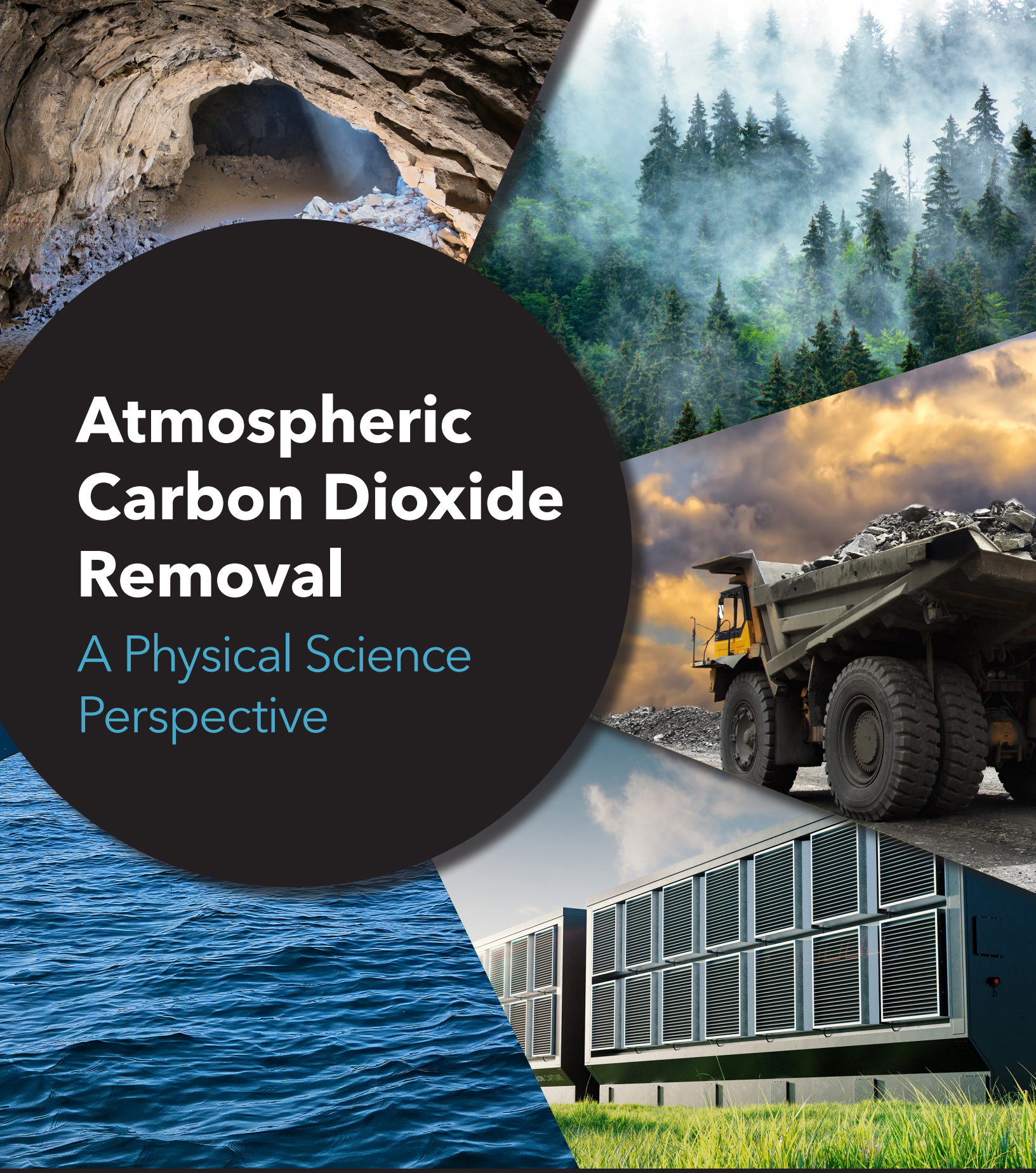

# Atmospheric Carbon Dioxide Removal

## A Physical Science Perspective

A SCIENCE POLICY REPORT ISSUED BY
the American Physical Society

**JANUARY 2025**

APS Advancing Physics

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This report was overseen by the APS Panel on Public Affairs. POPA routinely produces reports on timely topics being debated in government so as to inform the debate with the perspectives of physicists working in the relevant issue areas.

## Authorship

The American Physical Society has sole responsibility for the contents of this report and the questions, findings, and recommendations within. The views expressed in this report do not necessarily represent the views of the employers of the report authors and reviewers.


## Acknowledgments

The study group would like to express particular gratitude to Nina Anikeeva (MIT) for contributing to the research and writing of this report, to the APS POPA, Council, and Board members for their input and support, and to all the individuals consulted in the course of writing this report for providing us with their expertise.

We are very grateful to APS staff, particularly to Jorge Nicolás Hernández Charpak, who worked closely with the study authors throughout the report. Thanks also to Kendra Redmond for her help improving the report and to Travis Frazier for figures and graphic design.

## Publication Date: January 2025





Cover and report designed by Travis Frazier.


For additional information, including the participant bios and the report charge, please visit: https://www.aps.org/policy/reports/popa-reports/index.cfm

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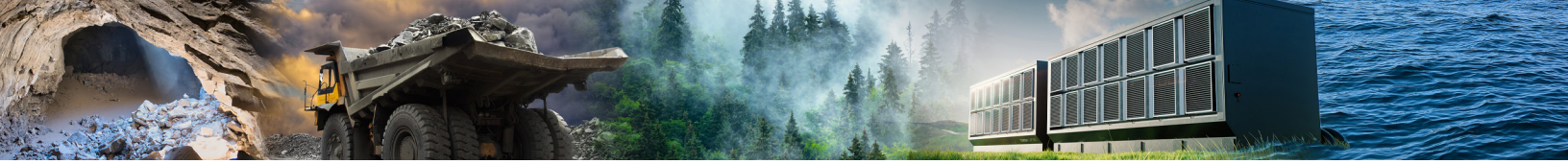

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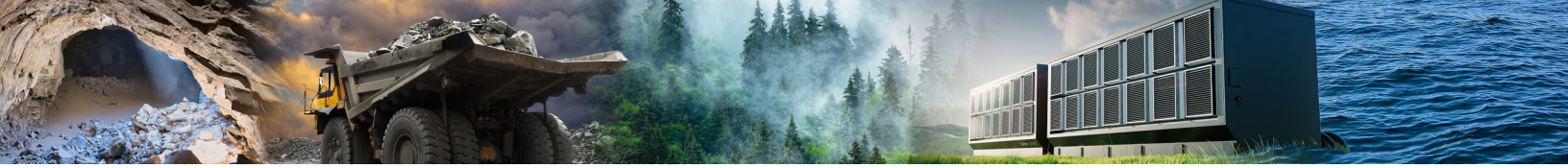


# Executive Summary

Humanity's activities currently lead to a net increase in atmospheric carbon dioxide ($CO_2$) by more than 15 billion metric tons per year. Concerns about the global consequences of this continuing rise, coupled with growing global energy needs, political constraints, and economic challenges that limit the rate of reduction of global $CO_2$ emissions, are driving strong interest from both the public and private sectors in the development of atmospheric carbon dioxide removal (CDR) methods.

CDR implementation methods, sometimes referred to as negative emission technologies, reduce the $CO_2$ levels in Earth's atmosphere by removing carbon dioxide molecules from the air and sequestering them for very long times. These approaches are distinct from carbon capture and storage (CCS) technologies that remove $CO_2$ at fossil fuel-powered sources, such as power plants or cement production facilities. CCS at fossil $CO_2$ point sources can help reduce emissions but does not remove $CO_2$ already in the atmosphere.

This report provides an overview of CDR methods and their fundamental physical constraints. We focus on CDR approaches that have the potential to contribute to $CO_2$ removal at the gigaton/year scale (where 1 gigaton = 1 Gt = 1 billion metric tons). This is the scale of effort that would be needed to have a meaningful global impact.

For those making and implementing policy, the general findings and recommendations of the report may provide a helpful science-based perspective. For those supporting or working directly on research and development efforts, and for those with some scientific background interested in a deeper understanding of the issues, the main text of the report includes concise but relatively self-contained descriptions of various technical and scientific points relevant to the broader conclusions of the report. The central findings and recommendations are outlined below. Section 7 provides more detailed guidance for policymakers and for potential funders of CDR research, pilot programs, and technology.

**Finding 1: CDR approaches divide loosely into two distinct categories, cyclic and once-through. CDR methods in these two categories have different features and challenges. Many approaches have been proposed in each category, although most approaches are still at early stages of development.**

Cyclic CDR systems, such as chemical direct air capture (DAC), use the same materials to capture atmospheric $CO_2$ over and over. These systems are necessarily energy intensive, but may enable carbon sequestration that is easier to verify and has less environmental impact than other approaches to CDR.

Once-through CDR systems, such as enhanced rock weathering (ERW), involve the single use of a resource. For example, many such approaches employ minerals extracted from the Earth to directly or indirectly absorb $CO_2$. These systems may use less energy, but they generally require extensive resource extraction and may have a significant environmental impact. Their effectiveness has been challenging to measure and verify.

**Finding 2: CDR at the scale of many gigatons of $CO_2$ per year requires large amounts of energy, particularly for cyclic processes.**

Capturing one gigaton of $CO_2$ from the atmosphere using cyclic CDR processes requires a lot of energy. The laws of physics dictate the absolute minimum energy needed, which is comparable to the combined electric consumption of 11 million average U.S. households in 2021 (14 gigawatt-years). The cyclic CDR approaches currently in development, including existing small-scale installations, require at least several times the theoretical minimum energy requirement for any amount of carbon captured.

**Finding 3: CDR at the scale of many gigatons of $CO_2$ per year requires processing large quantities of materials, particularly for once-through processes.**

For once-through processes, capturing several gigatons of $CO_2$ annually would require substantial effort. This would include mining, grinding, and widely distributing quantities of rock comparable to the mass of all cement produced globally, on a yearly basis. For cyclic DAC systems, capturing $CO_2$ at the rate of 1 Gt per year would involve processing a quantity of air at the scale of all AC/cooling systems currently operating on the planet.

**Finding 4: Unlike past experience with computing technology, there is no prospect of exponential growth in capability for any CDR approach.**

The semiconductor industry has notably doubled the number of transistors within a fixed space or at a fixed cost every two years since 1965. This rapid, exponential increase in computing capability is often referred to as Moore's law. There is no possible version of Moore's law for CDR. Physical constraints make it clear that there is no path by which the amount of carbon captured at a fixed energy or material cost will scale in a similar way. Note that achieving emission reductions through the replacement of fossil fuel energy sources by zero-carbon sources is similarly based on extensive material and infrastructure use and has similar constraints.

Based on the findings of this report, we recommend the following.

**Recommendation 1: Research and development on CDR should be selectively and prudently pursued, despite the many challenges described in this report.**

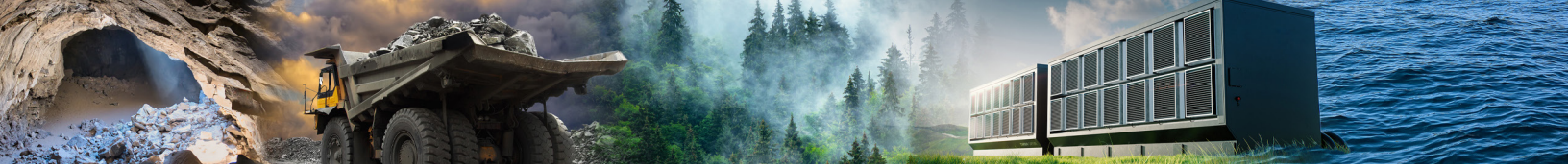
Even with sharp reductions in $CO_2$ emissions, CDR may be needed at the scale of 1-20 Gt/yr later this century to reach desired targets. Research investments and deployment of trial/demonstration projects can aid in the development of effective, economical CDR systems. In particular, they can improve understanding of the full life-cycle energy and material needs — along with possible negative consequences — of each of these approaches. Understanding these issues clearly can inform sound policy decisions. Funding agencies should carefully consider energy and materials needs, as well as scalability, for any proposed CDR project. A portfolio of approaches is likely necessary for a large-scale reduction in atmospheric $CO_2$. CDR technologies are rapidly evolving and new ideas continue to emerge. Thus, care should be taken not to commit to a specific approach that may limit innovation.

**Recommendation 2: The prospect of large-scale CDR should not compromise efforts to reduce carbon emissions.**

CDR at a large scale requires vast energy and material resources. Reducing carbon emissions is in general a more direct way of decreasing future atmospheric carbon levels. Emissions reductions can be achieved through changes in energy sources, systems, and infrastructure and by reducing carbon loss from natural and managed ecosystems (e.g., avoiding deforestation); efforts in these directions should not be reduced simply based on hypothetical future CDR scenarios without a fuller understanding and comparative analysis of cost, environmental impact, and lifecycle requirements.

**Recommendation 3: Ecosystem-based CDR approaches should be pursued when they are feasible and effective and do not compromise food production or other priorities.**

Carbon representing hundreds of gigatons of $CO_2$ has been lost from soil and terrestrial biomass through human activity. Reforestation, restoring wilderness, and changing agricultural practices can recapture some of this carbon at a relatively low cost without compromising other needs like food security. In the near term, this is likely the most immediate and cost-effective approach for reducing atmospheric $CO_2$ by one or more gigatons per year. However, the scale and net capture from these approaches are limited and depend



upon sustained land use changes over very large areas. In addition, measurement and verification can be difficult, and carbon can be returned to the atmosphere by fires, logging, erosion, and other disturbances.

**Recommendation 4: CDR planning must incorporate substantial additional carbon-free power sources, particularly for cyclic approaches. Such planning should also consider the land or sea area required and impacted.**

Implementing CDR at the scale of multiple gigatons of $CO_2$ per year using any cyclic approach should include installing dedicated carbon-free or low-carbon power sources. Direct air capture systems (DAC) at the several gigatons per year scale would require hundreds of gigawatts of additional power beyond what is needed for other existing and future energy uses. Using renewable sources to power any cyclic CDR approach may require substantial land areas. To reach scale, biologically based CDR approaches, such as bioenergy with carbon capture and storage (BECCS), may require vast land areas to gather the needed energy via photosynthesis. This may compete with food production, other land uses, and conservation. Nuclear power is a carbon-free power source that requires little land area, but raises issues of cost, safety, waste, and security. Once-through approaches also have significant energy requirements and the potential to impact large land and sea areas. Funding agencies should request that R&D plans for any CDR approaches seeking support should identify the expected energy demand and power source, and should provide a careful accounting of the land area needed and impacted.

**Recommendation 5: Once-through CDR approaches should not be deployed on a large scale until research determines their effectiveness and impact.**

Once-through approaches such as enhanced weathering and ocean alkalinity enhancement use large quantities of ground minerals to capture $CO_2$ directly through chemical reactions or indirectly by modifying ocean chemistry. While these may be promising large-scale solutions, their efficacy and environmental impacts are not yet clearly established. Further research and small-scale trial projects are needed before any attempt at large-scale implementation of these approaches.

**Recommendation 6: Reliable systems of measurement, reporting, and verification (MRV) must be developed for CDR systems. Robust standards for sequestration and for safety and environmental impact are also needed.**

Accurate measurement, reporting, and verification (MRV) mechanisms must be established for any $CO_2$ capture program to be effective. In a fast-moving technological landscape, MRV capabilities are crucial for assessing and comparing different approaches. The federal government, in partnership with the private sector, should establish clear standards and procedures for MRV. Standards are also needed to quantify and minimize or mitigate the risks and environmental impact associated with CDR. Evaluation of any proposed large-scale CDR system should include an assessment of its interactions with other components of Earth's coupled climate and biosphere systems.

**Recommendation 7: Economic and policy frameworks for carbon management should be designed and implemented. Such frameworks are needed to facilitate balancing the costs and benefits of large-scale CDR and emission reduction strategies.**

Large-scale carbon capture has enormous energy and material requirements, and would be quite costly, e.g., estimated at the level of hundreds of billions of dollars per gigaton for large-scale chemical DAC. Currently, there is no clear domestic or international framework for paying for large-scale CDR. Policies should be developed that enable an efficient balancing of CDR and emission reduction strategies. Such policies could include, for example, a mechanism for imposing a cost for carbon emissions that reflects the cost of abatement, and commitments by governments and/or private industries to bear these costs through an internationally equitable framework. Specific policy recommendations are outside the scope of this report.

# Glossary

A number of similar terms and acronyms are used to refer to carbon reduction strategies. This brief glossary describes how we use these and other relevant, common terms and acronyms in this report.

**BECCS:** Bioenergy with carbon capture and storage, §4.2.2.

**BiCRS:** Biomass with carbon removal and storage, §4.2.3.

**CCS:** Carbon capture and sequestration, refers to general approaches to carbon capture and sequestration, in particular including non-CDR approaches such as carbon capture (pre-/post-combustion) at point sources such as fossil fuel power plants, §1.1.

**CDR:** Carbon dioxide removal, refers to anthropogenic activities that remove $CO_2$ from the atmosphere and store it durably in geological, terrestrial, or ocean reservoirs, or in products, §1.

**DAC:** Direct air capture, primarily refers to chemical direct air capture, a specific class of approaches to carbon capture that can be used in combination with sequestration/storage for CDR (DACCS), §4.1.

**DACCS:** Direct air capture with carbon storage, see also DAC.

**DIC:** Dissolved inorganic carbon, refers to all inorganic carbon species in an aqueous solution, App. G.

**DOC:** Direct ocean capture, refers to the direct removal of $CO_2$ from the oceans, a specific class of approaches to CDR, §4.4.2.

**ERW:** Enhanced rock weathering, §4.3.

**LULUCF:** Land use, land use change, and forestry, App. H.

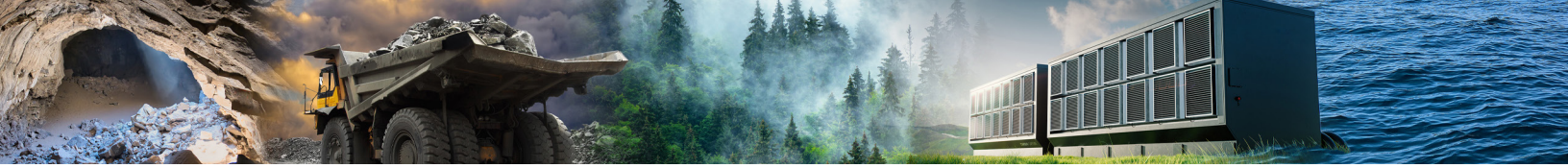
**mCDR:** Marine CDR, §4.4.

**MRV:** Measurement, reporting, verification, §6.1.

**NET:** Negative emissions technologies, see CDR, above.

**OAE:** Ocean alkalinity enhancement, §4.4.1.

**SOC:** Soil organic carbon, App. I.



# Units

For convenience, we list here some of the units that occur frequently in this report.

## Mass units for carbon dioxide and carbon:

One metric ton of $CO_2$ (1 t $CO_2$ = 1,000 kg $CO_2$ ≅ 2,205 lb $CO_2$) contains 272 kg of carbon (272 kg C) (see App. A).

1 Mt (megaton) = 1,000,000 t

1 Gt (gigaton) = 1 Pg (petagram) = 1,000,000,000 t

## Energy units:

Energy is often measured in joules (J) or kilowatt-hours (kWh), with 1 kWh = 3.6 MJ = 3,600,000 J.

1 MWh = 1,000 kWh

1 GWh = 1,000,000 kWh

1 MWy = 8,760 MWh = 8,760,000 kWh

1 GWy = 8,760,000 MWh = 8,760,000,000 kWh

1 MJ (megajoule) = 1,000,000 J

1 GJ (gigajoule) = 1,000,000,000 J

## Power units:

Units of energy and power are often confused.

Power is a rate of energy production, use, or transfer per unit time, and is measured in watts (W), with 1 W = 1 J/s, 1 kW = 1,000 J/s.

A kilowatt-hour is thus the amount of energy produced, used, or transferred over one hour at a rate of 1 kW.

1 kW (kilowatt) = 1,000 W

1 MW (megawatt) = 1,000,000 W = 1,000 kW

1 GW (gigawatt) = 1,000,000,000 W = 1,000,000 kW

1 TW (terawatt) = 1,000,000,000,000 W = 1,000,000,000 kW

# I. Introduction

Atmospheric carbon dioxide ($CO_2$) levels have continued to rise rapidly in recent years, due in large part to $CO_2$ emissions from fossil fuel use. As human energy needs and population continue to expand around the globe, efforts for significant reductions in $CO_2$ emissions in the coming decades face a number of political, economic, and technical challenges. Furthermore, even with a substantial shift toward carbon-free or carbon-neutral energy sources, some sectors will likely emit significant amounts of carbon from fossil sources into the atmosphere well into the future.

It is widely accepted that the continued rise in atmospheric $CO_2$ levels will have significant global consequences, although the precise extent of the impacts on Earth's complex climate and living systems is uncertain. As a result, the public and private sectors have expressed increasingly strong interest in developing methods and technologies for directly removing carbon dioxide molecules from the air. Such carbon dioxide removal (CDR) methods are also often referred to as negative emissions technologies. [1]

Large-scale deployment of CDR systems can in principle complement emissions reduction strategies to keep atmospheric carbon levels below thresholds of concern. While emission reduction can reduce the rate of growth of atmospheric $CO_2$ levels, CDR has the unique potential to directly reduce atmospheric $CO_2$ and realize net-negative emissions, and can also potentially offset ongoing emissions that are difficult or expensive to eliminate in the longer-term. Balancing the allocation of resources between emission reduction, CDR, and other pressing societal concerns, including energy needs, is a complex policy and values problem for which a clear understanding of the potential benefits and resource requirements of CDR is a crucial input.

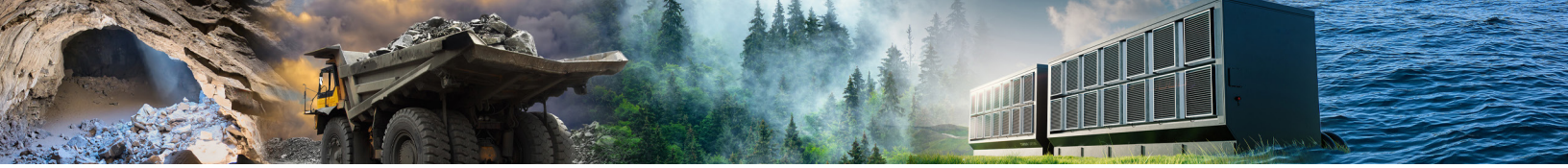
There are a number of possible approaches to CDR. Determining which approach(es) may be most effective and implementing these approaches pose substantial scientific and engineering challenges. Fundamental physical constraints, however, show that any carbon dioxide removal effort that would affect atmospheric $CO_2$ concentrations at a policy-relevant scale in the coming decades will require tremendous quantities of energy and materials.

This report is an effort by the APS Panel on Public Affairs (POPA) to provide a concise and clear summary of the main proposed approaches to carbon dioxide removal (CDR), with an emphasis on fundamental physical constraints, in order to provide policy guidance and perspective for a variety of stakeholders based on sound science.

## 1.1 Framing of This Report

Carbon dioxide reduction (CDR) methods have been studied extensively. There have been a number of recent studies and reports on CDR, including a prior POPA report on chemical direct air capture (DAC) [2], two National Academies studies [3, 4], books [5, 6], and online resources such as the "Carbon Dioxide Removal Primer," [7] the "Roads to Removal" report [8], and a "Strategy for NOAA Carbon Dioxide Removal Research" [9]. There are also reviews of many aspects of CDR in the "6th Assessment Report of the Intergovernmental Panel on Climate Change" (IPCC); see particularly [1]. A recent review of the status of various CDR approaches and technologies appears in [10]. This report builds upon these and other published reports and analyses as well as primary literature, and on discussion and consultation with subject matter experts. This report followed APS POPA report procedures and was peer-reviewed by experts in various relevant fields before approval by the APS Council.

[1]Following standard usage [1], in this report we define CDR as anthropogenic activities that remove $CO_2$ from the atmosphere and store it durably in geological, terrestrial, or ocean reservoirs, or in products.



This report focuses specifically on some of the fundamental physical limitations of CDR methods and how these limitations constrain existing and proposed CDR systems. The report considers a broad range of approaches to capturing atmospheric carbon in the form of $CO_2$; methane and other non-$CO_2$ greenhouse gases are discussed briefly in App. L. This report does not cover technologies for carbon capture at fossil fuel-based point sources. [2] Solar radiation management (SRM) efforts that would, for example, reflect some solar radiation without addressing the underlying atmospheric carbon issues are also outside the scope of this report. This report summarizes the basic physical limits and scaling aspects of several approaches to removing atmospheric carbon. It does not attempt a detailed comparison of specific technologies, nor recommend specific companies or enterprises in this domain.

Understanding the principles underlying the wide range of proposed CDR methodologies, and the constraints on these approaches, involves concepts and analyses from many fields, including physics, chemistry, biology, and earth sciences. To make the report as readable and self-contained as possible, we include a number of technical appendices with concise descriptions of some key scientific principles relevant for the systems described here, as well as a glossary of terms and acronyms. In various places we have included 'back-of-the-envelope' estimates that are intended to communicate the scales involved or plausibility of certain issues or assertions. These estimates are not intended to provide precise estimates or values.

This report is aimed at readers with a wide range of technical backgrounds. For readers primarily interested in policy-level questions, the executive summary, introduction (§1), conclusions (§7), and summary boxes may be of most interest. The main text expands on various points in more detail, and different sections may be of interest to different readers. The footnotes and appendices include more technical points and arguments, as well as detailed background on relevant subjects, and may be of greater interest to those with a more technical background.

[2] Carbon capture and storage (CCS) at fossil $CO_2$ point sources (§6.8) can be part of a low-carbon energy portfolio but does not directly reduce atmospheric carbon; when combined with biological energy sources, however, point-source capture can provide a net-negative carbon technology (BECCS, §4.2.2).

## 1.2 Anthropogenic $CO_2$ and the Motivation for CDR Research and Development


**Summary: Motivations for CDR**

As of 2023, human activity adds approximately thirty-five gigatons (35 Gt) of $CO_2$ to Earth's atmosphere yearly from fossil fuel sources. Roughly half of this added $CO_2$ is absorbed into the oceans and terrestrial biomass.

Limiting the total warming from anthropogenic $CO_2$ to 1.5 or 2 °C above preindustrial times would involve removing multiple gigatons of $CO_2$ per year of CDR later in this century, according to scenarios studied by the IPCC and others. Independent of such scenarios, estimates of climate sensitivity suggest that to limit warming to 1.5 °C, much or all of the net atmospheric $CO_2$ increase beyond current levels would need to be offset by CDR.

