. Here, priorities across the diverse goals can be defined at an appropriate level. Governments tend to be unwilling to wield their sovereignty over nationally determined policies, yet decision-supporting tools can nonetheless be developed e.g., by international institutions working on mitigation, international cooperation, technology transfer or capacity building based on the SDGs that enable transparent evaluations without precluding policy choices.

Based on the above, governance principles for CDR policy include (p) a contribution to sustainable development.

### **Potential Governance Principle on the Duty to Prevent Transboundary Harm and Preference for Rectifying Damage at Source**

States are to ensure that activities within their national jurisdiction or control do not cause significant damage to the environment of other states or areas beyond their national jurisdiction or control (UN, 1992). As a forward-looking legal principle, this may become relevant to some CDR activities, especially if taking place in the open ocean or at sufficiently large scale to result in transboundary effects. The principle itself remains rather abstract and might only indirectly inform decisions regarding local CDR applications. A related norm is the notion that damage should be rectified at its source, as stated

e.g., in the EU's guidelines for environmental policy (European Union, 2016). This concept expands the duty to avoid “exporting” environmental harm and/or damage from an international (as formulated in the duty to prevent transboundary harm) to a national or even local perspective. However, its relevance for CDR may depend on the operational definition of “damage.”

This suggests governance principles for CDR may include q) the duty to prevent transboundary harm and r) a preference for rectifying damage at source.

### **Potential Governance Principle Regarding Precaution**

There has been surprisingly little analysis of the precautionary principle – the most formal characterization of precaution – with respect to CDR, despite this norm often being raised in the context of “geoengineering.” Honegger (2020) finds precaution challenging for decision-problems on CDR, given risk-risk trade-offs and ambiguous interpretations (Wiener and Rogers, 2002; Sunstein, 2005). The PA also refers to the precautionary principle, evidently to call for mitigation action despite uncertainties over the severity of the climate change problem (Art. 3.3). This is in contrast to the “geoengineering” literature, where precaution signifies limiting deployment (Bodley, 2013).

Precaution can thus mean to proactively pursue a broad array of mitigation options including various forms of CDR, as well as caution in the reliance on individual options and in light of possible adverse effects. Governance principle could thus include the (s) proactive consideration of multi-risk trade-offs including policy or technology failure risks as well as countervailing risks of omitting policy steps.

## **Expectations From the CDR Literature**

Besides the previously discussed long-established norms on climate change mitigation as well as in other global environmental governance contexts, an emerging body of literature is identifying and proposing CDR-specific norms that can act – and sometimes are explicitly presented as – governance principles. We outline these in the following.

### **Potential Governance Principle on the Long-Term CDR Needs for Net-Zero Targets**

Much of the recent consideration of CDR has been fueled by a surge in net-zero targets among national and sub-national governments as well as in the private sector, including the EU, Scandinavia and Germany (Geden and Schenuit, 2020; Honegger et al., 2020). The European Commission has started a process for developing a Carbon Removal Certification Mechanism, and political dynamics around the role of CDR in the fair distribution of mitigation burdens are accelerating in discussions between EU member states and on the level of the UN climate regime (Geden et al., 2019; Pozo et al., 2020). In these developments CDR is ascribed a particular role – most frequently to offset residual, unavoidable emissions to reach net-zero. Congruently, a growing consensus narrative describes CDR as a necessity to fulfill ambitious climate targets (Otto et al., 2021).

This implies another governance principle (complementing (a) and (c)): (t) anticipate the longer-term CDR needs toward a stated net-zero or net-negative emissions target.

### Potential Governance Principle for Avoidance of Over-Promise and Under-Delivery

Some see CDR as an imaginary with little foundation in political and social realities (Low and Boettcher, 2020), but capturing public attention (Otto et al., 2021). Many fear a false sense of certainty regarding the feasibility and desirability of CDR (Low and Honegger, 2020). Avoidance of delaying, displacing or otherwise undermining decarbonization efforts is thus a prominent demand (also referred to as “mitigation deterrence,” or “moral hazard” (McLaren, 2016; McLaren and Markusson, 2020; Morrow et al., 2020). Scaling CDR to levels anticipated in IAM scenarios will take decades and is highly uncertain in light of limited technological maturity. Lenzi et al. (2018) indicate there is no time to wait for CDR to become ready to be deployed on a large scale yet increasing reliance on decarbonization also involves deep uncertainty. Thus, the long-term role CDR is expected to play in every nation’s long-term climate strategy – and corresponding short-term action – needs to be clear and the basis for policy-design (Morrow et al., 2020). Yet without dedicated policy accelerating CDR readiness, however, under-delivery is virtually certain (Honegger et al., 2021c). Mitigation policy overall and on CDR in particular must avoid over-promise and under-delivery.

Corresponding Governance Principles could include (u) communication of a strategy for preventing over-promise and under-delivery, (v) communication of intermittent targets and policy objectives, and (w) adapt policies if intermittent targets and objectives are missed.

### Potential Governance Principle for Specific (Separate) Targets

Calls for “separate” or specific targets for CDR and for emissions reductions are common in the CDR-policy literature (McLaren and Markusson, 2020; Morrow et al., 2020). A widely shared justification for focusing on CDR separately from emissions reductions is the need to incentivize action on both, without one (usually CDR) undermining the other. Yet a strict operationalization of such an expectation into siloed policies would generate trade-offs with other norms and ultimately act as a self-fulfilling prophecy, given that restricting fungibility of different mitigation options can cause inefficiency and render policies ineffective (Elkerbout and Bryhn, 2021). A “both-and” rather than an “either-or” toward defining targets and policy frameworks for CDR and other mitigation measures thus seems in order (Morrow et al., 2020). We postulate that “specific” targets rather than “separate” targets would better serve the underlying norm.

The above might suggest a potential Governance Principle to (x) formulate increasingly specific targets for various CDR and emission reduction methods within and across economic sectors.

### Potential Governance Principle for Specificity to CDR Cases

The term “CDR” covers a wide range of different methods ranging from so-called “nature-based solutions” such as afforestation to approaches such as DACCS. A further source of differences lies in their inherent socio-economic role: While DACCS is a pure mitigation measure with no other *raison d’être*, other approaches represent essentially a tweak of an ongoing productive activity – such as managing land and food production (forestry) or generating electric power and heat (BECCS). CDR types differ in other ways too – e.g., in regard to their resource needs and their potentials for co-benefits. In addition, regional and cultural differences also suggest that expectations and norms will vary across the globe. Contextualized, nuanced, and detailed analysis of conditions and circumstances on a case-specific basis thus appears to be an important condition for designing policy instruments that scale up different types of CDR (Morrow et al., 2020). Yet not every instrument needs to target every CDR method, so this norm may be met *via* an ensemble of instruments.

Governance Principles thus could include (y) consideration of each CDR methods specificities, and (z) pursuit of a sufficient policy ensemble that meets the needs of the targeted methods.

In the various applicable governance contexts and respective literatures outlined in Section *Established and Emerging Norms and Principles Relevant for CDR*, we have identified the governance principles summarized in **Table 1**. This represents by definition a debatable or incomplete list, given that other governance contexts can be evoked. They are – also by design – partly overlapping as they are designed to cover a specific range of governance contexts and point to particular interpretations thereof. They can nonetheless stand at the beginning of the policy design process for dedicated CDR policies and policy mixes. Governance principles should not be confused with a set of criteria (in the context of multi-criteria decision-making or specific assessments), where the priority is to ensure minimal overlap at the expense of normative breadth.

## APPLICATION TO POLICY DESIGN ISSUES FOR SPECIFIC CARBON DIOXIDE REMOVAL TECHNOLOGIES

In the previous sections, we have identified governance principles for CDR policy. In the next sections we explore whether and to what degree these governance principles could offer orientation in the policy design targeting three groups of CDR methods (BECCS, DACCS, and forestry). These three approaches cover a broad spectrum of CDR methods, but are of course only examples.

To acknowledge the broad range of policy options, we provide examples of potential outcomes, which might find heterogeneous solutions for trade-offs between the different pertinent norms. This outlines the large spectrum of possible compromises and approaches. Furthermore, we emphasize that the examples used here are far from exhaustive with regard to available and possible CDR methods and policy designs. They cover, however, a broad

**TABLE 1 |** The ABC of potential governance principles.

Governance context	Governance principles
Mitigation ambition	a) CDR should be considered in NDCs, b) CDR policies should not weaken other mitigation efforts c) Resulting CDR efforts should be commensurate with the long-term collective mitigation ambition
International support and cooperation on mitigation and adaptation	d) Policy mixes should include technology-transfer to help strengthen capacities for CDR e) Policy mixes should include international cooperation to improve CDR efficiency f) Policy mixes should include international climate finance transfers to mobilize CDR.
Environmental integrity	g) Policies should ensure consistent accounting for CDR results applying conservative baselines and including leakage h) Policies should apply robust MRV methodologies including on leakage
Equity and fair-share efforts	i) CDR policies should fulfill principles of inter- and intragenerational equity (e.g., Polluter Pays or Ability to Pay). <sup>a</sup> j) Efforts should internationally be differentiated per common-but-differentiated responsibilities k) Efforts should internationally be differentiated by respective capacities and (national) circumstances.
National appropriateness of policy and metrics	l) Policies should include a national determination of clear objectives, policies, and metrics for CDR m) Policies should consider both short- and long-term effectiveness and efficiency
Public deliberation and participation	n) Policies should be procedurally just o) The policy design process should involve public participation and stakeholder involvement
Sustainable Development Goals	p) Policies should contribute to sustainable development
Duty to prevent transboundary harm and preference for rectifying damage at source	q) Policies should prevent transboundary harm r) Policies should prioritize rectifying damage at source
Precaution	s) Policy designs should reflect multi-risk trade-offs including policy or technology failure risks as well as countervailing risks of omitting policy steps.
Long-term CDR needs for net-zero targets	t) Anticipation of longer-term CDR needs incl. toward net-zero or net-negative emissions targets
Avoidance of over-promise and under-delivery	u) Policy mixes should include strategies for preventing over-promise and under-delivery v) Policies should include intermittent targets and policy objectives w) Policies should be adapted upon missing intermittent targets and objectives.
Specific (separate) targets	x) Policies should involve increasingly specific targets for various CDR and emission reduction methods
Specificity of CDR cases	y) Policies should reflect CDR methods' specificities z) Policy ensembles should meet the needs of the targeted methods

<sup>a</sup>Polluter Pays and Ability to Pay represent two widely recognized principles of distributional justice. However, they serve here only as examples of principles of fair distribution. Which principles the policies have to fulfill in detail can neither be discussed nor determined here.

and heterogeneous spectrum of current CDR methods and therefore allow illustrating possible applications of governance principles presented in Section *Established and Emerging Norms and Principles Relevant for CDR*. Further cases, however, would require adjusted and detailed analysis – in specific (national, regional, or local) political contexts. In each sub-section, we list the letters of the principles that are relevant; space limitations prevent a discussion of each principle.

## Resulting Policy Considerations for Bio-Energy With Carbon Capture and Storage

In the following, we highlight examples of BECCS specific policy issues that may be further explored against the backdrop of the identified governance principles.

### Mitigation Ambition and Avoiding Over-Promises and Under-Delivery

BECCS is characterized by the tension between modeled potentials (Minx et al., 2018; Rickels et al., 2019; Low and Schäfer,

2020) and past controversy over biofuels as a mitigation measure (Beck and Mahony, 2018). Policy planning may thus need to proactively clarify the anticipated scale and role of BECCS to address inflated expectations and associated concerns. Careful interpretation of modeling results and realistic estimates should be made when policy targets for the short-, mid-, and long-term BECCS development are determined (governance principles: c, j, k, l, o, u, v, w, and x).

### Environmental Integrity

A key uncertainty for BECCS is the carbon-balance of biomass (which needs to be renewable, sustainably sourced, and not lead to depletion of biomass stocks elsewhere). To ensure this, MRV should consider the full value-chain (including biomass sourcing, carbon capture, transport and storage) and set a high bar of evidence especially regarding the biomass source. Storage permanence is also of relevance, but of lesser concern than for approaches resulting in lower inherent durability, and policy solutions exist at least in the EU (European Union, 2022). A newer method for storage through basaltic mineralization



promises virtually no reversal risk (governance principles g, h, and p).

### International Cooperation and Support

Policies can pursue international cooperation for BECCS as its value-chain can be split between countries. This is an opportunity for efficiency (principle e), but it introduces challenges for distributive fairness (principle j), and the transportation and storage infrastructure may be rejected due to perceived risks despite public participation (principle o; Gough et al., 2014; Pind Aradóttir and Hjálmarsson, 2018; Merk et al., 2022).

### Sustainable Development Goals

Land-use and biomass-use competition chiefly affects BECCS' sustainability. Policy instruments could pursue a (cascading) prioritization of biomass uses (prioritizing e.g., already existing plants and facilities, as well as waste feedstocks (agricultural residues, construction wastes). Biomass also holds important socio-economic dimensions: In low and middle income countries, biomass tends to serve low-income strata without access to commercial fuels. Policies thus must not divert biomass access from such populations to prevent severe social impacts and may need to address any such effects in their design (e.g., compensation or economic inclusion) which will require designing them with participation of such stakeholders (principles i, n, p, and s).

### Public Deliberation and Participation

Participatory policy design is relevant given the different types of industrial actors involved in the value chain (producers of biomass, energy producers, transport-providers, and storage-providers) and their likely heterogeneous level of influence (principles n and o).

### Specificity of BECCS Policy Trade-Offs

BECCS represents a range of applications: purely biomass power-plants or combined biomass- and fossil fuels, solid- or liquid waste incineration, liquid biofuel- or biochar production with energy generation. Policies will need to target each case specifically for their inherent differences (incl. implications from biomass-flows, land-use, or socio-economic).

An obvious trade-off lies in the discrepancy between efficient mitigation and socio-economic consideration. On the one hand, a policy could also focus on either the capture side, aiming at large amounts of biomass to be combusted and the resulting CO<sub>2</sub> captured, or on the transport and storage side, in which case leakages in transportation and storage would also be addressed<sup>3</sup>.

Policies with an overly strong focus on efficiently maximizing the amount of CO<sub>2</sub> captured would probably incentivize land-intensive production of biomass including fertilizers and pesticides, cheap capture technologies, transport, and storage sites. Besides the abovementioned consequences for SDGs focusing on biodiversity, land use conflicts and health issues stemming from the deployed chemicals might follow. More lenient transport and storage regulations and MRV could lead

to efficiency in reaching the policy target while compromising environmental effectiveness.

On the other hand, focusing solely on minimizing BECCS' impact on socio-economic aspects could lead to relatively little capacity for BECCS in settings where agricultural land is a limiting factor. Such an approach could incentivize outsourcing BECCS, thereby increasing international cooperation and, if low- and middle-income countries are targeted, financial support for host countries of BECCS installations. While NIMBY conflicts would be avoided in the country implementing the respective policy, societies with a high awareness of equity related issues would likely face a lack of public acceptance, as the outsourcing of potentially conflict-prone technologies like BECCS might not be perceived as acceptable.

We acknowledge that both examples are extreme cases, which are not likely to materialize in a real-world setting. However, they show the necessity for politics to develop a holistic view on BECCS and careful considerations regarding the setting of weights to the many principles to be considered in designing BECCS governance.

## Resulting Policy Considerations for Direct Air Carbon Capture and Storage (DACCS)

Here we address DACCS specific policy considerations against the backdrop of the governance principles.

### Mitigation Ambition, Long-Term Needs, and Under-Delivery

The mitigation contribution of DACCS hinges on the abundant availability of usable, low carbon energy and the provision of a sufficiently high carbon-revenue due to high costs. If solely included within carbon pricing and carbon market systems, in direct competition with other mitigation options, DACCS would not be competitive, so dedicated policies will need to address its near-term requirements for scaling and driving down costs for a meaningful medium to long-term contribution (principles m, t, u, v, w, and z).

### Environmental Integrity

As a highly technological process, MRV of DACCS is straightforward and uncontroversial. Embodied emissions from its significant material requirements can be considered in its net emissions. The main concern for environmental integrity of DACCS policies may relate to its low-carbon energy needs which risk displacement effects. Policies thus should ensure deployment takes place on sites with a structural surplus of zero-carbon energy (principles b, g, h, q, and x).

### International Cooperation and Support

Capture and storage in separate jurisdictions can entail some efficiency gains if energy and storage availability are separate. But the more likely case is international cooperation through purchases of removal units produced in a country that has significant potential for both through international carbon

<sup>3</sup>The question is, thus, what the target variable of a policy is, and how CDR is governed to contribute to national or voluntary targets.

markets<sup>4</sup>. Basaltic mineralization in volcanic areas often also features geothermal energy potentials, as is the case in Iceland and potentially other volcanic island environments. Dedicated policies in other world regions should ensure international collaborations also foster technology transfer and capacity building to counter a potential imbalance in DACCS capacities (principles d, e, i, and k).

### Specificity of the DACCS CDR Case

DACCS also includes several value chain elements (direct carbon capture from ambient air, transport, and storage). The main requirements are electricity and heat<sup>5</sup> as well as suitable storage. As such, DACCS can be placed where energy availability and storage capacities are high. Land requirements are non-negligible but substantially smaller than of biomass-based CDR. The costs of DACCS processes are high compared to other CDR options but may decrease through technology learning. So DACCS policies will need to deal with the initially high costs, as well as with continuing energy requirements and carefully balance these with other mitigation efforts.

### Avoidance of Over-Promise and Under-Delivery and Precaution

Due to its relative immaturity and lack of a non-carbon revenue source, the biggest concern for DACCS policy may be under-delivery: With only a handful of serious technology developers and 19 small-scale pilot plants currently being active worldwide (International Energy Agency (IEA), 2021), DACCS is a novel and relatively untested approach, and could not be upscaled if its current mitigation costs remained unchanged. A precautionary approach driven by mitigation urgency would view the high costs as an “insurance premium” (Morrow et al., 2020), but this argumentation crucially hinges on the ability of DACCS developers to bring costs down. DACCS technology suffers from the inherent thermodynamic limitations that impose a firm minimum on material and energy needs and costs. This cost threshold may, however, be lower than the long-term mitigation cost for limiting warming per the PA. For DACCS to play a meaningful role, policy ensembles thus need to ensure balancing short and long-term needs, technology development with cost-reduction, and mobilizing high carbon-revenues to enable the technology to compete (principles c, e, i, l, m, s, t, u, v, w, x, y, and z).

### Resulting Policy Considerations for Re- and Afforestation and Agroforestry

In stark contrast to blackbox engineering methods, land-use based CDR methods are the result of numerous actors' behaviors and resulting trends for soil and biomass carbon contents. Multiple dimensions of success can in this case only be viewed in a holistic ensemble and the policy considerations are highly particular to the methods, which is why in this case we inverse the order:

<sup>4</sup>Note that not only capture, but also storage site operators should be covered in potential CDR inclusion to carbon markets, as these entities need to ensure permanence.

<sup>5</sup>At least in most currently pursued technologies.

### Specificity of Re- and Afforestation and Agroforestry Case

In the climate debate, land uses play an ambivalent role, as effectiveness, fairness, environmental integrity, and sustainability dimensions are strongly intertwined for re- and afforestation as well as agroforestry. The land-users, their economic situations and resulting choices themselves are the key factor of success and failure on all accounts, as described in the following.

### Mitigation Ambition and Avoidance of Over-Promise and Under-Delivery

On the one hand, conversion of natural forests into agricultural uses is an important driver of climate change. This holds true especially for conversion for the purpose of cattle ranching and agro-industrial production of commodities. In addition to the massive release of carbon, deforestation has significant negative impacts in terms of biodiversity, and it poses a massive threat to the livelihoods of many highly vulnerable indigenous and traditional communities. This is particularly true for many tropical countries with weak economies and governance structures, where the destruction of forests is continuing at an unabated pace and is the main contributor to national emissions. Accordingly, the protection of the remaining natural forests is seen as one of the most (cost)effective measures to mitigate climate change.