Even with a higher bound on atmospheric $CO_2$ and significant emissions reductions, CDR at the scale of multiple gigatons of $CO_2$ per year may be desired to offset continuing emissions.


Human activity over the last century has led to an increase in atmospheric $CO_2$ levels from 280 ppm (parts per million by volume) in preindustrial times to around 420 ppm today (Fig. 1). This increase is well understood and well measured, and comes primarily from the combustion of fossil fuels (coal, oil, natural gas, etc.), with additional contributions from activities such as cement production and deforestation. Using fossil fuels releases carbon into the atmosphere-terrestrial carbon cycle that has been geologically sequestered for millions of years. Such changes in carbon levels are fundamentally different from effects such as methane production from biological activity, which modifies the form and distribution of carbon already in the system but does not add net carbon.

Current rates of fossil fuel use add approximately 35 Gt $CO_2$ [3] to the atmosphere each year. Roughly another 3-4 Gt $CO_2$/yr of emissions come from industrial processes such as cement production, and there is a comparable flux of about 3-4 Gt $CO_2$/yr into the atmosphere from other human activities such as land use and deforestation. Roughly half of this total anthropogenic carbon release is currently taken up by natural land and ocean systems (App. H), leading to a net rate of increase in the atmospheric $CO_2$ concentration of over 20 ppm each decade since 2000. Fossil fuels are compact and reliable sources of energy that have improved the human condition over the last century. However, the ongoing increase in atmospheric carbon levels will continue unless and until fossil fuel usage is substantially replaced by carbon-free alternatives such as solar, wind, other renewables, nuclear power, or fossil fuel plants with point-source carbon capture and minimal residual emissions. There is also significant uncertainty regarding how long the ocean and land sinks will continue to take up $CO_2$ at the current rate. A more complete picture of changes in atmospheric $CO_2$ over varying time scales is given in App. L.

[3] Note: Carbon dioxide fluxes are sometimes measured in (metric) tons of carbon (t C), and sometimes in tons of $CO_2$ (t $CO_2$); we primarily use Gt $CO_2$, but depending upon context we go back and forth between these units, which are related by ~12/44, the mass fraction of carbon in $CO_2$ (see App. A). Thus, for example, anthropogenic carbon emissions from fossil fuel usage are often alternatively quantified as 9-10 Gt of fossil carbon. One gigaton (Gt) is one billion tons (109 t), while one megaton (Mt) is one million tons ($10^6$ t), and one kiloton (kt) is one thousand tons ($10^3$ t).

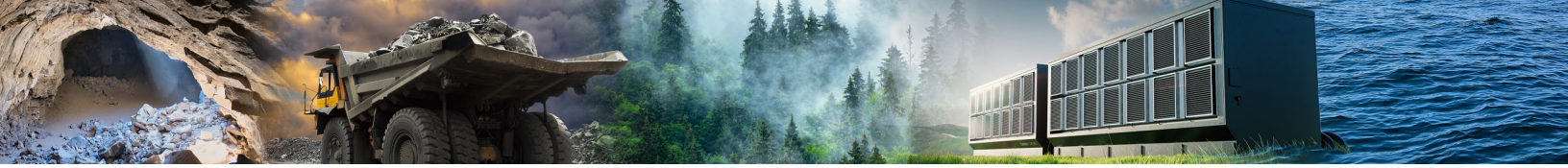
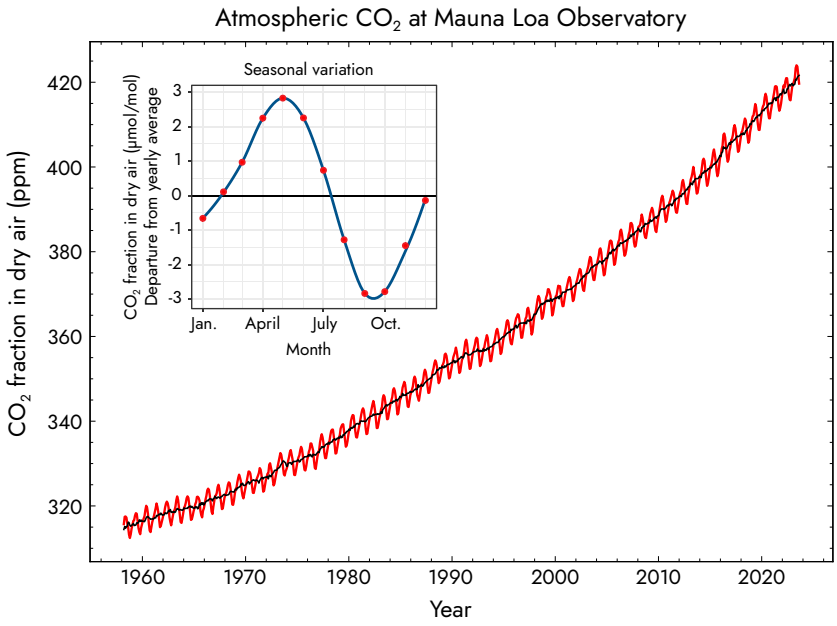


**Figure 1:** Atmospheric $CO_2$ levels from 1958 through mid-2023, as measured at the Mauna Loa Observatory; observations from 12/22-7/23 were taken at Mauna Kea. (Adapted from a figure by NOAA/Scripps [11].)




In projected scenarios where atmospheric carbon levels are managed so that Earth avoids a surface temperature rise of more than 1.5 ˚C or 2 ˚C without drastic and immediate cessation of $CO_2$ emissions, the IPCC and others incorporate CDR at the level of multiple gigatons of $CO_2$ per year in the latter part of this century [4, 12, 1, 10]. These scenarios depend upon estimates of climate sensitivity and assumptions regarding the future behavior of ocean and land sinks, both of which are difficult to predict with precision. Even with more optimistic (lower) estimates of climate sensitivity, CDR at the scale of multiple gigatons per year may be needed in the coming decades to keep temperatures within a desirable range. (See App. L for more details and references.) These considerations have recently led to substantial public and private efforts to understand and develop approaches and technologies for removing $CO_2$ from Earth's atmosphere.

Even with dramatic reductions in $CO_2$ emissions in the coming decades, it may be difficult or impossible to completely eliminate anthropogenic $CO_2$ emissions. Assessing the magnitude of these residual emissions is challenging. One analysis [7] based on a number of studies estimates 1.5-3.1 Gt $CO_2$/yr of difficult-to-avoid emissions. [4] While there is significant uncertainty in such estimates, in the longer (century) time scale — even if most fossil fuel usage is replaced with carbon-free energy sources — ongoing CDR at the level of one or several gigatons of $CO_2$ per year may be needed to keep atmospheric $CO_2$ levels relatively constant.

In this report we discuss CDR in units of 1 Gt $CO_2$/yr of atmospheric carbon dioxide removal. Efforts to stabilize atmospheric $CO_2$ may combine emissions reductions and CDR strategies in various ways, with approaches contributing at different levels. For example, [13] proposes a U.S. effort to achieve 1 Gt $CO_2$/yr of CDR by 2060.

[4] Difficult-to-avoid emissions may include, for example, emissions from shipping, trucking, and air transport that are difficult to completely eliminate, although there are, current efforts to develop alternatives such as electric, biofuel-powered, or hydrogen-powered airplanes. The estimate quoted also includes $CO_2$ equivalent greenhouse gas emissions from non-fossil sources such as nitrous oxide from agriculture; while such emissions can also cause net radiative forcing and associated warming, they are not involved in the long-term atmosphere-terrestrial carbon cycle, which is the primary focus of this report (see App. L for further discussion).

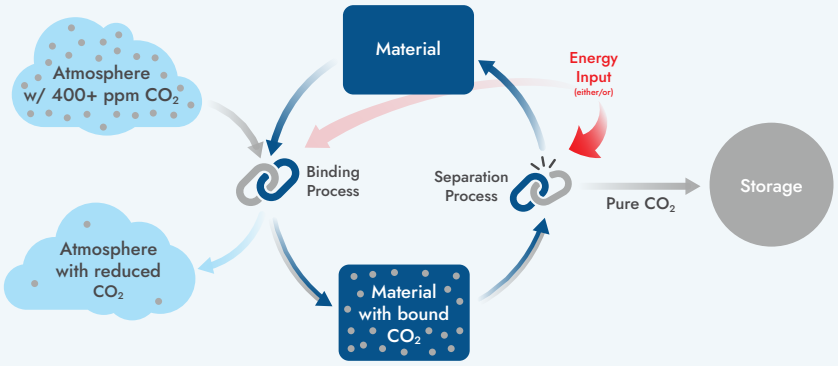
# 2. Classification of CDR Methods

This section gives an overview of CDR approaches. We begin with a conceptual classification of two distinct types of CDR systems (§2.1). We then give an overview of some of the main CDR approaches (§2.2); these are described in more detail in §4.

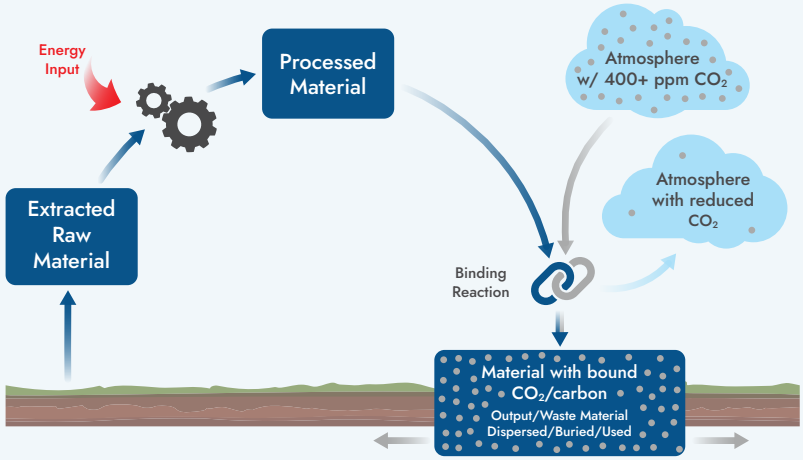
## 2.1 Classification: Cyclic vs. Once-Through Carbon Capture Processes

### Summary: Cyclic vs. Once-Through CDR Systems

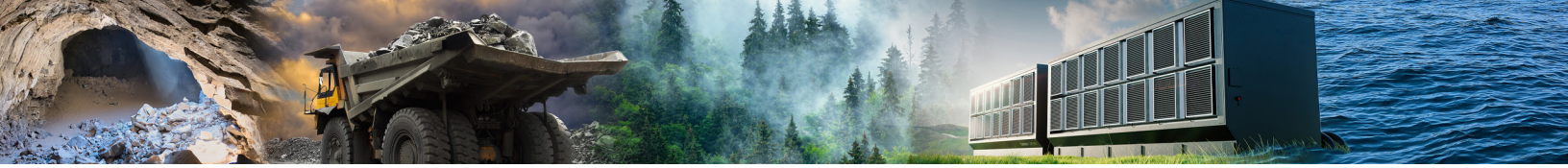
CDR systems can be conceptually divided into two basic types:




Cyclic CDR systems, such as chemical DAC, use a single batch of material to capture $CO_2$ over and over again in a cyclic process. After the material captures some $CO_2$, the $CO_2$ is extracted (typically using heat), the CO2 is separated into a relatively pure stream suitable for storage (geologic sequestration or mineralization), and the material is reused in the next cycle. The primary input (aside from air) for cyclic systems is energy, although some cyclic systems also require substantial amounts of water and, in application, materials degrade and must be replaced over time. For cyclic CDR systems, the second law of thermodynamics gives a lower bound on required energy.




Once-through CDR systems, such as enhanced weathering and ocean alkalinity enhancement (OAE), use materials that are out of equilibrium with the atmosphere, such as alkaline minerals extracted from the earth or certain kinds of industrial byproducts. After being processed, for example by grinding to a fine powder, this



material is distributed broadly and allowed to react with atmospheric $CO_2$ to bind the carbon. The reaction products are often left in place and/or dispersed into the environment, with potentially significant (negative or positive) environmental consequences. In more controlled situations the products may be disposed of or used in industrial processes (e.g., in construction materials).

The essential difference between these two types of systems is that the thermodynamic bound on minimum energy needs applies directly to all cyclic systems but not to once-through systems.

A wide variety of approaches for reducing atmospheric $CO_2$ concentrations have been proposed in recent years. Some involve using natural or engineered materials to directly bind $CO_2$ from the atmosphere (or from ocean surface waters) through chemical or biological processes. Other approaches involve modifying ocean chemistry or terrestrial biological systems to retain more carbon out of the atmosphere.

In some systems, the captured carbon is released in the form of (relatively) pure $CO_2$ that can easily be sequestered or used. In other systems the captured carbon is bound into solids, integrated into soil, tied into modified ocean chemistry, or ends up in other forms.

Carbon dioxide capture methodologies can be roughly separated into two distinct categories by distinguishing cyclic processes from once-through processes. The key difference between these categories of processes is that there are energetic constraints from thermodynamics that provide a clear lower bound on the energy needed for cyclic processes, while for many once-through CDR approaches, such as ERW and OAE, some of this energy has been supplied over millennia by natural processes. We characterize a cyclic carbon capture process as one in which the materials involved in the capture process are reused over and over as carbon is repeatedly separated from the atmosphere and then removed from the capture system. [5] In an ideal cyclic carbon capture process, the only inputs are energy and air (or $CO_2$-containing seawater), and the outputs are a stream of pure $CO_2$ and the residual air (or seawater) with reduced carbon content. Some cyclic systems also use water in each cycle, and in practice materials and system components must be replenished periodically. However, a once-through process generally uses a material only once in a chemical or biological reaction that captures molecules of $CO_2$. The material used in such processes may be extracted from a natural source or manufactured (including industrial byproducts). The outputs of a once-through process include a modified version of the input material that cannot easily be reused to capture more carbon.

Cyclic and once-through capture systems generally have different types of physical constraints as well as different environmental impacts. They also have different measurement, reporting, and verification (MRV) concerns and technical challenges. [6]

A principal characteristic of cyclic systems is that they have a minimum energy requirement given by the second law of thermodynamics, described further in §3.1. Most existing and proposed chemical DAC systems (§4.1) are cyclic-type systems. The output stream of relatively pure $CO_2$ from a cyclic capture system can be geologically stored or used in other ways (§5). Hybrid (engineered/biological) carbon capture systems such as BECCS (§4.2.2) where the output is basically a pure $CO_2$ stream are also, in essence, cyclic systems, when the full set of processes including initial photosynthesis by

[5] This parallels the notion of a cyclic ideal heat engine, as described in introductory physics and engineering courses.

[6] In the CDR literature, cyclic systems and once-through systems are sometimes referred to as 'closed systems' and 'open systems,' respectively, by analogy with these types of thermodynamic systems. We avoid this potentially confusing terminology because cyclic CDR approaches generally involve the transfer of $CO_2$ in and out of the system, while in standard physics, chemistry, and engineering terminology a closed system is defined as one that does not allow the transfer of matter in or out of the system.

plants is considered. The materials used are the ingredients of the natural ecosystem (e.g., water/$H_2O$), and the energy input comes directly from solar radiation. Note that in this case, however, the working material ($H_2O$) is drawn from and returned to the environmental pool, so the specific molecules involved vary from cycle to cycle.

In contrast, once-through systems can avoid the energy constraints imposed by the second law of thermodynamics if the input material is found in nature (or results from industrial processes) in a state that is out of equilibrium with the atmosphere. Generally, such materials bind with carbon dioxide through an exothermic (heat-releasing) reaction. Carbon removal proposals of this type include accelerated weathering of silicate rocks (ERW, §4.3) and ocean alkalinity enhancement (OAE, §4.4.1). While such systems do not have the same minimum energy input requirements (§3.1) as cyclic systems (the needed energy is already provided by geologic processes), they still have substantial energy needs (e.g., grinding and transporting the material). Furthermore, because the output of these systems is not pure $CO_2$, and in many cases ends up widely distributed on land or in the ocean, there are complicated environmental and ecological issues to consider. In particular, the mass of the input material is generally several times greater than that of the removed $CO_2$. That means, for example, that removing 1 Gt of $CO_2$ yearly through such systems would involve distributing something like one to several gigatons of material into the environment each year.

As a simplistic analogy, the distinction between cyclic and once-through CDR methods is like the difference between cleaning up a spilled liquid in the kitchen using a sponge/dishcloth or a paper towel. The sponge can be cleaned and re-used while the paper towel is discarded with the mess. While production of a sponge is more resource-intensive than that of a paper towel, just as for cyclic and once-through CDR approaches a full system analysis including number of cycles/duration of use would be required to fully evaluate the tradeoff between specific versions of these approaches. [7]

For ecosystem-based CDR approaches such as reforestation or biomass with carbon removal and storage (BiCRS, §4.2.1), the classification into cyclic versus once-through is slightly less clear. On the one hand, these approaches, which use solar energy to combine atmospheric $CO_2$ with water and other materials to form products that are retained in soil and biomass or are sequestered, are essentially once-through processes. On the other hand, unlike the other once-through processes considered here such as ERW and OAE, the materials used (primarily water) are more readily available, and these materials are not out of equilibrium with the atmosphere, so that the solar energy needed for carbon capture significantly exceeds the second law lower bound (App. E). Thus, from the point of view of energy requirements, ecosystem-based CDR fits most closely with the cyclic classes of processes considered here.

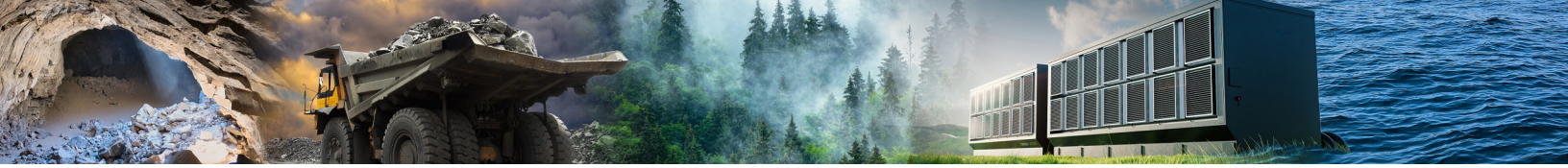
In this report we consider both cyclic and once-through types of carbon capture systems. We clarify further how some of the many ideas proposed in this arena fit into this classification and parse the resulting physical requirements and system issues in that context.

## 2.2 Approaches to CDR

In this section, we briefly describe some of the possible approaches to CDR (see Fig. 2). More detailed analyses of these and other approaches are found in §4.

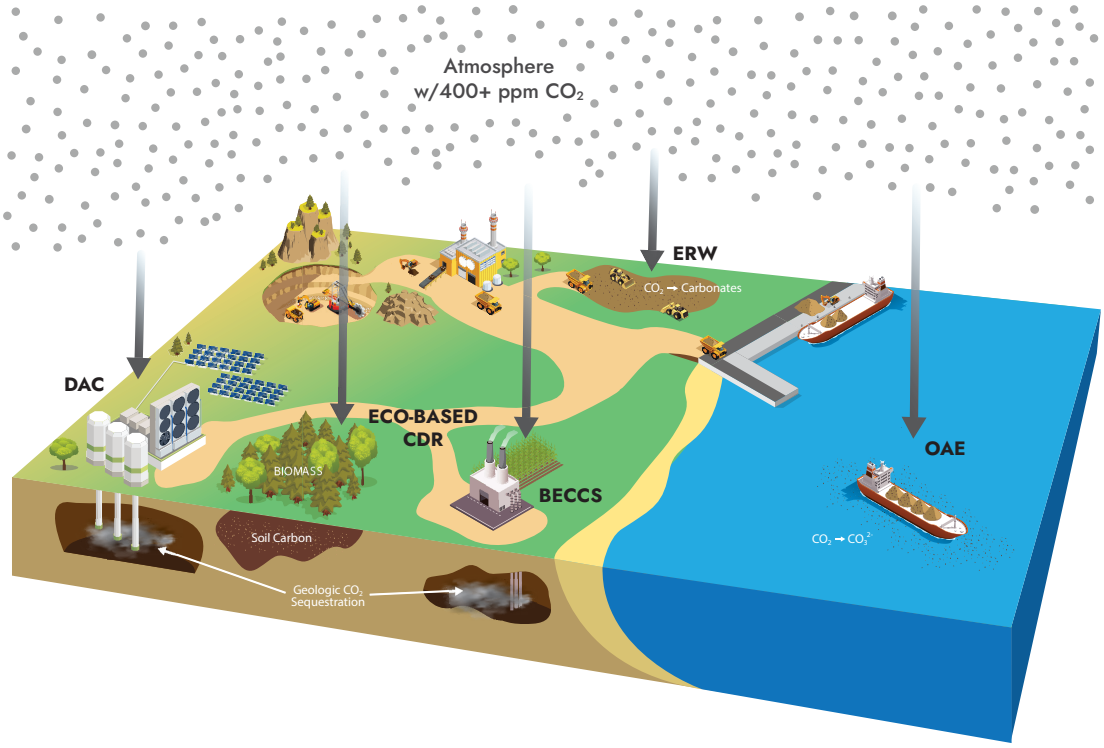
- **Chemical direct air capture (DAC):** In chemical direct air capture systems (§4.1), large volumes of air are brought into contact with materials that bind the $CO_2$. The $CO_2$ is then extracted and the process is repeated. Chemical DAC is a cyclic process in which the same batch of materials is used over and over

[7] Note that in this analogy, the second law bound on energy for CDR would correspond to energy needed for use and squeezing/cleaning of the sponge, not the energy used in its production.



to capture $CO_2$. DACCS (direct air capture with carbon storage) is a CDR method in which $CO_2$ from DAC is put into long-term storage, for example, in geological repositories.

**Figure 2:** Some of the main approaches that have been proposed for atmospheric CDR: direct air capture (DAC), enhanced rock weathering (ERW), ecosystem-based CDR, bioenergy with carbon capture and storage (BECCS), and ocean alkalinity enhancement (OAE).




- **Biological carbon capture:** Biological systems naturally capture $CO_2$ through photosynthesis, using solar energy. Ecosystem-based CDR (§4.2.1) harnesses this natural process to increase carbon stores in soil and plant material, decreasing atmospheric $CO_2$. Other biological approaches involve growing crops that capture $CO_2$ for long-term sequestration. In those approaches, the carbon is either sequestered directly or after extracting energy from the plant materials through, for example, biofuels or bioenergy with carbon capture and storage (BECCS, §4.2.2). Biological carbon capture processes such as BECCS that sequester carbon in the form of $CO_2$ are cyclic processes in the nomenclature of this report. As discussed above, other biological/ecosystem-based processes may be thought of as once-through processes with readily available materials but are subject to the same energy constraints as cyclic processes.

- **Enhanced rock weathering (ERW):** In enhanced weathering (§4.3), materials such as mined alkaline minerals are brought into contact with air containing $CO_2$, and the $CO_2$ binds with the materials. This process occurs naturally at geological rates. The idea of enhanced weathering is to artificially enhance the rate of this process. This is a once-through process because the material is not used again for $CO_2$ capture after binding with $CO_2$ to form relatively inert products.

- **Ocean-based CDR:** There are a number of approaches to ocean-based CDR (§4.4). The world's oceans contain much more $CO_2$ than the atmosphere, and in near-surface waters there is an approximate

equilibrium between dissolved inorganic carbon (DIC) and atmospheric $CO_2$ levels. By manipulating the chemistry of ocean surface water, for example, by ocean alkalinity enhancement (OAE), §4.4.1, the balance between different carbon-carrying DIC molecules is shifted so that more carbon is stored and atmospheric carbon is drawn into the surface waters. There are also ocean-based versions of chemical DAC (direct ocean capture or DOC) and accelerated mineralization where carbon is removed from ocean surface waters to indirectly reduce atmospheric carbon dioxide levels. DOC is a cyclic process for CDR, while OAE and ocean-based accelerated mineralization are once-through approaches.

# 3. Physical Limits on Capture of Atmospheric $CO_2$

## 3.1 Energy Constraints From Thermodynamics


**Summary: Energy Constraints on Cyclic CDR Systems**

The second law of thermodynamics gives a well-understood absolute lower bound on the energy cost of $CO_2$ capture for any cyclic CDR process:

Energy needed ≥ 120 kilowatt-hours per ton of $CO_2$

This lower bound describes the amount of energy needed regardless of its source. If a system uses thermal energy as its input, only a fraction of the input energy can be applied toward this bound. This absolute lower bound gives a useful metric for evaluating different approaches to carbon capture.

Most existing and proposed methods for chemical DAC require an energy input of at least three to ten times this lower bound, even when considering only the useful fraction of energy from thermal sources. Capturing $CO_2$ at the rate of one gigaton per year using energy at three to ten times the thermodynamic lower bound would require generating new carbon-free power at a level of 1-4% of 2021 global electric power generation.


The second law of thermodynamics imposes a fundamental lower limit on the energy needed by any cyclic direct air capture process. The mixing of carbon dioxide molecules with the dominant nitrogen and oxygen molecules in the atmosphere involves a high degree of disorder (entropy). Removing this disorder by sorting the molecules (i.e., separating out the $CO_2$) requires energy, because the entropy must be dumped back into the atmosphere through the transfer of thermal energy.

Quantitatively, the thermodynamic lower bound states that the energy cost of separating 1 ton of $CO_2$ from the atmosphere is at least 120 kilowatt-hours [8] (App. C). This means that the minimum energy cost of removing 1 Gt $CO_2$/yr would be equivalent to the full output of 14 nuclear power plants generating a continuous power output of 1 GWe each, [9] and comparable to the yearly energy used in mining bitcoin circa 2023 [14]. At a more realistic rate of energy use of three to ten times the thermodynamic minimum, the steady state power requirement for each gigaton of $CO_2$ per year of CDR would be 40-140 GWe. This is 1-4% of the current global rate of electric power generation. [10] This lower bound on energy requirements is very familiar to scientists and engineers working on carbon capture systems, but must be considered in any discussion of scaling and large-scale implementation of carbon capture systems.

[8] This assumes an ambient temperature of 300 K and $CO_2$ mixing at 420 ppm, with an output stream of pure $CO_2$. In standard physics and chemistry units, this energy cost is 120 kWh/t $CO_2$ ≅ 440 MJ/t $CO_2$ ≅ 20 kJ/mol $CO_2$.

[9] 4.4 × 1017 J ≅ 14,000 MWy, or ~ 14 GWe of average power. By 14 GWe, we mean 14 GW of electric power or equivalent. Power plants are often rated in thermal output, e.g., 1 GWt, but only a fraction of such thermal output represents 'useful energy' (exergy) that can be used toward the thermodynamic energy bound on CDR, as discussed in App. B. Elsewhere in the report we drop the appended e, but it is generally implied in discussing power needs for cyclic plants. Note that this does not include the energy needed to compress the $CO_2$ for storage.