Given the structural barriers, agroforestry CDR may be among the technologies with overly optimistic pathways in the IAM literature (Rickels et al., 2019). Expanding to all kinds of forestry may increase the potential but comes with important repercussions.

### Sustainable Development Goals

In practice, re- and afforestation efforts are frequently operationalized through the establishment of large-scale monocultures of exotic timber and pulp species such as Eucalyptus, Pinus, and Acacia operated by large private sector entities. This allows capturing the benefits from efficiency-oriented policies such as carbon pricing, provided robust monitoring is undertaken. Given that forestry-based CDR belongs to the lowest cost CDR options, this can mobilize significant mitigation (relates to principle e), if incentives are not set perversely to induce deforestation for later reforestation and if the plantations are operated in a rotation system that ensures permanence of the forest in the long run. A drawback of monocultural forests are their negative social and biodiversity-related impacts (Pokorny et al., 2010).

The (re-)establishment of environmentally and socially adequate land use systems like agroforestry can achieve multiple goals including carbon removal, biodiversity, and provision of environmental services (Waldén et al., 2020), forestry and agricultural goods, and benefits to small-scale land users (Jose, 2009; Montagnini, 2017). These benefits can also be realized in formerly deforested areas, which are often highly degraded as a result of inappropriate land uses. In poor countries, agroforestry is typically a highly distributed activity involving mainly small-scale, often subsistence farmers.

All vegetation-based CDR approaches can have negative repercussions on several SDGs (2, 3, 6, and 15), if a purely quantitative sequestration remuneration is undertaken that does not consider co-benefits of the vegetation cover, such as biodiversity, health issues and water quality (relates to principle p). Positive ecosystem services related to vegetation are rather unique to agroforestry. The ecosystem services of agroforestry should thus be specifically considered and ideally be remunerated by public policies; they may also increase public acceptance of biomass-based CDR.

### International Cooperation and Support

Afforestation and reforestation, as well as agroforestry projects have a long history in international development cooperation (Pandey, 2002) but have encountered difficulties regarding upscaling. This is often due to lacking capacities of the local population, which often ekes out a living by unsustainable land use and has no access to markets or agricultural extension services. Moreover, in a policy context that systematically promotes input-intensive mechanized agriculture and livestock production aimed at clearly differentiated global value chains of a few commodities, agroforestry systems are often not competitive. If these challenges were overcome, agroforestry would be well suited for international cooperation under the PA, be it through international carbon markets under Article 6 based cooperation or through international climate finance.

### Environmental Integrity

All policy instruments for forestry need to particularly consider permanence due to high reversal risks of biomass-based CDR. So far, no policy instrument has been able to address this in a way that safeguards both environmental integrity and efficiency. Temporary credits as applied under the CDM have not been attractive to potential carbon credit buyers, while buffer stock requirements will only be effective if applied on a highly aggregated level and being administered by an institution whose existence is assured in the long run. The recent approach of Verra to apply a 100-year monitoring for its nature-based CDR projects and to operate a global buffer stock over such periods may be a harbinger of similar approaches on the national level. In contrast, buffer stock requirements in the low double-digit percentage like specified by the ART TREES<sup>6</sup> standard under the multi-billion USD “LEAF” initiative seem inadequate and in conflict with environmental integrity.

Due to the large number of actors and differences in removal potentials on small spatial scales, agroforestry generates large uncertainties and high transaction costs, especially if high environmental integrity is to be guaranteed by robust MRV.

### Equity and Fair-Share Efforts

Exclusion of marginalized groups from land use needs to be prevented. Policy instruments could for example set minimum requirements for local socio-economic considerations. Considering the competition between small-scale agroforestry and large-scale plantations, remunerating CDR solely in a

carbon-results based approach can have negative distributional consequences, as activities might shift from current small-scale applications toward large-scale businesses, with potentially negative effects on local livelihoods.

## SYNERGIES AND TRADE-OFFS: TOWARD PRINCIPLED POLICY INSTRUMENT EVALUATION AND DESIGN

Looking beyond the case studies, evaluating completely new policy proposals seems rather straightforward. However, the more likely scenario is the adaptation of existing instruments – such as the EU ETS or other established carbon markets or mitigation subsidy programmes – to be extended to also cover CDR (or particular types of CDR). This complicates the assessment but should not deter it.

Provisions and adjustments to include CDR directly into the scope of the EU ETS or indirectly by creating units that can be sold into the EU ETS are already under discussion both in academic (Rickels et al., 2021) and political (Liese, 2022) contexts. This approach is appealing in terms of efficient implementation, avoidance of transaction costs, and utilizing political synergies. But the details will be contested: the policy design will have to consider whether CDR methods will be able to compete directly with more mature means of emission reduction and may therefore install separate subsidies or removal mandates linked with credit trading schemes. For example, Pozo et al. (2020) argue that current carbon trading systems should be revisited or parallel markets for CDR created to separate CDR targets from established mitigation targets. This illustrates the existence of competing norms and principles and the need for careful deliberation to achieve a balance of competing interests while securing the public good of effective mitigation.

Trade-offs also exist at the aggregate level of policy mixes: Lenzi et al. (2021) for example have demonstrated how three stylized policy approaches toward net-zero (including a stronger or weaker reliance on CDR) have different distributional implications, and that there may not be an unequivocally better or worse policy mix across multiple equity dimensions with small changes potentially yielding dramatically different outcomes. It is thus even more important to allow public deliberation regarding the performance of different policy instruments in an environment of multiple principles, weighted heterogeneously by different stakeholders, and to approach policy (mix) design in an adaptive manner that allows iterative improvement based on broad-stakeholder-based practical learning.

## DISCUSSION

CDR is a partially novel (DACCS, BECCS), partially long-standing (forestry) category of climate change mitigation. It is developed within a pre-existing landscape of formal as well as informal, but nonetheless important norms used as principles that guide mitigation efforts. While their interpretation leaves room for specific political, regional, and socio-economic contexts, they may already offer some high-level orientation in

<sup>6</sup><https://www.artredd.org>

deliberation, planning, and policy implementation that aim at putting in place ensembles of CDR alongside emissions-reducing measures to mitigate climate change rapidly enough to “prevent dangerous anthropogenic interference with the climate system” (UNFCCC, 1992, Art. 2).

Given that the feasibility and ultimate scalability of all mitigation technologies and measures – particularly more novel ones – to the levels commensurate with the long-term goals of the PA is fraught with social, economic, and political uncertainties, a portfolio approach appears sensible (Michaelowa et al., 2018). This is aligned with provisions of the international climate policy regime regarding ambition and the need for comprehensiveness in NDCs.

Ultimately, no policy design may be considered universally good or bad on its own. Such judgment must be done contextually – in the socio-economic and political as well as the geographical and environmental context in which a measure is being proposed. The same is true for CDR methods themselves as Honegger et al. (2021b) have emphasized. Technology arguably cannot be judged innately good or bad outside of its social and political context. This calls for contextualized and method-specific assessment of CDR-policy proposals in the national (and sub-national) contexts in which they are considered (and by the respective local or national actors and stakeholders).

Since the uncertainties mentioned above, as well as the broad range of situation-specific framework conditions for implementation of CDR measures characterize the planning and implementation of CDR action, a precautionary approach to limiting the risk of extreme emission overshoot could involve a two-pronged strategy: avoiding over-promise as well as under-delivery. This would mean to plan mitigation action on the assumption that individual CDR methods will fail to deliver, thus potentially “underselling” their potentials. Simultaneously, governments would have to work explicitly toward a broad portfolio of CDR options each becoming cornerstone methods – through appropriate research and development support and by generating effective long-term policy frameworks. Many forms of CDR do not generate revenues from the sale of goods or services, so they will require substantial amounts of funding, either from public or private sources. Public policy also needs provisions to “cut off” support for CDR methods that consistently underperform regarding economic effectiveness and overall SDG impacts compared to their peers. Experiences from the multi-decadal funding of underperforming technologies like nuclear fusion and fission should not be repeated. Given the scarcity of public resources, policy instruments can follow more detailed sets of principles regarding economic dimensions as laid out in Grubb (2014), and for mature technologies requiring scale prioritize efficiency.

The bottom-up nature of the PA suggests that each nation may need to figure out its own nationally determined and most appropriate pathways toward its mitigation targets and the policies utilized along the way. This implies that each party also needs to go through deliberative processes at the national level (based also on their respective actor-constellations and interests) in order to make legitimate decisions on how the public good of CDR is acted-upon. This includes identifying

nationally appropriate prioritizations for the use of key resources. In the cases of agroforestry and BECCS this would mean to identify priority functions in ecosystems, living biomass, and harvested wood – to ensure policy incentives result in systemically sustainable mitigation action.

Since these differing national policies will lead to differing subsidy rates per unit of CDR, efficiency requires the availability of international carbon markets to harness cost differentials. In the past, such markets have shown their effectiveness by e.g., the CDM mobilizing over 7000 mitigation projects in over a 100 countries in <10 years (Michaelowa et al., 2019a). While we acknowledge the drawbacks and problems associated with the CDM, e.g., its relatively low carbon price and uneven spatial distribution of projects, the overall quantity and high utilization of credits generated under the CDM speak for its effective functioning. CDR has the inbuilt advantage that most CDR options can clearly show their “additionality” to business-as-usual, which has been difficult for emission reduction technologies that usually generate revenues from the sale of goods or services (Michaelowa et al., 2019b). This means the use of CDR under Article 6 of the PA needs to be rapidly operationalized. The development of CDR-specific methodologies, as pursued e.g., by the CCS+ initiative<sup>7</sup>, could help to increase the number of CDR projects realized under Article 6. Possible, detrimental effects of such collaboration need to be avoided by robust governance on the international level, namely through the Article 6.4 Supervisory Body. The recent decision to require reporting of sustainable development implications of Article 6.4 collaboration serves as good basis but needs consistent operationalization in the next years.

Besides international carbon markets, international climate finance is essential for ambitious and fair mitigation action, yet could also result in over-promise and under-delivery, given past mixed outcomes of international transfers (Lenzi et al., 2021).

Identifying high-level norms and using them as governance principles to guide deliberations on CDR or proposed CDR policies is one way of approaching the policy challenges posed by CDR. Identifying operationalized criteria ready to be applied to specific policy decision problems is another. In identifying general norms, we have remained at a level of abstraction that allows painting a large picture with broad strokes. By illustrating some of the possible interpretations in CDR method specific cases we sought to point to possibilities for operationalization of principles into policy design guidelines. Such an approach allows approaching policy design and evaluation on a normatively transparent basis that can and should be contested.

However, assessment of CDR or climate policies is not always done in such a manner. In fact, a considerable body of literature already provides assessments of “large-scale” CDR (as a category of “Climate Engineering”; National Research Council, 2015; Schäfer et al., 2015). Yet much of that literature is not explicit as to its normative basis, and most of those evaluations are not situated within any particular governance context (e.g., the mitigation of climate change under the PA or national mitigation action in a particular country). Both – normative transparency

<sup>7</sup>see <https://ccsplus.org/>



and governance contextualization – are, however, key conditions for generating orientation knowledge. Another set of literatures offers criteria for the assessment of climate finance policies (see e.g., Gewirtzman et al., 2018; Michaelowa et al., 2020; respective chapters on (inter-)national climate policy in the IPCC Assessment Reports). While this is more explicitly rooted within a particular policy field, it largely remains unspecific to CDR to date.

Both types of literatures might be leveraged for moving toward normatively transparent evaluation of CDR policy options. Nonetheless, this necessarily politically deliberative move will take time, and the operationalization into criteria for CDR measures and CDR policies requires numerous contestable interpretative steps and inclusion of CDR-specific concerns. The work, however, does not stop with the development of operationalized assessment criteria, but political assessment will require a framework that clarifies how interdependencies of various assessment criteria are to be dealt with: Do several criteria – for example regarding efficiency and alignment with SDGs – jointly constitute necessary conditions (for sound policy)? Are there any hierarchies among criteria, in case of conflicts or trade-offs between them? For example, what should one do if a policy instrument is highly efficient but generates concentrated instead of widely shared co-benefits or revenues? Do criteria ultimately draw on commensurable or incommensurable value-systems, and, in case of the latter, how can policymakers and academics address genuine reasons for disagreement based on fundamental differences in worldviews? To which extent can such evaluations be done with a global claim to relevance and on which choices will countries or regions exercise their sovereign right to shape and pursue mitigation in a nationally determined manner? We offer our tentative identification of principles in form of various norms and expectations as another very early step on a long road toward holistic assessment frameworks for CDR policy, in which both normative and non-normative criteria transparently shape the evaluation of different options and pathways (see Baatz, 2017, 2018).

## CONCLUSIONS

Currently, net-zero emission targets are emerging in several jurisdictions, indicating the anticipated scaling-up of CDR. In this situation, we observe a move away from the abstract and “sanitized” view on CDR derived from the IAM top-down calculated “requirements” (Low and Honegger, 2020) toward a confrontation with real-world policy challenges. In this paper, we have argued that understanding CDR policy options in the

light of structuring governance principles is required to make meaningful progress in this ongoing process.

Based on our observations of (potentially) pertinent governance principles stemming from the policy fields of climate change mitigation, other global environmental governance contexts, and CDR-specific governance literature, we view the political feasibility of CDR policy options to be intimately linked to their performance across such (interlinked) governance principles (Mace et al., 2018; Waller et al., 2020). These are increasingly understood to jointly shape a policy proposals’ acceptability, social and institutional support, and ultimately political feasibility. Public support for CDR policy proposals depends on their (perceived) contribution to fair decarbonization transitions (Gough and Mander, 2019). A less holistic perspective on CDR policy proposals focusing on techno-economic factors alone would fail to capture such connections.

These observations indicate that a robust understanding of governance principles and the trade-offs between such principles is a precondition to the effective discussion and sound design of CDR policies. For such policies to contribute meaningfully and continually to climate mitigation action and, eventually, the achievement of national targets and global objectives their normative basis needs to be established within their respective local, national or regional context. Eventually, there will have to be a convergence of understanding which principles drive CDR policies and how a constructive debate addressing inevitable trade-offs can be held, both on the international and the national level.

## AUTHOR CONTRIBUTIONS

MH: initial conceptualization and writing–editing. MW: conceptual development and writing–editing. CB, SE, AH-C, AM, BP, and MP: writing–editing. All authors contributed to the article and approved the submitted version.

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# Carbon Removal Using Coastal Blue Carbon Ecosystems Is Uncertain and Unreliable, With Questionable Climatic Cost-Effectiveness

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Mangrove forests, seagrass meadows and tidal saltmarshes are vegetated coastal ecosystems that accumulate and store large quantities of carbon in their sediments. Many recent studies and reviews have favorably identified the potential for such coastal “blue carbon” ecosystems to provide a natural climate solution in two ways: by conservation, reducing the greenhouse gas emissions arising from the loss and degradation of such habitats, and by restoration, to increase carbon dioxide drawdown and its long-term storage. The focus here is on the latter, assessing the feasibility of achieving quantified and secure carbon removal (negative emissions) through the restoration of coastal vegetation. Seven issues that affect the reliability of carbon accounting for this approach are considered: high variability in carbon burial rates; errors in determining carbon burial rates; lateral carbon transport; fluxes of methane and nitrous oxide; carbonate formation and dissolution; vulnerability to future climate change; and vulnerability to non-climatic factors. Information on restoration costs is also reviewed, with the conclusion that costs are highly uncertain, with lower-range estimates unrealistic for wider application. CO<sub>2</sub> removal using coastal blue carbon restoration therefore has questionable cost-effectiveness when considered only as a climate mitigation action, either for carbon-offsetting or for inclusion in Nationally Determined Contributions. Many important issues relating to the measurement of carbon fluxes and storage have yet to be resolved, affecting certification and resulting in potential over-crediting. The restoration of coastal blue carbon ecosystems is nevertheless highly advantageous for climate adaptation, coastal protection, food provision and biodiversity conservation. Such action can therefore be societally justified in very many circumstances, based on the multiple benefits that such habitats provide at the local scale.

**Keywords:** negative emissions, carbon dioxide removal, blue carbon, mangroves, saltmarsh, seagrass, restoration, carbon offset



## INTRODUCTION

The overall desirability of large-scale protection and restoration of coastal vegetated coastal ecosystems (primarily mangrove forests, saltmarshes, and seagrass meadows) and their carbon-rich sediments is now well-accepted. Although their global role in carbon storage has only relatively recently been identified (Duarte et al., 2005; Laffoley and Grimsditch, 2009; Nellemann et al., 2009), such “blue carbon” ecosystems have subsequently attracted considerable scientific attention, with >1,000 papers on their biogeochemical and socio-economic importance (2009–2021; Web of Science, with “blue carbon” as search term). The many services provided by coastal blue carbon ecosystems (hereafter CBCEs) include contributions to climate mitigation and adaptation (McLeod et al., 2011; Alongi, 2018; Bindoff et al., 2019; Windham-Myers et al., 2019), thereby providing a nature-based solution that could contribute to the Paris Agreement and the Glasgow Climate Pact (UNFCCC, 2015, 2021). The potential of those contributions is considered sufficiently large to justify the substantive use of CBCEs in climate policy by many studies and reviews (e.g., Herr and Landis, 2016; Crooks et al., 2018; Hoegh-Guldberg et al., 2019; Seddon et al., 2020; UNESCO, 2020; Hilmi et al., 2021; Macreadie et al., 2021; UNEP and IUCN, 2021), with the associated concepts of blue carbon farming (Duarte de Paula Costa et al., 2022) and blue carbon markets (Claes et al., 2022). The further development of such approaches has been prioritized for US research on CO<sub>2</sub> removal (NASEM, 2019).

Until recently, coastal vegetation management has not been separately considered as a mitigation lever in IPCC climate assessments, being subsumed within afforestation and reforestation in Integrated Assessment Models (IPCC, 2018). It has also been omitted from several comparative assessments of negative emissions (e.g., Minx et al., 2018; GESAMP, 2019), or grouped with freshwater wetland and peatland restoration (McLaren, 2012; Royal Society Royal Academy of Engineering, 2018), or considered as a soil or land-based mitigation technique (Bossio et al., 2020; Roe et al., 2021). The IPCC AR6 cycle does, however, specifically discuss CBCEs in all three Working Group reports, with WG II coverage emphasizing their co-benefits (Parmesan et al., 2022) and vulnerability to climate change and direct anthropogenic impacts (Cooley et al., 2022). The AR6 WG III report considers “blue carbon management in coastal wetlands” and “peatland and coastal wetland restoration” as separate approaches in a NET summary table, assessing their status and role in mitigation pathways. However, costs (USD tCO<sub>2</sub><sup>-1</sup> removed) are not given in the table for these approaches, due to “insufficient data” (Babiker et al., 2022). In other syntheses, the restoration of coastal vegetation has been considered to have low effectiveness in reducing global warming and its impacts, providing a “low regret” or “no regrets” mitigation action (Gattuso et al., 2018, 2021; Bindoff et al., 2019).

To resolve the varying opinions on the usefulness of CBCEs in meeting climate policy goals, there is recognized need for “robust scientific evidence along with accountability of the contribution [of blue carbon options] to greenhouse gas mitigation, including meeting the requirements of additionality and permanence of this benefit” (Macreadie et al., 2021).