[10] 3.2e TW in 2021 [15].

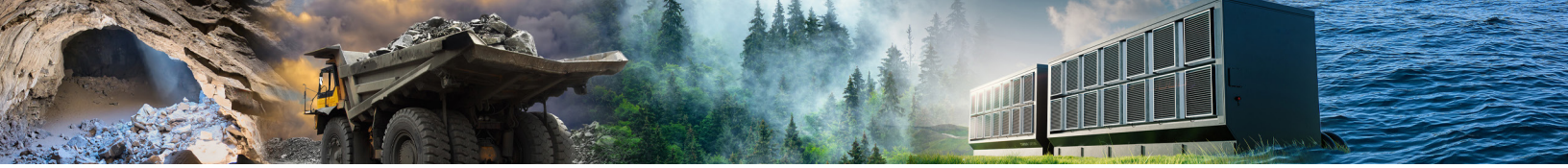
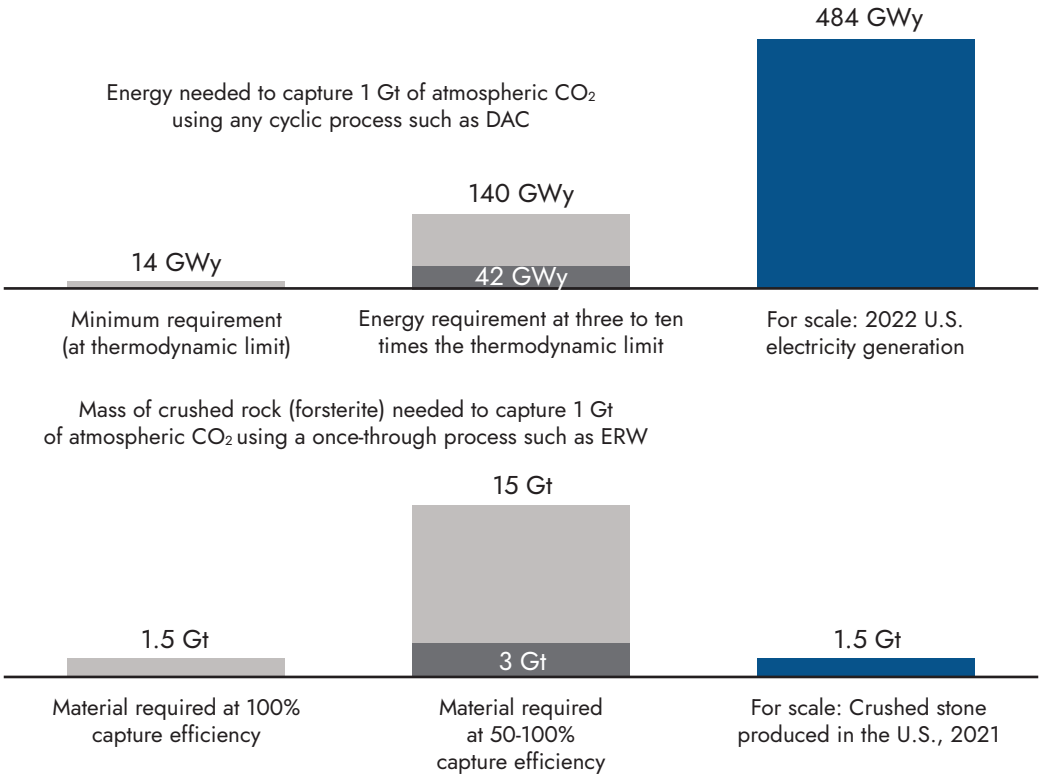


**Figure 3:** Rough estimates of the energy and materials needed to capture 1 Gt of atmospheric $CO_2$ using cyclic and once-through processes, respectively. Energy is given in gigawatt-years (GWy, see units definitions). Capturing multiple gigatons per year would require proportionally more energy and materials. Note that while the ratio of materials for the once-through process example is greater than the ratio of energies for the cyclic process example, the production of crushed rock represents a relatively small fraction of total energy used in the United States.




Existing or foreseeable direct air carbon capture systems will likely require at least several times as much energy as the thermodynamic lower bound. [11] As discussed in §2, the thermodynamic lower bound on energy requirements applies to any $CO_2$ capture process that uses a closed cycle of material inputs (aside from the $CO_2$-containing atmosphere, of course), such as most chemical-based DAC systems (§4.1). This bound also applies to biologically based CDR approaches, where the power needed is provided by solar energy through photosynthesis. For biological systems, the second law constraint thus translates into a requirement for large land areas for CDR at large scale (see further discussion in §3.2, App. E). Note also, however, that the minimum energy required by any cyclic CDR process is reduced if the output stream after capture is not pure $CO_2$ [12] (see discussion in App. C).

Many $CO_2$ capture approaches can directly use thermal energy (such as waste heat from industrial processes) for key processes. However, the second law imposes a further limit that restricts the fraction of thermal energy that can be used to overcome the energy constraint on $CO_2$ capture. As discussed in detail in App. B, the thermal energy required to provide a fixed amount of useful energy (exergy) increases rapidly as

[11] Due to the dilute nature of atmospheric $CO_2$, approaching the thermodynamic limit for cyclic capture processes is likely to be harder than for processes like thermal-to-electric energy conversion in power plants, which can have efficiencies of 75% or more of the second law theoretical bound.

[12] For example, if the output stream is only 5% $CO_2$, the energy required is reduced to roughly 62% of the energy needed to capture a pure $CO_2$ stream (although the volume of gas that must be sequestered grows by a factor of 20).

the temperature of the thermal resource drops toward the ambient temperature. This issue adds to the challenge of using the limited thermal energy available from waste heat and near-surface geothermal sources to power large-scale CDR deployment.

In addition to the energy needed to separate out $CO_2$ from the atmosphere, energy is needed for storage. In one primary sequestration paradigm (§5.1), the $CO_2$ is compressed to a supercritical fluid at a pressure of at least 72.8 atm. Compressing 1 ton of $CO_2$ from atmospheric pressure to 72.8 atm requires roughly 250 MJ (70 kWh) of energy. This brings the total minimum energy required for the capture and compression of 1 ton of $CO_2$ to about 700 MJ (190 kWh). [13]

As discussed in §2, the energy constraints described here do not apply directly to once-through processes such as enhanced weathering or OAE. The energy needs for those approaches, such as for grinding rock, are discussed separately (§4.3). Note that much of the energy used in mining and transporting raw materials, such as would be needed for extensive once-through CDR efforts, currently comes from fossil fuel sources such as diesel fuel. This may present challenges to fully decarbonizing in the near future. Also note that the thermodynamic constraint on energy use does hold for cyclic systems like electrochemical DOC systems that extract $CO_2$ from seawater (§4.4.2), because the carbon in surface waters is in equilibrium with the atmosphere (App. G).

## 3.2 Material and Volume Constraints for CDR Systems

**Summary: Material and Volume Requirements**

Capturing gigatons of carbon dioxide requires processing gigatons of material, in one way or another. Once-through CDR approaches (enhanced weathering, OAE) require mining, grinding, and distributing multiple tons of rock or other alkaline materials per ton of $CO_2$ captured. Cyclic DAC systems require moving large quantities of air. Biologically based systems such as BECCS require growing biomass over large areas (at the scale of a thousand square kilometers for each megaton of $CO_2$ captured, App. E), competing with agriculture for land use.

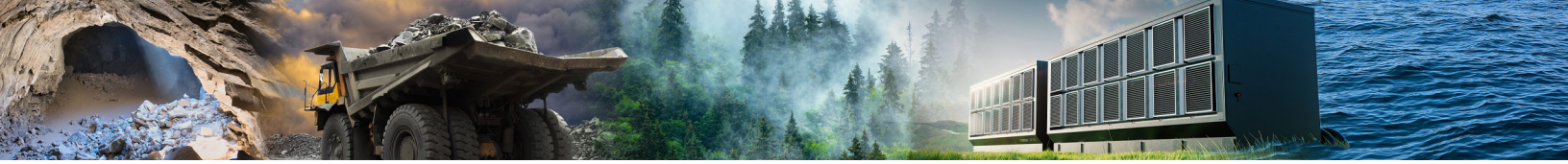
In this section we give some order of magnitude estimates for the mass and volume of materials that must be processed for some approaches to large-scale $CO_2$ capture. More precise statements are given in §4. First, note that extensive amounts of material are involved in the fossil fuel combustion that produces $CO_2$ emissions in the first place (1,100 pounds of coal, 18,000 cubic feet of natural gas, or 120 gallons of gasoline per ton of $CO_2$ emitted [16]). It is thus perhaps not surprising that the recapture of any substantial part of this $CO_2$ from the atmosphere requires similar levels of materials and infrastructure.

For once-through approaches such as enhanced weathering (§4.3) or ocean alkalinity enhancement (§4.4.1), material such as silicate rocks must be mined and ground in quantities commensurate with the quantity of $CO_2$ captured. Thus, to capture 1 Gt of $CO_2$, roughly 1 Gt or more of rock must be extracted, processed, and distributed. [14] By comparison, the quantity of iron ore mined worldwide (2015-2019 average) is more than 3 Gt/yr [17].

For carbon capture in systems such as chemical DAC plants (§4.1), large volumes of air must be circulated

[13] Note that the compression process is not particularly difficult technically and it should be possible to carry out with energy inputs reasonably close to the thermodynamic limit.

[14] While the molecular weight of alkaline minerals is generally several times larger than the molecular weight of $CO_2$, up to two molecules of $CO_2$ can be captured for each magnesium or calcium ion, so there is roughly a one-to-one correspondence between the mass of material used and the mass of $CO_2$ captured.



past the capture materials. For example, at a typical airflow rate of 2 m/s, a system with an intake height of 20 m would require a total horizontal intake length on the order of 1,000 km to capture 1 Gt $CO_2$/yr (see App. D for details). [15] To capture 1 Gt of $CO_2$, the amount of air that would need to be moved is comparable to the amount moved by all air conditioning systems in the world yearly. This is roughly the volume of air over a region the size of the state of Georgia.

Sequestering gigatons of $CO_2$ also requires large storage reservoirs. One gigaton of supercritical $CO_2$ has a volume of several cubic kilometers. [16]

Note that the carbon density (per unit volume) of seawater is higher than that of air by a factor of roughly 100 (App. G). Therefore, in ocean direct carbon capture systems (§4.4.2), the minimum volume and corresponding intake sizes are reduced by roughly a factor of 100 compared to air capture systems with a similar flow rate. [17] (However, removing a large fraction of the carbon may be more challenging for ocean DOC (DAC-like) approaches.)

Biological approaches to CDR require large land areas and large quantities of fresh water (in some cases naturally supplied) to produce large quantities of biomass. Capturing and removing 1 Gt $CO_2$/yr using photosynthesis-based processes would require sequestering the carbon in about 1 Gt of biomass yearly. For comparison, global cereal grain production (maize, wheat, rice, etc.) is about 3 Gt/yr. A more detailed analysis of energy and land needs for photosynthesis-based approaches to CDR is given in App. E.

## 3.3 Rate Constraints

The rate at which chemical reactions occur is not limited by any principle as fundamental as the energy and material constraints from thermodynamics and mass conservation. In general, reaction rates are controlled by a variety of factors, such as concentration, temperature, diffusion, surface area, turbulent mixing, and the presence of catalysts. For example, in many cases a chemical reaction rate can be increased by either increasing the temperature or lowering the energy barrier. [18] Nonetheless, it may not be easy to speed up many naturally slow reactions. This may be a limiting factor for some CDR processes. For example, the weathering of silicate rocks is limited by the diffusion of ions. This process only occurs on geologic timescales unless significant energy is spent on grinding the rock to small enough grains (typically on the order of 10 micrometers (10 μm), smaller than the thickness of a human hair). As discussed in §4.3, this grinding process can be the single largest energy cost for enhanced weathering schemes.

## 3.4 Carbon Budget

**Summary: Carbon Budget**

For CDR systems to effectively remove net carbon dioxide from the atmosphere, the energy inputs should be as carbon-free as possible.

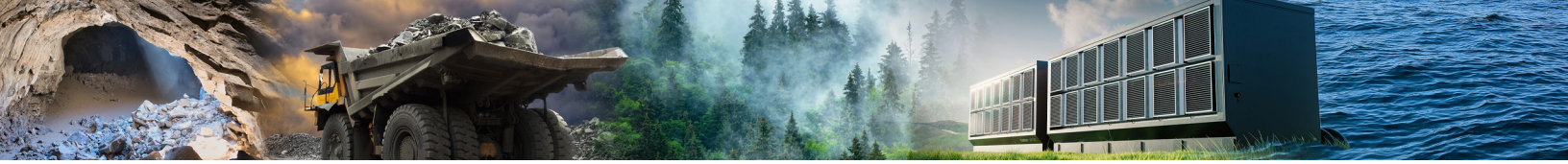
One important consideration for atmospheric $CO_2$ removal systems is the extent to which generating the energy to run them will emit additional carbon dioxide. Direct air capture systems should be powered by

[15] Higher flow rates are possible for some systems, and would reduce the intake area, but require more energy.
[16] At the critical point, T = 304.13 K and pressure 72.8 atm, 1 gigaton of $CO_2$ has a volume of 2.14 $km^3$.
[17] 2 m/s is about the average flow rate of the Gulf Stream.
[18] This follows from the Arrhenius equation for a reaction rate: $k \propto \exp(-E_b/k_BT)$, where Eb is the energy barrier and T is the temperature ($k_B$ is the Boltzmann constant defined in App. B).

renewable or low-carbon energy sources to be effective, due to the substantial overhead for carbon intensive energy sources.

As a simple and extreme example, consider a coal plant (burning subbituminous coal or lignite [16]) that operates at 33% efficiency. This plant will emit roughly 1 kg of $CO_2$ for every 3.3 MJ (0.9 kWh) of electrical energy that it produces. Imagine that this example coal plant powers a $CO_2$ capture and sequestration system that uses three to five times the thermodynamic minimum of capture energy, and that it compresses the $CO_2$ to a supercritical fluid. The power plant would need to produce 3 GJ or 9.5 GJ, respectively, for every net decrease of 1 ton in atmospheric $CO_2$. A carbon-free energy source would need to produce only 1.6 GJ or 2.45 GJ, respectively, for the 3× and 5× cases. Note that the increase in energy needs is highly nonlinear in the efficiency factor. [19] For energy sources with lower carbon intensity, and efficient direct air capture systems, the overhead (extra energy needed) is less; for example, for a combined cycle natural gas plant operating at 60% efficiency, the power plant would need to produce 1.85 GJ for the 3× DAC plant, with an overhead of about 15%, and 3.1 GJ for the 5× plant, with an overhead of about 27%.

[19] In general, if the energy source of a $CO_2$ capture system produces additional atmospheric $CO_2$ that represents a fraction $x$ of the $CO_2$ captured by the system it is powering, then the net energy needed to capture a fixed net quantity of $CO_2$ is increased by a factor of $1/(1 - x)$. In this example, for the 3× case, the energy requirement to capture and sequester 1 kg of atmospheric $CO_2$ would be roughly 1.6 MJ. (This comes from multiplying the capture energy 0.44 MJ by 3 and adding the compression energy, 0.25 MJ.) The emission fraction would be $x \cong 1.6/3.3 \cong 0.48$, giving $1/(1 - x) \cong 1.9$, and leading to the result quoted above. A similar computation gives the 9.5 GJ result for the 5× case. Note that this calculation is a very rough approximation, intended only to illustrate the rapid growth in energy need as $x$ increases toward 1.



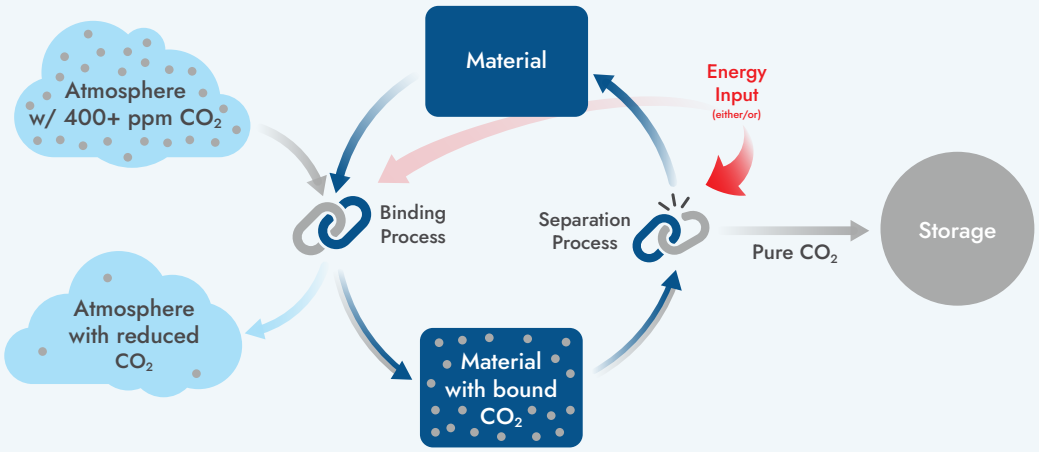
# 4. Approaches to CDR

This section gives an overview of many of the principal approaches to CDR that have been suggested. While we do not cover every possible technology, most of the approaches that we encountered during this study fit reasonably well into these frameworks.

## 4.1 Chemical Direct Air Capture (DAC) Systems

### Summary: Chemical DAC systems

Chemical DAC systems are cyclic systems in which $CO_2$ is brought in contact with a working material that easily binds the $CO_2$ molecules. The $CO_2$ is then dissociated from the working material in a separate process by using a change of temperature, pressure, humidity, or other conditions, or by using electrochemistry. Then the same material is reused for additional cycles. The output $CO_2$ is typically a relatively pure gas that can be sequestered or used for other purposes.




Process type: Cyclic

Inputs: Air (with $CO_2$ concentration 0.04%), energy (more than 120 kWh/t $CO_2$), water (for some processes), and, as needed, replacement materials.

Output: Relatively pure $CO_2$ stream.

Advantages: Simple output, less land area is impacted than in many other CDR systems, manageable measurement, reporting, and verification (MRV).

Potential drawbacks: Energy requirements.

Uncertainties and research opportunities: Energy optimization, material optimization, water and other material requirements, scaling.

Chemical DAC systems remove $CO_2$ from the air using a working material, such as a solid sorbent or a liquid solvent, that combines with $CO_2$ at the molecular level in a process called sorption. When the sorbent/solvent is sufficiently saturated with $CO_2$, the material is isolated from the air and the $CO_2$ is extracted. This regenerates the working material for use in another extraction cycle. The $CO_2$ is then sequestered (DACCS) or, perhaps, utilized for an industrial application (DACCU). The last decade has seen an explosion of research [18, 19, 20, 21, 22] on DAC science and engineering, and there has been a signifi-

cant increase in the number of companies operating in the DAC field. Detailed introductions to aspects of DAC systems can be found in [5, 6].

Removing multiple gigatons per year of $CO_2$ from the atmosphere using chemical DAC systems would be a major infrastructure project and have significant challenges. Among them are energy demands, water usage, sorbent and other material consumption, land use (land is needed both for removing $CO_2$ and for new energy sources), capital and operating costs, and environmental impacts.

The largest energy demand in DAC systems is for extracting $CO_2$ from the sorbent or solvent. The thermodynamic minimum energy for separating 1 Gt $CO_2$/yr (see §3.1) requires roughly 14 GW average power. Pumps, compressing $CO_2$ for storage, and fans for moving air through the facility require additional power. Energy estimates for existing and larger-scale planned facilities are in the range of three to ten times the thermodynamic limit on exergy (§3.1) needs. [20]

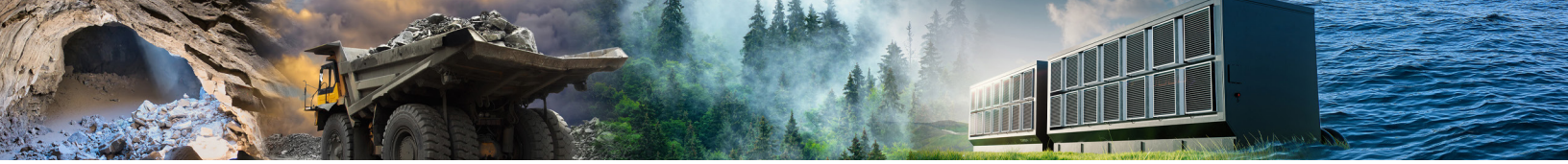
One important question in DAC is to what extent the chemical sorption and desorption processes can approach the thermodynamic limit in large-scale, deployable systems. The energy needed to dissociate $CO_2$, for any given process, has a lower bound given by the Gibbs free energy of the relevant chemical reaction. This is generally at least three times the thermodynamic bound of 20 kJ/mol for current systems. Further research may lead to more effective materials and processes that lower the energy requirement, such as in battery technology. There, a system of chemical reactions can be performed with an efficiency reasonably close to the theoretical limit (lithium-ion batteries have an energy efficiency of more than 90%). However, the highly diffuse nature of atmospheric $CO_2$ may make it much harder to come close to the thermodynamic limit for carbon capture systems.

The removal of $CO_2$ at the gigatons per year level requires large facilities. A moderate continuous air flow of about 2 m/s corresponds to an intake area of roughly 20 m × 1,000 km for 1 Gt $CO_2$/yr, even assuming 100% carbon removal (see §3.2). Using higher flow rates would reduce the plant size (for the same $CO_2$ removal rate) but the fans would require more energy.

No single solution to these complex challenges has emerged as the preferred chemical DAC technology choice. The two main approaches use a solid sorbent or liquid solvent as the working material and heat input to separate $CO_2$. More recent concepts that use electrical energy instead of heat offer potential improvements. In particular, electrochemical systems could be powered by solar photovoltaics (PV). This may be economically favorable as PV prices continue to drop. Further details on these approaches to chemical DAC, with further references, are given in App. J.

Chemical DAC has some potentially attractive features. Facility locations can be selected that optimize the availability of waste heat or renewable energy sources, water requirements, proximity to $CO_2$ sequestration sites, operating environments (such as temperature and humidity), and geopolitical considerations, among other factors. Measurement, reporting, and verification (MRV, §6.1) should be more straightforward than in most other large-scale CDR systems that have been proposed, because chemical DAC facilities would produce easy-to-measure outputs of concentrated $CO_2$. While chemical DAC plants would be large, they would impact significantly less land area than most terrestrial once-through or biological CDR systems with comparable capacity. This is the case even for DAC systems powered by dedicated renewable energy systems, such as a field of solar thermal or PV collectors (§6.5). The land area need would be even smaller for a DAC plant powered by a nuclear plant (with attendant concerns) or by a fossil-fuel power plant that itself would produce additional $CO_2$ unless equipped with CCS technology (§3.4).

[20] Solvent/causticization approaches are around ten times the thermodynamic limit (10× TL) ([23] Table 1); sorbents are lower, but at least 3.5-5× TL ([24] Table 2); the moisture swing process of [25] has a theoretical minimum energy need of at least 2.5 × TL, just for the desorption reaction, not including water needs or other energy requirements.



## 4.2 Biological Carbon Capture

Biological systems across the planet capture atmospheric carbon dioxide every day through photosynthesis. Using solar energy, they transform $CO_2$ and water ($H_2O$) into hydrocarbons such as sugars (e.g., $C_6H_{12}O_6$) and other molecules used in plant metabolism and structure. Most of the captured carbon is eventually released back into the atmosphere as $CO_2$ or methane, either when the energy is used in metabolic activity or the biological material decays and breaks down. The balance between these carbon capture and release processes can be seen in the seasonal variation of atmospheric $CO_2$ in Fig. 1, dominated by the annual cycle of northern hemisphere vegetation.

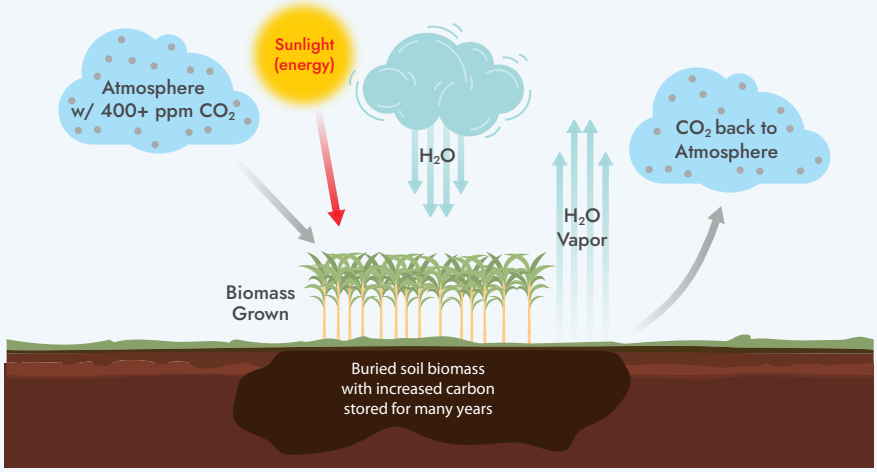
Biological systems provide opportunities to decrease atmospheric carbon levels over longer time scales through this natural carbon capture process. One approach is through ecosystem restoration and other changes in land use that promote increased carbon storage in soil or terrestrial biomass (§4.2.1). We refer to these approaches collectively as 'ecosystem-based' CDR methods. Another approach to biological CDR is using natural or engineered biological systems to capture carbon and then sequestering it as a liquid or solid in perpetuity. In BECCS (bioenergy with carbon capture and storage, §4.2.2) the chemical energy in biomass containing carbon is released and used, and the resulting $CO_2$ is sequestered. Alternatively, biomass can be sequestered directly or used in long-lasting built systems, such as in construction materials (§4.2.3). We address these different types of systems separately.

There are ancillary benefits to many ecosystem-based CDR approaches, and they likely represent the most economical immediate approach to CDR. Indeed, CDR through land use changes and BECCS are the dominant forms of CDR incorporated in, for example, scenarios explored by the IPCC [12, 1] for the remainder of the twenty-first century. However, ecosystem-based CDR has limited capacity and is difficult to guarantee over longer time scales. While many of the technical components of BECCS systems are reasonably well-developed, deployment of BECCS is so far still quite limited. Energy constraints and associated land area issues may limit the potential contribution of biological CDR in the coming decades.

## 4.2.1 Ecosystem-Based Biological CDR

### Summary: Ecosystem-Based Biological CDR

Ecosystem-based biological carbon dioxide removal generally involves modifying land use and management practices to promote increased carbon storage in soil or biomass.