Here we review the issues (primarily biogeochemical) affecting the reliability of carbon accounting for CBCE restoration, considered broadly, and the implications of these constraints, uncertainties and risks in determining climatic benefits—and hence the cost-effectiveness of this approach for CO<sub>2</sub> removal, acknowledging that there are many non-climatic benefits. The scope for emission avoidance, by protection of coastal blue carbon stores, is not assessed (notwithstanding its importance), nor is any evaluation made of the considerable benefits that CBCE restoration can undoubtedly provide through improved climate adaptation (e.g., coastal protection) and through other ecosystem services (e.g., fisheries and biodiversity conservation). Comprehensive valuation estimates for the very many ecosystem services provided by CBCEs are given elsewhere (e.g., Vegh et al., 2019; Laffoley, 2020), with the wider socio-economic challenges associated with CBCE restoration reviewed by Thomas (2014), Abelson et al. (2020), and Macreadie et al. (2022).

Long-term carbon accumulation and storage primarily occurs in the sediment of CBCEs, rather than their above-ground biomass (that, once established, is not expected to significantly increase). CBCE restoration can involve actions of three kinds, each potentially providing negative emissions through increased carbon burial fluxes and storage. First, by resource management to improve local environmental conditions and promote natural functioning. For example, by restoring natural hydrodynamics to increase freshwater flows, tidal exchanges and sediment supply; by reducing pollution, particularly by nutrients; and by reinstating natural predatory control of bioturbators (Kroeger et al., 2017; Macreadie et al., 2017a). Second, by re-establishing such habitats where they had previously been lost as a result of land-use change or other coastal developments. This action involves planting seagrass or mangrove seedlings in subtidal or intertidal sediments; many such initiatives have already been carried out (van Katwijk et al., 2016; López-Portillo et al., 2017). Third, by the creation of entirely new habitats, such as purposeful coastal flooding to stimulate saltmarsh formation (Adam, 2019). This is not strictly a “restoration” process, but has been widely considered as such in terms of policy action.

In all cases, it is the combination of sustained photosynthesis and long term carbon storage that provides the climatic benefits of CBCE restoration. The aerial vegetation of mangroves and saltmarshes removes CO<sub>2</sub> directly from the atmosphere; for continually-submerged seagrasses, the mitigation effect is indirect, mediated through seawater CO<sub>2</sub> uptake (subsequently affecting air-sea CO<sub>2</sub> fluxes).

## ACCOUNTING ISSUES

For CBCE restoration to provide net carbon removal (negative emissions) that are valid contributions to national climate strategies (Herr and Landis, 2016; Hilmi et al., 2021) and/or used for carbon trading purposes (Ullman et al., 2013; Vanderklift et al., 2019), it is necessary for there to be additional carbon uptake and storage by such systems that is the unequivocal consequence of management action. It is also necessary for the expected magnitude of that climatic benefit to be forecast with

reasonable confidence (preferably  $\pm 10\%$ ), and for those benefits to be verifiable. Thus the projected extra CO<sub>2</sub> removal from the atmosphere has to be initially estimated, together with any other associated climatic effects (e.g., changes in fluxes of non-CO<sub>2</sub> greenhouse gases), and then reliably determined according to internationally-agreed standards (IPCC, 2014; Needelman et al., 2019; Eger et al., 2022). Furthermore, the additional carbon sequestered should be securely stored, and therefore monitored, for many decades, preferably “permanently” (CEC, 2014; Brander et al., 2021). From a climatic perspective, permanence is generally considered as  $>100$  years (Fearnside, 2002). Whilst shorter storage may help meet near-term policy targets (Ruseva et al., 2020), it does not contribute to the UNFCCC goal of long-term climate stabilization and may actually be counter-productive (Kirschbaum, 2006).

These requirements are extremely challenging for CBCE restoration. To provide confidence that they are achievable, a Verified Carbon Standard (VCS) for Tidal Wetlands and Seagrass Restoration has been developed (Needelman et al., 2018; Verra, 2021), primarily in a US context. The VCS methodology is complex and technically demanding, with  $>30$  parameters involved. To facilitate its operational application, a range of alternative sources can be used to obtain many of the basic datasets: proxies, published values, default factors, models, field measurements and historical (chrono-sequenced) data (Verra, 2021). These alternative data sources have different qualities and involve different assumptions; as a result, the intended standardization would seem compromised. The most reliable estimates for carbon removal are those directly derived from *in situ* measurements, ideally including comprehensive baseline data collected up to 4 years before the start of restoration (Verra, 2021). It may then take a further 10–20 years after the start of the restoration project before its carbon burial rates match those of adjacent mature ecosystems (Carnell et al., 2022), and the magnitude of sustained removal of additional carbon can then be determined.

The main unresolved uncertainties and risks that jeopardize the reliability of carbon accounting (and hence the associated climatic benefits of CBCE restoration) are discussed below. Although these issues are presented separately, there are many potential interactions between them. For example, much of the variability in reported carbon burial rates seems likely to be a consequence of the variability in sedimentation rates and the relative importance of bioturbation effects and of microbial decomposition. Similarly, the rate at which recalcitrant carbon increases in deeper sediments is a function of the composition (and decomposition) of recently-deposited organic carbon.

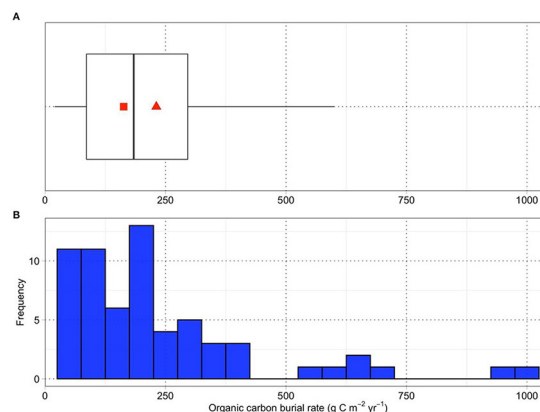
## High Variability in Carbon Burial Rates

Multiple biological, chemical and physical factors strongly influence CBCE carbon burial rates through their interacting effects on primary production, sedimentation, decomposition and preservation (McLeod et al., 2011; Macreadie et al., 2019; NASEM, 2019). As a result, there is very high variability in site-specific estimates of natural carbon burial rates: for saltmarshes, there is a 600-fold range between lowest and highest reported burial rates ( $7.7\text{--}4,693\text{ mg C m}^{-2}\text{ day}^{-1}$ ); for seagrasses, the

### BOX 1 | Low confidence in blue carbon confidence limits?

The literature review by McLeod et al. (2011) calculated global mean carbon burial rates and their standard errors to be  $226 \pm 39\text{ gC m}^{-2}\text{ yr}^{-1}$  for mangroves,  $218 \pm 24\text{ gC m}^{-2}\text{ yr}^{-1}$  for saltmarshes and  $138 \pm 38$  for seagrasses, based on 34, 96 and 123 sites respectively. This study has been highly influential ( $\sim 2,400$  citations) and was used by Griscom et al. (2017) for their coastal wetland synthesis, including associated confidence limits. Breithaupt's et al. (2012) used a closely similar primary dataset for mangrove carbon burial, and found a closely similar arithmetic mean:  $231\text{ gC m}^{-2}\text{ yr}^{-1}$ . However, their uncertainty (standard error) estimate was considerably higher, at  $\pm 209$ , as a result of high skewness (a long right tail of extreme values) in the dataset; i.e., a non-normal distribution. When recalculated from log-transformed values, Breithaupt et al.'s geometric mean for carbon burial was  $\sim 30\%$  lower, at  $163\text{ gC m}^{-2}\text{ yr}^{-1}$ , with 95% confidence range of  $131\text{--}203\text{ gC m}^{-2}\text{ yr}^{-1}$ .

The frequency distribution of Breithaupt's et al. (2012) dataset and its basic statistics are shown in **Figure 1**. Whilst we are unaware of equivalent statistical analyses for saltmarsh and seagrass data, or of more up-to-date compilations for mangroves, the problem of non-normal distributions in carbon burial rates was recognized relatively early (Duarte et al., 2005) and seems deep-seated, strongly favoring the use of geometric means or medians to determine the central tendency. This data skewness may genuinely reflect the global reality; however, the occurrence of “anomalous” high values could also arise from unrepresentative geographical sampling (Breithaupt's et al., 2012), and/or from methodological errors, potentially of more than an order of magnitude (Johannessen and Macdonald, 2016). Such issues contribute to the high variability, and possible overestimation, of the global mitigation potential of CBCE restoration (see **Table 1**), frequently calculated using the arithmetic mean values of McLeod et al. (2011).



**FIGURE 1 | (A)** Box-and-whisker plot of 65 reported estimates of the burial rates of organic carbon in mangroves compiled by Breithaupt's et al. (2012). The box represents the middle 50% of the data, between the 1st and 3rd quartiles. The median, represented by the vertical line across the box, is the middle value of the data set; half the values are greater than the median and half are less. The whiskers represent the “minimum” and “maximum” values, based on the inter-quartile range and excluding reported data considered as outliers. The red triangle and square represent the arithmetic and geometric means respectively, for all the data. **(B)** Histogram of the frequency distribution summarized in **(A)**.

range between lowest and highest is 76-fold ( $6.9\text{--}521\text{ mg C m}^{-2}\text{ day}^{-1}$ ), and for mangroves, 19-fold ( $155\text{--}2,940\text{ mg C m}^{-2}\text{ day}^{-1}$ ) (Rosentreter et al., 2021, updated to include Hatje et al., 2021). Ranges rather than means and their confidence limits are given

**TABLE 1** | Global mitigation potential estimates for CBCE restoration (excluding emission reductions through conservation).

Source	Annual mitigation potential (Gt CO <sub>2</sub> yr <sup>-1</sup> )	Percent annual mitigation potential (compared to 2020 emissions)	Cumulative mitigation potential (2025–2100) (Gt CO <sub>2</sub> )
Griscom et al. (2017):			
“Cost effective”	<b>0.202</b>	0.6%	15.2*
“Maximum potential”	<b>0.841</b>	2.4%	63.1*
Gattuso et al. (2018)	1.27**	3.9%	<b>95.4</b>
NASEM (2019): “safe”	<b>0.13</b>	0.4%	9.8*
Hoegh-Guldberg et al. (2019):			
Rate by 2030	<b>0.064–0.11</b>	0.02–0.3%	13.6–22.6***
Rate by 2050	<b>0.20–0.33</b>	0.6–0.9%	
Macreadie et al. (2021):	<b>0.621–1.064</b>	~3% (abstract)	46.6–79.8*
Annual mitigation range based on 95% confidence limits from Griscom et al. (2017)’s “maximum potential” estimates		<b>2.3–2.5% (text)</b> 1.8–3.1%	
UNEP and IUCN (2021)	1.22–2.14**	3.8–6.6%	<b>91.7–160.3</b>

The percent annual mitigation potential is scaled to 2020 emissions of CO<sub>2</sub> (34.8 Gt yr<sup>-1</sup>; Friedlingstein et al., 2022), and is assumed to be achievable by 2025. Also note that: 2020 CO<sub>2</sub> emissions were temporarily decreased by ~5% as a result of Covid-19, and annual mitigation potential could take up to a decade to achieve, by which time current annual emissions ought to be greatly reduced through UNFCCC goals and national commitments. Conversion factor: C/CO<sub>2</sub> mass ratio of 0.27 (12/44).

\*Yearly mitigation potential assumed to be constant between 2025 and 2100.

\*\*Cumulative potential has been equally distributed across 75 years.

\*\*\*Based on 10 years at 2030 rate and 65 years at 2050 rate, for lowest and highest values of the range.

Values in bold are reported figures, others are derived estimates. Estimates that are derived from, or closely similar to, the sources provided here (e.g., those in IPCC reports: Bindoff et al., 2019; Babiker et al., 2022) are not included.

here because of unresolved statistical problems relating to data distributions; see **Box 1**.

Much effort is currently being made to account for this high variability in carbon burial through process-based research and multi-factorial modeling (e.g., Morris and Callaway, 2019; Kim et al., 2022), with increasing focus on rates and time-scales rather than standing stocks—as discussed in greater detail below. However, comprehensive understanding of its causality has yet to be achieved, and progress on this front is likely to require the collection of much more site-specific environmental data, with significant cost implications for CBCE restoration projects. Until then, mean or median values derived from the global literature provide a highly uncertain default estimate for the projected outcome of any new restoration initiative. Whilst national or regional data would seem inherently more acceptable (by limiting the influence of large-scale environmental and biogeographic factors), local variability in carbon burial rates can also be very high. For example, Hatje et al. (2021) found that the variability in measured burial rates for six intertidal transects of mangrove forest in a single Brazilian estuary was as great as the global range.

## Errors in Determining Carbon Burial Rates

Most estimates of carbon burial rates in CBCEs to date have been indirect, based on the sediment carbon inventory (soil carbon stock) in near-surface layers, typically the top 1 m, rather than using direct flux measurements. However, inventory-based approaches can result in order-of-magnitude errors in carbon burial rates (Johannessen and Macdonald, 2016), unless there is also reliable information on sediment accumulation rates, derived directly or using isotopic tracers (Arias-Ortiz

et al., 2018a; Callaway, 2019; Jennerjahn, 2020). Such accretion rates may have changed markedly in recent decades due to anthropogenic and climatic influences on local water flows and sediment delivery (Ladd et al., 2019). In estuaries, these parameters will be strongly influenced by upstream land-use and water quality changes at catchment-wide scale. Since restoration projects are almost always located at sites that have been subject to substantive anthropogenic disturbance, interpretation of their sediment core data (and also from nearby “control” sites) requires particular care.

Bioturbation (sediment mixing by burrowing fauna) can significantly compromise tracer-based measurement of sediment accumulation, and hence carbon burial rates, resulting in the linked over-estimation of these parameters (Silverberg et al., 1986). Microbial decomposition of organic carbon can also continue, albeit slowly, in more recently-deposited sediment layers. In seagrass sediments, both these processes can occur to ~40 cm depth. If not measured or otherwise allowed for (Gardner et al., 1987), each effect can result in an over-estimation of carbon burial rates by 50–100% (Johannessen and Macdonald, 2016).

When restoration is carried out, initial measurements of organic carbon in sediment profiles will primarily provide information on carbon deposition, rather than long-term carbon burial. This effect is similar to the achievement of steady state conditions for above-ground biomass; both processes may take several years, if not decades, to (re-)establish the sustained carbon dynamics of the mature system. The time-dependence of such processes should, ideally, be determined on a site-specific basis (Carnell et al., 2022).

## Lateral Carbon Transport

For CBCE-based climate mitigation, it is necessary to know the source of the carbon buried in coastal sediments, with subsequent exclusion of most, if not all, of the allochthonous (non-local) carbon originating from terrestrial or atmospheric sources, or other marine ecosystems. That is because the CBCE did not remove allochthonous carbon from the atmosphere (neither directly nor indirectly), and its long-term storage may have occurred anyway. The contributions of different organic materials to long-term carbon burial in a CBCE restoration site can be estimated by identifying their taxonomic origins using “fingerprinting” techniques (Geraldi et al., 2019), thereby determining the proportion of the total that can be ascribed to the restoration action. The criteria for inclusion can be wider than just material arising from the dominant, habitat-structuring species; for example, benthic microalgae can be considered as part of a mangrove ecosystem, and therefore their contribution to carbon storage is autochthonous rather than allochthonous. Even for non-local carbon, full exclusion may not be necessary if it can be shown that its decomposition has been significantly slowed or prevented by the anoxic conditions of CBCE sediments. However, this factor introduces additional site-specific uncertainties that cannot easily be resolved.

The proportion of carbon buried in CBCE sediments that is allochthonous is highly variable. For mangroves, allochthonous carbon has been estimated as 24–55% of the total for a range of mangrove sites in Vietnam (Hieu et al., 2017), 3–73% in China (Xiong et al., 2018), and 59–79% in Ecuador (Suello et al., 2022). For seagrasses, the allochthonous contribution to carbon burial can be as high as 70–90% in Australian estuaries (Ricart et al., 2020). For saltmarshes, allochthonous sources may also dominate (Saintilan et al., 2013; Van de Broek et al., 2018), although again with high variability. For example, the relatively low proportions of 12 and 39% were estimated for two saltmarsh sites in Germany (Mueller et al., 2019), and of 11–27% in undisturbed and 58–68% in restored US saltmarsh sites (Drexler et al., 2020).

Recalcitrant black carbon, including soot from burning biomass or fossil fuels, can be an important component of allochthonous carbon, comprising 9–25% of total carbon burial in a range of deltaic CBCEs in China (Li et al., 2021) and up to 43% in sandy seagrass environments in Australia (Chew and Gallagher, 2018).

Most of the above values are depth-averaged, including near-surface layers. Such measurements are, however, likely to underestimate the contribution of allochthonous carbon to long-term carbon storage (and hence further overestimate the climatic effectiveness of CBCE restoration). That is because the locally-derived organic carbon can be expected to be fresher and more labile, decomposing more rapidly than material from elsewhere, likely to be mostly land-derived (at least in estuaries) and either recalcitrant or already partly re-mineralized (Van de Broek et al., 2018). Thus a typical 50:50 ratio of allochthonous to autochthonous carbon in near-surface saltmarsh deposits may change to a 80:20 ratio for more deeply-buried buried carbon (Leorri et al., 2018; Van de Broek et al., 2018). Similar effects can be expected for mangrove and seagrass sediments, but have not to our knowledge been investigated.

The rate at which allochthonous carbon proportionally increases with sediment depth can be expected to vary between sites, since it will be affected by site-specific sedimentation rates and bioturbation processes.

A counteracting consequence of lateral carbon transport (that would result in an underestimation of climatic benefits) is that there is also likely to be significant carbon export from CBCEs, a proportion of which may be subject to long-term storage, either as dissolved inorganic or organic carbon in deep ocean water, or as particulates that are buried in other depositional systems. The scale of these export processes may be as great as, or exceed, direct carbon burial (Maher et al., 2018; Santos et al., 2019, 2021). However, export cannot be directly equated with long-term sequestration, and the decomposition rates and fate (on decadal time-scales) of carbon transported from the coast to very large areas of the open ocean is poorly constrained, spatially heterogeneous and difficult to quantify (Legge et al., 2020). Furthermore, it is relevant to note that the initial identification of the importance of CBCEs in the global carbon cycle was based on the estimate that around 50% of carbon burial in the global ocean occurred in their sediments (Duarte et al., 2005).

Cost considerations mean that it is very unlikely that the scale of the potential benefits arising from carbon export and subsequent long-term sequestration from restored CBCEs could be reliably determined through their routine monitoring.

## Fluxes of Methane and Nitrous Oxide

The anaerobic conditions in CBCE sediments that are responsible for long-term carbon storage also favor the production and emissions of two potent greenhouse gases of increasing climatic concern: methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O; Rosentreter et al., 2021). Contrary to previous indications (Poffenbarger et al., 2011) and assumptions (IPCC, 2014), saline conditions do not fully inhibit CH<sub>4</sub> production in coastal sediments (Conrad, 2020; Marchand et al., 2022). Such emissions can show high temporal variability (Roth et al., 2022); furthermore, biogenic structures such as bioturbation burrows can enhance CH<sub>4</sub> fluxes by an order of magnitude (Kristensen et al., 2022), and CH<sub>4</sub> can be released directly to the air from plant stems (Zhang et al., 2022).

Total global CH<sub>4</sub> emissions from CBCEs have been estimated at >5 million tons yr<sup>-1</sup> (Al-Haj and Fulweiler, 2020). This total has the potential to fully counteract the climatic benefits of carbon burial by these ecosystems. However, statistical methods affect these estimates and comparisons, with lower global values for CH<sub>4</sub> emissions obtained when based on the median, rather than the arithmetic mean, of site-specific studies (Rosentreter and Williamson, 2020). As discussed in **Box 1**, the arithmetic mean is best avoided for highly skewed distributions.

Further methodological uncertainties relate to the timescales and assumptions used to estimate the global warming potential (GWP) of CH<sub>4</sub>, for comparison of its warming effects to those of CO<sub>2</sub>. Because of the relatively short atmospheric lifetime of CH<sub>4</sub>, higher GWP values apply when considering medium-term mitigation targets (e.g., Nationally Determined Contributions by 2030, or net zero by 2050) rather than global radiative flux budgets in 2100. For long-established natural CBCEs, their CH<sub>4</sub> emissions (even if high) can have neutral radiative



balance, since such emissions are counteracted by steady-state atmospheric removal processes, i.e., they do not result in any further atmospheric accumulation (Neubauer and Verhoeven, 2019).

Key issues for CBCE restoration is therefore whether that action increases  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions (likely from new habitat formation, e.g., saltmarsh creation) or decreases them, and the magnitude of those changes. Decreases could occur when pre-restoration conditions are strongly methanogenic, e.g., rice paddies, shrimp ponds or wet pastures (Chauhan et al., 2017; Yang et al., 2017; Iram et al., 2021) or might be achieved if restoration involves increasing water flows to existing habitat (Kroeger et al., 2017). Nevertheless, it is invalid to assume that restoration will necessarily reduce  $\text{CH}_4$  emissions, or to assume there will be no effect, since there is also potential for increased  $\text{CH}_4$  emissions to significantly offset future climate benefits (Rosentreter et al., 2021).

Emissions of  $\text{N}_2\text{O}$  from CBCEs seem climatically less important than  $\text{CH}_4$ , and uptake can also occur as a result of denitrification when nitrate availability is low (Foster and Fulweiler, 2016). However, the net global  $\text{N}_2\text{O}$  efflux from CBCEs is not well-constrained (Wilson et al., 2020; Rosentreter et al., 2021), with high spatial and temporal variability resulting in a 40-fold range between upper and lower quartiles for estimated global values for the three ecosystems combined (Murray et al., 2015). Although  $\text{N}_2\text{O}$  and  $\text{CH}_4$  have similar radiative forcing effects on a weight-for-weight basis,  $\text{N}_2\text{O}$  has a much longer atmospheric lifetime, resulting in a 100 year GWP (for a pulse emission, with feedbacks) around 9 times higher than  $\text{CH}_4$ , and nearly 300 times higher than  $\text{CO}_2$  (Myhre et al., 2013). As a result, relatively small changes in  $\text{N}_2\text{O}$  emissions can significantly affect the climatic benefits of CBCE restoration.

Long-term site-specific monitoring of both  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes would therefore seem necessary for CBCE restoration for climate mitigation purposes, with sufficient baseline data to determine the changes arising from restoration. Site-specific knowledge of previous land-use changes (i.e., when the historical habitat degradation or loss occurred) is also relevant to  $\text{CH}_4$  emissions, since associated “switchover times” (Neubauer and Verhoeven, 2019) affect the scale of net radiative warming or cooling prior to the restoration, and hence the magnitude and direction of the change arising from management action. Proxy measurements (salinity, hydrology, and plant community composition) have been proposed as alternatives to direct measurements of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes at restoration sites to reduce costs (Derby et al., 2022); their reliability and wider effectiveness have yet to be demonstrated.

## Carbonate Formation and Dissolution

Calcium carbonate ( $\text{CaCO}_3$ ) precipitation (including the biological process of calcification by corals, many other benthic invertebrates and coralline algae) releases  $\text{CO}_2$ , whilst its dissolution has the opposite effect. Both these processes can occur in CBCEs (Macreadie et al., 2017b; Howard et al., 2018; Kennedy et al., 2019), and there is ongoing debate regarding the overall direction and magnitude (and hence climatic consequences) of such effects.

Uncertainties are greatest for mangrove and seagrass habitats that accumulate relatively high levels of biogenic  $\text{CaCO}_3$  in their sediments. If the initial stage of calcium carbonate formation occurred centuries earlier or took place elsewhere, e.g., adjacent coral reefs, the  $\text{CO}_2$  emissions associated with calcification cannot be ascribed to the CBCE, whether natural or restored. Under those circumstances, calcium carbonate dissolution occurring within CBCE sediments has the potential to significantly enhance their climate mitigation role (Saderne et al., 2019). In Red Sea mangroves with relatively low organic carbon burial rates, such mitigation enhancement is estimated to be as much as 23-fold (Saderne et al., 2021). The opposite effect has been reported for one of the largest seagrass-dominated ecosystems in the world, in Florida Bay, USA. At that site, local calcification apparently exceeds dissolution; as a result,  $\text{CO}_2$  emissions are estimated to be around three times  $>\text{CO}_2$  removal through the burial of organic carbon (Van Dam et al., 2021). This unexpected outcome was shown by an atmospheric eddy covariance technique that directly measures air-water  $\text{CO}_2$  exchanges; it contradicts earlier evidence for net carbonate dissolution, based on water sampling and benthic chambers, with less extensive spatial and temporal coverage (Van Dam et al., 2019). However, there may be a contribution to  $\text{CO}_2$  emissions from high- $\text{CO}_2$  groundwater discharge, that has been shown to be important elsewhere (Sadat-Noori et al., 2016).

Whilst both the Red Sea and Florida sites may be considered untypical, for different reasons, they show that the calcium carbonate dynamics can potentially override the climatic role of organic carbon burial in CBCEs. Such effects have been neglected to date by standard blue carbon accounting, such as the VCS methodology (Verra, 2021). There is now urgent need to determine their wider applicability and implications for CBCE restoration.

## Vulnerability to Future Climate Change

The effects on CBCEs of future warming (including marine heatwaves), sea level rise, increased storminess and ocean acidification have been assessed by IPCC (Bindoff et al., 2019; Cooley et al., 2022) and elsewhere (Macreadie et al., 2019; Lovelock and Reef, 2020; Williamson and Guinder, 2021). Overall effects are considered to be damaging, threatening the continued viability of such ecosystems (Bindoff et al., 2019). Nevertheless, Macreadie et al. (2019) identified 11 potentially positive climate change effects for mangrove, saltmarsh and seagrass ecosystems considered separately, in addition to 11 potentially negative effects and 11 effects where the outcome was uncertain. The main factor that could result in beneficial effects is landward range expansion, with a potential increase of 1.5 GtC in net global carbon storage by 2100 (Lovelock and Reef, 2020) for the high emissions scenario, RCP 8.5. If such landward movement is not possible, losses of 3.4 GtC by 2100 were estimated as the worst-case scenario.

Seagrasses are considered the CBCE that is most sensitive to higher temperatures (Nguyen et al., 2021), particularly episodic extremes: their ecological structure and functioning are already subject to moderate impacts, attributed to warming with medium confidence (Bindoff et al., 2019). High impacts for seagrasses

are projected if global mean surface temperatures increase by  $> \sim 2.3^{\circ}\text{C}$  relative to pre-industrial, likely to involve the loss of their carbon stores (Arias-Ortiz et al., 2018b; Chefaoui et al., 2018). For saltmarshes, the thresholds for moderate and high impacts are estimated to be global temperature increases of  $\sim 1.2$  and  $3.1^{\circ}\text{C}$ , respectively; for mangroves,  $\sim 2.0$  and  $3.7^{\circ}\text{C}$  (Bindoff et al., 2019). Although mangroves therefore seem relatively resilient, severe and widespread mortalities have already occurred in Australia, linked to drought conditions, unprecedented high temperatures and a temporary drop in sea level (Duke et al., 2017).

The possibility of range expansion provides a counteracting effect, with expansion of mangroves into saltmarsh habitat already occurring on several continents (Saintilan et al., 2014; Cavanaugh et al., 2019). For CBCE restoration sites, the consequences of climate-driven species replacements are difficult to predict. Whilst they might be either increase or decrease carbon burial rates, the latter would seem inherently more likely. Future warming impacts for restored CBCEs could however be minimized, although not completely averted, if restoration effort is focused on the higher latitude (i.e., cooler) parts of the distributional ranges of the species that structure such ecosystems.

The implications of future changes in sea level rise, storm events, wave energy, and sea level rise for CBCEs are not well-understood, and are likely to show greater local and regional variability. Thus, vulnerability—and resilience—will also be affected by sediment erodibility and sediment resupply, as well as by vegetation type, the frequency of extreme events, the rate of local sea level rise, and whether there is space for landward relocation (Schuerch et al., 2018; Valiela et al., 2018; Rogers et al., 2019; Hanley et al., 2020). The possibility of landward migration would seem inapplicable to most CBCE restoration projects, unless land suitable for such re-location is either initially included or can be added later; either option would have significant cost implications.

## Vulnerability to Non-climatic Factors

Assuming (optimistically) that climate change impacts can be minimized by limiting global warming to  $\sim 1.5^{\circ}\text{C}$ , then would still be many non-climatic factors that might adversely affect the viability and long-term survival of restored CBCEs. The success of such restoration projects has been variable to date (Cunha et al., 2012; van Katwijk et al., 2016; Zhao et al., 2016; Kodikara et al., 2017; Duarte et al., 2020). A key issue is whether the (human) factors causing original loss/degradation or land use conversion have been properly addressed. The concept of social-ecological restoration (Abelson et al., 2020) is applicable here, taking account of all relevant anthropogenic drivers and decision-making processes, from local to international level. These include “opportunity costs” (not usually included in the restoration costs discussed below); i.e., the potential economic benefits that are lost when CBCE restoration excludes other uses of the coastal land, for agriculture, aquaculture, industry, or settlement (Stewart et al., 2003; Herr et al., 2019; Zeng et al., 2020).

Other causes for CBCE restoration failures include poor site selection or poor choice of species introduced, particularly for

saltmarshes (Konisky and Burdick, 2004; Berck and Gustafson, 2012). Larger-scale plantings, natural regeneration, and strong local stakeholder engagement all increase the likelihood of long-term survival for the restored system (Bayraktarov et al., 2016; van Katwijk et al., 2016). Additional “lessons learnt” from greater implementation of CBCE restoration projects can be expected to increase their future success (Wylie et al., 2016). There is also evidence for natural recovery of CBCEs due to reduced anthropogenic pressures, at least at the local and regional scale (Almeida et al., 2014; Jia et al., 2018; de los Santos et al., 2019; Duarte et al., 2020). Nevertheless, adequate resourcing for long-term monitoring and strong protection of restored CBCE habitats will be needed, to ensure that they function as intended, for long enough to deliver their expected climate mitigation benefits.

## CLIMATIC COST-EFFECTIVENESS AND SCALABILITY

The cost-effectiveness of using CBCEs for climate mitigation through carbon removal is considered here in terms of US\$ per ton of  $\text{CO}_2\text{e}$  removed from the atmosphere as a result of restoration action carried out for climate mitigation purposes, either as an integral part of national climate policy (Gallo et al., 2017; Kelleway et al., 2020) or primarily for carbon trading purposes (Vanderklift et al., 2019; Sapkota and White, 2020). The cost-effectiveness for CBCE restoration can also be determined more holistically, based on both climatic and non-climatic benefits; that wider context is briefly considered in Discussion and Conclusions below. Whichever approach is taken, many uncertainties affect the determination of cost-effectiveness, limiting the usefulness of CBCE restoration as a reliable and significant component of climate policy.

These uncertainties not only relate to the accounting issues discussed above, but also due to the very high variability in restoration costs, both between and within the three main vegetation types. Based on 91 studies of CBCE restoration (environmental improvements, re-planting and new habitat creation, as considered here), Bayraktarov et al. (2016) estimated that median total restoration costs for mangroves were US\$  $2,500\text{ ha}^{-1}$ ; for saltmarsh, US\$  $151,100\text{ ha}^{-1}$  and seagrasses, US\$  $383,700\text{ ha}^{-1}$  (2010 prices). The wide range of costs within each CBCE is shown by their arithmetic mean values being 2–7 times higher than their medians, at US\$  $15,000\text{ ha}^{-1}$ , US\$  $1,042,100\text{ ha}^{-1}$ , and US\$  $699,500\text{ ha}^{-1}$ , respectively. Causes for cost variability included: whether or not monitoring costs were included; the differences in labor costs between developed and developing countries (responsible for a  $\sim 30$ -fold difference in median values for mangroves); and the planting method (responsible for an 8-fold difference for saltmarshes).

Taillardat et al. (2020) combined data on CBCE restoration costs (expressed annually) with estimated  $\text{CO}_2$  removal and  $\text{CH}_4$  emission rates: they estimated global median costs of US\$  $469,100\text{ t}^{-1}\text{ CO}_2\text{e}$  for climate mitigation using saltmarsh restoration and US\$  $560\text{ t}^{-1}\text{ CO}_2\text{e}$  for mangrove restoration. There was insufficient data to estimate equivalent values for seagrasses. Very

**TABLE 2 |** Summary of biogeochemical issues affecting the reliability of using CBCE restoration for climate mitigation.

Issues and factors	Potential effect on realized climate mitigation		Relative cost to resolve	Comments
	Decrease	Increase		
Carbon burial rates				
<i>Large range of global data</i>	<<	>>	\$	Need for site-specific data
<i>Inappropriate statistics</i>	<		-	Data skewness effects
<i>Indirect C flux estimates</i>	<<		\$	Reliable methods are complex and demanding
Lateral carbon transport				
<i>C imports (allochthony)</i>	<		\$	Need for site-specific data
<i>C exports</i>		>	\$\$	
Other greenhouse gases				
<i>CH<sub>4</sub> and N<sub>2</sub>O</i>	<<	>	\$	Need for site-specific data
Carbonate dynamics				
<i>CaCO<sub>3</sub> formation</i>	<<		\$	Need for site-specific data
<i>CaCO<sub>3</sub> dissolution</i>		>>	\$	
Climate change impacts				
<i>Direct temperature effects</i>	<<		\$	Need for long-term monitoring; effects are scenario-dependent
<i>Sea level rise</i>	<	>	\$	
Other factors affecting restoration success				
<i>Range of direct and indirect human pressures</i>	<<		\$\$	Need for long-term site protection

The direction and relative magnitude of the potential effects of specific factors on realized climate mitigation over the next 20–50 years are based on information given in text. Two categories are given for magnitude: low-to-medium (single chevron) and high (double chevron). For increases, high includes the possibility that site-specific specific climatic benefits might be more than doubled; for decreases, that they may be reduced by an order of magnitude relative to what might be expected from global estimates, e.g., McLeod et al. (2011) and derived analyses. The potential cost to resolve these uncertainties is also indicated on a relative basis, as low-to-medium (\$) or high (\$\$).

large differences in median costs were confirmed for mangrove restoration between developing and developed nations (at US\$ 990 ha<sup>-1</sup> yr<sup>-1</sup> and US\$ 100,860 ha<sup>-1</sup> yr<sup>-1</sup>, respectively); global cost estimates should therefore not be used as site-specific default values.

Griscom et al. (2017) applied a different approach for their cost-effectiveness analysis, using a Marginal Abatement Cost (MAC) curve to estimate that 24% of lost or degraded coastal wetlands could be restored at a target cost of less than US\$ 100 t<sup>-1</sup> CO<sub>2</sub> (quoted as CO<sub>2</sub>e to indicate C to CO<sub>2</sub> conversion, but without allowing for CH<sub>4</sub> emissions). Their Supplementary Data showed that this proportion was based on 30% of “maximum restoration with safeguards” for the area of lost mangrove habitat and 60% of the area of lost saltmarsh, but no seagrass restoration (since the MAC-assessed cost of seagrass restoration was unable to meet the <US\$ 100 criterion). There could, however, be bias in Griscom et al.’s estimates, that are based on the reported cost range of CBCE restoration projects to date. These initiatives are highly likely to have favored relatively straightforward and successful situations (Bayraktarov et al., 2016), thus their costs are likely to be unrepresentative (and lower) than average for the total restorable area where CBCE losses have occurred. The “safeguards” used by Griscom et al. (2017) did not seem to address this issue: their exclusions for cropland and timber production are generally inapplicable to CBCEs, where habitat losses are frequently due to mariculture, major hydrological changes, pollution, the construction of hard sea defenses and coastal development related to tourism, port infrastructure and

human settlement. For most coastal development, restoration costs are prohibitively high; as a result, such data are not included in the reported cost range.

In South East Asia, the region often considered as a coastal blue carbon “hotspot” with high restoration potential for mangroves (Thorhaug et al., 2020), a combination of financial, land-use and operational factors were estimated to limit reforestation (of all kinds) to 0.3–18% of the maximum theoretically possible (Zeng et al., 2020). Macreadie et al. (2021) recognized this scalability problem: “The scope for global-scale coastal wetland restoration is constrained by multiple socio-economic considerations. This constraint is especially important in countries where a large proportion of the restorable habitat is on small agricultural land holdings, where restoration efforts could conflict with livelihoods and food security of local communities.” Nevertheless, Macreadie et al. (2021) used Griscom et al.’s (2017) values for maximum restoration, without any cost considerations, as the basis for their estimates of the potential climatic benefits of CBCE restoration (see Table 1).

## DISCUSSION AND CONCLUSION

All IPCC pathways that limit global warming to 1.5°C with limited or no overshoot use negative emissions with a cumulative total of 100–1,000 Gt CO<sub>2</sub> over the twenty-first century (IPCC, 2018). Coastal and ocean-based CO<sub>2</sub> removal has attracted attention because there are multiple feasibility and sustainability

constraints for land-based mitigation (Williamson, 2016), and the ocean has a very large potential for carbon uptake and storage due to its huge surface area and volume as well as distinct chemical characteristics.

Nature based solutions involving marine processes, such as CBCE restoration, are attractive not only for climate mitigation but also in the context of their other benefits, that include improved food security, reduced coastal erosion, and rebuilding marine biodiversity. They also enjoy stronger public support than ocean-based technological measures regarded as geoengineering (Bertram and Merk, 2020), although the natural/artificial distinction is arguably over-simplistic (Osaka et al., 2021). **Table 1** gives estimates by different studies of the potential climatic effectiveness of CBCE restoration, considered as the additional CO<sub>2</sub> that might be removed and stored per year with a ramp-up time of a few years. The range of these estimates is large, from 0.06 to 2.1 Gt CO<sub>2</sub> per year, equivalent to 0.02 to 6.6% of 2020 CO<sub>2</sub> emissions (Friedlingstein et al., 2022), reflecting the key uncertainties reviewed above.

Several of the effectiveness estimates are upper theoretical limits that are unrealistic for policy planning and implementation. For example, the relatively high estimate of Gattuso et al. (2018) assumes that historic losses of blue carbon ecosystems (Waycott et al., 2009; McLeod et al., 2011; estimated as 50% for mangroves since the 1940's, 29% for seagrass since 1879, 25% for saltmarsh since the 1800's) can all be restored or compensated for elsewhere, and uses arithmetic mean values for global carbon burial rates (McLeod et al., 2011). The former assumption is not feasible, since a large fraction of surface area of lost CBCEs, particularly mangrove and saltmarsh habitats, has been developed in ways that would make restoration prohibitively expensive and/or societally unacceptable (e.g., for settlement, port facilities or tourism infrastructure). Furthermore, as discussed above, carbon burial rates seem likely to be overestimated; fluxes of other greenhouse gases and carbonate dynamics are not taken into account; and the long-term security of storage may be jeopardized by future climate change or direct anthropogenic impacts.

**Table 2** summarizes the specific uncertainties discussed in this paper that affect carbon accounting and wider effectiveness of using CBCE restoration for climate mitigation. On the basis of the groupings given, most effects are directional: six decrease and two increase the potential for CBCE-based climate mitigation, whilst three could have effects in either direction.