Process type: Once-through but subject to second law energy bounds; can also be thought of as cyclic with extended carbon retention time.

Inputs: Air and water.

Output: Air and water.

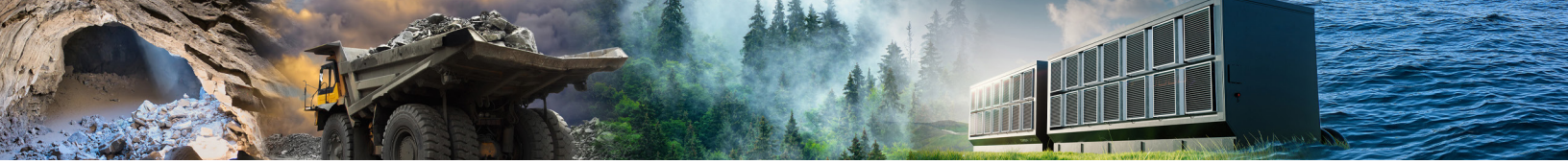
Advantages: Often associated with increased ecosystem health, biodiversity, and improved ecosystem services such as clean water and air; low cost and minimal material and infrastructure investment.

Potential drawbacks: Approaches somewhat site-specific, scale of CDR is limited in any given location, challenging to ensure durability/permanence of storage.

Uncertainties and research opportunities: MRV is difficult, in particular difficult to verify additionality and assess leakage rates; changing climate conditions may reduce carbon storage potential in some systems.

Forests, grasslands, and other terrestrial ecosystems store several times as much carbon in their soil and biomass as is present in the atmosphere (App. H). This carbon stock has been depleted over centuries by human patterns of land use, such as deforestation and agricultural practices. Terrestrial ecosystem-based approaches to CDR (a.k.a. natural climate solutions) involve modifying land use in ways that increase the amount of carbon stored in soil and biomass. (Similar approaches for oceanic systems are described in §4.4.3.) CDR approaches through land stewardship include ecosystem restoration and improved land management practices in forests, wetlands, grasslands, and agricultural lands.

In most accountings of current CDR efforts (e.g., [10]), ecosystem-based CDR removes more atmospheric $CO_2$ than any other method. An overview of ecosystem-based CDR [26] suggests that these approaches could, in principle, reduce atmospheric $CO_2$ by something like 3-10 Gt/yr at relatively low cost over the coming decades, without compromising other human needs such as food and fiber security. However, an impact at this scale would involve modifying land management practices over very large areas globally. Furthermore, the rate of CDR could not be sustained indefinitely in any given place, because soil and terrestrial biomass have limited potential for carbon storage.



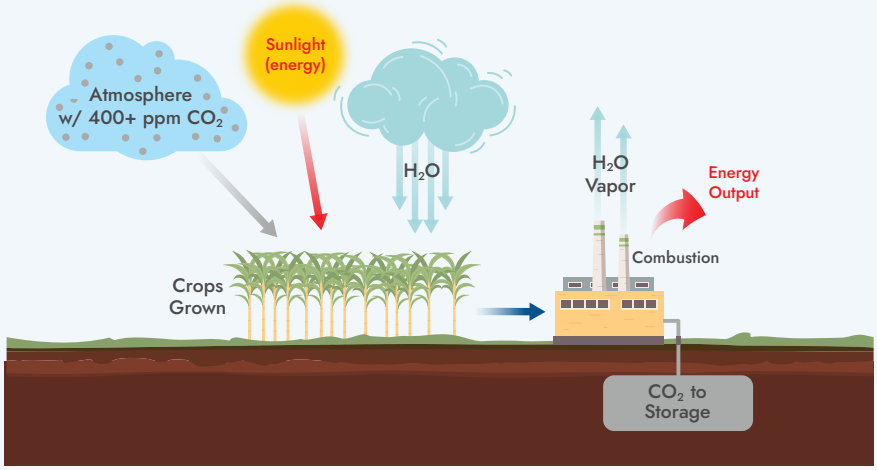
As discussed in §2.1, ecosystem-based CDR approaches can be thought of in some sense as once-through processes where water is the working material, although the process is driven by solar energy and must respect the second-law energy bound. If the processes in which the water is again separated from the carbon are included, ecosystem-based CDR can also be thought of as a cyclic approach where the carbon is retained for some time. The residence time of carbon in soil and biomass can range from 10-1,000 years (App. H); for ecosystem-based approaches requiring active management, even to maintain a given level of storage, once achieved, would require continued activity. Additional details on the potential of these approaches are given in App. I.

Ecosystem-based CDR approaches often have other benefits such as preserving biodiversity, increasing ecosystem health, and promoting ecosystem services such as fresh water. However, ecosystems are complex, methods must be tailored to individual locations, and it can be difficult to predict and evaluate the results of specific changes in land use (§6.1). In particular, it can be difficult to determine the additionality of carbon gains due to specific activities and to measure rates of leakage over time. Furthermore, ecosystem-based CDR has limited potential in any given land area and is only effective when the added carbon storage is maintained over long times; this requires long-term intentional land use practices, and durability of storage can be compromised by many difficult-to-control factors including fire, insects, and changes in climate. On a related note, avoiding deforestation and the degradation of existing ecosystems will help reduce net anthropogenic $CO_2$ emissions.

### 4.2.2 Bioenergy With Carbon Capture and Storage (BECCS)

**Summary: BECCS**

BECCS involves growing crops or trees over large land areas, using that biomass to produce energy though direct combustion or via fermentation to biofuel, and then capturing the released $CO_2$ for sequestration.




Process type: Cyclic

Inputs: Air, water, likely nitrogen-based fertilizer

Output: Relatively pure $CO_2$ stream, energy

Advantages: Simple output, negative-carbon energy

Potential drawbacks: Land use competes with agriculture and natural ecosystems, loss of biodiversity, energy inputs for fertilizer

Uncertainties and research opportunities: Scaling

Bioenergy with carbon capture and storage (BECCS) generally refers to growing crops or trees that capture biomass, using that biomass in power plants with point-source carbon dioxide capture, and sequestering the $CO_2$ (through, e.g., direct geologic storage or integration into long-lived products that are used or sequestered, as in §5). In the carbon management scenarios considered by the IPCC [1], the largest contribution to atmospheric $CO_2$ removal was assumed to come from BECCS. The NASEM report [4] states that BECCS "has come to be viewed as a key carbon dioxide removal approach." BECCS is often viewed as the least expensive technological approach to large-scale CDR. However, as emphasized in NASEM [4], BECCS is not yet widely deployed, and there are a number of obstacles to implementing this approach at large scales.

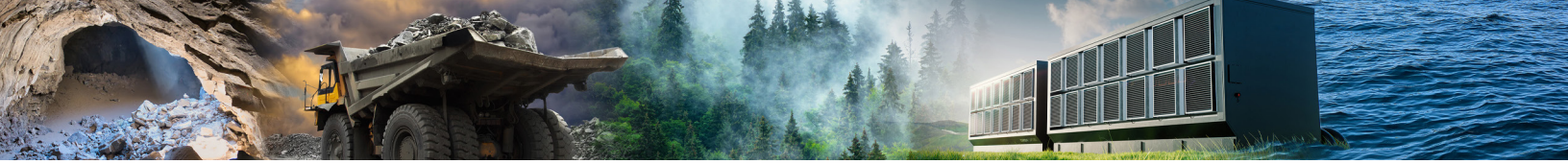
BECCS is essentially a cyclic process, as discussed in §2.1, when the energy capture process through photosynthesis is included. The primary molecule that binds with atmospheric $CO_2$ is water ($H_2O$). Energy is input (through photosynthesis) to bind carbon into a sugar/carbohydrate molecule. The energy is released through combustion, and the water and carbon dioxide are separated in a controlled environment where the $CO_2$ can be captured and sequestered.

Primary approaches to BECCS involve either direct combustion or fermentation (as in ethanol production), followed by combustion of the biomass. The outputs of these processes contain much higher concentrations of $CO_2$ than the atmosphere. The thermodynamic lower bound on capture energy at this stage of the process is therefore significantly lower than the amount needed for atmospheric capture. [21] There are currently no existing power plants based on the combustion of purpose-grown biomass that perform carbon capture and storage (CCS) [4]. However, CCS is incorporated in several ethanol plants to capture the $CO_2$ output in the fermentation process, for which the practical technology and frameworks are more commercially ready [27]. CCS is also used at several municipal solid waste plants around the world.

Substantial land area in the United States and other countries is used, at least in part, for biofuel (e.g., ethanol) production, but point-source capture is much harder to implement (and may be intractable) when use is widely distributed, such as in automobiles. Thus, achieving BECCS at the gigatons of $CO_2$ per year scale would involve technical innovation and revamping existing energy infrastructure (e.g., replacing direct biofuel use in transportation with combustion in power plants and electric vehicle use). Another approach to BECCS involves biomass conversion to fuels with the co-production of biochar (§4.2.3).

Any BECCS scenario using conventional crops would need large amounts of land to capture $CO_2$ at the gigatons per year scale, as well as potentially substantial energy inputs for fertilization. Large-scale BECCS also requires extensive freshwater inputs [28], which could be challenging in areas where water is already limited. Carbon capture and storage at this scale using plants with a photosynthesis efficiency near that of current agricultural crops would require land on the scale of 1 million $km^2$, representing about 20% of U.S. farmland; it has been suggested that 'second-generation' biomass such as perennial grasses could reduce the needed area somewhat (see §3.2 and App. E for more details). By using waste and residues from existing agricultural processes, BECCS could be realized in principle by using some fraction of the photosynthetic productivity on a given land area without displacing other agricultural land uses. Other approaches, such as growing species of algae that are more efficient at photosynthesis, may eventually reduce the amount of land required for BECCS and related CDR methods. However, in any scenario using conventional terrestrial crops, large-scale BECCS would compete with other land uses such as growing food and ecosystem conservation.

[21] The energy for atmospheric capture has already been supplied from solar energy through the photosynthesis process.



### 4.2.3 Other Biological Approaches to CDR

**Direct biocapture and storage.** One strategy that has been suggested in a variety of contexts is to simply capture carbon in biomass and then sequester the biomass, often after some processing that increases its carbon density and/or improves its potential for long-term storage. This is sometimes referred to as BiCRS (biomass with carbon removal and storage). Like BECCS, any such approach needs large land areas for biomass crops (App. E), competing with agriculture and reducing biodiversity. Energy-intensive fertilizers may also be required. One approach that has been considered is large-scale cultivation of high-productivity crops such as switchgrass, with stable sequestration in biolandfills [29]. Another approach that has been suggested is planting large areas with specific tree species followed by regular harvesting and processing of the wood (e.g., through gasification) for long-term sequestration [8]. While such monocrop tree plantations may remove more carbon dioxide over the long term, they come at the cost of displacing more diverse and healthy ecosystems.

Using wood and other carbon-containing natural materials in long-lasting building materials, such as for houses, can sequester $CO_2$ potentially for centuries. There is also some research on using biological approaches to directly produce carbonates for storage. This research is still at an early stage, but it may be an effective way to sequester carbon.

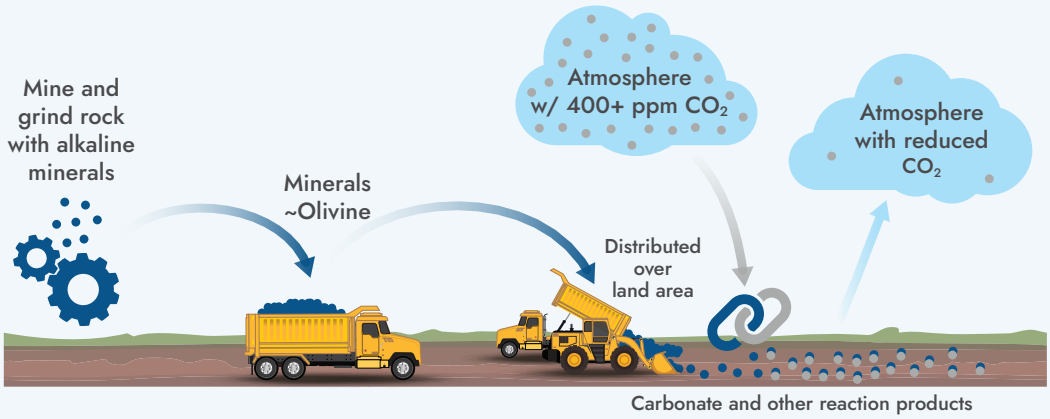
**Biochar and bio-oil.** Biochar is a solid material with a high carbon content. It is produced by thermochemically processing biomass in an oxygen-limited environment. It has been suggested that adding biochar to soil can positively impact crops and vegetation, particularly in tropical areas, and may increase long-term soil carbon retention. The extent of these benefits is uncertain, however. Some energy is released when biochar is produced, so biochar production can be incorporated into BECCS-type systems. MRV is difficult with biochar as with other approaches to increasing soil carbon; more research is needed to understand the impact and long-term benefits of biochar, as well as its stability and longevity in soil. This is complicated by the fact that different processes can produce different forms of biochar with widely varying properties. Further comments and references regarding biochar can be found in App. I.

Bio-oil is a liquid that is also produced from biomass through pyrolysis. Bio-oil can be used in biofuel, and there are currently efforts to directly sequester bio-oil as a form of CDR.

## 4.3 Enhanced Rock Weathering

### Summary: Enhanced Rock Weathering

The idea of enhanced rock weathering (ERW) is to capture atmospheric $CO_2$ using minerals that are out of equilibrium with the atmosphere. This can be done by mining, crushing, and grinding rock containing alkaline minerals and spreading them over large areas. There, they react with $CO_2$ to form relatively stable carbonates or carbonate ions in solution. In some cases, the process may lead to increased agricultural yields, but there are many environmental concerns.




Process type: Once-through

Inputs: Ground rock containing alkaline minerals

Output: Carbonate rock and/or carbonate ions and other products, e.g., silicon dioxide

Advantages: Total energy requirements (including for mining and processing) likely less than for cyclic systems

Potential drawbacks: Requires large amount of mining and processing

Uncertainties and research opportunities: Rate and effectiveness unclear; measurement, reporting, and verification is difficult; environmental consequences of distributed reaction products, some that may include heavy metals

When exposed to the atmosphere, rocks are weathered by wind, rain, and other physical, chemical, and biological mechanisms. Some minerals react exothermically with atmospheric and soil $CO_2$, the latter of which is derived from root and microbial respiration, producing carbonate ions or solids. Over geologic time scales, some of these carbonates accumulate in deep ocean sediments and lock away atmospheric $CO_2$. Enhanced rock weathering (ERW, also known as 'accelerated weathering' or 'accelerated mineralization') attempts to speed up these natural weathering processes in order to capture significant quantities of atmospheric $CO_2$ over time scales of a few years.

More generally, a variety of approaches have been proposed that use alkaline minerals to remove carbon dioxide. Campbell et al. [30] classify the approaches into three groups: surficial (alkaline minerals at the surface of the Earth react with atmospheric $CO_2$ or fluids containing $CO_2$), in situ ($CO_2$ is injected into underground rock formations), and ex situ (alkaline minerals are brought to sites of highly concentrated carbon dioxide production). Here we focus on surficial processes. The other two types of processes are discussed in §5.2.

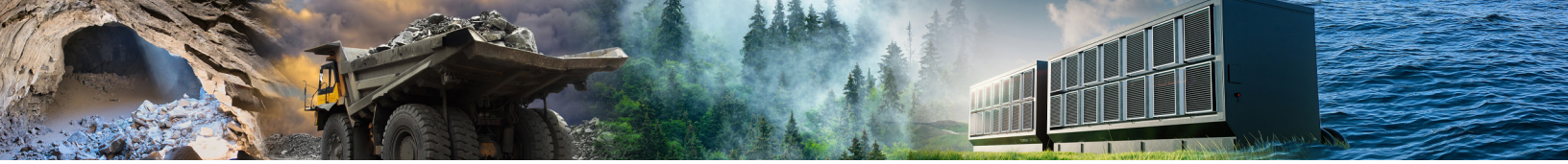


**Figure 4:** Peridotite in the Trinity Alps of northern California, showing orangish weathered surfaces and freshly exposed dark surfaces. (Photo credit: Brad Marston.)

Natural weathering has played an important role in determining Earth's current climate and $CO_2$ levels. [22] The present-day net global $CO_2$ absorption rate by natural rock weathering has been estimated [32] to be roughly 1.1 Gt $CO_2$/yr but with substantial uncertainty. Much lower estimates have also been reported [33, 34]. The mineralization of $CO_2$ is concentrated in hot spots around the globe where ultramafic rocks are exposed, especially in moist tropical regions and in some mine tailings. Ultramafic rocks are silicate rocks with relatively low (< 45%) silica content that are rich in magnesium and iron. A detailed study of hot spots may shed light on how to best engineer enhanced rock weathering.

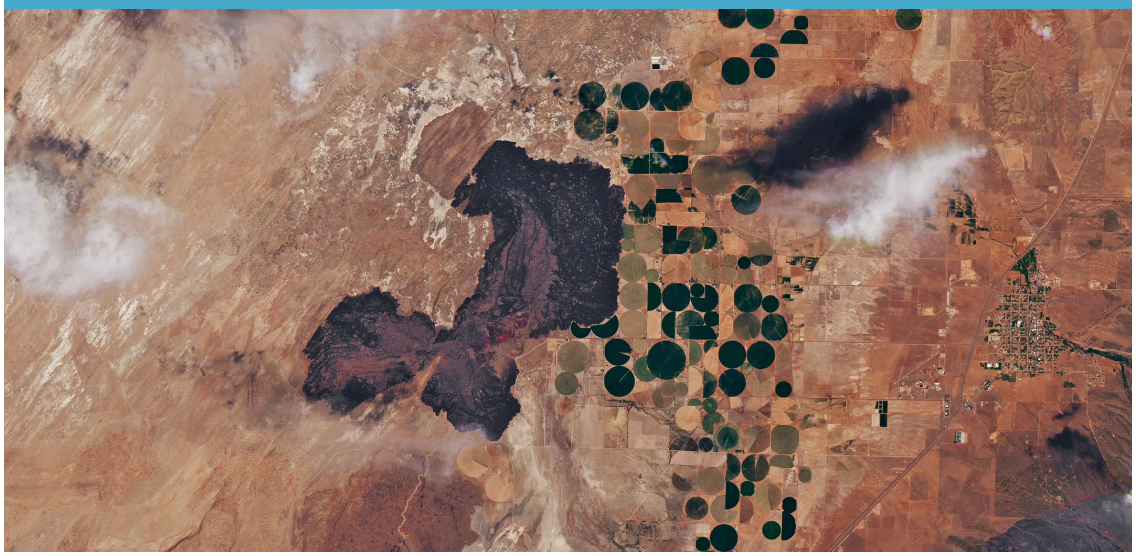
A number of specific rocks and minerals have been proposed for enhanced weathering processes. Peridotite, an ultramafic rock that makes up much of Earth's upper mantle, consists mostly of the minerals olivine and pyroxene (Fig. 4). Forsterite ($Mg_2SiO_4$), one of the olivine group of minerals, reacts energetically with carbon dioxide to form magnesium carbonate and silicon dioxide [35]:

$$Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2 + 90\ \text{kJ/mol} \qquad (1)$$

(See App. F for the basic chemistry of such reactions.) Serpentine rocks are composed of hydrothermally altered ultramafic minerals and react less energetically, releasing about 64 kJ/mol. Mafic rocks such as basalt react less energetically still but are more abundant, forming much of the ocean floor and flood basalts (Fig. 5) such as those found in eastern Washington state. [23]

[22] For example, geologists have proposed that the uplift of the Tibetan and Colorado plateaus exposed enough fresh rock to weathering that the resulting drop in carbon dioxide levels played an important role in the development of the ice ages in Earth's climate [31].
[23] Though silicate rocks react exothermically with carbon dioxide, the chemical reaction rate with $CO_2$ dissolved in water is limited by the breaking of ionic bonds between the cations ($Mg^{2+}$ or $Ca^{2+}$) and silicate groups [37], as well as the formation of clay passivation layers around the mineral grains. Olivine dissolves faster than crystalline basalt over a wide temperature range, but glassy basalt reacts even more quickly than olivine showing that free energy differences are not the only, or even most important, factor [4].

**Figure 5:** View from space of the 45 km² Ice Springs basalt lava flow in Utah. This is part of the Cinders volcanic complex, which has a total volume of $5.4 \times 10^8$ m³. Assuming the basalt has a density of 2.9 g/cm³, the flow has a mass of about 1.5 Gt. At a conversion rate of 0.3 t $CO_2$/ton basalt [36], this source could remove about 0.5 Gt $CO_2$. (Photo credit: NASA Earth Observatory.)



The concentration of $CO_2$ in soils can be much greater than that of the atmosphere, enhancing mineralization. Wet locations are best for accelerating weathering, because carbon in the form of carbonic acid reacts with silicate minerals. To maximize reaction rates, the rock should be ground to a fine powder [38] through a process called comminution. The ideal grain size is on the order of 10 micrometers — about one-fifth the width of an average human hair. The energy required for comminution down to that size exceeds the energy required for mining and transporting the rock [36, 39, 40].

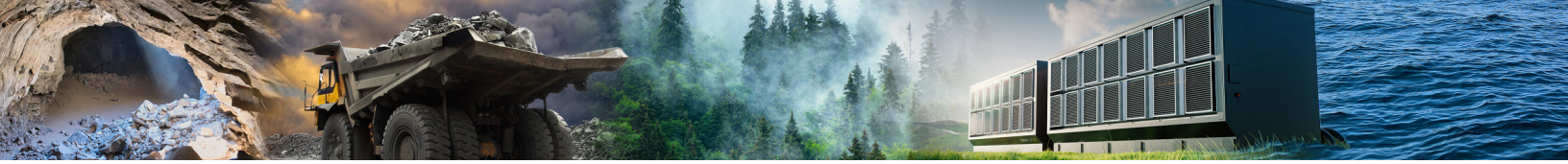
Technology for mechanical comminution has a long history, because it is the first step in processing ore [41]. The estimated energy efficiencies of mechanical comminution starting at the level of covalent molecular bonds [42, 43, 44] are on the order of 1%, thus there is potential for improvements in efficiency [45]. [24] Comminution can also be carried out with electromagnetic mills, electric pulses, and microwaves. It is unclear if these technologies are more efficient than standard mechanical methods [47].

Transporting large amounts of material from quarries and mines to farmland or coasts (for ocean alkalinity enhancement) might appear to be a major problem, but the technology and some of the infrastructure already exist. As mentioned, the energy cost of transporting the material is small compared to fine comminution [46], although it will require effort and time to fully decarbonize the transportation system. Accelerated weathering of silicate rock to form solid carbonates sequesters up to 1 $CO_2$ molecule per $Mg^{2+}$ or $Ca^{2+}$ ion per reaction, as in Eq. 1. [25] Thus, 1.5 Gt of forsterite could capture up to 1 Gt $CO_2$. The United States already mines and transports quantities of crushed rock at a scale comparable to that required for substantial enhanced weathering. About 142 Mt of coal was mined in the United States during 2021 [48] and 1.5 Gt of crushed stone was produced. [26]

[24] The trade-off between higher reaction rates and greater energy consumption is complicated but explored in [46, 39] where it is noted that surface areas of 1 – 10 m²/gm (corresponding to grain sizes on the order of 10 μm) can currently be achieved with an energy input of 10-100 kWh per ton of rock.

[25] The molecular weight of forsterite is 140 g/mol compared to 12 g/mol for carbon; because a mole of forsterite contains 2 moles of magnesium ions, roughly 5 Mt of forsterite would need to be mined to compensate for the added $CO_2$ from burning 1 Mt of coal under the assumption that the forsterite completely weathers.

[26] Of this crushed stone, 70% was limestone and dolomite and 15%, granite rock [49].



If the energy needed for comminution can be kept significantly below 100 kWh/t of rock, and weathering and carbon retention rates are high, the energy cost of this CDR approach may be lower than the thermodynamic bound on the energy input for cyclic processes. Therefore, the total energy requirements for ERW may be significantly lower than that of cyclic approaches requiring an energy of three to ten times the thermodynamic bound.

Ground rock can fertilize soils, which is potentially an attractive co-benefit of this approach as long as contamination with heavy metals can be avoided [50, 40]. Silicate rock can move soil pH levels into desirable ranges and potentially replace the lime additives often used for this purpose. However, some ultramafic rocks should not be used for this, because they contain nickel and chromium. Rock dust can be harmful to the lungs; serpentine rock in particular often contains asbestos. Additionally, magnesium from ultramafic rocks can interfere with plant uptake of calcium.

Determining the effectiveness of ERW is a complex problem. Soils are diverse and complex. Field experiments designed to measure the uptake of carbon dioxide by soils treated with crushed rock have found greatly varying results [51, 52, 53]. One recent experimental study, for instance, finds limited removal of carbon dioxide by soils in the United Kingdom treated with ground basalt for reasons that were not clear [54], but another study in the U.S. Corn Belt shows more promising results [55]. A 2024 meta-analysis of ERW field studies illuminates uncertainties in quantifying CDR and the wide range of measured responses [56]. Note that a rate of absorption of $CO_2$ by treated soils on the high end of 10 t/ha/year translates to one million square kilometers of land that would be required to remove 1 Gt $CO_2$ per year. Before the viability of ERW can be determined, a better understanding of how different soils respond to treatment with silicate rock is needed, including an understanding of the fraction of silicate minerals that react and the rate of the reactions. Possible changes in the amount of light reflected by the surface (its albedo) and increased emissions of nitrous oxide (a potent greenhouse gas) from treated soils [57] should also be considered, as well as the fraction of carbon dioxide that escapes from streams and rivers as dissolved inorganic carbon (DIC) makes its way to the sea [58].