The above concerns do not mean that CBCE restoration cannot be worthwhile, and do not contradict Gattuso et al.'s (2018, 2021) assessments that CBCE restoration is a “low regret” measure. The underlying rationale of that conclusion is that, despite low effectiveness in increasing carbon removal (cumulative removal of, at most, 2–3 years of current CO<sub>2</sub> emissions by 2100; **Table 1**), CBCE restoration has a high level of technological readiness, high governability at the local scale, high-to-very-high levels of co-benefits, and a low-to-very-low level of disbenefits (Gattuso et al., 2021; Babiker et al., 2022). Measures from the “low regret” cluster were therefore recommended by Gattuso et al. (2021) as high priority for

#### BOX 2 | Caution needed for blue carbon comparisons.

Comparisons of CBCE carbon stocks (that are relatively easy to measure) with their carbon burial rates (that are not) can be misleading (Jennerjahn, 2020). An inadvertent example is provided by Macreadie et al. (2021), where the accumulation of sediment carbon resulting from widespread mangrove restoration in the Mekong Delta is stated as being climatically equivalent to three times Vietnam's 2013 greenhouse gas emissions. That claim is based on Dung et al. (2016), who measured depth-profiled carbon stock in 2.5 m sediment cores at a single cluster of sites in 2013.

However, sediment accretion rates were not determined, and the “accumulation” value would only be valid if there was zero carbon before restoration began (in 1978). Initial sediment carbon stocks do not seem to have been measured, but nearby unforested mudflats might reasonably be considered to provide “control” values for comparative purposes. On that basis, the carbon increase, and associated climate benefit, would be reduced to 7.1% of claimed values over the 35 years of the restoration project—hence 0.20% when expressed as an annual rate. Further scaling-down would seem necessary due to: the presence of non-local (allochthonous) carbon in the sediment; the non-permanent storage of organic carbon in upper layers; and the possibility of enhanced CH<sub>4</sub> and N<sub>2</sub>O emissions. Taking account of just the first factor could result in the actual annual benefit being 0.12% of the amount claimed (assuming the proportion of allochthonous carbon is 0.4, a mid-range value as measured at another Vietnamese mangrove restoration site; Hieu et al., 2017). Overall, these considerations could therefore reduce the climatic benefits of the mangrove restoration by as much as two orders of magnitude, from three times Vietnam's 2013 emissions to around 0.3% of those emissions, when both rates are expressed annually (although subject to several unresolved uncertainties).

Nevertheless, wetland habitat restoration in the Mekong Delta, and its long-term protection, can be considered worthwhile for a wide range of other reasons.

implementation, recognizing that they may prove to be more advantageous at the national, rather than global, level (Herr et al., 2018; Taillardat et al., 2018).

Yet caution is clearly needed, even at the national level (**Box 2**). The many uncertainties associated with using CBCE restoration for carbon removal (**Table 2**) mean that there is a very real risk of non-delivery of the expected climatic benefits. For that reason, and because of the well-recognized seriousness of failure to meet national and global mitigation targets in the near future, we consider that it is premature to “operationalize” marketable blue carbon (Macreadie et al., 2022) with associated effort to include CBCE restoration in carbon offset trading (Claes et al., 2022). CBCE restoration should therefore be in addition to, not as a substitute for, near-total emission reductions. Where CBCE restoration projects are carried out primarily for their intended climatic benefits, they need to include comprehensive long-term monitoring and protection, involving additional costs.

Additional governance precautions are also likely to be necessary (Jakovac et al., 2020). Such precautions include the avoidance of loopholes, mis-reporting and perverse incentives (Climate Analytics, 2017), as have widely occurred when complex financial incentives have been used as part of UNFCCC's REDD+ mechanism under the Kyoto Protocol (Correa et al., 2019; Badgley et al., 2021) with its goal of reducing emissions from deforestation and forest degradation in developing countries.



## AUTHOR CONTRIBUTIONS

PW and J-PG drafted and finalized the content of this paper. All authors contributed to the article and approved the submitted version.

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# Geochemical Negative Emissions Technologies: Part II. Roadmap

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Geochemical negative emissions technologies (NETs) comprise a set of approaches to climate change mitigation that make use of alkaline minerals to remove and/or permanently store carbon dioxide (CO<sub>2</sub>) as solid carbonate minerals or dissolved ocean bicarbonate ions. This roadmap accompanies the comprehensive review of geochemical NETs by the same authors and offers guidance for the development and deployment of geochemical NETs at gigaton per year (Gt yr.<sup>-1</sup>) scale. We lay out needs and high-priority initiatives across six key elements required for the responsible and effective deployment of geochemical NETs: (i) technical readiness, (ii) social license, (iii) demand, (iv) supply chains, (v) human capital, and (vi) infrastructure. We put forward proposals for: specific initiatives to be undertaken; their approximate costs and timelines; and the roles that various actors could play in undertaking them. Our intent is to progress toward a working consensus among researchers, practitioners, and key players about initiatives that merit resourcing and action, primarily focusing on the near-term.

## KEYWORDS

carbon dioxide removal, enhanced weathering, CO<sub>2</sub> mineralization, ocean alkalinity enhancement, geochemical CDR, CDR roadmap, negative emissions technologies, negative emissions roadmap

## Introduction

The recent IPCC AR6 report reflects a broad consensus that, even under optimistic decarbonization scenarios, a non-trivial amount of negative emissions will be necessary within a generation to minimize catastrophic warming, irreversible damage to ecosystems, and reduced quality of life (Shukla et al., 2022). Geochemical NETs are a set of technologies that use alkaline minerals for removing and/or permanently

storing atmospheric CO<sub>2</sub> (Campbell et al., 2022). Research suggests that geochemical NETs offer potential scalability within time periods relevant to climate targets, an ability to simultaneously capture and store CO<sub>2</sub> (Campbell et al., 2022) and an ability to permanently store CO<sub>2</sub> away from the active carbon cycle (Carton et al., 2021). However, the possible externalities and impacts of geochemical NETs have not been fully explored, nor rigorously tested in the field. A rapid large-scale deployment of geochemical NETs as they stand today could therefore be accompanied by unacceptable environmental or social burdens. Moreover, geochemical NET projects will take time to site and scale. In our view, this suggests there is no time to waste: we must fast-track the research, development, and stakeholder engagement required for potential deployment of geochemical NETs at scale.

In our review of the current body of knowledge on geochemical NETs, various gaps were identified between the present state of research and activity, and what is required for a fully functioning, effective, safe and responsible deployment at the scale of megatons (Mt), and eventually gigatons (Gt), annual removal of atmospheric CO<sub>2</sub> (Campbell et al., 2022). Here, we provide suggestions for closing those gaps in the form of a roadmap for the responsible and expeditious development and deployment of geochemical NETs. We begin with a needs assessment, outline opportunities at the research and innovation frontier, and then present recommendations covering initiatives, approximate resource needs, and roles for different actors, including researchers, governments, startups, industry, investors, philanthropists and non-profits.

## Needs assessment

A safe, high-functioning, Gt yr.<sup>-1</sup> scale ecosystem for geochemical NETs would require at least six supporting elements: technical readiness, social license, demand, supply chains, human capital, and infrastructure. The gaps between what is needed across these elements and what exists today are profound.

## Technical readiness

Geochemical NETs harness chemical reactions between alkaline minerals and CO<sub>2</sub> to form stable carbonate compounds or dissolved ocean bicarbonate anions. Approaches include CO<sub>2</sub> mineralization (Power et al., 2013; Sanna et al., 2014), enhanced weathering (Renforth et al., 2015; Montserrat et al., 2017; Rigopoulos et al., 2018), electrochemical seawater splitting (de Lannoy et al., 2018; Eisaman et al., 2018), ocean liming (Renforth and Henderson, 2017; Caserini et al., 2021), and hybrid direct air capture (DAC) systems utilizing solid minerals (McQueen et al., 2020).

Further research and development (R&D), including additional studies across all geochemical NETs in different environments, are critical to achieving requisite technology readiness levels (TRLs) for deployment. Current TRLs for *in situ* mineralization approaches range from 2 to 6 (i.e., from “technology concept formulated” to “technology demonstrated in relevant environment” on a scale of 1–9) (Kearns, 2021). Other techniques, such as enhanced weathering in soils and ocean alkalinity enhancement, rank even lower (e.g., TRL 1; “basic principles observed”). Substantial work is required in the laboratory, in small-scale field studies, and finally larger scale demonstration studies in industrial settings, before geochemical NETs achieve TRL 8 or TRL 9, required for deployment.

There are significant gaps in particular in our understanding of the underlying kinetics of geochemical NETs, the effects of secondary minerals and their re-dissolution, and their impacts on ecosystems, the environment, and public health, as well as pore clogging and cracking for *in situ* methods (Kelemen et al., 2019; Fuhr et al., 2022). Work is required to demonstrate that geochemical NETs can be effective, reliable, and safe, to a high degree of confidence, and to lay the foundation for robust methods for measurement, monitoring, and verification (MMV) that can support result-based investment decisions, such as carbon credit procurement.

## Social license

The active removal of Gt of CO<sub>2</sub> from the atmosphere, equivalent in scale in terms of infrastructure and resource use to major global industries, will require non-tacit acceptance from wide publics, stakeholders, and policymakers. Today, public awareness of geochemical NETs is generally low. The public's views on these approaches are at a formative stage and are beginning to be investigated by researchers (Cox et al., 2020; Spence et al., 2021).

This provides a challenge for the geochemical NETs research community in how it develops its research agenda with growing exposure of the field. An opaque research effort led primarily by commercial actors, effectively isolated from stakeholders and wider publics, may struggle to secure broad-based, durable support from the public and policymakers. In contrast, co-development through principles of responsible research and innovation (Owen et al., 2012) may provide the means by which the eventual costs, benefits, and other trade-offs of scaled-up approaches are accurately defined, broadly understood, and equitably shared.

Awareness among policy makers is also generally low and compared to other NET approaches geochemical NETs have the lowest ratio of policy coverage to potential impact, highlighting a further gap (Sandalow et al., 2021). Most relevant existing policy frameworks (e.g., 45Q, California Low Carbon Fuel Standard, EU Emissions Trading System

[Federal Register, 2021; EU Emissions Trading System (EU ETS), 2015; Low Carbon Fuel Standard]] include criteria that leave out geochemical NETs unintentionally. Furthermore, international agreements disallow the release of alkaline minerals into the ocean beyond contained research experiments (Verlaan, 2013), constraining the deployment of ocean-based geochemical NETs. New frameworks for projects involving multiple jurisdictions and/or the oceans are of particular priority. Overall, for geochemical NETs, gaps in demand, infrastructure and other areas will not be met without policy intervention on local, national and international levels, which in turn will be reliant on our ability to address gaps in social license.

## Demand

Today, demand for geochemical NETs is generated primarily through voluntary carbon markets and corporate commitments (Battersby et al., 2022). Demand for durable forms of carbon removal has been increasing, with the most significant recent commitment being the \$925 million advance market commitment by Frontier (2022). Geochemical NETs offer permanent CO<sub>2</sub> storage and may be competitive applicants for such funding (Joppa et al., 2021). However, the potential volume of these voluntary funding flows is unknown, and they may be unsuitable funding mechanisms for some of the work that needs to be done. For example, voluntary payments to commercial companies for carbon credits or carbon removal delivery may well be insufficient to support geochemical NETs at a Gt or even Mt-scale, and an inappropriate or ineffective tool for funding research, development, and stakeholder engagement in a manner that is open, inclusive, robust, and aligned with the principles for responsible research and innovation.

Given this, philanthropic and public sources of funding and demand must be increased dramatically, if geochemical NETs are to be researched in a robust manner and then deployed at meaningful scale. Specifically, government procurement of carbon removal from geochemical NETs and/or the inclusion of geochemical NETs into compliance-based regimes more significant than those that exist today will be necessary (Rickels et al., 2021). Geochemical NETs have considerable potential scalability, coupled with tangible durability of the resulting minerals (Lackner, 2003), and as such, we anticipate that significant demand for durable carbon removal outcomes generally would translate into significant demand for geochemical NETs specifically, particularly as other NETs that are more established but perhaps limited in their potential scale, such as BECCS, saturate.

Beyond demand for the carbon removal outcomes generated by geochemical NETs, there may be opportunities to expand demand for the physical products and byproducts of geochemical NETs, which will improve their economics as carbon removal solutions. For instance, calcium carbonate is a

product itself (chalk, talc), but more importantly, it is also an additive in numerous materials: paper, plastic, paint, adhesives, sealants, and coatings, among many others. Current demand for CaCO<sub>3</sub> is ~125 Mt yr.<sup>-1</sup> (~\$45 billion) and has been growing >5% a year driven largely by packaging, shipping and hygiene applications (Grand View Research, 2022). In particular, new high-volume products such as carbon-negative CaCO<sub>3</sub>-based cements could bring demand to Gt yr.<sup>-1</sup> (Hargis et al., 2021).

MgCO<sub>3</sub> is used in flooring, fire-retardant materials, rubber, and in food and cosmetics. It has lower demand than CaCO<sub>3</sub>, at ~2 Mt yr.<sup>-1</sup> (\$300 million). Exciting growth opportunities include Mg-rich alloys for automotive and aerospace applications (Magnesium Carbonate Market) and Sorel cement (“magnesium cement”), which holds certain advantages over Portland cement for specific applications and can be prepared from relatively impure materials (Jurišová et al., 2015).

Finally, demand for silica (SiO<sub>2</sub>), another product of mineral carbonation, is immense (~500 Mt. yr.<sup>-1</sup>, ~\$6 billion) and growing rapidly (>8% annually)—fueled largely by the semiconductor and glass industries. Silica may also find increased applicability at relevant scales in concrete, where its use as a substitute cementitious material could potentially enhance concrete strength while reducing emissions from cement manufacture (Gadikota et al., 2015). Altogether, there are many potential market opportunities for the products of geochemical NETs.

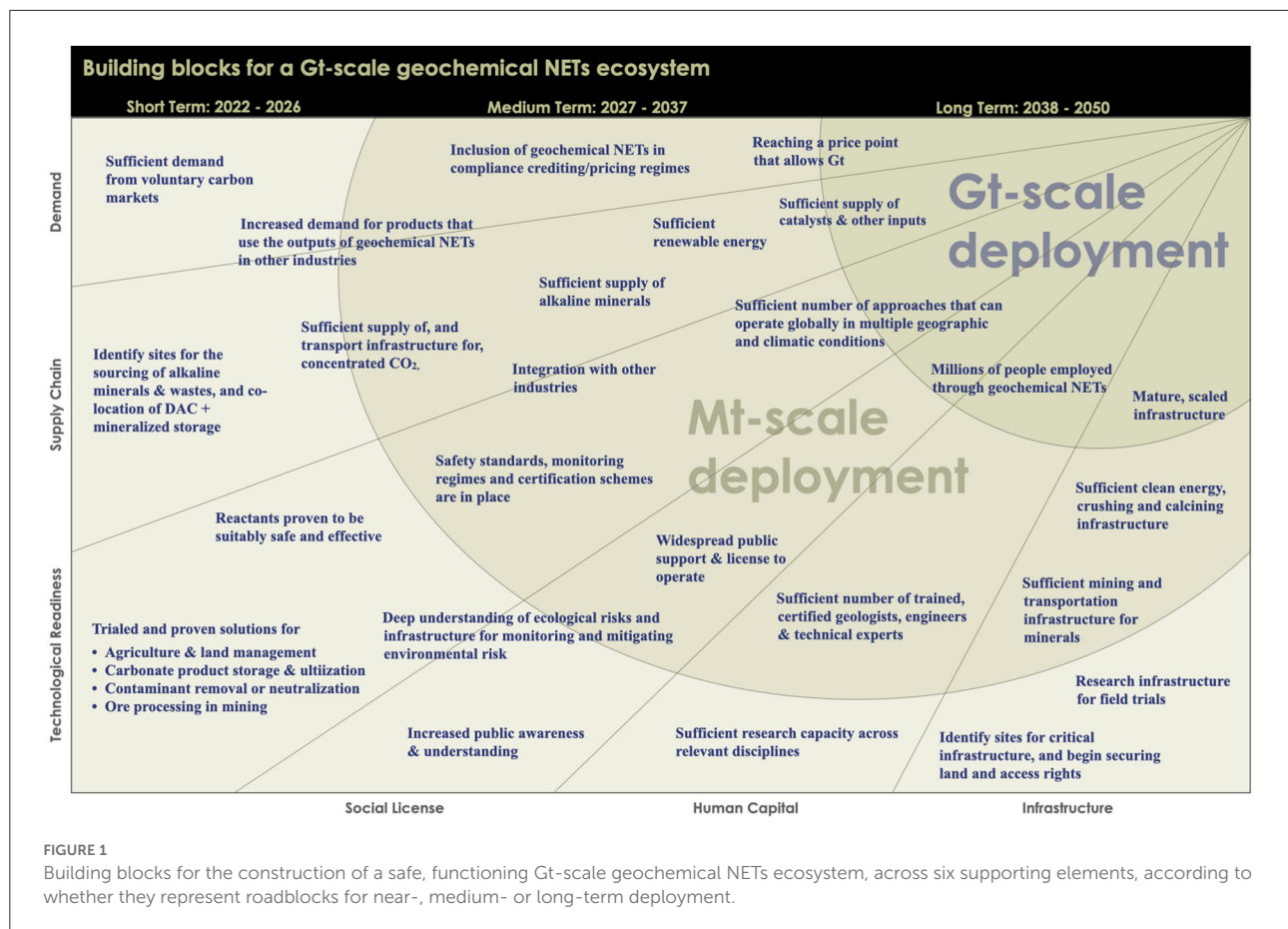
It is likely that reaching Gt yr.<sup>-1</sup> scale by 2030 will require mechanisms beyond creation of markets for value-added products. Nonetheless, these sources of demand warrant development.

## Supply chain

Supply of alkaline mineral feedstock is not expected to be limiting, as such minerals comprise the bulk of the Earth’s crust. However, at Gt yr.<sup>-1</sup> scale, the availability of low-cost, low-carbon electricity for grinding may be a major limitation for some approaches (Strefler et al., 2018; Eufrazio et al., 2022). Land based *in situ* mineralization methods may require significant amounts of water, up to 25–30 tons of H<sub>2</sub>O per ton CO<sub>2</sub> (Gunnarsson et al., 2018). All *in situ* methods will require access to enough concentrated CO<sub>2</sub>, which may become a bottleneck if DAC or bioenergy with carbon capture and storage (BECCS) do not scale alongside CO<sub>2</sub> mineralization efforts. For surface geochemical NETs (*ex situ* and surficial), the need for enhancements—e.g., acid, heat, grinding, catalysts, microbes, *etc.*—is the focus of ongoing research (Campbell et al., 2022), and whether these enhancements are inhibited by, or met with, new bottlenecks at greater deployment scales is a key uncertainty.

A complicating factor is that alkaline minerals are not uniformly distributed across the Earth’s surface. However, they





are widely available, with several noteworthy hotspots of surface accumulation. *In situ* mineralization could store  $>1$  Gt CO<sub>2</sub> yr.<sup>-1</sup> in peridotite massifs of Oman alone (Kelemen and Matter, 2008). At regional levels, megatons of alkaline wastes are available (Riley et al., 2020), which could also potentially be used by geochemical NETs. Use of waste products such as mine tailings and industrial slag for carbonation serves the added purpose of potentially reducing the toxicity of such wastes, through fixation of heavy metal ions and stabilization of pH (Gomes et al., 2016). For large-scale projects, a balance must be struck between minimizing excavation and transport with the efficiency and throughput of industrial-scale processing, the latter of which will likely require centralized infrastructure.

## Human capital

While the current number of researchers, engineers, skilled technicians and other experts dedicated to geochemical NETs is certainly not sufficient for wide-scale deployment, these approaches employ similar skill sets to the mining, cement manufacture and concrete production, agricultural liming, fossil

fuel and other industries, creating opportunities for the transfer of human capital into the deployment of geochemical NETs. It is of paramount importance that programs focused on talent transitions to “green” jobs involve transfer into NET industries. University modules focused on training students in the science, engineering processes and impacts behind geochemical NETs should be created to increase available talent. Such programs would also benefit current and future generations of students who will likely be increasingly required to work in the climate mitigation sector, since geochemical NETs provide opportunities for a wide range of interests, skill sets and geographies.

## Infrastructure

Mining, transport, and clean energy infrastructure is not expected to be sufficient in the near-term for Mt yr.<sup>-1</sup> scale, and deployment at Gt yr.<sup>-1</sup> scale would require massive investment and buildout, especially in non-fossil energy. For some geochemical NETs, particularly applications in *ex situ*, *in situ*, and surficial carbon mineralization, pre-concentration of CO<sub>2</sub> from air will be required, suggesting

that energy requirements might become tangled with those of DAC. For *in situ* methods in particular, transport of CO<sub>2</sub> may become a challenge both logistically and in terms of public support, especially where pipelines are involved (Teng et al., 2021). Furthermore, existing mining, crushing, and calcination infrastructure cannot cope with the needs of geochemical NETs at scale and therefore will need to be substantially expanded (Caserini et al., 2021). Depending on the geochemical NETs employed, increased transport, storage, burial or utilization of the reaction products would be required.

## Gap assessment

The most significant specific gaps across the six supporting elements above are depicted in Figure 1 according to whether they represent roadblocks for near-, medium- or long-term deployment. This figure assumes that the next 4 years constitute a continued research phase, Mt yr.<sup>-1</sup> deployment will be reached between 5 and 10 years from now, and Gt yr.<sup>-1</sup> deployment before 2050.

In the near term, addressing many of the gaps for reaching a Mt yr.<sup>-1</sup> deployment should be achievable, as they are primarily focused on the research required to vet geochemical NETs and establish their effectiveness. As such, they require limited amounts of funding, people, land, and social license, relative to the amounts required for subsequent stages of deployment.

However, if the timescale for this vetting stretches too far, we risk two things: First, geochemical NETs may be deployed anyway due to high demand for permanent removals, but without full knowledge of how to manage impacts. This could create problems in terms of human and environmental wellbeing and could also jeopardize any future deployment of geochemical NETs, similar to the observed controversy surrounding ocean iron fertilization (Fuentes-George, 2017). Second, we risk deployment too far in the future to achieve the primary goal of geochemical NETs, which is to reduce the atmospheric CO<sub>2</sub> concentration in time to avoid climate catastrophes and tipping points (Boers and Rypdal, 2021; Fewster et al., 2022; Shukla et al., 2022).

The gaps between the Mt and Gt yr.<sup>-1</sup> scenarios are far more considerable. The energy and transport infrastructure required is a substantial increase from that which exists today (Renforth, 2012; Lefebvre et al., 2019), and mining activity would be expected to increase significantly (Goll et al., 2021). Both government procurement and inclusion in compliance carbon markets would be required to support demand, though unique problems will emerge in bringing down price, and a greater number of efficiencies come under consideration. The approximate cost estimates and minimum timescales required

to close gaps are explored further in the section Action plan and shown graphically in Figure 2.

## An agenda for advancing Geochemical NETs

We propose an agenda for research and innovation that includes essential priorities and other opportunities that merit exploration and investment. Many of these opportunities lie at the intersections among different research disciplines, or among NETs, or large-scale industries, such as concrete or steel production. Our priorities build on those put forth by the non-profit organization Ocean Visions (Ocean Visions, 2021) and the Innovation for Cool Earth Forum (Sandalow et al., 2021), among others.

## First-order priorities for research and development

Research advances essential for the safe, timely, and effective demonstration and deployment of geochemical NETs include:

- Integration of the principles of responsible research and innovation (Owen et al., 2012) into research agendas, including the co-development of programs with wider stakeholders and publics.
- Confirming net carbon fluxes in surface systems, accounting, e.g., for groundwater effects (Sandalow et al., 2021).
- Identification of the ecological effects of enhanced weathering and surficial mineralization techniques, and downstream effects on natural assets, economic activity, and public health. Further work is also needed to address the impacts of enhanced weathering on, for example, the eutrophication of aquatic systems, biodiversity, biosphere-atmosphere feedbacks, and air, water and soil pollution (Strefler et al., 2018).
- Development of a range of techniques, technologies, and protocols for the modeling, monitoring, measurement, and verification aspects of geochemical NETs. Development of standardized methodologies for the sampling and analysis of solid alkaline materials to determine the mineralized CO<sub>2</sub> and facilitate regulatory oversight and carbon crediting for geochemical NETs. These will enable authorities to ensure proper carbon accounting.
- Resolution of legal challenges pertaining to ocean dispersion to enable rapid scale-up of ocean-based geochemical NETs once the ecological impacts have been determined and can be properly managed.



FIGURE 2 (Continued)



FIGURE 2

Proposed priority initiatives and relevant actors, based on assessment of our work presented in Campbell et al. (2022), along with previous reports and roadmapping efforts (National Academies of Sciences, Engineering, and Medicine, 2019; Sandalow et al., 2021; Ocean Visions, 2021). Solid blue squares indicate a role in which leadership is essential, striped squares indicate a role in which there is significant opportunity to contribute, and open squares indicate a role in which there is some opportunity to contribute. The provided cost estimates represent best guesses based on available information, where "Hi" >\$100 M by 2030, "Med" = \$10–100 M by 2030, and "Low" <\$10 M by 2030. The rough time estimates for reaching Mt CO<sub>2</sub> yr.<sup>−1</sup> and Gt CO<sub>2</sub> yr.<sup>−1</sup> scales are provided to help illustrate the overall scope of work.



## Other research and development opportunities that merit exploration and investment

Other initiatives that merit exploration and investment because they could yield important and valuable new gains include:

- Research infrastructure to enable the cross-disciplinary study across physics, geology, chemistry, biology, ecology, engineering, social sciences, law and governance necessary to bridge gaps and address problems in tangible and actionable ways.
- Selection and optimization of mineral deposits/feedstocks to maximize the labile  $Mg^{2+}$  and  $Ca^{2+}$  content and enhance reaction rates, while reducing the release of toxic metals and limiting environmental impacts. This work may benefit from computer modeling and artificial intelligence, to identify potential candidate feedstocks and materials.
- Advancement of methods for studying and manipulating soil and root microbiomes to promote weathering in soils of various biomes, including agricultural fields.
- Advancement of understanding of the mechanisms of chemical catalysts of dissolution of alkaline minerals, identification of optimal catalysts, and assessment of their utility, including development of platform technologies for their study and engineering.
- Advancement of understanding of the biological mechanisms influencing mineral weathering and  $CO_2$  mineralization and their utility, through development of new platform technologies for their study and engineering (e.g., methods for high-throughput study of relevant mechanisms and biomolecules in microbes and microbial communities).
- Design and techno-economic assessment of methods leveraging chemical and biological enhancement.
- Improvement in understanding potential co-benefits of enhanced weathering in soils, including whether release of key limiting nutrients (e.g., P or K) from basalt weathering has the potential to boost biomass production, resulting in additional  $CO_2$  removal (Goll et al., 2021).
- Further lab and field testing of the complex interactions and feedbacks between permeability, reaction rate, reactive surface area, fluid flow dynamics, and in particular, the effects of pore clogging and cracking, during circulation of  $CO_2$ -rich fluids through subsurface mafic and ultramafic rocks (Kelemen et al., 2020).
- Creating databases and maps of sub-surface mafic and ultramafic rock resources, as well as alkaline material deposits at Earth's surface, to help identify, with the aid of multi-criteria decision analysis or other decision-making

tools, prime sites for negative emissions *via* mineralization (Raza et al., 2022).

- Greater exploration of the potential synergies between mineralization technologies and DAC and BECCS, and in particular, the benefits of producing and using gases with a range of  $CO_2$  purities, with their potential to lower costs compared to systems requiring pure  $CO_2$  (Wilcox et al., 2017; Kelemen et al., 2020). Co-location of *in situ*  $CO_2$  mineralization activities with geothermal energy production.
- Further development of solid mineral DAC systems, which utilize abundant, low-cost natural and artificial alkaline minerals (e.g., lime, magnesia, or other alkaline materials), to remove  $CO_2$  from the air. In particular, there is potential for new materials, other than lime, which can capture ambient  $CO_2$  at comparable rates, but which have lower kiln temperatures for regeneration (*i.e.*, more facile carbonate decomposition) thus decreasing energy consumption and enabling integration with renewable energy (e.g., enabling a move away from natural gas-based kilns toward renewable-energy powered kilns) (Nikulshina et al., 2008; Campbell, 2019; Kelemen et al., 2020; McQueen et al., 2020).
- Innovation in developing new products or disposal techniques for the carbonates produced, as well as in commercial and societal side-products, such as fertilizers, soil remediation, coastal protection, or hydrogen production (Beerling et al., 2018; Hargis et al., 2021).
- Creation of Focused Research Organizations (FROs), which exist outside traditional research infrastructure to streamline and fast track research, in order to tackle some of the outstanding challenges listed in this and the previous section (Marblestone et al., 2022).

## Opportunities for innovation in partnership with existing industries

Other opportunities for innovation in partnership with many of the world's largest existing industries include:

- Deployment of enhanced weathering in soils in combination within agriculture, forestry, and soil management, including in combination with biochar.
- Integration of  $CO_2$  mineralization into cement/concrete and construction industries as a storage option for distributed DAC.
- Integration of  $CO_2$  mineralization into ore mining and processing. Mining activities that deliver carbon removal and produce metals and minerals for other uses, such as the extraction of rare earth metals, could enhance economics and sustainability. Carbonation could provide the mining

industry with valuable carbon offset opportunities, while simultaneously reducing safety risks from mine tailings (Harrison et al., 2013).

- Integration of CO<sub>2</sub> mineralization into other existing large-scale industries, such as iron and steel production, could provide feedstocks, reaction sites, or storage solutions for carbon mineralization in a manner that is relatively economical and highly scalable.
- Innovations in the decarbonization of processes like calcination, e.g., development of non-fossil fuel-powered kilns, which could simultaneously advance decarbonization in cement production (Fennell et al., 2021) and the prospects for ocean liming, or hybrid metal oxide looping approaches (Kelemen et al., 2020; McQueen et al., 2020).
- Many large commercial companies, in construction, mining, and other sectors, produce waste streams that could be used as CO<sub>2</sub> mineralization reactants. Demonstration trials with these wastes may be relatively economical and low-hanging fruit for deployment-led innovation, with the potential for upscaling.

## Action plan

Requisite scales of carbon removal through geochemical NETs will never be reached without engagement across multiple domains. Coordination and collaboration across sectors and domains are required to advance the field on relevant timescales, similar to the accelerated development of multiple COVID-19 vaccines (Bok et al., 2021). Ensuring efficient information exchange between groups in a way that leads to relevant and timely action may require creativity in terms of strengthened interactive discussions and sharing of informational resources and datasets, as well as accelerated technology transfer from laboratories to the public or private sector.

Near-term, many required actions focus on the research and development needed to assess risks, improve efficiencies, and establish robust MMV systems that enable public acceptance and market systems for carbon removal outcomes. All actions undertaken will need to consider environmental justice and equity concerns, as public acceptance will only build as successful projects including geochemical NETs are seen to benefit communities, through means such as increased employment, revenue-sharing schemes, environmental restoration, or combinations of these.

While action is required across all sectors, local, national and international governments deserve special mention, as it is their role to provide leadership in addressing climate change. They are uniquely able to establish laws and regulations, assemble and distribute certain types of data, and incentivize behavior and markets through policy decisions. They can act as major leaders in the marketplace through procurement decisions and setting of geochemical NET targets. They can also ensure that data

and information are collected and shared openly. Furthermore, governments can develop integrated regional strategies for industrial and economic development based on assessments of local assets, such as mineral deposits, clean energy sources, deployment locations, R&D capabilities, and transportation infrastructure. They can identify locations for Gt yr.<sup>-1</sup> clusters and lead early planning and stakeholder engagement. Additionally, governments can make foundational investments in infrastructure, energy systems, feedstock sourcing, and/or storage sites to catalyze those clusters. They can lead R&D funding on integral and structurally important technologies across domains, e.g., calcination for cement manufacturing and for various geochemical NET pathways, construction materials, energy systems, and mining practices. Governments should also lead on setting and harmonizing standards and permitting regimes, defining acceptable ecosystem impacts, and providing targeted subsidies and carbon pricing.

Figure 2 lists detailed recommendations for actions by all actors in addressing gaps across the six elements in the section Needs assessment, as well as levels of requisite engagement by each actor, highlighting specific initiatives for which specific actors have opportunities to lead or to contribute. These recommendations are presented along with rough cost estimates in order to signal the level of effort required to undertake these recommendations and the degree to which certain actors can spark activity through funding mechanisms [e.g., the recent Ocean Alkalinity Enhancement Engineering Award funded through philanthropy (Additional Ventures, 2022)].

Also depicted in Figure 2 are rough estimates for the minimum time necessary for each recommended action to help bridge gaps between now and Mt yr.<sup>-1</sup> and Gt yr.<sup>-1</sup> deployment scenarios, which are included as a means of reckoning with the overall scope of work required. For example, we generally expect that lab research and field trials might require ~2–5 years, but larger scale demos might require at least ~5–8 years, and any significant deployment cannot take place before then. And while we may be only partially confident we have cost and time estimates for each initiative correct, we are confident that these are initiatives that must begin happening now if we are to achieve Mt yr.<sup>-1</sup> deployment of geochemical NETs by 2030, with a path toward Gt yr.<sup>-1</sup> deployment during the following two decades.

## Discussion

With increasing consensus on the need for NETs to limit warming to 1.5°C (Shukla et al., 2022), the question is no longer whether negative emissions are necessary to stay within carbon budgets, but how we develop and implement them. Geochemical NETs are a potential opportunity to not only remove atmospheric CO<sub>2</sub>, but to address wider environmental and sustainability issues, such as ocean acidification. This roadmap highlights gaps in current technical readiness, social

license, demand, supply chains, human capital, infrastructure for geochemical NETs, and that required for  $\text{Gt yr}^{-1}$  deployment, and provides our initial ideas for how to address these gaps.

Decarbonizing industry while developing NETs is a daunting challenge. Geochemical NETs could potentially reduce the need for land- or energy-intensive carbon removal approaches, as well as enhance synergetic approaches, such as coupling with biochar or DAC (Buss et al., 2021). Further, wide-scale deployment of geochemical NETs could help enable some of the world's largest industries, including agriculture, concrete, mining, shipping, and steel, to reduce their emissions, toward net zero and perhaps even achieve net negative, in line with global climate targets. Application of geochemical NETs should not be considered a parallel track to reach these climate targets separate from decarbonization efforts; rather, the two should complement and reinforce one another and reciprocally unlock possibilities.

Geochemical NETs are generally characterized by low technology/system readiness, and there are significant gaps in our understanding of their kinetics and impacts. In our view, this only highlights the imperative to accelerate and expand research and development efforts in this field.

For geochemical NETs, large quantities of material will need to be safely extracted, transported, and transformed, and large quantities of low-emissions energy will be needed. Generating economies of scale, scope, and learning may require the development of regional clusters, in which mineral resources, modeling and monitoring tools, energy and transportation infrastructure, processing facilities, deployment sites, and labor pools can be strategically developed and maximally utilized and provide a basis for continuous learning and improvement. This suggests a need for geospatial assessments to map available mineral and infrastructure resources and ecological considerations and facilitate intelligent long-term regional planning and cluster development.

So how do we suggest prioritizing next steps? Near term, R&D is the priority. It should be accelerated across all aspects of geochemical NETs, including through multiple pilot projects and research experiments to evaluate the efficacy of carbon drawdown and the ecological impacts across geographies, climates and feedstocks. Activities advancing MMV of all geochemical NETs will be critical to getting them off the ground. Dedicated activities to address the lack of public understanding should begin now. The synergism with existing mining and mineral processing and distribution activities, critical for large scale-up of geochemical NETs, should be exploited as a way to make use of existing feedstocks and infrastructure. Finally, we encourage leaders in government, philanthropy, finance, and in relevant industries—such as agriculture, lime and cement/concrete, mining, shipping, and iron/steel—to assess what they can uniquely add to advance geochemical NETs, given the assets, capabilities, and opportunities of each specific region or organization.

Ultimately, society must be confident that it can deploy geochemical NETs safely, and that these technologies are competitive with other possible climate remediation efforts in terms of efficacy, cost, and other less tangible impacts and benefits, such as biodiversity, impacts on land and ocean ecosystems, social equity, economic activity, and political acceptability. And while there is a growing community of research and practice, dominated by North America and Europe, a considerably more global set of actors and efforts are needed to advance the field. We propose a concerted effort to engage and cultivate this global community, using this roadmap and others that follow it, to coordinate and strategically grow the field of geochemical NETs in a way that is safe, scalable, effective, and timely.

## Author's note

This novel roadmap work is a collaboration between academics and NGO The Climate Map, which brings a fresh perspective to an academic-heavy field.

## Data availability statement

This perspective article is based on a comprehensive review on Geochemical NETs, available here: Campbell et al. (2022).

## Author contributions

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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# Assessing the optimal uses of biomass: Carbon and energy price conditions for the Aines Principle to apply

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In the context of climate mitigation, biomass has traditionally been viewed as a means to deliver low-carbon energy products. Adding carbon capture and sequestration (CCS) to a bioenergy production process can yield net-removals of CO<sub>2</sub> from the atmosphere, albeit at an increased cost. Recently, the Aines Principle was established, stating that at some carbon price, the revenue generated from CO<sub>2</sub> removal will exceed the revenue generated from energy production from a given bioconversion process. This principle has only been illustrated for the theoretical conversion of a non-specific biomass source, and has not yet been demonstrated to show real carbon prices that can tip the scale for biomass carbon removal to be more economically favorable than bioenergy production. In this study, we demonstrate the Aines Principle at work in two specific examples of biomass conversion. The first case involves a Chinese municipal solid waste incineration plant, with and without CCS. The second case compares using forestry residue solely for energy production (*via* gasification), solely for carbon removal (*via* burial) or both. By comparing the energy and carbon revenue streams under a range of carbon prices, we show that carbon removal revenue can exceed energy revenue at currently available carbon prices below \$200/tCO<sub>2</sub>.

## KEYWORDS

biomass conversion, carbon removal, carbon capture, bioenergy, climate change, resource allocation

## Introduction

Biomass has been a quintessential part of the climate solution for decades. Traditionally, the use of biomass in the climate context has been focused on the production of bioenergy as a lower-emitting solution to fossil fuels. While burning biofuels still produces CO<sub>2</sub>, the technology is considered carbon neutral because the biomass is the result of photosynthetically removing an equivalent amount of CO<sub>2</sub> from the atmosphere. If the bioenergy production is coupled with carbon capture and storage (BECCS), the system can become carbon negative and achieve carbon dioxide removal (CDR). With the increasing realization of the need for CDR to meet climate goals and

avoid 2°C warming, the role of biomass has received increased attention. After all—biomass is one of nature’s own CDR systems, drawing down a net 7.6 GtCO<sub>2</sub>e per year today in forests alone (Harris et al., 2021) and even up to thousands of gigatons of CO<sub>2</sub> in the Arctic during the middle Eocene period (Speelman et al., 2009).

In December 2020, a new term was introduced to shift the emphasis of biomass utilization from primarily bioenergy production to primarily CDR: BiCRS (Biomass Carbon Removal and Storage) (Sandalow et al., 2020). With this, the Aines Principle was introduced, stating that for some carbon price, a ton of biomass can become more valuable for its use in CDR than for its use to produce bioenergy. To demonstrate this principle, a simple chart was produced that relates carbon price to the value of carbon removal, with references to the values of oil, gas, coal, and wood pellets (Figure 1). This chart demonstrates that, for example, at a carbon price of about \$35/tCO<sub>2</sub>, the carbon within biomass—representing carbon removed from the atmosphere—is more valuable than the energy it can provide at a value equivalent to that of natural gas. Similarly, if the energy provided by the biomass is valued at an equivalent price to oil (or coal) the breakeven carbon price is higher (or lower).