## 4.4 Ocean CDR

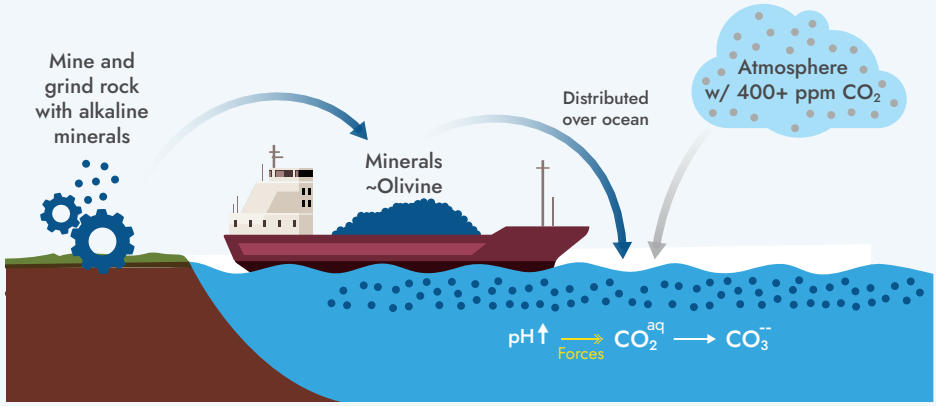
The world's oceans contain roughly 40 times as much carbon as the atmosphere, and have a carbon density per unit volume roughly 100 times that of air. Most of the dissolved inorganic carbon (DIC) in ocean surface waters is in the form of bicarbonate (~ 90%, $HCO_3^-$) and carbonate (~ 9%, $CO_3^{2-}$). Only about 1% is in the form of dissolved $CO_2$ ($CO_2^{aq}$ +$H_2CO_3$). On rapid timescales, these forms of carbon equilibrate in surface water, in ratios that depend on the pH or alkalinity of the water, temperature, pressure, and other factors. Related aspects of ocean chemistry are described briefly in App. G. While carbon levels equilibrate quickly between the atmosphere and ocean surface waters, it takes thousands of years for surface waters and the carbon they contain to mix and reach equilibrium with the deep ocean.

Because the ocean contains such vast quantities of carbon, it is natural to consider ways that this carbon store can be used to capture and hold additional carbon from the atmosphere. A wide range of approaches have been proposed, including modifying ocean chemistry to move atmospheric $CO_2$ into the form of carbonate ions dissolved in surface waters (ocean alkalinity enhancement) and electrochemical approaches. We briefly describe some of the main ideas here. More comprehensive reports on a broad range of approaches to ocean CDR (also known as marine CDR or mCDR) can be found in [ 30, 59, 60].

### 4.4.1 Ocean Alkalinity Enhancement (OAE)

**Summary: OAE**

The idea of ocean alkalinity enhancement (OAE) is to use alkaline minerals or other materials to change the chemistry of ocean surface waters so that they uptake and store atmospheric $CO_2$ in the form of carbonate ions. In time, this captured carbon is brought into the deep ocean by global ocean circulation.




Process type: Once-through

Inputs: Ground rock containing alkaline minerals

Output: Dissolved alkaline material, increased carbonate in ocean surface waters

Advantages: Total energy requirements likely less than for cyclic systems, reduction in ocean acidity may positively impact some ecosystems

Potential drawbacks: Requires a large amount of mining and processing

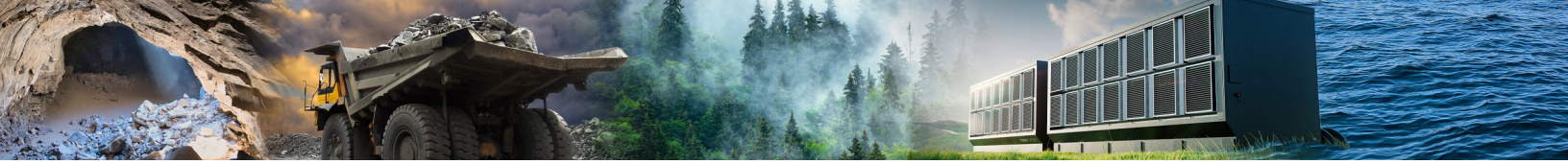
Uncertainties and research opportunities: Rate and effectiveness unclear, measurement and verification difficult, environmental consequences of adding large quantities of alkaline materials and trace elements to oceans are not understood

Atmospheric carbon can be removed by increasing the alkalinity of ocean waters. [27] With an increase in alkalinity, the DIC composition shifts. The fraction of dissolved $CO_2$ decreases in favor of carbonate ions. As the water's concentration of dissolved $CO_2$ is rapidly brought back into equilibrium with the atmosphere through mixing by, e.g., wave action, the water pulls in more atmospheric $CO_2$. Using this mechanism for CDR is known as ocean alkalinity enhancement (OAE).

One approach for increasing ocean alkalinity is depositing tons of ground silicate minerals, such as olivine, into ocean surface waters. Limestone and calcium carbonate derivatives have also been considered. Using this mechanism for CDR is a type of once-through carbon capture process (§2), so the mass of material needed is similar to the mass of the carbon that is removed. For example, at least 2 Gt of olivine is required to remove 1 Gt of $CO_2$ [61]). Implementing this approach on the gigatons per year scale would thus require adding billions of tons of alkaline minerals yearly to ocean systems.

The energy requirements for OAE are similar to those for enhanced rock weathering (§4.3). Energy needs are likely dominated by the energy for grinding rock, but also include transportation and distribution needs. In principle, if done in appropriate places, the surface water bearing extra alkalinity would equil-

[27] Alkalinity is related to but distinct from pH, see App. F and App. G.



ibrate with atmospheric carbon dioxide and then sink into the deep ocean, where the captured carbon would remain over thousands of years [62]. [28] Regions of strong deep convection, such as the ocean south of Iceland, where the surface waters may sink before carbon capture from OAE has a chance to fully take effect should thus be avoided.

OAE may be a promising approach to large-scale atmospheric carbon removal, similar in spirit to enhanced weathering. However, many aspects of the ocean chemistry and biological impacts of this approach are poorly understood and need further research before any effort to implement at scale. [29] In particular, as with ERW there are questions about the overall efficiency of the approach in terms of the quantity of $CO_2$ captured for a given amount of material added. Some recent research [64] indicates that adding alkaline minerals may lead to much less change in total alkalinity (TA) and associated $CO_2$ uptake than expected [30], although other studies suggest that these effects may be controllable [65, 66]. There are also questions regarding the environmental impact of OAE. [31] Distributing the alkaline material as broadly as possible to minimize the magnitude of local changes may help to reduce adverse effects, but would increase the cost of transport and distribution. The long-term effects of adding gigatons of alkaline minerals to the ocean and the time scale of mixing also need to be better understood. Possible changes to ocean albedo due to plankton blooms or through the release of dimethyl sulfide that can form cloud condensation nuclei should be considered. There may also be issues with a decrease in the Meridional Overturning Current driven by climate change [67, 68] that could reduce the sinking of carbon-rich waters.

Just as for enhanced weathering, before any effort to undertake OAE at large scale, it is important to better understand the issues associated with the full implementation cycle, including the energy costs of extraction, transport, and grinding, as well as the environmental impacts and other externalities for gigatons of materials (for comparison, the global cement industry uses roughly 7 Gt of material annually). It has also been suggested that some industrial waste may be used as a source of alkalinity, but this would likely raise increased issues of toxic trace elements.

There are close connections between the chemistry involved in enhanced weathering (§4.3) and OAE. [32] The enhanced weathering process removes atmospheric carbon through the formation of carbonates. If the carbonates enter the ocean and dissolve, the originally captured carbon is released but the increase in alkalinity causes the uptake of other atmospheric $CO_2$ through OAE. This is the reverse of the process noted in App. G, where the precipitation of calcium carbonate leads to the expulsion of $CO_2$ from the ocean into the atmosphere.

### 4.4.2 Electrochemical Ocean CDR

Various electrochemical approaches to ocean carbon dioxide removal have been considered. Many of them either modify ocean alkalinity as in §4.4.1, or are essentially cyclic $CO_2$ removal processes (§2) like the chemical DAC systems described in §4.1. Reviews of some of these approaches can be found in [30, 59, 60].

[28] It is worth noting that an additional consequence of adding alkalinity to ocean water that sinks into the deep ocean is that it offsets the extent to which ocean acidification increases dissolution of calcium carbonate in the deep ocean floor — such dissolution would add alkalinity while bringing deposited carbon back into the ocean system.
[29] This is a very active area of investigation. Some of the issues are outlined in more detail in, e.g., [63].
[30] This is attributed to precipitation of, e.g., calcium carbonate (which reduces pH, see App. G), Mg-Si precipitates, and buffering effects of seawater.
[31] For example, in addition to the magnesium that enhances alkalinity, reaction products from olivine include silicic acid and iron, which can significantly impact marine biology, as well as trace amounts of nickel, chromium, and other potentially harmful metals.
[32] In particular, enhanced weathering on beaches has aspects of both approaches (see, e.g., [63]).

In many cases, electrochemical OAE uses electrodes to separate acid and base components from seawater or brine from desalination plants. The acid is then neutralized (typically through reaction with a silicate mineral), and the base is returned to the ocean. This increases the alkalinity and leads to a shift in carbonate chemistry that pulls $CO_2$ from the atmosphere as described in §4.1. This approach shares many of the issues with the general OAE approach. The impacts and effectiveness of these approaches have similar uncertainties in terms of ocean chemistry. In electrochemical OAE, however, the material output (e.g., salt and silica) is contained and not distributed in the ocean, so it can be disposed of or used in other ways. The energy cost of separating the acid and base can be high, with estimates of 3-18 GJ per ton of $CO_2$ [63].

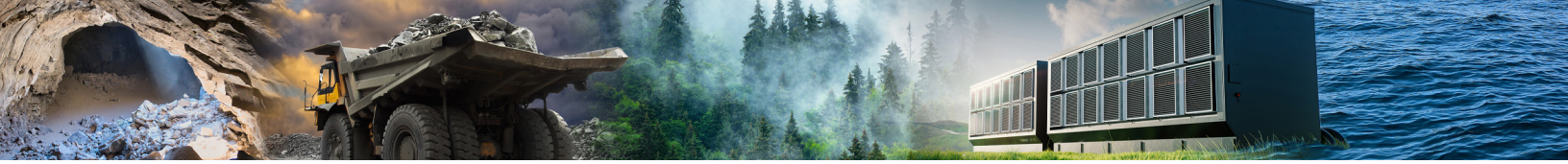
Two kinds of electrochemical ocean carbon capture (often called direct ocean capture or DOC) systems are being considered. In the 'acid process,' an acid is combined with a contained quantity of seawater, promoting the release of $CO_2$. That $CO_2$ is then captured and stored. In the 'base process,' a base is added to the seawater and carbonates are precipitated as solids and removed. Precipitation of the carbonates can also shift the chemistry so that $CO_2$ is released (App. G), and both the solid carbonate and $CO_2$ gas can be sequestered and stored. In both electrochemical ocean DOC processes, the acting acid or base is typically produced by an electrochemical process like electrolysis that generates both an acid and a base. The acid and base streams are recombined before they are returned to the ocean, so there is no net change in ocean alkalinity. These approaches are cyclic (§2), so second law constraints apply to the energy inputs. These approaches do not modify ocean chemistry as significantly as OAE approaches, but electrochemical DAC/DOC processes are not yet well developed. The much higher carbon density of seawater per unit volume (compared to air) suggests that such approaches may be effective. Note, however, that seawater is also much more dense than the atmosphere (by a factor of about 1,000), so the mass of water that needs to be processed to capture a given amount of $CO_2$ is roughly 10 times greater than the mass of air needed.

### 4.4.3 Biologically-based Ocean CDR

There are a wide range of approaches to using oceanic biological systems to capture and store atmospheric carbon. We briefly describe some of the main ideas here.

**Restoration of coastal and marine ecosystems (coastal blue carbon).** Just as in terrestrial ecosystems, human activity has led to a substantial loss of carbon stored in coastal and marine ecosystems — including salt marshes, mangroves, seagrasses, and others — through environmental degradation. Protecting and conserving existing coastal ecosystems is important for maintaining carbon stores. Restoring damaged ecosystems can effectively remove carbon dioxide and store it for hundreds or thousands of years, as long as the ecosystem is preserved. Similar to the CDR approaches based on the restoration of terrestrial ecosystems described in App. I, restoring coastal and marine ecosystems can provide a moderate amount of carbon recapture and storage with relatively minimal risks and potential auxiliary benefits (e.g., water quality, biodiversity, flood control). While such efforts are highly location-specific and must be considered in the broader context of human activity, land use, and biodiversity conservation, very rough estimates suggest that 0.1-1 Gt/yr of $CO_2$ could be captured and sequestered in this way [59].

**Seaweed cultivation.** Large-scale cultivation of seaweed has been proposed for carbon capture, similar to the idea of terrestrial direct carbon biocapture and storage. This approach and relevant literature are reviewed in [59]. A significant challenge to large-scale implementation of the seaweed approach is that coastal waters are essentially one-dimensional. Furthermore, seaweed photosynthesis is less efficient than microalgae photosynthesis and seems harder to optimally configure. For these reasons, seaweed farms capable of sequestering 0.1 Gt $CO_2$/yr would require roughly 60% of global coastline [59]. There are also substantial uncertainties and concerns about the environmental impact of this approach and proposed sequestration methods such as by sinking the seaweed to the deep ocean.



**Artificial upwelling and downwelling.** It has been suggested that artificially enhanced upwelling, bringing up nutrient-rich ocean water (e.g., by large banks of pumps), could enhance phytoplankton production and associated carbon capture. This idea is unproven and there are serious questions about its effectiveness and global impacts [59]. There are related suggestions for accelerating downwelling, increasing the rate at which the higher surface carbon content is brought into the deep ocean. It seems unlikely that these ideas could lead to effective large-scale atmospheric carbon removal, and much more research on their feasibility and potential impacts is needed before they should be considered possible CDR approaches.

**Nutrient fertilization.** Another proposed approach to increasing the rate of carbon capture by oceanic biological systems is adding nutrients (e.g., iron) to surface waters. The idea is to enhance phytoplankton photosynthesis and help the resulting carbon-based biomass reach the deep ocean for sequestration. It has been suggested that such fertilization efforts may have the co-benefit of enhancing some fisheries. While the cost and effort involved in implementing such processes may be relatively low, the effectiveness and environmental impact of such large-scale activities are complex and not well understood. For example, large-scale nutrient fertilization in the southern oceans could remove nutrients that would otherwise be consumed by organisms in northern oceans later (see, e.g., [69]). Research in this area has been complicated by poorly-designed experiments. [33] Some of the issues and related literature are reviewed in [59, 70]. This area is experiencing a revival in interest and government funding, but we do not focus on it in this report due to the uncertainties and potential large-scale global impact on ecosystems and human activity.

### 4.5 Other Approaches

As illustrated, a wide range of ideas have been proposed for CDR, and further creative solutions to the carbon capture problem may emerge. While not all ideas are treated directly here, we expect the general principles underlying this report to be relevant for most approaches. In App. K, we briefly describe a few more speculative directions beyond the main approaches treated in this section.

## 5 Approaches to Storing Captured Carbon


**Summary: Carbon Storage and Sequestration**

A straightforward approach to carbon storage and sequestration involves compressing the $CO_2$ to a supercritical fluid and injecting it into an underground reservoir, where it remains trapped. Under some circumstances the $CO_2$ can gradually mineralize underground. Studies suggest that underground storage sites are available that could accommodate many thousands of gigatons of supercritical $CO_2$. There do not seem to be any fundamental obstacles to implementing this type of $CO_2$ sequestration at large scales, but concerns such as leaks and earthquakes need to be addressed.

Other approaches for dealing with captured $CO_2$ include mineralization and use in long-lived construction materials such as concrete. These methods are less developed.


A number of approaches have been suggested for using, storing, and sequestering captured $CO_2$. Currently, most captured $CO_2$ is used in enhanced oil recovery or for various industrial purposes (§5.3). To affect

[33] In addition, in 2012 the Haida Salmon Restoration Corporation added five times more iron sulfide than previous experiments to the Pacific Ocean off the coast of British Columbia without approval by Environment Canada, setting back further research.

actual net carbon emissions over the long term, however, the carbon in the $CO_2$ must not reenter the atmosphere/biosphere carbon cycle for thousands of years — using $CO_2$ to produce carbonated beverages, for example, does not count. The primary approaches to long-term sequestration can be broken down into (1) storing $CO_2$ permanently underground, (2) binding captured carbon into solid materials that may be buried or sequestered, and (3) incorporating captured carbon into relatively permanent material structures for human use. While there is some overlap, we briefly discuss each of these separately. A comprehensive report on these topics is given in chapters 4-7 of [71].

## 5.1 Compressed Underground Storage

The primary approach envisioned for handling large quantities of $CO_2$ captured in cyclic CDR approaches, such as chemical DAC, is compressing the $CO_2$ to a supercritical fluid and sequestering it in geologic reservoirs. [34] The supercritical fluid has a high density that reduces buoyancy and allows more $CO_2$ to be sequestered. This approach can be effective on millennial time scales. Over time, depending upon conditions, the $CO_2$ can become mineralized (§5.2) — further increasing the stability of this approach.

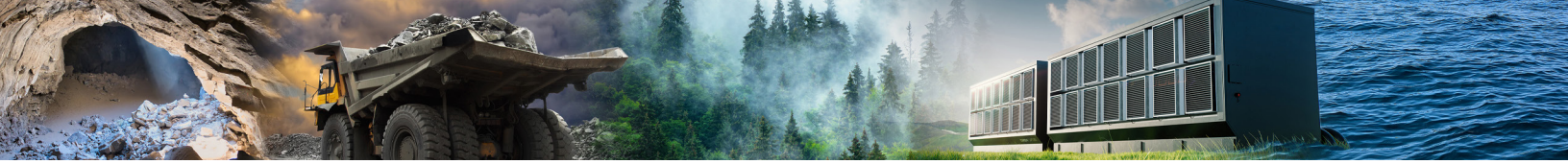
Geologic formations suitable for long-term $CO_2$ storage generally consist of porous and permeable rock that can serve as a reservoir and a seal, such as a 'cap' of impermeable rock. Many depleted oil and gas fields meet these criteria. There are also many deep saline formations that satisfy these conditions [74], with a range of trapping mechanisms, including residual trapping and dissolution trapping in addition to structural trapping [75]. Currently, $CO_2$ is used in enhanced oil recovery (EOR) to push out the last parts of a fossil fuel reservoir, although the $CO_2$ in this context is gaseous and long-term storage is not an explicit goal. For large-scale storage and sequestration of supercritical $CO_2$, it will likely be necessary to go beyond current and depleted fossil fuel reservoirs. [35] A 2013 meta-analysis of published literature on available geologic storage estimates a 'practical' capacity of more than 3,900 Gt $CO_2$ [74], much in deep saline formations. A more recent report estimates 8,000-55,000 Gt [78]. Considering these and other estimates, there is likely more than enough capacity to store tens of gigatons of $CO_2$ per year for the rest of the century. At the current time, roughly 30 Mt/yr of $CO_2$ is being sequestered in a combination of enhanced oil recovery projects and saline aquifers [79].

Some concerns have been expressed regarding the potential for leakage or seismic events from $CO_2$ injections. A high rate of leakage could compromise the permanence of storage. Because $CO_2$ is denser than air, and can pool in low-lying areas, a large-scale rapid leak of compressed $CO_2$ could also have tragic local consequences (as in the 1986 Lake Nyos disaster [80] and Mammoth Mountain $CO_2$ leakages [81], both involving natural subsurface $CO_2$ accumulations). Some studies [82] have estimated low risks from natural $CO_2$ seeps. While experience and studies related to fossil fuel exploration and extraction offer some perspective on seismic risks, a more thorough understanding of these issues in the context of large-scale injection of pressurized $CO_2$ is desirable. Recent summaries of these risks and how they may be addressed are given in [83, 84].

Large-scale sequestration can likely be carried out at many sites with safety precautions that reduce the likelihood of disastrous leakage or a catastrophic seismic event to a very small possibility. Nonetheless, accurate measurement and evaluation methods, including establishing baseline conditions and monitoring both subsurface and surface changes, are needed to detect leaks or reservoir weaknesses in a timely fashion. There have been a number of experimental tests of $CO_2$ release from shallow sources, and quantifying leakage is challenging using current methodologies [85]. Some further discussion of measurement, reporting, and verification (MRV) approaches for geologic storage is given in §6.1. It would be prudent, however, to do

[34] This includes offshore storage in geologic reservoirs beneath the sea floor [72]. Deep ocean storage has also been considered but is not currently viewed as a favorable option [73].

[35] However, it is worth noting that, for example, the yearly volume of reinjected produced fluids from the U.S. oil and gas industry — ~ 15-25 Gbbl ≅ $24\text{-}40\times10^{11}$ liters [76, 77] — is equivalent to that of several Gt of compressed $CO_2$.



further study and risk assessment and to develop robust MRV protocols while moving toward large-scale implementation of underground $CO_2$ sequestration. It is worth noting that the consequences of leaks and seismic events are substantially attenuated for geologic reservoirs beneath the sea floor.

## 5.2 Mineralization

Carbon dioxide can react with alkaline minerals to produce solid carbonates such as calcium carbonate ($CaCO_3$) and magnesium carbonate ($MgCO_3$) (as described in §4.3 and App. F). In addition to surficial approaches, like ERW, carbon mineralization can be carried out in situ in subsurface reservoirs, or ex situ in controlled situations such as engineered reactors. The resulting carbonates store the carbon in a stable and compact form. While we focus primarily on chemistry-based mechanisms for mineralization, there is also current research on using biological systems to enhance or implement mineralization processes. This is a promising avenue for further research and development. A more complete review of many of these processes and approaches can be found in [30].

In situ mineralization can occur naturally in geologic repositories when the surrounding rock types are sufficiently alkaline, such as basalt or other mafic or ultramafic rocks. The mineralization rate is enhanced in the presence of water. [36] At the Climeworks/Carbfix Orca DAC plant in Iceland, $CO_2$ from direct air capture is dissolved in water and mixed with brine before injection, and the reaction rate is enhanced by geothermal energy (see, e.g., [87]); in this operation, around 25 t of water are needed for each ton of $CO_2$. Studies based on models [88] suggest a high rate and capacity for mineralization, although there are substantial uncertainties regarding many of the processes involved (for further discussion and references see [30]).

Ex situ approaches to mineralization have been studied in the context of point-source carbon dioxide capture, but could potentially be applied in DAC scenarios as well. In ex situ mineralization, natural rock or industrial alkaline waste or byproducts are reacted with $CO_2$-rich gases in controlled environments, such as engineered reactors. Ex situ approaches may enable better control of toxic or environmentally hazardous trace elements, particularly for industrial output, compared to surficial approaches such as simply spreading alkaline industrial output or mine tailings over large land areas. The quantity of industrial alkaline waste material produced globally has been estimated at 7 Gt/yr, with a potential for capturing 3-8 Gt $CO_2$/yr, [89]. An overview of the carbonation of silicate minerals and alkaline industrial wastes is given in [90]. Ex situ approaches may also be desirable in situations where $CO_2$ is integrated into a solid that may have other uses (see next section).

## 5.3 Integration of Captured $CO_2$ Into Value-added Materials

Carbon dioxide has extensive uses in industry. Aside from its previously mentioned use in enhanced oil recovery, $CO_2$ is used to produce urea for fertilizer, in food and beverage packaging and production, and for many other purposes. About 150 Mt of $CO_2$ is used in these ways each year [91], but much of this $CO_2$ is released to the atmosphere within a few months or years. [37] However, there has been recent interest in integrating captured $CO_2$ into building materials such as concrete, where it can be sequestered for potentially hundreds to thousands of years. Global cement production occurs at a rate of roughly 4.2 Gt/

[36] The effectiveness of injecting water with the supercritical $CO_2$ has been investigated, see, e.g., [86]. Other approaches to in situ mineralization include pre-dissolution of $CO_2$ before injection, even in the absence of a caprock or containing structure; mixing of the $CO_2$ with alkaline geological fluids before injection to form DIC; and circulating seawater through ultramafic rocks in coastal regions [30]. The last of these approaches is closely related to surficial weathering and coastal OAE approaches that also capture carbon through alkalinity enhancement (§4.4.1).

[37] And, in the case of urea used in fertilizer, there is associated release of other greenhouse gases ($N_2O$). see, e.g., [91].

yr [92]. Because producing cement for concrete contributes substantially to anthropogenic $CO_2$ emissions, integration of captured $CO_2$ is often studied in conjunction with other approaches to producing building materials with a low carbon footprint. There are also efforts underway to incorporate $CO_2$ into the stony aggregate that constitutes more than half of the mass of most concrete mixtures (vs. ~ 10-15% for cement). These efforts are at an early stage of development, but may be a promising approach to long-term sequestration of multiple Gt of $CO_2$ yearly and further research and development in these directions should be pursued. Further details on such approaches can be found in [90, 93]. Other uses for captured $CO_2$, such as fuel production, are also being explored. [38] An overview of technological and economic aspects of a number of pathways for $CO_2$ utilization is given in [94].

# 6. Systems Considerations and Frameworks


**Summary: Measurement, Reporting, Verification, and Other System Issues**

Good measurement, reporting, and verification (MRV) methodologies need to be developed for most approaches to atmospheric carbon dioxide removal. MRV for geologic sequestration of compressed $CO_2$ is somewhat developed already. For ecosystem-based CDR and once-through processes such as enhanced weathering and OAE, the issues are more subtle and systematic methodologies have yet to be developed.

Good MRV is needed in order to create an effective economic framework that supports large-scale CDR. Atmospheric carbon capture at the Gt/yr scale will be difficult and costly, but if the price can be reduced to $100-$200 per ton of $CO_2$ for engineered CDR systems, as some estimates suggest, that would correspond, for example, to a cost of $1-$2 to capture the carbon released when an automobile burns a gallon of gasoline.

Building a coherent and effective portfolio of CDR systems requires a better understanding of the risks and potential environmental impacts of different approaches. It also requires a coordinated approach to implementing CDR systems in appropriate geographical locations, in combination with energy sources and storage sites, and with consideration of land use and energy issues.

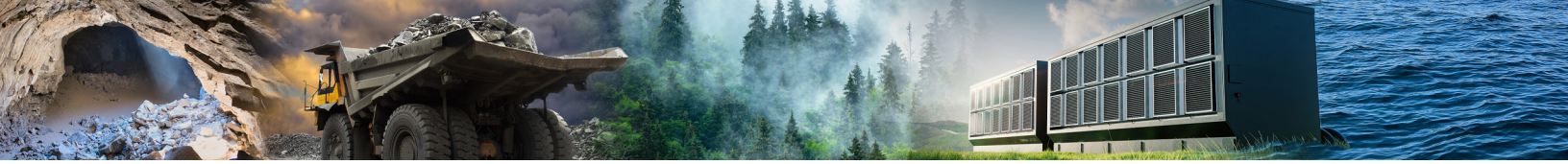

In this section we consider some systems aspects of CDR approaches and technologies, as well as questions related to measurement, verification, safety and environmental impact, costs, and policies.

## 6.1 Measurement, Reporting, Verification (MRV), and Standards

If a serious effort is made to implement CDR at a large scale, it is vital to have accurate and reliable methods for verifying that carbon has actually been removed from the atmosphere and is stored permanently with minimal leakage, and that the process has not had serious negative impacts on ecosystems or human welfare. We briefly discuss here some issues of measurement, reporting, and verification (MRV), with a focus on the challenges of measuring and verifying that carbon has actually been stored and is retained; such systems are needed for any functional carbon management policy. Issues of risk, safety, and impact are discussed briefly in the following subsection.

A number of third-party organizations have developed guidelines for MRV practices, particularly in the context of geologic storage [95, 96, 97], following the ISO International Standard for GHG Emissions Inventories and Verification [98]. Some private companies developing direct air capture systems have made preliminary progress in verification systems. The US Environmental Protection Agency (EPA) has developed an MRV framework for geologic sequestration sites (Class VI wells), that is designed to protect public health

[38] See, e.g., [91].



and underground sources of drinking water [99]. There is, however, no general, systematic, and well-defined framework in the United States for setting standards or vetting carbon capture and storage efforts for most potential CDR methods. The lack of systematic MRV has led to claimed carbon offsets of questionable validity (see, e.g., [100]). Uncertainty in carbon storage measurements has been cited as a contributing factor in the failure of one U.S. attempt to form a voluntary carbon market (the Chicago Climate Exchange) [101]. In the European Union, a carbon Emissions Trading System (EU ETS) has been developed around allocations for carbon emissions from various industries. This has motivated some of the MRV efforts for carbon capture systems. Nonetheless, the technical components and institutional structures needed for a robust MRV framework for CDR implementations are still at an early stage of development. Such systems are crucial to enabling governments and private organizations to work in consort with market forces to effectively develop and harness large-scale carbon capture systems. One effort to organize the current state of MRV frameworks for a variety of proposed CDR pathways can be found online at [102].

At a technical level, the questions involved in MRV are perhaps clearest for the geologic storage of compressed $CO_2$ gas. In this context, measurement and verification procedures are needed to assess the rates of storage, leakage, and mineralization, and other factors such as the subsurface distribution of $CO_2$. For such assessments, it is crucial to have accurate baseline measurements. A review of some state-of-the-art systems involved in such monitoring and verification is given in [96]. For geologic storage, monitoring techniques are roughly divided into atmospheric monitoring techniques, near-surface monitoring techniques, and subsurface monitoring. [39] While further research is desirable on these (and other) MRV approaches to underground storage, many of the ingredients exist for a robust measurement framework that can be integrated into a uniform system of standards and regulation around geologic $CO_2$ sequestration (like the EPA Class VI framework mentioned above). MRV frameworks should also be relatively manageable for the long-term storage of compact material containing carbon as in BiCRS (§4.2.3), and there are recent efforts in this direction.

Monitoring and verification for other carbon capture approaches, particularly once-through (§2) systems such as enhanced weathering and OAE, are at a much less developed stage. [40] One primary challenge is to separate CDR gains from large natural fluxes that may occur — to identify $CO_2$ that is removed beyond what would happen in the absence of CDR. Measurement alone may not suffice. Accurate modeling (with its inherent uncertainties) will likely be required [62]. This makes verification and standardization of such systems a significant challenge. For ERW, one MRV challenge is studying how carbon captured on land may be lost as the carbon in reaction products runs off the land, through rivers, and to the ocean. In OAE, particular challenges for accurate MRV include clarifying the dispersal of alkalinity, the rate of $CO_2$ uptake, and the extent to which the inorganic precipitation of carbonates occurs, leading to the release of carbon dioxide gas [105].

MRV for changes in soil organic carbon (SOC) in ecosystem-based CDR that alters agriculture and grazing practices is challenging for similar reasons, as is ensuring the longevity of CDR realized through reforestation. Land use practices lead to large fluxes associated with the emission as well as removal of

[39] Atmospheric monitoring techniques include relatively inexpensive optical sensors, with lower precision, and eddy covariance flux monitors, which are more expensive but more precise, among other technologies. Space-based monitoring techniques may also play a role here. Near-surface tools and techniques include geochemical monitoring of soil and groundwater and surface displacement monitoring. Subsurface monitoring techniques are generally based on well-developed approaches used in the oil and gas industries, and include wellbore-based monitoring tools, active and passive seismic methods, and gravity and electrical measurements. One primary role of subsurface methods is to track the motion of an injected $CO_2$ plume in a geologic formation. Note that a number of new sensor systems are also under development for detecting methane emissions from oil and gas infrastructure [103]. The standard near-IR bands for sensing methane are relatively close to bands for sensing $CO_2$, so new methane detection technologies might be repurposed for $CO_2$ detection.

[40] There is a great deal of work on this and progress has been made on some approaches, such as isotope dilution inductively coupled plasma mass spectrometry (see, e.g., [104]).

atmospheric carbon, and changes in terrestrial biomass and soil carbon content come from both human land management choices and changing environmental conditions. [41] In addition, it is difficult to ensure the longevity of ecosystem-based CDR in the face of a still-growing human population, changing human resource needs, and climate change.

Developing MRV approaches for once-through and ecosystem-based CDR that honestly verify and evaluate net carbon capture, as well as confirm that such carbon removal is additional (an added gain that would not have occurred naturally), real, and permanent, is an important challenge for integrating these CDR approaches into a broad carbon management strategy.

## 6.2 Safety, Risk, and Environmental Impact of CDR

Because any approach to CDR at the gigaton per year scale will involve processing billions of tons of material, it is important to carefully assess the risks and potential impact of any CDR process before implementing at scale. MRV efforts (as described in, e.g., [102]) are often primarily focused on measuring and verifying the amount of carbon captured and that the sequestration is real and permanent. However, a systematic approach and associated regulations and standards are also needed to ensure that large-scale CDR efforts do not have an undue impact on humans or the environment. We have mentioned some of these issues in connection with various CDR approaches in §4. For chemical DAC and associated geologic storage, some of the concerns are of large $CO_2$ leaks and induced seismic activity (see section4.1). While these concerns can likely be managed, active and effective monitoring is important for a reliable and robust large-scale geologic sequestration effort. For ERW and OAE, in addition to the environmental impact of mining billions of tons of alkaline minerals, the primary concerns revolve around the environmental impact of adding billions of tons of reaction products to the land or ocean. For both these once-through approaches, the reaction products will gradually disperse into the environment, and their full life cycle should be understood and modeled. In some cases, ecosystem-based CDR can have more environmental benefits than negative impacts, but poorly planned or poorly managed projects can do harm both to ecosystems and ecosystem services for humans. Trade-offs in land use are important in any approach to CDR, particularly for biologically based approaches, as discussed further in §6.5.

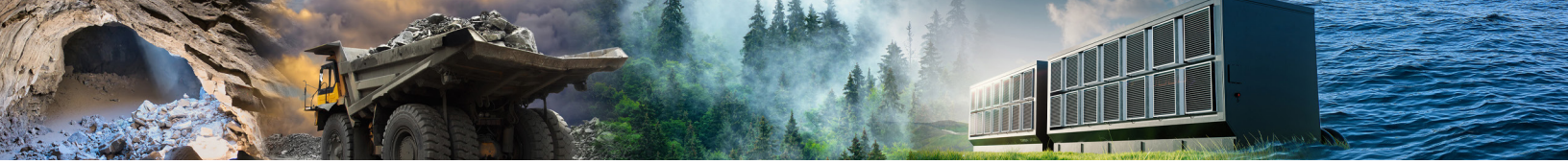
## 6.3 Systems Considerations

There are a number of system-level issues to consider for large-scale implementation of CDR. Here we focus on some aspects relevant to cyclic terrestrial DAC systems. More distributed approaches, such as ERW, OAE, and ecosystem-based capture, also have important life-cycle and system issues, but the issues are perhaps more diffuse and complex.

**Energy sources.** Capturing a few gigatons of $CO_2$ per year using cyclic CDR systems would likely require hundreds of gigawatts of power, and capturing 10-20 Gt/yr would require power at the terawatt (TW) scale (§3.1). This power cannot come from the existing grid without displacing existing energy uses, so significant new power production capacity would be necessary for any large-scale CDR effort of this type. [42] Because fossil-fuel based power generation without point-source carbon capture lowers the effectiveness of any carbon capture system (§3.4), the power supply should be carbon-neutral or as low-carbon as possible.

[41] Examples of $CO_2$ fluxes from changing environmental conditions include the increased fertilization effect associated with increased atmospheric $CO_2$ and possible enhancement of soil organic carbon decomposition in a changing climate [106, 107])

[42] As renewable energy sources power increasingly large fractions of the grid, it may be possible to consider using intermittent renewable overcapacity to power CDR, although, for example, running a DAC plant only during daytime hours may be economically inefficient.



Many existing and proposed systems rely heavily on thermal energy, for example, those that use thermal swing chemical processes to capture and release $CO_2$. It is thus natural to consider directly using energy from thermal sources. As of 2023, the largest existing $CO_2$ capture plant was located in Iceland and makes use of that country's abundant supply of geothermal energy. In other locations, potential sources of carbon-free thermal energy include solar thermal energy and waste heat. While in principle using waste heat from power plants or industry is an appealing approach, the quantity available may be insufficient for gigaton-scale CDR, and the fraction of useful energy (exergy, §3.1) is relatively low. This is an important area for further research and development, which is onging. That being said, solar energy is widely available. The technology for concentrated solar power has long been developed and could potentially play a role in powering cyclic DAC systems with high-temperature thermal energy. The rapidly dropping costs of photovoltaic (PV) solar panels, however, may make PV a simpler and more economical energy source. PV power may be particularly effective with new electrochemical DAC methods (see further discussion in §6.5), although it would require storage for 24/7 operation. [43] Nuclear power is another potential carbon-free option with low land requirements, although it has its own issues.

Because the energy and materials needed for CDR are so extensive, new capacity will be needed to power CDR systems. This should be additional to the development of carbon-free energy sources to replace existing fossil-fuel based infrastructure and to expand energy availability. In many circumstances, even without considering economics, using new carbon-free power generation to replace existing fossil fuel infrastructure is more effective than using it to power CDR. [44] For either application, storage and intermittency issues for renewables such as solar and wind need to be addressed systematically.

**Siting.** It is widely appreciated that proximity to energy sources and geological storage sites is an important consideration in siting CDR plants such as chemical DAC systems. One question that needs to be understood better is how densely placed $CO_2$ capture systems can be while remaining effective. Similar to the need to place wind turbines sufficiently far apart to avoid interfering with one another, there is likely an optimum density for different DAC plant designs. It is not clear if this imposes any practical limitations on large-scale implementation of these technologies, but the question should be studied further. Additionally, lower $CO_2$ downstream may adversely affect sensitive ecosystems or agricultural areas, which may also affect siting decisions.

## 6.4 Time Scales of Different CDR Approaches

Different CDR methods may be more appropriate for different time scales and goals. Many factors should be considered in this context, such as the timescale over which $CO_2$ removal is needed, the natural lifecycle time of different CDR systems, their predicted removal rate, and the time to vet and develop them fully. Some technologies may take longer to develop, or may operate at a slower rate, but may remove atmospheric $CO_2$ in a more inexpensive, safe, and sustainable way over long time scales. For example, the natural weathering of silicate rocks removes around 1 Gt $CO_2$/yr (§4.3). Doubling the rate with ERW would not remove the multiple gigatons of $CO_2$ per year that may be desirable in the coming decades, but

[43] For DAC processes that primarily require thermal energy, solar thermal energy can be more easily stored over the diurnal cycle.
[44] For example, using a 100 MW solar plant to replace a coal plant with typical emissions of ~ 1 t $CO_2$/MWh results in a similar net energy output and $CO_2$ emission reduction profile as powering a chemical DAC plant operating at eight times the thermodynamic limit for carbon capture energy needs. Replacing the coal plant, however, removes the cost of operation of the coal plant and fuel (as well as avoiding any adverse health effects from coal plant emissions), whereas there would be additional costs to building the DAC plant and for compression and storage. While issues such as intermittency and storage would need to be addressed in a more complete analysis, energy and carbon considerations alone suggest that in a case like this replacing the coal plant should be prioritized, even without considering the details of costs. In any realistic situation, of course, particularly for DAC systems with lower energy costs, or power plants like combined cycle natural gas plants with lower emissions, a more detailed economic analysis would be needed.

if this could be done safely the approach might play a significant role in keeping $CO_2$ levels constant on a century time scale. Similarly, other relatively low-tech approaches, like OAE, may play a useful role in the long-term management of atmospheric $CO_2$.

## 6.5 Land Use

One important issue for many CDR methods is land use. In particular, biologically based CDR such as BECCS requires large land areas for growing crops (App. E), and the total amount of carbon that ecosystem-based CDR can capture and sequester is limited to a fixed total that is proportional to the land area over which these strategies are applied. By contrast, chemical DAC plants based on cyclic processes require less land. Their main land area requirement may be primarily what is needed to power the system with renewable energy. As a back-of-the-envelope estimate, consider powering a set of chemical DAC plants capturing 1 Gt $CO_2$/yr using three times the thermodynamic minimum of energy. Generating sufficient power using solar collectors would require about 4,000 $km^2$. [45] This is a large area but smaller — by at least an order of magnitude — than the area necessary for similar capture using BECCS (App. E). The area needed for a chemical DAC plant itself is comparatively smaller, but still substantial; there are likely some constraints on how closely such plants can be placed to remain fully effective (§6.3).

## 6.6 Cost of Atmospheric Carbon Capture

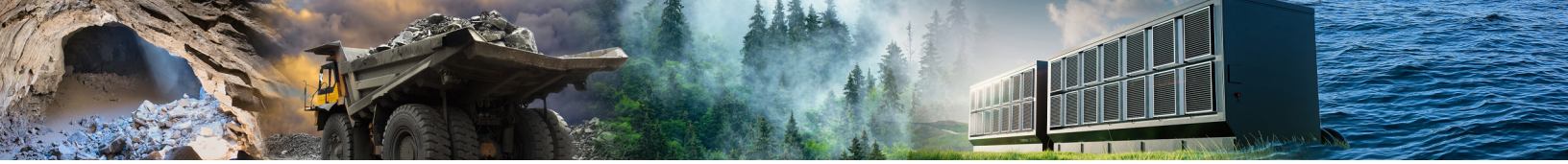
In the bulk of this report we have avoided discussing specific cost estimates for atmospheric CDR technologies. A complete cost analysis must include the cost of all aspects of the life cycle, including extraction of materials, construction of systems, energy and other inputs, transportation, storage, and externalities. Such an analysis is outside the scope of this report. Some estimates suggest that chemical DAC systems currently under development may be able to achieve costs in the range \$100-\$200/t $CO_2$ (see [20, 4] for a more extensive discussion and references). [46] A back-of-the-envelope computation [47] shows that even at several times the thermodynamic minimum, the basic energy costs for a cyclic CDR approach like DAC can, in principle, be on the order of \$20-\$40/t $CO_2$. This suggests that the basic energy constraints given by thermodynamics are not incompatible with the goal of reducing the cost of capturing carbon with chemical DAC systems to \$100-\$200/t $CO_2$.

The relatively high cost of carbon capture makes it clear that it will often be less expensive to avoid emis-

[45] 42 GW of sustained power is necessary. We assume solar collectors operating at a typical gross efficiency of 5%, and land areas with an average surface irradiation of 200 W/$m^2$. While typical modern solar cells have an efficiency of ~ 20% for normally incident light, for large solar fields the relevant parameter is 'gross conversion efficiency,' given by average power output divided by average irradiation, which takes into account the substantial fraction of land that is covered by infrastructure and not solar panels. For example, the largest U.S. solar plant, the Copper Mountain Solar Facility, in a location with irradiation ~ 270 W/$m^2$, has a site area of 16 $km^2$, a capacity of 802 MW, and a capacity factor (ratio of average actual production to full capacity) of 27.9%, so gross efficiency is close to 5%. Note that land area is smaller if irradiation is above 200 W/$m^2$; we have chosen this typical number as representative because many factors beyond irradiation will go into siting large chemical DAC installations, we have also used this number of 200 W/$m^2$ elsewhere for analogous calculations, e.g., App. E. Similarly, for systems with larger gross efficiency, land area reduces correspondingly.

[46] One commonly used benchmark is the goal of direct air carbon capture and storage at a cost of \$100/t $CO_2$; this is the target cost, for example, of DOE's Carbon Negative Earth Shot program [108]. The full cost of current chemical DAC systems, including all stages of the life cycle, seems to be substantially higher than this (e.g., Climeworks has stated prices of \$500-\$600/t, [20]), and the target of \$100/t $CO_2$ may be difficult to reach [109]. In this context, it is worth noting that it has been estimated that nature/ecosystem-based CDR approaches can potentially remove up to several Gt $CO_2$/yr over the next several decades at a much lower cost of ~ \$10/t $CO_2$ [26, 110], although there are limitations regarding scale and MRV, and ensuring permanence of $CO_2$ removal can be difficult.

[47] Using an estimate of \$40/MWh [111] for the levelized cost of electricity (which includes life-cycle costs such as power plant construction, etc.), and assuming the $CO_2$ separation process can be implemented at three to six times the thermodynamic limit, i.e., 1.5-3 MJ/kg, and assuming that additional energy costs for compression and/or air movement raise this to 2-4 MJ/kg, this gives a cost of roughly \$20-\$40/t $CO_2$.



sions than to recapture carbon. In particular, it would make economic sense to replace power plants and other systems that generate extensive $CO_2$ emissions with carbon-free or lower-emitting alternatives when the cost lies below that of the CDR alternative. For example, before installing a DAC system at a cost of \$200/t $CO_2$, it would make sense to replace any coal plant emitting on the order of 1 t $CO_2$/MWh with alternative zero-carbon power sources having levelized energy costs below \$200/MWh (see, e.g., [111]).

## 6.7 Economic Framework for Atmospheric Carbon Management

Removing multiple gigatons of carbon dioxide from the atmosphere on a yearly basis using approaches such as chemical DAC and once-through processes like OAE and enhanced weathering would be quite costly. At a cost of \$100-\$200/t $CO_2$, the cost of CDR would amount to \$100-\$200 billion a year for each Gt/yr of $CO_2$ captured. Emissions reduction strategies also have associated costs, and a rational strategy for atmospheric carbon management would involve developing policies that provide comparable economic incentives for these approaches, while also considering environmental and social costs.

A conceptually simple framework correlates the cost of carbon capture with the cost of $CO_2$ emissions. CDR at the rate of \$100-\$200/t $CO_2$ would represent a cost of roughly \$0.90-\$1.80 to capture the $CO_2$ emitted in burning a gallon of gasoline. [48] Whether the cost of atmospheric carbon management through emissions reductions and CDR is borne by some kind of cap and trade system, as has been developed in the European Union, or is shouldered by governments (as proposed in, e.g., [13]), is a policy question that goes beyond the framework of this report. It seems logical, however, if CDR is to be implemented at any substantial scale, that an economic and policy framework should be put in place to impose an effective cost on carbon emission that reflects the abatement cost. [49] Such a framework would allow the market to naturally implement trade-offs like those discussed above. [50] Note that this kind of framework can apply even in scenarios with net negative emissions, although in such scenarios the question of who pays for the atmospheric carbon reduction (which we do not address here) becomes particularly difficult.

How efforts to limit atmospheric carbon levels and associated costs are distributed internationally is a difficult and important question. Some equity considerations may suggest that developed countries that have already emitted substantial amounts of carbon have greater responsibility for carbon removal, and balancing the rights and responsibilities of different countries in this regard will be a challenging aspect of managing global atmospheric carbon levels.

## 6.8 Other Technologies: Point-Source Capture and Solar Radiation Management

This report has not focused on direct air capture at fossil $CO_2$ point sources (as in CCS), such as pre- or post-combustion capture in fossil fuel power plants. For such systems the concentration of $CO_2$ in, for example, flue gases is much higher than in the atmosphere. [51] Thus, the energy costs of point-source capture are significantly smaller than DAC systems per ton of $CO_2$, although point-source capture at fossil fuel power plants does not directly reduce atmospheric carbon and must be tailored to the specific parameters of different sources. While this approach may be useful in reducing carbon emissions in the coming years, efforts to date have been less successful and more costly than anticipated [113].

[48] Burning a gallon of gasoline releases roughly 8.9 kg $CO_2$ into the atmosphere.
[49] A similar conclusion was expressed in a special report on CDR in *The Economist* [112]: "The idea of a market where the cost of emitting carbon dioxide is the price you have to pay to have it removed is very appealing." They continue, however, with "Actually creating one will be very hard."
[50] Recent enhancements to 45Q tax credits for carbon capture in the Inflation Reduction Act incentivize both point-source and direct air carbon capture by U.S. companies.
[51] e.g., 13-15%/3-4% for coal/gas plants for post-combustion, 50% or higher in syngas in pre-combustion capture systems.

This report has also not considered solar radiation management (SRM) approaches that would offset warming without reducing atmospheric $CO_2$ levels by, for example, seeding the stratosphere with reflective aerosols or brightening marine clouds. The impacts of such approaches are still poorly understood, and consideration of these issues is outside the scope of this report. We note, however, that some approaches to CDR, such as forestation and possibly ERW and OAE, may have significant secondary radiative impacts [114]. It is also important to note that methods proposed for solar radiation management do not actually reduce atmospheric carbon. Thus, in addition to other possible negative impacts on the environment, if these technologies are implemented and anthropogenic carbon emissions are not significantly reduced, $CO_2$ will continue to build up in the atmosphere and oceans, requiring even greater offsets and leading to increasingly strong repercussions if/when the radiation management effort is ended or interrupted. This could create a situation in which the disruption of human activity due to any global crises (pandemic, war, etc.) could be exacerbated by an artificially maintained climate system in an unstable state. Additionally, offsetting warming does not mitigate other aspects of anthropogenic $CO_2$-related climate change such as increased ocean acidification. Further discussion of the issues and concerns with solar radiation management appears in [115].

To develop a sensible carbon management policy that reflects the potential costs and benefits of all available options, careful evaluation and risk assessment of solar radiation management approaches to offset warming should be pursued. Some specific recommendations along these lines are given in [116].

# 7. Overview, Conclusions and Recommendations

## 7.1 Overview

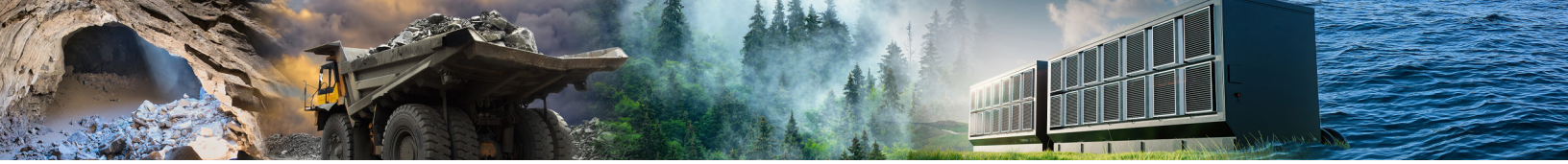
Modern society depends crucially on access to extensive and reliable energy, and fossil fuels have provided a compact and economical source of such energy over the last century. There are various strategies — depending on wealth, resources, politics, and societal values — that countries (independently or through international agreements) may use to balance energy needs with concerns about the ongoing rapid increase in atmospheric carbon dioxide levels.

**(A)** **Emissions reductions:** Replace existing fossil fuel-based energy systems that emit $CO_2$ with low-carbon or zero-carbon alternatives, and use low- or zero-carbon energy sources for new additional energy systems. Pursue energy efficiency measures and conserve carbon-rich ecosystems.

**(B)** **CDR:** Use carbon dioxide removal (CDR) methods, as described in this report, to extract excess $CO_2$ from the atmosphere and/or balance ongoing emissions from difficult-to-transition technologies.

**(C)** **Solar radiation management (SRM):** Implement large-scale SRM to reduce warming. Various concepts have been proposed for reducing the warming effects of atmospheric $CO_2$ by offsetting incoming solar radiation without directly addressing atmospheric $CO_2$ levels. Some of these ideas are speculative, and the direct and indirect impacts of solar radiation management strategies are not yet well understood. While these are not the subject of this report, a brief discussion of such technologies and potential issues is given in §6.8.

**(D)** **Adaptation:** Where feasible, offset the undesirable consequences of warming and adapt systems to accommodate changes in climate. Such strategies, and whether they are practical, is outside the domain of this report, but they may be expensive and would only address specific problems at a local level.



These strategies are not mutually exclusive, and likely some combination of approaches will be taken. These choices are set against a backdrop where global energy consumption continues to grow. Increasing energy availability could enable a wider global population, including those currently without substantial access to energy resources, to enjoy the benefits energy can provide. In this context, keeping $CO_2$ levels below 500 ppm, for example, while increasing the equity of energy use would likely require both strategies A and B to be vigorously implemented.

A principal goal of this report is to provide guidance on the challenges and realistic expectations of strategy B, CDR. The report aims to inform sensible policy decisions regarding balancing the extent to which approaches A-D are taken, given the costs, challenges, and impacts of each as understood in the framework of current scientific and technical knowledge, and in a global context with many concurrent challenges — international conflicts, a large and still growing human population, the need for increased energy and food security for substantial parts of that population, and critically endangered species and ecosystems around the planet.

The conclusions summarized in this section are broken into guidance for policymakers (§7.2) and guidance for funding experimental or pilot projects for different CDR approaches and technologies (§7.3). Because this report starts from a fundamental physics perspective, we focus primarily on questions related to this general domain; however, it is important to emphasize the broad and interdisciplinary nature of CDR research and technologies. Developing effective and economical approaches to large-scale CDR will likely require extensive research and development efforts involving physics, chemistry, biology, materials science, earth sciences, agricultural and ecological sciences, and other technical areas. It will also require work on the economic and societal frameworks for the large-scale transitions that may be needed to implement these methods at scale.

Note that this report focuses on the fundamental principles underlying broad classes of CDR methods and does not attempt to pick winners and losers. Different CDR approaches have different features and issues, most are at an early stage of development, and it seems likely that a portfolio of different CDR methods will be most effective as a component of a comprehensive carbon management strategy. For other perspectives on the relative advantages of different approaches, see, for example, [4, 117]. Note also that the DOE Office of Fossil Energy and Carbon Management is currently engaged in efforts to support and facilitate research and development at various levels on the full range of carbon capture and storage methodologies discussed here [118], in the context of the DOE Carbon Negative Earth Shot [108]. These efforts are broadly compatible with the perspective and conclusions of this report in the context of the larger policy framework in which the United States and the international community seek to understand and balance strategies A-D.

We briefly summarize here types of CDR approaches that can potentially contribute to a large-scale global CDR effort. There is a key distinction here between cyclic processes where thermodynamics places a lower limit on the energy needed, and once-through processes, which may have smaller energy needs but greater material needs and environmental impact.