Figure 1 is a helpful tool to illustrate the point of the Aines Principle that at some carbon price, the carbon content of biomass is worth more than the energy content. Moreover, the carbon prices at which the biomass carbon value exceeds the value of traditional energy products (shown in Figure 1 to be about \$17–65/tCO<sub>2</sub>) are sufficiently low that they are in the range of existing public subsidies and private purchases, as shown in Table 1. The US Section 45Q tax credit was recently updated under the Inflation Reduction Act, now providing \$85/tCO<sub>2</sub> for carbon emissions captured from energy production and industrial facilities (Bright, 2022). From July 2021 to July 2022, credits provided by the California Low-Carbon Fuel Standard (LCFS) were in the range of \$117–188/tCO<sub>2</sub> (California Air Resources Board, 2022), and permits from the EU Emission Trading System (ETS) ranged from about €54–97/tCO<sub>2</sub> during the same period (Tradingeconomics, 2021). Several other nations across the globe are considering implementing carbon taxes or ETS schemes, or have already scheduled them for a later date (World Bank, 2022). Recently, corporations such as Microsoft, Shopify and Stripe have begun directly procuring carbon removal offsets as part of a nascent voluntary market; Stripe announced purchase prices ranging from \$200 to 2,000/tCO<sub>2</sub> (Stripe, 2021). As carbon prices in this range become an ever clearer reality, the time is ripe to re-think the way we view biomass within the portfolio of climate solutions, and shift from a viewpoint of producing bioenergy with some “bonus” carbon removal to a new viewpoint that prioritizes achieving carbon removal with some “bonus” bioenergy.

However, in order to validate this shift in thinking it is important to address the complexities of real-world biomass

carbon removal and energy conversion. Figure 1, while broadly useful for visualization, is based on the simplified assumption that one bone-dry ton of biomass contains half a ton of carbon (0.5 tC/bdt), and is agnostic to different biomass types and conversion technologies. In fact, these factors are very influential in determining the actual realized revenue of energy production, the cost of capturing and removing CO<sub>2</sub>, and the production of process emissions. Therefore, the generalized approach of the previous treatment of the Aines Principle needs to be put on a firmer quantitative footing by including these important process-specific factors.

On a related note, some authors have begun to argue for a shift in thinking about the best role of biomass in climate for other reasons. A report by Material Economics observes that bioenergy is becoming uncompetitive with lower-cost solutions in road transport, low-temperature heat, power generation, and shipping, and concludes that higher-value uses of bioenergy will soon be preferred, like providing high-temperature industrial heat, liquid fuels for long-haul aviation, or negative emissions (Material Economics, 2021). In assessing biomass conversion processes for higher value purposes such as these, Patrizio et al. (2021) demonstrated that the processes that are most efficient at producing bioenergy do not necessarily provide the largest carbon mitigation, showing higher mitigation is associated with capturing CO<sub>2</sub> at higher rates and displacing more carbon-intensive alternatives. A similar observation was made previously by Mac Dowell and Fajardy (2017), who showed that low-efficiency BECCS power plants can deliver more carbon removals at a lower cost than high-efficiency plants by requiring more biomass to produce one MWh of energy (producing more CO<sub>2</sub> per MWh), and by having lower initial investment costs than high-efficiency plants. Due to their advantages, these low-efficiency plants are shown to yield a faster turnaround time on their initial investment (assuming a fixed carbon price of about \$100/tCO<sub>2</sub>). While these publications indirectly validate the Aines Principle by discussing the notion that carbon removal is a more valuable use of biomass than bioenergy, they do not directly demonstrate the principle by comparing the revenue streams generated by energy and CO<sub>2</sub> to identify a breakeven carbon price.

A closer, process-specific assessment of the balance of the energy and carbon values of biomass is needed for two primary reasons. First, it will help to inform policies on carbon prices in public and private offset markets by providing better insight into relevant carbon values that tip the scale for specific biomass conversion technologies. While some markets may already have carbon prices that incentivize carbon removal over bioenergy production (as demonstrated here), others looking to follow suit could benefit from this sort of analysis. Second, it could inform technology decisions, where processes can be optimized for carbon removal rather than energy production.

In this study, we demonstrate the Aines Principle at work in two specific examples of biomass conversion. By comparing the

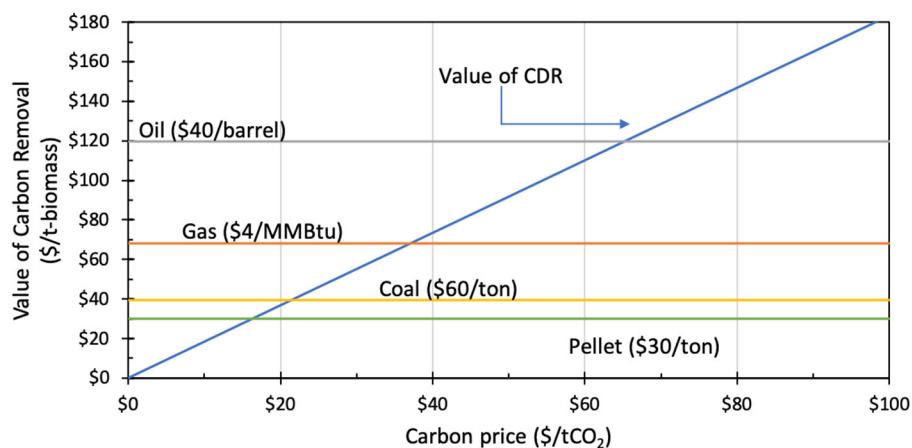


FIGURE 1

Change in the CDR value of biomass with increasing carbon price, compared to the value of reference energy sources. Based on figure in ICEF BiCRS report (Sandalow et al., 2020).

revenue generated from energy production with the potential revenue from carbon removal under a range of carbon prices, we show that carbon removal revenue can exceed energy revenue at currently available carbon prices below  $200/\text{tCO}_2$ . Both of the cases we consider focus on conversion of waste biomass, taken as the more sustainable option over dedicated “energy crops” (Welfle et al., 2017). The first case will compare municipal solid waste (MSW) incineration, with and without CCS. The second case will compare using forestry residue for solely energy production (*via* gasification), solely carbon removal (*via* burial), or both (BECCS/BiCRS). The demonstration of the Aines Principle through these two cases will be followed by a discussion of the implications to broader biomass usage and energy systems, along with limitations of the study. Afterward, the conclusion will summarize the key findings of the study and provide suggestions for how the work shown here can be used in the future. To facilitate a faster dissemination of the key ideas in this work, the methodology used to construct the demonstration figures is explained after the conclusion.

## Demonstration of the Aines Principle

### Case 1—Municipal solid waste

The first case considers the generation of electricity through incineration of municipal solid waste (MSW). About 11% of the world’s 2 billion tons of MSW is incinerated annually, primarily in high-income countries with land constraints (Kaza et al., 2018). Globally, there are more than 2,400 MSW plants with incineration in operation, and this number could increase to more than 2,700 by 2027 (Kearns, 2019). Capturing the  $\text{CO}_2$  emitted from an incineration plant can yield carbon

TABLE 1 Carbon prices currently paid by public policies and private corporations.

Entity/policy	Carbon price ( $\text{tCO}_2^{-1}$ )	Applicable process
U.S. Tax Credit 45Q (Bright, 2022)	\$85	Power/industrial facilities
California LCFS (California Air Resources Board, 2022)	\$117–188	Various fuel-related processes
EU Emission Trading System (Tradingeconomics, 2021)	€54–97	Power/industrial facilities, aviation
Stripe Offset Purchases (Stripe, 2021)	\$200–2,000	Various removal processes

removals, although to varying degrees depending on the MSW composition. Rosa et al. use a wide range of 42–71% to represent the biogenic portion of  $\text{CO}_2$  emissions in European incineration plants (Rosa et al., 2021).

A recent analysis of a Chinese MSW incineration plant analyzes the economic and environmental impact of capturing  $\text{CO}_2$  emissions through three different CCS technologies (i.e., MEA, P/VSA, and oxyfuel) (Tang and You, 2018). Without CCS, the incineration plant produces electricity at a rate of slightly more than 272 kWh per ton of MSW and sells it to the grid for about  $\$0.105/\text{kWh}$ . This process emits 586  $\text{kgCO}_2$  per ton of MSW, and assuming the carbon content of the waste is 57%



biogenic, there are 334 kgCO<sub>2</sub> that could be captured to yield carbon removals from the atmosphere.

The oxyfuel CCS case was demonstrated by Tang and You to capture the most CO<sub>2</sub> at the least cost (Tang and You, 2018). Adding oxyfuel CCS to the incineration plant captures 95% of the incinerator's emissions, but reduces the electricity output to 111 kWh/tMSW and adds capital expenses (with a capital charge factor of about 8.4%) and operating expenses, as described in the original analysis by Tang and You (2018). Capturing the CO<sub>2</sub> from the incinerator allows the plant to sell carbon removal credits in addition to its electricity product. The two revenue streams at different carbon prices are shown in Figure 2, with a fixed rate at which electricity can be sold to the grid (\$0.105/kWh).

When no carbon price is available, installing oxyfuel CCS on the incineration plant yields a net-expense, illustrated by a negative value of CDR in Figure 2. This led the original authors to conclude that CCS technologies are currently too immature to be installed on Chinese incineration plants (Tang and You, 2018). However, Figure 2 demonstrates that a carbon price of about \$35/tCO<sub>2</sub> rectifies the added expense of CCS installation. Further, a carbon price of about \$70/tCO<sub>2</sub> makes the removed CO<sub>2</sub> more valuable than the electricity produced by an incineration + CCS plant, and at a breakeven price of about \$130/tCO<sub>2</sub> the CO<sub>2</sub> revenue exceeds the electricity revenue that would be realized by the incineration plant without installing CCS. While this full range of carbon prices is currently available in public and private US carbon markets, they are not yet available in China. Although China recently introduced a cap and trade carbon market, the opening day carbon price was <\$8/tCO<sub>2</sub> (Nakano and Kennedy, 2021), and it does not yet have a market dedicated to carbon removals.

## Case 2—Forestry residue

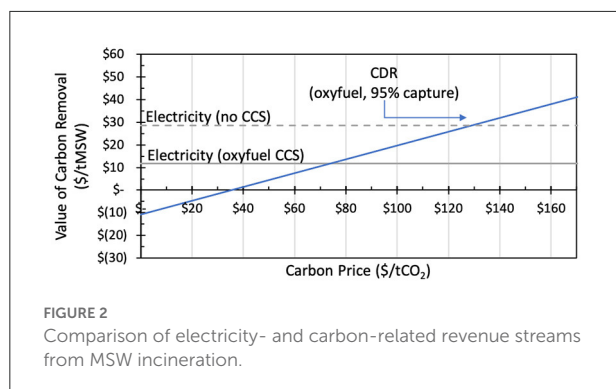
A more complex case considers various ways to remove carbon using forestry residue, which is generated from forest operations such as clearcutting, logging, and thinning, as well as

from natural disturbances like fires and wind throw (Braghiroli and Passarini, 2020). Several national-level assessments show that substantial amounts of forestry residue are generated each year in countries including but not limited to Canada (~61 Mt) (Mansuy et al., 2017), Mexico (~1.4 Mt) (Honorato-Salazar and Sadhukhan, 2020), and Europe (~320 Mt, primarily in Sweden and Finland) (Hamelin et al., 2019).

Forestry residues present a suitable feedstock for gasification because of their relatively low moisture content. Larson et al. describe a gasification process that produces a combination of electricity and fuel—either Fischer-Tropsch (FT) fuels or liquid hydrogen (Larson et al., 2009). There are two hydrogen-producing variations: one where hydrogen and power are produced in a nearly-even split (H5050), and one where hydrogen production is maximized while producing minimal power (HMAX). Assuming this process is located in California, as assessed by Baker et al. (2020), the three energy products of electricity, FT fuels, and hydrogen can be sold at wholesale prices of \$0.06/kWh, \$2.35/gal, and \$2.90–3.80/kg. Alternatively, the forestry residue could be directly buried, as proposed by Zeng (2008) and Zeng and Hausmann (2022). While this method would not provide any energy-based revenue, it avoids the conversion of carbon to CO<sub>2</sub> emissions or fuels, presenting the potential to sequester all biogenic carbon from the forestry residue.

The comparison of potential energy and carbon revenue streams from burying or gasifying forestry residue is shown in Figure 3. Just as discussed with Case 1, implementing CCS on a gasification process brings capital and operational expenses and decreases the net energy output of the plant. The costs of gasification with CCS are calculated by estimating the cost of biomass conversion, H<sub>2</sub> liquefaction, and the capture, drying, compression, transport and injection of CO<sub>2</sub> as estimated previously (Larson et al., 2009; Baker et al., 2020). Capital expenses are assumed to be paid with a capital charge factor of 15%, and the cost of gathering forestry residue feedstock is estimated at \$50 per dry ton (Baker et al., 2020). The cost of wood burial involves land purchase, construction, and operation estimated to be about \$13–22 per tCO<sub>2</sub> sequestered (Zeng and Hausmann, 2022), in addition to the \$50/bdt feedstock costs. For a conservative estimate, we use the upper bound cost of \$22. Additional information on these cost estimations is provided in the methods section.

One point to note is that the y-axis values are much higher than in Figure 2 as a result of producing higher-value energy products. However, installing carbon capture to the gasification process adds considerable costs. Where producing power and FT fuels generates a revenue of about \$165/bdt, the net profit is estimated to be about –\$60/bdt. Similarly, while the hydrogen production cases generate energy revenues of up to about \$200/bdt (H5050) and \$310/bdt (HMAX), the net profit is –\$76/bdt and –\$59/bdt, respectively, at the hydrogen wholesale price of \$3.80/kg (see Table 6).



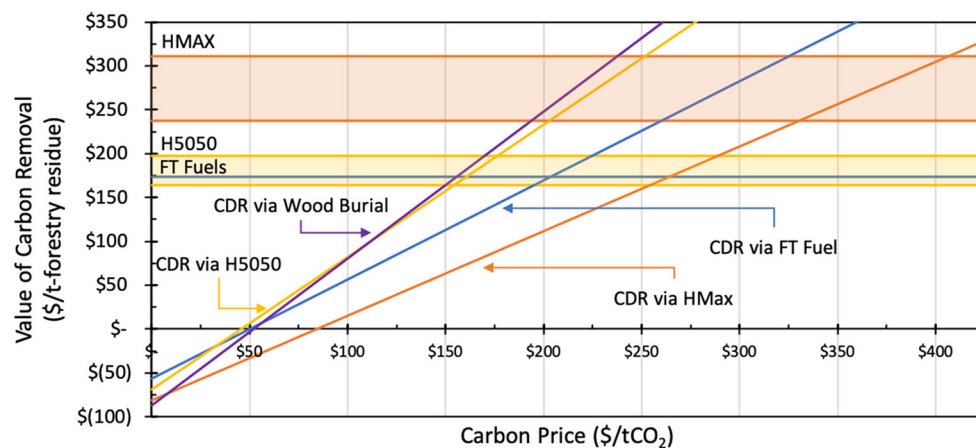


FIGURE 3

Comparison of the energy- and carbon-related revenues from gasification of forestry residue. Also shown is the estimated carbon revenue from burying an equivalent amount of forestry residue. A range of liquid hydrogen prices is given, as described in the text.

As demonstrated in Case 1 with MSW incineration, adding CCS to gasification requires a non-zero carbon price to become economically feasible. Specifically, the carbon price that allows the gasification + CCS processes to become economically feasible is about \$50/tCO<sub>2</sub> for FT fuels and H5050, and about \$85/tCO<sub>2</sub> for HMAX. This is within the range of BECCS gasification costs of \$30–150/tCO<sub>2</sub> used in integrated assessment models (IAMs) (Fuss et al., 2018; Butnar et al., 2020). The energy products in Case 2 are higher value, meaning higher breakeven carbon prices are required for the carbon-based revenue to exceed the energy-based revenue and demonstrate the Aines Principle. These carbon prices are about \$205/tCO<sub>2</sub> (FT fuels), \$150–175/tCO<sub>2</sub> (H5050), and \$325–400/tCO<sub>2</sub> (HMAX).

Without any energy-related revenue, CDR *via* wood burial bears all feedstock and facility costs. However, because it is assumed to retain all of the biogenic carbon, a carbon price of \$50/tCO<sub>2</sub> enables burial to generate a net profit. At a carbon price of about \$155/tCO<sub>2</sub>, one ton of forestry residue is more valuable for CDR *via* wood burial than it is for production of power and FT fuels; and at a carbon price range of about \$150–170/tCO<sub>2</sub>, forestry residue burial becomes more economical than the H5050 process. These prices are well-within the range of what has been purchased on the voluntary offset market and through the California LCFS.

Also illustrated in Figure 3 is the choice of optimizing a process for energy production vs. carbon removal. Without a market for carbon removal, one would likely decide to use gasification to produce hydrogen *via* the HMAX configuration, as this process generates more energy-related revenue and is more profitable than the other two gasification configurations (shown explicitly in Table 6). Producing hydrogen generally results in a higher volume of CO<sub>2</sub> that is captured and stored

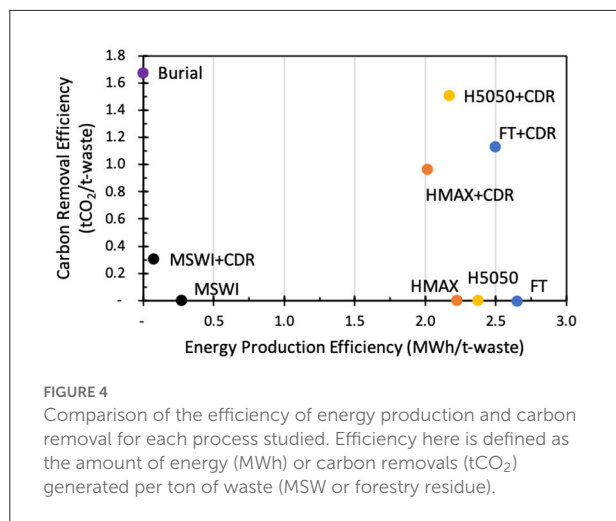
because all of the biogenic carbon is available for capture (as opposed to only 75% when producing FT fuels). However, the lack of power production in the HMAX configuration results in using carbon-intensive power from the grid to power carbon capture, resulting in a lower net-removal of CO<sub>2</sub> compared to the H5050 process. Thus, although the H5050 configuration yields less energy-related revenue, its enhanced production of net carbon removals yields higher carbon-related revenue at carbon prices as low as \$50/tCO<sub>2</sub>.

## Discussion

In this study we demonstrated the Aines Principle for two different cases of biomass conversion technologies. That is, these processes generate revenue streams from carbon removal that exceed those from energy production at carbon prices that have already been paid in public and private markets. The Aines Principle has implications for technology decisions, municipal waste management, and climate policy.

## Energy, removals, or both?

Biomass presents many potential pathways to achieve emission reductions, including production of electricity, materials, fuels, and heat; if coupled with CCS, it can yield net-carbon removals. While at the surface this might present a “best of both worlds” scenario, where energy can be produced while removing CO<sub>2</sub> from the atmosphere, it is not that simple. Not only does adding CCS to a bioconversion process add operating and capital expenses, it also reduces the net output of energy products and their associated revenue streams. This



is evidenced by both cases presented here, where additional electricity production is sacrificed to power a carbon capture unit and achieve carbon removals.