**Engineered cyclic CDR approaches,** in particular chemical direct air capture with carbon storage (DACCS) (§4.1), can in principle be scaled up to capture and sequester atmospheric $CO_2$ at the gigatons per year scale and have simple primary inputs (energy) and outputs (relatively pure $CO_2$ stream). The current challenge is to develop energy-efficient methods with system life cycles, material and water needs, and other requirements that are feasible and economical at the necessary scale. A key to scaling such systems is developing integrated carbon-free energy sources (e.g., solar thermal, PV, or nuclear power) so that these CDR systems do not take energy away from other uses. Bioenergy with carbon capture and sequestration (BECCS) is essentially a cyclic system where the energy input is from solar radiation. It has the co-benefit of producing some useful biomass energy. The low efficiency of photosynthesis in converting energy to

biomass, however, means that a lot of land would be required to implement BECCS on a large scale. This land use would compete with agriculture and biodiversity efforts. In addition, further energy inputs in the form of nitrogen-based fertilizer would generally be needed.

**Ecosystem-based biological CDR,** such as reforestation and other ecosystem restoration efforts, as well as changing agricultural and grazing practices to increase the retention of carbon in soil, may provide relatively low-cost opportunities for CDR at the gigatons of $CO_2$ per year scale. Potential side benefits include positive impacts on ecosystems and ecosystem services; however, total carbon capture is limited to a fixed amount for any given ecosystem. Furthermore, ecosystems are highly diverse, MRV is difficult, efforts must be tailored to local conditions, persistent storage is difficult to guarantee, and accurate predictions are challenging due to the complexity of ecosystems and human interactions.

**Once-through CDR approaches,** such as enhanced rock weathering (ERW) and ocean alkalinity enhancement (OAE) involve extracting and processing material that is out of equilibrium with the atmosphere (e.g., alkaline rock). These approaches thus avoid the fundamental energy constraints on cyclic processes given by the second law of thermodynamics; this energy has already been provided by natural processes. The effectiveness of these approaches, however, is still a subject of basic research, as is their potential impact on land and ocean ecosystems. The latter consideration is particularly complicated for once-through ocean CDR approaches, because ocean waters are generally considered part of the global commons.

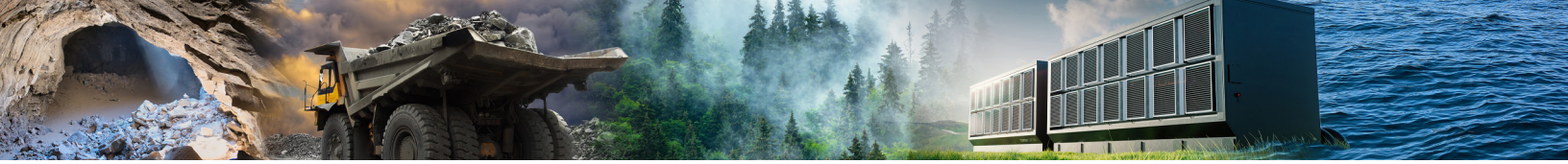
## 7.2 Science-based Conclusions and Guidance for Policymakers

A summary of recommendations for the different types of CDR approaches is given in Table 1.

- **Large-scale CDR requires substantial energy inputs and massive material infrastructure.** This is a primary message of this report. These facts are well-known to experts on CDR, and they are important to keep at the forefront of any discussion on incorporating large-scale CDR into global efforts to stabilize atmospheric carbon levels. In particular, the second law of thermodynamics gives an absolute lower bound to the energy needed to capture carbon with any cyclic system, such as chemical DAC or BECCS. [52] Similarly, basic chemistry constraints dictate that the mass of rock or other material needed for any once-through process is comparable to the mass of $CO_2$ captured. This means that there is no analogue of Moore's law for any approach to CDR. In other words, unlike the computing industry, which has benefited from an exponential growth in computational capacity for a fixed chip size or cost, there is no possible technological route in which the amount of carbon captured will scale exponentially in time for a fixed cost or fixed energy and material inputs. Note that similar considerations hold for any effort to replace fossil fuel power sources with zero-carbon power: any known energy source requires an amount of material infrastructure that is proportional to the rate of useful energy production. [53]

- **CDR at the Gt $CO_2$/yr scale may be desirable in the coming decades to meet specific climate goals, so research and development on a variety of CDR approaches is recommended despite their extensive energy and material needs.** Keeping atmospheric carbon dioxide below a specific level, such as 500 ppm, would almost certainly require deploying CDR at the scale of multiple gigatons of $CO_2$ per year, even with aggressive efforts to curtail emissions and/or allowing for somewhat higher atmospheric $CO_2$ levels. Cyclic systems currently in development require substantially more energy than the second law minimum. The *additional* power needed for such extensive atmospheric carbon removal using cyclic approaches such as chemical DAC would likely approach the terawatt scale (a noticeable

[52] For BECCS the energy requirement translates into a land area requirement.
[53] For nuclear power systems, however, the power source can be much more compact because nuclear interactions instead of chemical interactions are involved.



fraction of global electric power generation — 3 TW [15]). The material needed for once-through CDR approaches at this scale would require mining and processing rock at a scale comparable to global cement production. Investments in CDR research and deployment of trial/demonstration-scale projects can help enable the development of effective, economical systems and improve understanding of the full life-cycle energy and material needs — along with possible negative consequences — of each of these methods and technologies, in order to inform sound policy decisions. A portfolio of different approaches would likely be needed to achieve capture and sequestration at very large scales. These technologies are rapidly evolving and new ideas for CDR continue to emerge, so care should be taken not to prematurely commit to a specific approach that may soon be superseded.

- **Efforts to reduce carbon emissions should not be compromised by the possibility of future large-scale atmospheric carbon dioxide removal.** It is important to prioritize reducing emissions and converting to zero-carbon energy sources (i.e., strategy A) in all situations where the effective cost (monetary, societal, and environmental) is lower than the (appropriately weighted) cost of capturing the associated atmospheric carbon. Because CDR at large scales will be expensive and require extensive energy and/or material resources, the potential for implementing CDR systems should in no way replace or diminish efforts to decarbonize current energy systems and reduce the carbon losses in natural systems. [54]

- **Ecosystem-based CDR approaches ('natural climate solutions') should be pursued where feasible and effective in the near term** while developing other larger-scale, longer-term CDR approaches. As discussed in §4.2.1, ecosystem-based approaches are estimated to have the potential to remove several gigatons of $CO_2$ per year for several decades at costs well below \$100/t $CO_2$ without compromising other important human priorities such as food and water security. These approaches have additional benefits such as generating clean water and air and other ecosystem services, and helping to preserve biodiversity. Quantifying the effects of many natural CDR approaches presents an outstanding challenge, however. Additional challenges are that restoring lost soil and biomass carbon at the gigaton scale requires site-specific changes in land use practices over large areas and that it is difficult to ensure that carbon is retained over long times.

- **Any substantial development of CDR systems, particularly cyclic systems such as DAC, should be done in combination with dedicated low-carbon or carbon-free energy sources.** Large-scale CDR efforts would require additional power sources beyond those needed for other existing and growing world energy needs. This additional energy supply should be based on low-carbon or carbon-neutral power sources, because powering CDR systems with carbon-intensive energy sources dramatically decreases their effectiveness. While much of the energy for certain CDR systems, such as chemical DAC systems, can come from low-temperature waste heat, the usable fraction of this energy is limited by the laws of thermodynamics. For any large-scale direct air capture system, the source of energy is thus a central consideration.

- **Economic and policy frameworks should be put into place to balance emissions with removal costs.** This report has avoided detailed analysis of monetary costs and related economic issues. At the present time there is no clear U.S. domestic or international framework for paying for large-scale CDR. The essential purpose of CDR, however, is simply to remove carbon dioxide from Earth's atmosphere. [55] This suggests that to maintain atmospheric carbon at or below any fixed level, the cost of capturing and sequestering a ton of $CO_2$ (with appropriate adjustments for timing and other external factors) should

[54] Concerns that the investigation or implementation of CDR approaches may be used to justify continuing to emit $CO_2$ from fossil sources is often framed as a 'moral hazard' of CDR.
[55] However, some approaches such as ecosystem-based CDR, BECCS, and, e.g., use of carbon in construction materials may provide auxiliary benefits.

eventually determine the effective cost of emitting a ton of $CO_2$ through some rational economic policy that balances the incentives for CDR and emission reduction. While current costs are much higher, some projections anticipate that the cost of direct air carbon capture systems may come down to the range of \$100-\$200/t $CO_2$ in the foreseeable future. This translates, for example, into a cost of roughly \$1-\$2 to capture the amount of $CO_2$ that would be emitted by burning a gallon of gasoline. Specific policy recommendations based on these observations are outside the scope of this report.

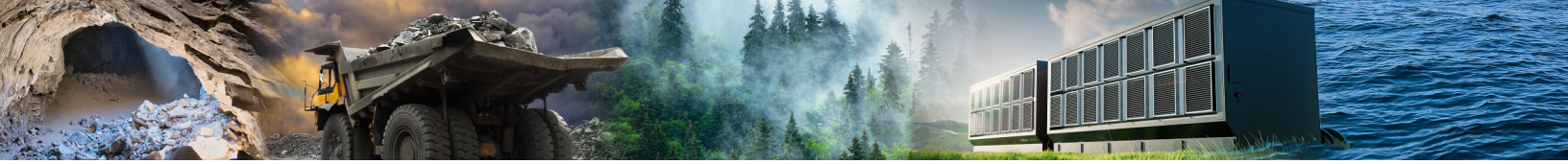
- **It is important to develop reliable systems of measurement, reporting, and verification (MRV)** to quantify the effectiveness of CDR systems and confirm that they are reliably capturing and permanently sequestering atmospheric $CO_2$. This is particularly important for market transparency, given the broad and varied set of potential CDR approaches and technologies. Currently, MRV methodologies are particularly weak for many once-through approaches (e.g., enhanced weathering, OAE) and ecosystem-based approaches (e.g., reforestation/afforestation). It may be difficult to incentivize a rigorous and effective MRV system with purely voluntary offsets. If CDR is implemented at large scale, governments will need to regulate emissions in some way, which would allow the market to manage the problem, or governments will need to foot the bill for much of the CDR deployment (as advocated in, e.g., [13]). In either case, policies should be considered that balance the role of government with market efficiency and promote efforts that are equitable internationally. Developing a system of government-approved standards that can be applied across the range of CDR methodologies may be helpful in any scenario where large-scale CDR is deployed.

**Table 1:** Summary of issues and recommendations for different types of CDR.

| Type of CDR | Primary Physical Constraint | Other Potential issues | Recommendation |
|---|---|---|---|
| **Engineered Cyclic (e.g. Chemical DAC)** | Energy | Cost, scale | Pursue research, development and pilot projects to develop cost-effective and scalable approaches |
| **Once-through Approaches (e.g., ERW, OAE)** | Materials | Uncertain effectiveness, environmental impact | Pursue further research on impact and effectiveness before considering large-scale deployment |
| **Ecosystem-based Approaches (e.g., reforestation)** | Energy → Land | Limited capacity, verifying effectiveness and longevity | Pursue further research on MRV; deploy where effective and economical, particularly where there are co-benefits for biodiversity, conservation, and ecosystem services |

## 7.3 Science-based Guidance for Funding Pilot Programs

This section is aimed at funding agencies and investors who are considering proposals for pilot programs and/or demonstration plants implementing potentially scalable technologies for carbon capture. We provide a set of key questions that should be considered in evaluating the possible challenges facing any CDR program or implementation, including both cyclic and one-through approaches.



*How much energy does the underlying set of physical processes require?*

*What is the estimated actual energy cost for the full system?*

*Where will the energy come from?*

*What carbon output will be associated with producing this energy?*

*If low-temperature thermal energy is used, are expectations for energy needs compatible with second-law bounds and underlying energy requirements?*

*What are the material needs?*

*Is there a realistic framework for the integration of all parts of the system, from the material needs in production to deployment to the sequestration of captured carbon?*

*What are the plans for scaling up from a pilot program to tens or hundreds of Mt $CO_2$/yr?*

*What MRV approaches are available and how reliable are they?*

*What are the risks and potential impacts on land and oceans and how well are they understood?*

For cyclic systems, questions regarding energy inputs are particularly relevant, although material needs, impact, and scaling issues should also be considered.

For once-through systems, questions regarding material needs, verification, effectiveness, longevity, and environmental impact of the output stream are particularly important; energy needs and scaling issues should also be considered.

## 8. List of Experts Consulted for This Report

The study group would like to thank the individuals consulted in the course of writing this report.

We are much indebted to all of these experts for sharing their insights on the science and technology related to carbon capture: Peter Agbo (LBNL), Isabel Barton (U. Arizona), Daniel Bodansky (Arizona State), Bill Casey (UC Davis), Pamela Chu (NIST), William Collins (LBNL), Don DePaolo (UC Berkeley), David Des Marais (MIT), Michael Follows (MIT), Baylor Fox-Kemper (Brown), Inez Fung (UC Berkeley), Brad Hager (MIT), Tim Herbert (Brown), Greg Hirth (Brown), Dan Ibarra (Brown), David Keith (U. Chicago), Peter Kelemen (Columbia), Jeff Long (UC Berkeley), Noah McQueen (Heirloom, Inc), Steve Pacala (Princeton), Dan Rothman (MIT), Robert Socolow (Princeton), Mike Tamor (Arizona State), Cesar Terrer (MIT).

# Technical Appendices

## A. Quantifying Carbon and Carbon Dioxide

In this appendix we include a few relevant notes regarding carbon and carbon dioxide quantities and concentration.

In some situations and in some references, carbon dioxide is quantified by the mass contained in $CO_2$ molecules, while in other places only the mass contained in the carbon atoms is included; atmospheric carbon dioxide levels are also frequently indicated by a concentration in parts per million (ppm) by volume, while sometimes the concentration is given in ppm by mass.

The conversion factor between the mass of $CO_2$ and the mass of the carbon contained therein is given by the mass fraction ~ 12/44 of carbon in $CO_2$, so we can approximately relate

$$1\,\text{kg C} \leftrightarrow 3.67\,\text{kg CO}_2\,. \tag{2}$$

The mass density of air (at sea level) is approximately 1.275 kg/m$^3$. At 420 ppm by volume with a molecular weight of approximately 44 for $CO_2$ and 29 for the atmosphere, the carbon concentration is 640 ppm by mass, so this gives an atmospheric mass density of carbon dioxide of 816 kg $CO_2$/(10$^6$ m$^3$).

## B. Second Law of Thermodynamics

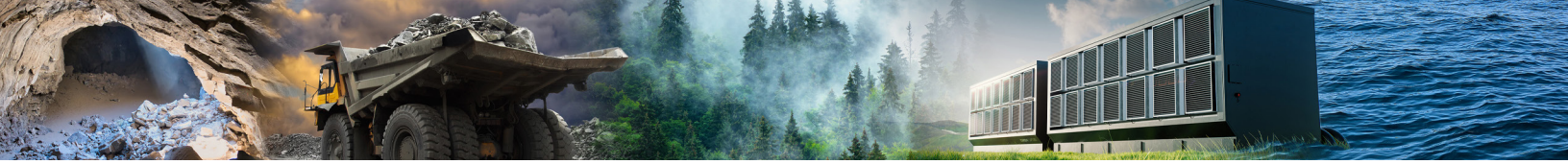
Here, we briefly review the essence of the second law of thermodynamics. [56] A more thorough introduction to these ideas can be found in [119]. Note that the description here is simplified in various ways, omitting details that are not relevant for the issues at hand.

The notion of entropy is fundamental to thermodynamics. Entropy is a measure of the randomness of a state, i.e., the extent to which the detailed positions and velocities of particles (or the quantum equivalents) are not fixed by the known thermodynamic quantities. For example, take a gas that is initially at a fixed temperature and pressure in a partition of size $V$ within a box of size $2V$, separated by a wall from a vacuum in the remaining volume $V$. If we suddenly remove the wall so that the gas expands freely to fill the rest of the box, after the system comes to equilibrium the gas molecules will be uniformly distributed throughout the larger volume. Their average kinetic energy does not change, but there is essentially one extra bit (0/1) of information for each molecule in the random state of the gas, corresponding to which half of the box that molecule is in after the gas has freely expanded. This leads to an entropy increase of $\Delta S = k_B \ln(2)$ for each of the $N$ molecules of the gas, where $k_B \equiv 1.381 \times 10^{-23}$ J/$K$ is the Boltzmann constant. [57]

The essence of the second law of thermodynamics is that the amount of entropy in a given closed system, or in a system along with the environment to which is coupled, cannot decrease. This fundamental principle essentially follows from the reversibility of the laws of physics. If a system has 100 possible unknown states, and one performs a fixed series of reversible manipulations on the system without considering the state of the system, then the system will still have at least 100 possible unknown states at the end of the process. Thus, the second law of thermodynamics states that for any closed system, $\Delta S \geq 0$.

[56] The first law of thermodynamics simply refers to conservation of energy: the fact that in any closed system the amount of energy does not change with time, although it can change in form.

[57] The factor of ln(2) arises because when entropy was first discovered, it was standard to use natural logarithms rather than measuring information in units of bits. Note that this direct information-theoretic description of the entropy change for free expansion is possible because no thermal energy is transferred and no work is done; for more details see [119], § 8.4.2, 8.4.4.



In general, in many important energy systems (such as a car engine or direct air capture device), the desired output of the system (mechanical energy or captured carbon) has less entropy than the initial state. In such a situation, entropy must be jettisoned to the environment. This is done by expelling energy to the environment, which is at an ambient temperature $T$. The minimum amount of energy that must go to the environment along with a transfer of entropy $\Delta S$ is $\Delta E \geq T\Delta S$. This relation is so fundamental that it is in many cases used to define temperature in terms of energy and entropy, and also underlies the principle that there is a minimum energy cost to decreasing the entropy of a system.

In this report, entropy and the second law play a fundamental role in governing several aspects of direct air capture systems.

**Entropy of mixing**

When molecules of two distinct substances are fully mixed, as carbon dioxide is mixed into Earth's atmosphere, there is an entropy associated with this mixing. For example, if we have 100 red marbles and 100 blue marbles mixed together in a bag, there is an entropy of mixing associated with the number of random ways in which the 200 marbles are distributed into red and blue. When the total number of molecules is large, the entropy of mixing associated with $N_A$ molecules of a substance A mixed into a larger system with a small concentration fraction c « 1 is just

$$S \cong N_A k_B \ln 1/c\,. \tag{3}$$

This can be understood heuristically in terms of the above discussion by noting that, for example, if the concentration $c$ is $1/K$ for some large number $K$, this means that $1/K$ of the molecules is a molecule of substance $A$, and the number of bits of information needed to identify each such molecule is roughly $\log_2 K = \ln K/\ln 2$, and multiplying $N_A \times (\ln K/\ln 2) \times k_B \ln 2$ gives precisely (3). The formula (3) for the entropy of mixing is used in the next section to compute the energy needed to 'unmix' $N$ molecules of $CO_2$ from the atmosphere at a concentration $c = 420/10^6 = 420$ ppm. [58]

**Thermal energy and usable energy**

Many energy systems, from power plants to car engines, rely on the conversion of thermal energy to mechanical energy. Because of the second law of thermodynamics, there is a fundamental limit to the fraction of thermal energy that can be used to produce mechanical energy or any other effect that is essentially entropy-free. Basically, because thermal energy carries entropy, to convert thermal energy into something like mechanical energy, the excess entropy in the original thermal energy must be jettisoned to the environment along with some of the energy. Therefore, not all of the energy can be used for the intended purpose. Known as the Carnot limit, this fundamental physical limit states that the maximum efficiency $\eta = E_{\text{useful}}/E_{\text{used}}$ is capped at

$$\eta \leq \frac{T_+ - T_{\text{env}}}{T_+} = 1 - \frac{T_{\text{env}}}{T_+}\,, \tag{4}$$

where $T_+$ is the temperature of the thermal energy resource being used, and $T_{env}$ is the energy of the environment. One convenient way to think about this limit is captured by the notion of exergy, or 'usable energy' associated with thermal energy. Given a small portion [59] $\Delta E_{\text{thermal}}$ of the thermal energy in a fluid at temperature $T_+$, the fraction of this portion of energy that can be used for processes without residual entropy is thus at most $\Delta B = (1 - \frac{T_{\text{env}}}{T_+})\Delta E_{\text{thermal}}$. The thermal energy used in processes that sepa-

[58] Note that the formula (3) also gives the exact entropy change for separating $N_A$ molecules of species $A$ from an atmosphere at concentration $c$, even when the concentration factor is not small, in the ideal gas approximation and in the limit of an infinite atmosphere.

rate carbon dioxide from the atmosphere is subject to this same bound relative to the energy requirement associated with the entropy of mixing. Note that the bound stated on available energy relies on a continuous supply of thermal energy at the high temperature; for a finite volume of material at a higher temperature, the exergy is actually lower because the fraction of thermal energy that can be extracted drops steadily as the material cools.

## C. Energy Cost of Carbon Capture From Atmosphere: Computation

In this appendix we compute the minimal energy cost for separating carbon dioxide from the atmosphere.

Using the formula (3) for the entropy of mixing, and recalling the basic mathematical relation that $\ln 1/c_1 - \ln 1/c_2 = \ln c_2/c_1$, we see that the decrease in entropy of $N$ molecules of carbon dioxide as they are concentrated from $c_1$ to $c_2$ is given by [60]

$$\Delta S \cong N k_B \ln\left(\frac{c_2}{c_1}\right). \tag{5}$$

(The Boltzmann constant $k_B$ is defined in App. B.) At an ambient temperature $T$, the minimum energy required to separate out the $CO_2$ in this way is

$$\Delta E \cong T\Delta S, \tag{6}$$

because the decrease in entropy of the $CO_2$ must be made up for by increasing the entropy of the environment at temperature $T$ by at least this much.

For 1 kg of $CO_2$, we can compute that the number of molecules is

$$N \cong 1\ \text{kg}/44\text{u} \cong 1.4 \times 10^{25}, \tag{7}$$

where $\text{u} \cong 1.66 \times 10^{-27}$ kg is the atomic mass unit, and the molar mass of $CO_2$ is 44.01 u. The absolute minimum energy cost to extract 1 kg of $CO_2$ from an atmospheric concentration of 420 ppmv (parts per million by volume, elsewhere abbreviated ppm) at an ambient temperature $T \cong 300\ K$ ($\cong 26.85\ ^\circ\text{C}$), with a final concentration of $c = 1$ (pure $CO_2$) is then given by

$$\Delta E = T\Delta S \cong (300\text{K}) N k_B \ln\left(\frac{10^6\ \text{ppm}}{420\ \text{ppm}}\right) \cong 441\ \text{kJ}. \tag{8}$$

Because of the logarithm in this equation, the energy cost reduces only slowly as atmospheric $CO_2$ levels rise. At a concentration of 500 ppm, the energy cost is reduced by only a few percent, to 431 kJ.

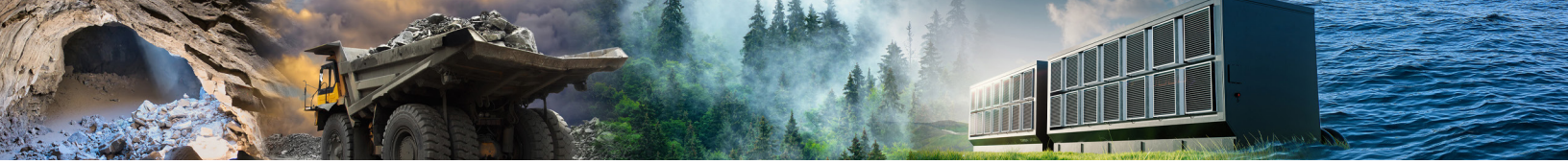
We use the round number 440 kJ elsewhere in this report. This is easily converted into kWh using 1 kWh = 3.6 MJ, so 441 kJ/kg ≅ 122.4 kWh/t. In chemistry units, one mole ($6.022 \times 10^{23}$ molecules) of $CO_2$ weighs 44.01 g, so the energy cost is 19.39 kJ/mol, approximated elsewhere in the report as 20 kJ/mol.

Using this estimate, the steady state power needed to capture 1 Gt of $CO_2$ over a year is

$$P \cong 10^{12}(\text{kg/Gt}) \times 441\ \text{kJ/kg}/(3.15 \times 10^7\ \text{s/yr}) \cong 14\ \text{GW}/(\text{Gt/yr}). \tag{9}$$

[59] Technical note: We need to consider a small portion $\Delta E$ of all the energy because once we use some of the energy, the temperature goes down.

[60] This formula gives the change in the full entropy of mixing for a finite size system in the ideal gas approximation when the concentrations $c_1, c_2$ are small. To completely separate out a pure stream of $CO_2$, we take $c_2 = 1$, and the formula gives an exact measure of the entropy change needed in the assumption of an infinite atmosphere. Note that when $c_1$ is not small, and the separation is done in a finite volume, the entropy change is slightly larger. For atmospheric concentrations $c_1 \cong 0.0004$, corrections to this formula are negligible.



If the final concentration is lower than $c_2 = 1$, the entropy difference (5) is reduced, and the minimum energy cost is similarly decreased. The implications of this for carbon capture systems are discussed briefly in §3.1.

Note that the calculations in this appendix do not involve any unknown or uncertain quantities; instead, they rely only on basic physical principles that have been well understood since early in the last century.

## D. Energy and Volume of Air Movement Needs for Cyclic CDR

In this appendix we briefly describe the energy and volume requirements for moving air through a large-scale cyclic CDR system.

Some energy is needed to move air through any direct air capture system. Unlike in the preceding appendix, physics gives no fundamental lower bound on the energy needed for this. If the air moves through the system extremely slowly, very little energy is required, but then the system will not process $CO_2$ very quickly and needs to be larger for a given capture rate. Thus, there is a trade-off between energy used for the fans and the size of the system.

Let us consider as an example a system with a flow rate of $v = 2$ m/s. [61] From App. A, we recall that the concentration of $CO_2$ by mass is 640 ppm. Thus, the mass of atmosphere that must be moved through the system to capture 1 kg $CO_2$ at 100% efficiency is $M \cong 10^6/640$ kg $\cong$ 1560 kg. The kinetic energy of this mass of air would be

$$Mv^2 \cong (1560\ \mathrm{kg}) \times (2\ \mathrm{m/s})^2 \cong 6\ \mathrm{kJ}\,. \tag{10}$$

This is less than 2% of the energy needed for the unmixing process (8). Because this energy scales quadratically with the velocity, however, at much higher airflow speeds like 10 m/s, the energy requirement for moving the air would become a significant fraction of the thermodynamic minimum for the carbon capture process.