Thus, the Aines Principle has implications on technology optimization decisions—where energy production efficiency can be decreased in order to increase carbon removal efficiency, as noted previously (Mac Dowell and Fajardy, 2017) and illustrated in Figure 4. Take Case 2, where the increased hydrogen production of HMAX enables the process to generate a higher energy-related revenue stream than H5050 despite its lower electricity production (shown in Figure 3). However, when adding CCS, the lower electricity production of HMAX causes the process to require external grid electricity and yield less net-removal of CO<sub>2</sub> per ton of forestry residue, making H5050 the superior option in terms of carbon removal.

Decisions on optimizing toward energy production vs. carbon removal can be taken a step further in deciding whether biomass should be used for *only* carbon removal. In Case 2, burial of forestry residue was compared to gasification processes, proving to be a simpler and often economically superior method of generating carbon removal revenue. This is a result of sequestering 100% of the biomass carbon rather than converting a portion to fuels or uncaptured emissions to the atmosphere. This type of carbon removal has been proposed for other forms of biomass like leaves and municipal waste (Amelse, 2020), and has even spawned a few companies. Charm Industrial converts biomass into bio-oil and injects the oil into underground wells (Pontecorvo, 2021), and Running Tide grows kelp and allows it to sink to the bottom of the ocean (Benveniste, 2021). However, decisions to completely forego energy production must also consider the potential for future changes in energy prices. While a gasification + CCS process might be able to adapt to increased energy value by optimizing back to increased energy efficiency, a project that involves only carbon removal would not have this option.

At the carbon prices identified in Case 1, cities may change the way they look at the treatment of MSW. When MSW is deposited to landfills, it undergoes a slow degradation process that results in CH<sub>4</sub> and CO<sub>2</sub> emissions, although the extent of this degradation is debated (O'Dwyer et al., 2018; Zhao, 2019). These emissions can be managed by directing the gas to a flare or an energy recovery system at the landfill, or they can be avoided altogether by incinerating the MSW. Many cities incentivize diverting waste from landfills by charging landfill taxes to disposers (CEWEP, 2021), which in turn can provide extra revenue for the incineration plant that burns the waste to produce power. Here, there is already a clear incentive to convert the waste, but not necessarily to capture and remove the CO<sub>2</sub>. In Case 1, the MSW treatment revenue (\$27/tMSW) (Tang and You, 2018) is nearly the same as the electricity revenue without CCS shown in Figure 2, meaning that the carbon revenue exceeds both electricity and MSW treatment revenue at about the same carbon price of \$130/tCO<sub>2</sub>. While there are other important considerations when choosing among different waste management practices, especially their impact on human health and the environment (Giusti, 2009; Iqbal et al., 2020), the potential carbon removal and associated revenue stream generated from capturing biogenic emissions at an incineration plant should be considered as new important factors.

## Limitations of this study

It is important to note that some assumptions made in this study present limitations that might change the comparison among different technologies. One limitation is the important uncertainties around biomass burial that might make it a less favorable option than presented in Case 2. Wood burial is still in its early stages and its costs are still not well-characterized (Zeng and Hausmann, 2022); this was addressed by using a conservatively high cost estimate. Further, while geologic storage of CO<sub>2</sub> has been performed for decades and is coupled with established methodologies for monitoring and verification of secure storage (US Department of Energy, 2017), the same cannot be said for biomass burial. Although burying biomass does not involve the risks associated with high CO<sub>2</sub> pressures (e.g., caprock hydraulic fracturing) (Kelemen et al., 2019), other unique factors exist (e.g., burial pit design, conditions to hinder biomass decomposition) (O'Dwyer et al., 2018; National Academies Press, 2019) for which standards are being established to ensure long-term carbon sequestration (Puro.earth, 2022a,b). Ensuring sequestration of all of the biogenic carbon is important for evaluating this technology, as just a small decrease in the retention of carbon from 100 to 90% would cause the carbon revenue from wood burial to be less than that from the H5050 process in the full domain of carbon prices evaluated (see Figure 5). This demonstrates the importance of

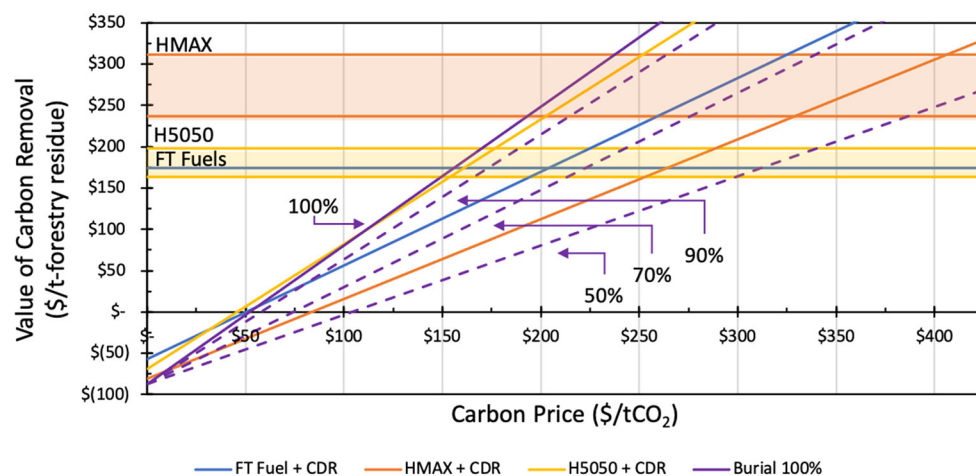


FIGURE 5  
Sensitivity of the carbon-related revenue stream of biomass burial to its carbon removal efficiency, which is varied from 50 to 100%.

storage reliability in all cases, whether stored as biogenic carbon or pure CO<sub>2</sub>.

Other limitations exist due to assumptions around biomass feedstocks and energy products. While waste biomass was considered here, the results would differ for other biomass sources like dedicated energy crops which introduce substantial emissions from things like fertilizers and land use change. This would decrease the net-removal of CO<sub>2</sub>, resulting in a higher breakeven carbon price (Fajardy and Mac Dowell, 2017). Changes in energy price assumptions would also impact the results. For example, the two cases used different electricity prices to represent China (\$0.105/kWh) and California (\$0.05/kWh). Using the latter electricity price for Case 1 would effectively cut the electricity revenue in half, significantly decreasing the breakeven carbon price and making carbon removal a more favorable alternative to energy production. The same effect would occur for decreases to wholesale fuel prices.

## Conclusions and future applications

In this study, we assess the recently introduced Aines Principle and demonstrate it for two specific biomass conversion cases—showing that revenue generated from capturing and permanently storing biogenic carbon emissions can exceed revenue from the associated bioenergy production at currently available carbon prices below \$200/tCO<sub>2</sub>. This principle has implications for technology optimization, meaning that energy production efficiency can be sacrificed to achieve increased carbon removal efficiency and, ultimately, a larger net revenue for a project. As carbon removal and storage technology develops, this principle might in some cases lead to the decision

of completely forgoing bioenergy production in exchange for pure carbon removal *via* biomass burial.

The two cases considered here are each associated with specific biomass feedstocks, conversion processes, energy products, and representative economics. There is still a vast array of other cases with different parameters that would alter the ultimate results (i.e., the breakeven carbon price). This study serves as an initial demonstration of the Aines Principle, and offers two simple equations to develop the breakeven carbon price (see Section Methods). Future analyses of biomass conversion processes could use these simple equations to determine the breakeven carbon price under localized conditions of energy prices, carbon prices, and local infrastructure. A larger pool of breakeven carbon prices can help inform policymakers on what levels of carbon incentives can become transformative for carbon removal.

## Methods

### Case 1—Municipal solid waste

Case 1 calculations were performed using data from Tang and You, who performed an economic and environmental assessment on the addition of three different CCS options to a Chinese MSW incineration plant (Tang and You, 2018). The authors report the various costs of the process, the revenue stream from electricity production, and the net profit. In cases with CCS, the net profit is negative. The authors also report the CO<sub>2</sub> captured with each CCS option. In the oxyfuel case, 95% of the emissions from the incineration plant are captured. The authors do not account for biogenic emissions, so a 57% fraction was assumed (median value of the range reported by



**TABLE 2** Data used to generate plots for Case 1, based on data from [Tang and You \(2018\)](#).

	Base incineration plant	Incineration + oxyfuel CCS
Treatment cost (\$/tMSW)	48.1	49.9
Waste treatment revenue (\$/tMSW)	27.0	27.0
Electricity revenue (\$/tMSW)	28.6	11.7
CO <sub>2</sub> transport/storage (\$/tMSW)	0.0	8.9
Profit at \$0/tCO <sub>2</sub> (\$/tMSW)	7.5	−20.2

**TABLE 3** Carbon balance for Case 1.

	Base incineration plant	Incineration + Oxyfuel CCS
Generated in process (kgCO <sub>2</sub> /tMSW)	586.5	586.5
Captured and stored (kgCO <sub>2</sub> /tMSW)	0.0	557.1
Initial biogenic uptake (kgCO <sub>2</sub> /tMSW)	−334.3	−334.3
Net emissions (kgCO <sub>2</sub> /tMSW)	252.2	−305.0
Net removed (kgCO <sub>2</sub> /tMSW)	0.0	305.0
Net removed (MtCO <sub>2</sub> /yr)	0.0	0.20

[Rosa et al., 2021](#)). One other modification to the data of Tang and You is the additional cost of CO<sub>2</sub> transport and storage, assumed to be \$16/tCO<sub>2</sub> based on the transport and storage cost tiers identified for China by [Smith et al. \(2021\)](#). The relevant cost values are displayed in [Table 2](#), and the carbon emissions balance is displayed in [Table 3](#).

In [Figure 2](#), the horizontal lines for electricity revenue use the two electricity revenue values shown in [Table 2](#). The diagonal line representing CDR revenue in [Figure 2](#) is plotted using Equations (1) and (2):

$$C_{\text{added}}(\text{tMSW}) = [\text{Treatmentcost}]_{\text{CCS}} - [\text{Treatmentcost}]_{\text{Base}} + [\text{CO}_2\text{transport/storagecost}] \quad (1)$$

$$R_{\text{CDR}}(\text{tMSW}) = [C_{\text{added}}] + [\text{CO}_2\text{Removed}][\text{CO}_2\text{price}] \quad (2)$$

**TABLE 4** Data used to generate plots for Case 2, based on data from [Larson et al. \(2009\)](#) and [Baker et al. \(2020\)](#).

	FT fuel production	H <sub>2</sub> production (HMAX)	H <sub>2</sub> production (H5050)
Electricity generation efficiency	23.1%	4.6%	31.5%
Fuel generation efficiency	34.1%	58.9%	26.9%
Gasification CAPEX (\$MM)	800.8	587.6	608.3
CO <sub>2</sub> capture CAPEX (\$MM)	228.6	272.5	272.5
CO <sub>2</sub> drying/compression CAPEX (\$MM)	32.0	39.9	39.9
H <sub>2</sub> liquefaction CAPEX (\$MM)	0.0	827.6	442.1

**TABLE 5** Power demand for Case 2, based on data from [Larson et al. \(2009\)](#) and equations from [Baker et al. \(2020\)](#).

	FT fuel production	H <sub>2</sub> production (HMAX)	H <sub>2</sub> production (H5050)
Conversion (MW <sub>e</sub> )	20.6	56.1	16.1
CO <sub>2</sub> capture (MW <sub>e</sub> )	13.7	18.2	18.2
CO <sub>2</sub> drying/compression (MW <sub>e</sub> )	15.6	20.8	20.8
H <sub>2</sub> liquefaction (MW <sub>e</sub> )	0.0	135.5	61.9

where  $C_{\text{added}}$  is the additional treatment cost introduced by adding CCS to the base plant, and  $R_{\text{CDR}}$  is the net revenue stream from CO<sub>2</sub> removal credits. Thus, the intersection of electricity revenue and  $R_{\text{CDR}}$  indicates the breakeven carbon price, where the revenue from CDR exceeds the revenue from energy production despite costs added from CCS.

## Case 2—Forestry residue

Case 2 calculations were performed using an economic model based on a gasification process that converts 4,536 dry tons per day into Fischer Tropsch (FT) fuels or hydrogen, along with varying amounts of electricity ([Zeng and Hausmann, 2022](#)). In both cases, the gasification process consists of a fluidized-bed gasifier fed with oxygen from an air separation unit (ASU). The gasifier produces a mixture of light combustibles (CO, H<sub>2</sub>, and CH<sub>4</sub>), heavies (tars and oils), and minor contaminants at about 1,000°C. After this gas is cleaned and cooled, it is processed to

TABLE 6 Data used to generate plots for Case 2, based on data from [Larson et al. \(2009\)](#) and equations from [Baker et al. \(2020\)](#).

	FT fuel—Base	FT fuel—CCS	HMAX—Base	HMAX—CCS	H5050—Base	H5050—CCS	Burial
Total energy revenue (low) (\$/tbio)	173.8	164.5	237.2	237.2	176.2	163.8	0.0
Total energy revenue (high) (\$/tbio)	173.8	164.5	310.8	310.8	209.8	197.4	0.0
Biomass treatment cost* (\$/tbio)	167.9	224.3	288.6	369.6	204.7	273.3	86.9
Profit (low) at \$0/tCO <sub>2</sub> (\$/tbio)	6.0	−59.8	−51.4	−132.4	−28.5	−109.5	−86.9
Profit (high) at \$0/tCO <sub>2</sub> (\$/tbio)	6.0	−59.8	22.2	−58.8	5.1	−75.9	−86.9

\*Treatment cost for CCS includes cost of CO<sub>2</sub> transport and storage; values rounded to nearest tenth.

TABLE 7 Carbon balance for Case 2.

	FT fuel—Base	FT fuel—CCS	HMAX— Base	HMAX—CCS	H5050—Base	H5050—CCS	Burial
External electricity (MtCO <sub>2</sub> /yr)	0.00	0.00	0.51	0.72	0.00	0.00	0.00
Emitted <i>via</i> conversion (MtCO <sub>2</sub> /yr)	1.66	1.66	2.22	2.22	2.22	2.22	0.00
Emitted upon fuel use (MtCO <sub>2</sub> /yr)	0.55	0.55	0.00	0.00	0.00	0.00	0.00
Captured and stored (MtCO <sub>2</sub> /yr)	0.00	−1.50	0.00	−2.00	0.00	−2.00	0.00
Initial biogenic uptake (MtCO <sub>2</sub> /yr)	−2.22	−2.22	−2.22	−2.22	−2.22	−2.22	−2.22
Net emissions (MtCO <sub>2</sub> /yr)	0.00	−1.50	0.51	−1.28	0.00	−2.00	−2.22
Net removed (MtCO <sub>2</sub> /yr)	0.00	1.50	0.00	1.28	0.00	2.00	2.22
Net removed (tCO <sub>2</sub> /tbio)	0.00	1.13	0.00	0.96	0.00	1.51	1.68

produce the desired products of electricity and either FT fuels or hydrogen. The model was originally developed by [Larson et al. \(2009\)](#) and was more recently adapted for forestry residue by [Baker et al. \(2020\)](#).

Like with [Figure 2](#) (Case 1), [Figure 3](#) (Case 2) is based on the energy revenue streams, added cost, and net CO<sub>2</sub> removal of the process. However, the original model of Larson et al. was meant for switchgrass, so the analysis here is slightly adapted for the characteristics of forestry residue. Specifically, the forestry residue is assumed to have a carbon content of about 46% and a calorific value of about 16.7 MJ/kg, based on an average value of wood wastes tested by [Greinert et al. \(2019\)](#). Electricity and fuel generation efficiencies reported by Larson et al. ([Table 4](#)) were applied to the forestry residue calorific value to estimate the generation of energy products.

The CAPEX also required some adaptations. The reported overnight CAPEX for the gasification plant was converted to 2019 USD using the chemical engineering plant cost index (CEPCI). Additionally, the equipment was scaled down linearly to reflect the slightly lower calorific value of forestry residue

compared to switchgrass (plant thermal capacity of 875 MW<sub>th</sub> compared to 893 MW<sub>th</sub> of the original model). The original model by Larson et al. does not include carbon capture or hydrogen liquefaction, so additional CAPEX was added using capacity-dependent equations reported by [Baker et al. \(2020\)](#), also converted to 2019 USD using CEPCI. Final CAPEX values are shown in [Table 4](#).

The biomass treatment cost (i.e., operating expense) also deviates from the original model of Larson et al., and more closely follows the method of Baker et al. A fixed capital charge factor of 15% and a fixed operating and maintenance rate of 4.5% were both applied to the total CAPEX for each case. The feedstock cost was assumed to be \$50 per dry ton ([Baker et al., 2020](#)). Power demand for the conversion process (shown in [Table 5](#)) is based on Larson et al. (linearly scaled as described for CAPEX), while the power demanded by CO<sub>2</sub> capture and H<sub>2</sub> liquefaction processes are calculated using equations given by Baker et al. Any power needs are initially satisfied by the power produced *via* the gasification process, and necessary excess power is taken from the grid. The cost of transporting

the compressed CO<sub>2</sub> 100 miles is estimated to be \$5/tCO<sub>2</sub> for the FT fuel process and \$4/tCO<sub>2</sub> for the hydrogen processes based on cost curves from Smith et al. (2021) (where the latter case is cheaper per ton because more CO<sub>2</sub> is transported). Injection costs are assumed to be \$11/tCO<sub>2</sub> (Psarras et al., 2020). All treatment costs are incurred based on a plant capacity of 4,536 dry tons of forestry residue per day with an annual plant throughput of 1,325 kt/year (assuming an uptime of 80%).

The revenue streams from the three energy products of electricity, FT fuels, and hydrogen are estimated based on selling prices of \$0.06/kWh, \$2.35/gal, and \$2.90–3.10/kg, respectively, based on values from Baker et al. (2020). The profit of each case is the difference between total treatment cost and energy-based revenue. These final values (shown in Table 6) are used in Equations (1) and (2) to generate the plot shown in Figure 3. The cost of CO<sub>2</sub> transport and storage (required for Equation 1) is included in the biomass treatment costs shown in Table 6.

Also shown in Table 6 are parameters for forestry residue burial. These were modeled using the same annual throughput (1,325 kt) and feedstock price (\$50/t) as the gasification process. The operating expense is based on the cost of land purchase, construction, and operation, which (Zeng and Hausmann, 2022) estimate to be about \$13–22/tCO<sub>2</sub> sequestered. For a conservative comparison, we choose the upper bound of this estimated range.

The final parameter necessary for Equation (2) is the net-removed CO<sub>2</sub>. The carbon balance for processes in Case 2 was estimated using the biomass carbon content. For gasification products, FT fuels retain 25% of the feedstock carbon, while H<sub>2</sub> retains 0%; the remaining carbon content is emitted in the flue gas, with 90% of the emissions being captured. The carbon retained in the FT fuel is still included in the carbon balance, as it would later be emitted upon fuel combustion. Meanwhile, burial in specially engineered facilities is assumed to retain 100% of the carbon within the forestry residue (Zeng and Hausmann, 2022). The HMAX process does not produce enough electricity to satisfy all of its power demand, so it has some emissions from grid electricity with an emissions intensity of 170 gCO<sub>2</sub>/MJ (Baker et al., 2020). The emissions of each process are summed to determine the net emissions as shown in Table 7. If the energy products replace carbon-intensive products, additional emission reductions will be realized that could further reduce the net emissions and increase the net removed CO<sub>2</sub>. However,

because the emissions intensity of the counterfactual energy product varies with time and location, it is not included in the carbon balance.

## Data availability statement

The raw data supporting the conclusions of this article will be made available by the authors upon request.

## Author contributions

CW and CM contributed to conception and design of the study. CW performed the spreadsheet analysis, performed extensive review, suggestive revision of the spreadsheet analysis, and wrote the first draft of the manuscript. All authors contributed to manuscript revision, read, and approved the submitted version.

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## Conflict of interest

Author CM was employed by company Carbon Direct.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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