For a simple estimate of the volume of air involved in large-scale carbon capture systems, note that 1 t of $CO_2$ represents about $1.2 \times 10^6$ m³ of atmosphere. Thus, to capture 1 Gt $CO_2$, even using a 100% effective removal process, over $10^{15}$ m³ of air would need to be moved through the system. To move this much air per year through a system at 2 m/s the intake area would need to be roughly 20,000,000 m², requiring, for example, an intake height of 20 m and a linear horizontal intake length of around 1,000 km. To get another perspective on the scale involved, we can compare with the amount of air moved globally in air conditioning systems. In rough numbers, 1 ton of air conditioning in constant operation draws roughly 3.5 kW, and involves an airflow typically on the order of 600 m³/h. As a result, 1 kWh of AC involves moving roughly 200 m³ of air. Roughly 10% of electrical capacity worldwide, or 2,800 TWh, goes to residential and commercial cooling [120]. Finally, this means that if we outfitted all cooling units in the world with devices enabling complete capture of all carbon in all the air flowing through them, we would get less than 1 Gt $CO_2$/yr of carbon capture. [62]

## E. Energy and Land Area for Biological Carbon Capture

Virtually all life on Earth depends directly or indirectly on the energy and carbon captured by photosyn-

[61] We assume here a continuous flow rate; some DAC approaches such as solid sorbent systems (App. J) turn the fan off for parts of the cycle; in this case the same net flow rate can be realized at the cost of a slightly higher v when the air is flowing.
[62] Note that this is a very rough back of the envelope estimate; cooling systems include not only AC but also 'swamp fans' and other systems, and the air movement through different cooling systems varies fairly widely, this is just intended to give a very rough sense of the scale of effort involved.

thesis. Although this process evolved over many millions of years to be as efficient as possible for the organisms involved, there are both fundamental and practical limits to the rate at which any given biological system in a fixed area can use photosynthesis to produce biomass storing carbon. A simple estimate of the scale of effort needed to capture gigatons of $CO_2$ in biomass comes from considering the carbon content of typical agricultural crops such as grains, which is roughly 40% of dry weight [121]. Because carbon mass ratio for $CO_2$ is $12/44 \cong 27\%$, roughly speaking 1 Gt of carbon dioxide corresponds to the carbon in 2/3 of a Gt of biomass. Thus, capturing 1 Gt of $CO_2$ in biological systems will require a comparable effort that needed to grow 2/3 of a billion tons of crops.

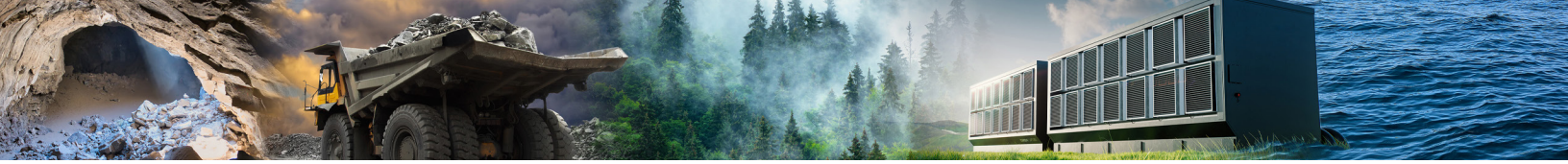
Considering the energy aspect of these processes, available solar energy (insolation) is abundant in many locations. Thus, while the energy bound from the second law of thermodynamics plays some role in limiting biological carbon capture, there are two further factors that limit the effectiveness of biological carbon capture. One fundamental issue is that in terms of the conversion of incident solar energy to energy in available excess biomass (i.e., not used in metabolism), the efficiency of photosynthesis is rather low. In this sense, the photosynthetic efficiency of most crops is generally below 1% and often closer to 0.25-0.5% [122]. Beyond this, the biomass energy associated with a given amount of carbon is significantly above the thermodynamic bound on energy needed for capturing the carbon. Taken together, these factors make biological carbon capture a very land-intensive process in any scenario using conventional terrestrial crops.

To get a sense of the order of magnitude, if we assume a photosynthesis energy efficiency of 0.25%, and a typical average irradiance of 200 W/m², the rate of carbon-containing biomass production [63] will be roughly 1 kg $C_6H_{12}O_6$/m² per year. This corresponds to a capture rate of roughly 1.5 kt $CO_2$/yr over an area of 1 km². At this rate, the area needed for a capture of 1 Gt $CO_2$/yr would be roughly 0.7 million km², or 0.35 million km² at a photosynthetic efficiency of 0.5%. This can be compared to the area of all farmland currently in the United States, roughly 3.6 million km².

The above estimates are based on a photosynthetic efficiency of 0.25-0.5%, which is comparable to that realized for many crops with current intensive agriculture methods. It is certainly possible, however, that with the right organism and an engineered environment, much higher efficiencies and capture rates may be possible. One recent estimate suggests that one gigaton of $CO_2$ per year could be captured on 0.25 million km² using high-productivity crops such as switchgrass [29]. For certain species of microalgae, in controlled environments such as suspended or attached growth reactors, a net photosynthetic efficiency of 3-6% can be achieved [122]. At this rate, the area needed to capture 1 Gt $CO_2$/yr would be roughly 30,000-60,000 km². Such systems would have large water requirements, but may be a useful way of implementing CDR at a significant scale, particularly in conjunction with BECCS systems. Note that even these numbers are still larger by an order of magnitude than the areas needed to supply solar energy to something like a chemical DAC system operating at three times the thermodynamic limit (§6.5).

One way to think about these numbers is that, roughly speaking, the lower energy efficiency of photosynthesis of typical crops compared to, e.g., a solar field, increases the area needed by one order of magnitude from energy considerations alone (see, e.g., [123]). The biomass energy per kilogram (~ 15 MJ/kg) is also much larger than the thermodynamic energy bound for capture (~ 0.44 MJ/kg), increasing the land area needed for biological capture by another order of magnitude. Thus, the land area and associated solar energy needed for atmospheric carbon capture through photosynthesis by typical crops is roughly two orders of magnitude greater than what would be needed for, e.g., a solar field to power a CDR plant operating at three times the thermodynamic lower bound described in §3.1.

[63] For this calculation we assume the carbon and energy content of the biomass is in the form of simple sugars, e.g., glucose, $C_6H_{12}O_6$, with an energy density of 15.5 MJ/kg; the numbers for starches or similar compounds in other biomass are not too different.



## F. Basic Chemical Reactions

In this appendix we review some basic chemistry relevant for ERW (§4.3) and chemical DAC (§4.1). The following appendix goes further into the chemistry relevant for ocean CDR (§4.4). It is instructive to first consider the chemistry of carbon dioxide dissolved in pure water without the complicating effects of other ions. (The role of other ions is discussed later in App. G.) The equilibrium concentration of aqueous carbon dioxide, $CO_2{}^{aq}$, in otherwise pure water, is given by Henry's law:

$$[\mathrm{CO_2^{aq}}] = p(\mathrm{CO_2})\, K_H \tag{11}$$

where $K_H = 3.4 \times 10^{-2}\ \frac{\text{moles/liter}}{\text{bar}}$ at 25°C and $p(CO_2)$ is the partial pressure of gaseous $CO_2$ in bars (1 bar is nearly the mean atmospheric pressure at sea level). The notation [X] indicates the concentration of molecules of X in a solution (here in moles/liter).

Aqueous $CO_2$ partially reacts with water to yield carbonic acid ($H_2CO_3$), and we introduce the notation $CO_2$* to refer to a mixture of aqueous $CO_2{}^{aq}$ and $H_2CO_3$. Carbonic acid then reaches an equilibrium with bicarbonate ions $HCO_3{}^-$ (note that 'bicarbonate' is a misnomer and is used for historical reasons; more properly it is called 'hydrogencarbonate'):

$$\mathrm{H_2CO_3} \leftrightarrow \mathrm{HCO_3^-} + \mathrm{H^+}\ . \tag{12}$$

Finally, the bicarbonate ion partially dissociates into carbonate ions and protons:

$$\mathrm{HCO_3^-} \leftrightarrow \mathrm{CO_3^{2-}} + \mathrm{H^+}\ . \tag{13}$$

Equations 12 and 13 each have associated equilibrium constants. Each proton liberated by the forward reactions increases the acidity of the water as quantified by a decrease in the pH:

$$\mathrm{pH} \equiv -\log_{10}([H^+])\ . \tag{14}$$

The addition of hydrogen ions (e.g., by adding acid) forces the reactions to the left, eventually reducing the concentrations of carbonate and bicarbonate ions in favor of aqueous carbon dioxide (see Fig. 6). The concentration of total dissolved inorganic carbon (DIC) is given by

$$[\mathrm{DIC}] = [\mathrm{CO_2^*}] + [\mathrm{HCO_3^-}] + [\mathrm{CO_3^{2-}}] = [CO_2^{aq}] + [\mathrm{H_2CO_3}] + [\mathrm{HCO_3^-}] + [\mathrm{CO_3^{2-}}]\ . \tag{15}$$

Silicate minerals such as forsterite ($Mg_2SiO_4$), when dissolved in water, can react with rainwater to form magnesium ions, bicarbonate ions, and silicic acid:

$$\mathrm{Mg_2SiO_4} + 4\mathrm{CO_2} + 4\mathrm{H_2O} \rightarrow 2\mathrm{Mg^{2+}} + 4\mathrm{HCO_3^-} + \mathrm{H_4SiO_4}\ . \tag{16}$$

Thus each magnesium atom can remove two atoms of carbon, in contrast to the solid reaction of Eq. 1 where they are in a 1:1 ratio. If, however, the water is then removed by evaporation, the magnesium ions precipitate out as magnesium carbonate (magnesite) but at the cost of releasing $CO_2$ per Eq. 1. Similar reactions involving calcium ions result in calcium carbonate (calcite). Formations of ultramafic rocks interpenetrated with calcite veins show that such reactions occur in nature and participate in the carbon cycle on geologic timescales. Carbon dioxide injected into mafic basalt formations can also solidify to form carbonates, thus immobilizing the carbon.

Perhaps the simplest reactions for chemical DAC, such as the one employed by Heirloom Carbon Technologies, are the exothermic combination of solid calcium oxide ('quicklime') or magnesium oxide with carbon dioxide to form calcite or magnesite [30]:

$$\begin{aligned} \mathrm{MgO} + \mathrm{CO_2} &\rightarrow \mathrm{MgCO_3} + 76\mathrm{kJ/mole} \\ \mathrm{CaO} + \mathrm{CO_2} &\rightarrow \mathrm{CaCO_3} + 141\mathrm{kJ/mole}\ . \end{aligned} \tag{17}$$

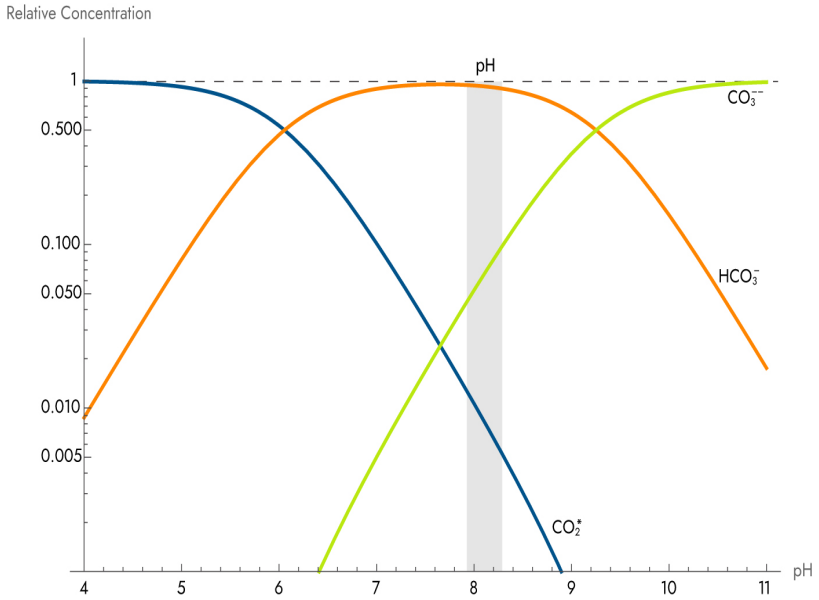
Spreading MgO over large areas has been proposed as a way to absorb $CO_2$; the resulting $MgCO_3$ could then be calcinated, regenerating the MgO and releasing a concentrated stream of carbon dioxide that could be stored [124].

## G. Ocean Chemistry

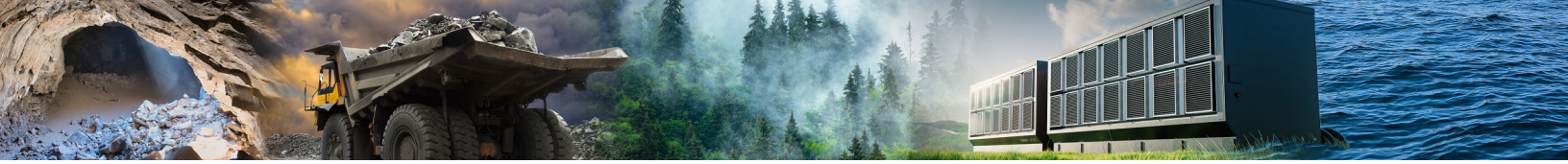
We review here some basic aspects of ocean chemistry relevant for §4.4. More detailed treatments of these topics can be found in, for example, [125, 126, 127].

As described in App. F, the ratios between concentrations (typically measured in moles/liter or μmol $kg^{-1}$) of dissolved $CO_2$ ($[CO_2^*] = [CO_2^{aq}] + [H_2CO_3]$), bicarbonate ($[HCO_3^-]$), and carbonate ($[CO_3^{2-}]$), as well as hydrogen ions ($[H^+]$), are determined by chemical equilibrium equations with equilibrium coefficients that vary with temperature, salinity, and pressure. For typical conditions near the ocean's surface, where the system is roughly in equilibrium with the atmosphere, the concentrations of the DIC (dissolved inorganic carbon) constituents [CO2*], $[HCO_3^-]$, and $[CO_3^{2-}]$ are roughly in the ratios 1:90:9, but vary strongly with changes in pH; see Fig. 6. [64] The pH of ocean surface waters measured in various locations has decreased from around 8.1 to 8.05 over the last 30 years; while this change appears numerically small, due to the logarithmic definition of pH, this corresponds to an increase in hydrogen ion concentration of ~ 20% to ~ 35%, depending upon the specific location sampled [128, 129]. With a concentration of DIC in surface waters around 2 mmol $kg^{-1}$, one cubic meter of surface seawater (≅ 1,020 kg) contains roughly 24 g of carbon, or 88 g of $CO_2$ equivalent; this is roughly 100 times the volumetric carbon density of Earth's current atmosphere.

**Figure 6:** Bjerrum plot of the concentrations of different constituents of the DIC as a function of pH for typical surface temperature and salinity (after Wikipedia article: carbonic acid). The gray region indicates recent pH range in surface waters; the recent trend is downward (more acidic).




[64] In the discussion here we neglect ion-pairing effects in describing DIC for clarity; ion pairing complicates the speciation of seawater, but does not have a significant impact on the conclusions discussed here.



The total alkalinity $A_T$ of a solution describes the sensitivity of its pH to the addition of protons. For seawater, total alkalinity can be defined as the (charge-weighted) difference between the sum over conservative positive ions (cations) and the sum over conservative negative ions (anions), where conservative ions are those relatively unaffected by variation in pH, temperature, or pressure. Thus, for a net charge-neutral solution,

$$A_T = [\mathrm{Na}^+] + 2[\mathrm{Ca}^{2+}] - [\mathrm{Cl}^-] + \cdots = [\mathrm{HCO}_3^-] + 2[\mathrm{CO}_3^{2-}] + [\mathrm{B(OH)}_4^-] + [\mathrm{OH}^-] - [\mathrm{H}^+] + \cdots , \quad (18)$$

where the right-hand side contains all the non-conservative ions. Many aspects of ocean carbon systems are most conveniently understood in terms of the variation of DIC and $A_T$ (rather than pH) [125]. In fact, the carbonate system acts as a kind of buffer moderating changes to the pH; the addition of an acid or alkaline substance to seawater leads to a change in the relative balance within the carbonate system that moderates the expected change in pH. Increased DIC due to anthropogenic $CO_2$ increases leaves $A_T$ unchanged, because all the associated charged ions appear on the right-hand side of Eq. (18) and cancel, although it decreases the pH. In contrast, dissolution in seawater of, e.g., olivine into $2Mg_2^{++}$+ $SiO_4^{4-}$ increases $A_T$, because the magnesium ions are conservative while the $SiO_4^{4-}$ are not.

An important process in ocean carbon chemistry is the precipitation of carbonates such as calcium carbonate ($CaCO_3$). Precipitation of 1 mol of $CaCO_3$ per liter decreases $A_T$ by two units, and decreases DIC by one unit. While this removes carbon from the system, it also lowers the pH and pushes the carbonate system toward a higher fraction of [$CO_2$*]; in surface waters this pushes $CO_2$ out of the ocean and back into the atmosphere. In many places the surface ocean is supersaturated with calcium and carbonate ions, meaning that the concentrations present are greater than the equilibrium concentration in the presence of solid calcium carbonate (which has several forms). Calcium carbonate that forms through precipitation in the upper ocean sinks downward; below a depth known as the 'saturation horizon,' it begins to dissolve again and is completely dissolved below the 'carbonate compensation depth.' Some of the calcium carbonate accumulates into sediments on the ocean floor above the carbonate compensation depth; some of these sediments are buried deeply and leave the ocean-atmosphere carbon cycle.

## H. Global Carbon Budget and Carbon in the Terrestrial Biosphere

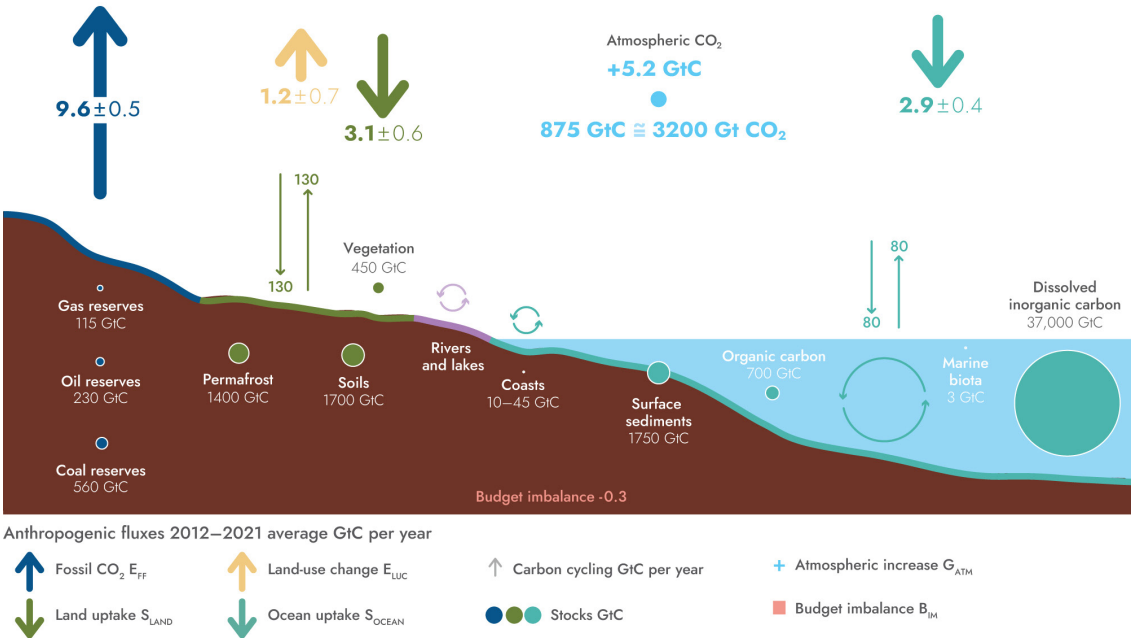
A comprehensive review of the global carbon budget is given in [130]; best estimates of carbon stores in the ocean, atmosphere, soil and biomass, etc., and the net fluxes for the decade 2012-2021 are shown in Fig. 7. Similar estimates appear in [131].

The net increase in atmospheric $CO_2$ from 2021-2022 (see Fig. 1) measured by NOAA [11] was roughly 2.1 ppm/yr, representing roughly a 0.5%/yr increase in the current level of 420 ppm, or an increase of roughly 4.4 Gt C/yr ≅ 16 Gt $CO_2$/yr; slightly larger estimates from [130] give ≅ 19 Gt $CO_2$/yr.

Of the 9-10 Gt of carbon added to the atmosphere yearly from fossil sources, roughly 30% is absorbed by the ocean sink. Another 30% is currently taken up by land ecosystems through natural processes associated with increased $CO_2$ and warming ($CO_2$ fertilization effect, longer growing season), although human land use offsets some of this by releasing over 1 Gt of carbon from terrestrial systems back to the atmosphere. [65] Biomass in forests is estimated to give a net sink of roughly 1.1 Gt C/yr, including both natural drivers and human land use (deforestation, afforestation, etc.) [132]; grasslands and changes in soil carbon also contribute to the net land sink. There is, however, substantial uncertainty regarding the longevity of both the ocean and the terrestrial carbon sinks.

[65] The combined effect of land use, land-use change, and forestry is sometimes referred to as LULUCF. Roughly another 1,500 Gt of C is stored in frozen/permafrost soils [134, 130].

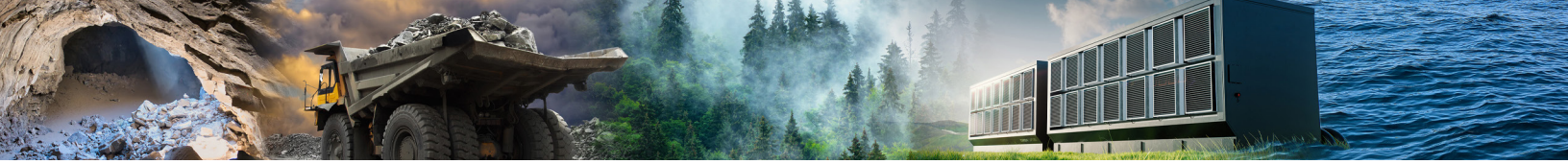
**Figure 7:** The global carbon cycle, from [130]: Anthropogenic changes to the global carbon cycle (mean annual fluxes for the decade 2012-2021) are illustrated by thick arrows at the top of the figure. Current carbon stores are at the bottom; the natural cycle is illustrated by thin background arrows. Note that changes in $CO_2$ are related to changes in C by a factor of 44/12 ≅ 3.67.




Considering the terrestrial part of the carbon budget in more detail, carbon circulates constantly between the atmosphere and the biosphere. Terrestrial biomass uptakes roughly 130 Gt C/yr from the atmosphere each year. Roughly half of this is returned to the atmosphere directly through plant respiration, and the other half through microbial respiration and decomposition of decaying organic matter. The annual cycle of spring growth and fall decay is apparent in the detailed atmospheric measurements of $CO_2$ in Fig. 1, dominated by seasonal change in the Northern Hemisphere. A small fraction of the carbon that circulates through terrestrial biomass is retained over a longer time in soil. The residence time of carbon in tropical soils is roughly 10 years or less, while in temperate soils there are organic carbon components with residence times closer to 100 and 1,000 years [133]. Total carbon in terrestrial ecosystems is roughly 3,000 Gt [66], most of which is contained in the soil (~ 1,500 Gt organic (SOC) [134] to a depth of 1 m, ~ 1,000 Gt inorganic, ~ 500 Gt biomass/vegetation). This is several times larger than the quantity of carbon in the atmosphere (~ 900 Gt C, equivalent to 3,200 Gt $CO_2$).

Human land use patterns significantly affect the total quantity of carbon stored in terrestrial biomass and soil. Current estimates suggest that the total carbon currently stored in terrestrial biomass is roughly 450 Gt while biomass carbon stocks of potential vegetation are over 900 Gt C. This means that human land use practices, primarily agriculture, grazing, and forest management, reduce the net carbon storage potential of biomass by a factor of roughly two [135]. Human activity has also broadly decreased the carbon content of soils globally. A recent estimate suggests a loss of 116 Gt of SOC (~ 400 Gt $CO_2$ equivalent) through agricultural land use (grazing + cropland) [136] since pre-agricultural times.

[66] Roughly another 1,500 Gt of C is stored in frozen/permafrost soils [134, 130].



## I. Ecosystem-based CDR: Details and Further References

A broad variety of ecosystem-based approaches to CDR exist. Ecosystem restoration can include simple land abandonment: allowing land that has been cleared for cultivation or other usage to regrow naturally; more active measures can also be used to remove artificial constructions such as dams or to restore lost species to enable natural processes to repair damaged ecosystems ('rewilding'). Regrowth of forests in the northeast United States in land previously used for agriculture over the last 150 years, for example, contributes to the current land sink (App. H). Reforestation and ecosystem restoration fit with other national and international goals such as the 30 × 30 Initiative [137, 138] to protect 30% of Earth's land and ocean areas by 2030. Other ecosystem-based CDR approaches involve more active changes in land management practices, such as modifying agricultural or grazing methodology to increase persistent carbon stores in biomass and/or soil. An overview of ecosystem-based approaches for carbon storage and avoided emissions is given in [26]. This appendix contains a few further details and references regarding these approaches.

**Carbon retention in soil.** It has been estimated that over the last 12,000 years, human activity has reduced the net quantity of soil organic carbon (SOC) and terrestrial biomass by hundreds of Gt C, with SOC losses from agricultural land use exceeding 100 Gt C (App. H). Some fraction of this could be restored by selective abandonment and rewilding, and another fraction could be regained by changing agricultural land use approaches, such as overgrazing and monoculture practices that degrade the soil. A recent second-order meta-analysis of current and future human and climate impacts on SOC is given in [107], with the overall conclusion that the impacts on SOC of land-use change in the coming decades will be much larger than the direct effects of climate change. Some estimates suggest that restoring broader biodiversity in grasslands and modified grazing approaches could lead to carbon sequestration at the rate of up to several gigatons per year [139]. There are also suggestions that modified 'no-till' agricultural practices, use of cover crops, and addition of biochar (§4.2.3) to soil can enhance the accumulation of organic carbon in soil (see, e.g., [140] for a meta-analysis), although the extent to which no-till agriculture can significantly impact soil carbon content has been disputed [141] and understanding of the longevity of the carbon in biochar in soil is still limited, with half-life estimates ranging from $10^2$ to $10^7$ years depending on composition [142].

A recent synthesis of peer-reviewed literature on SOC sequestration in agroecosystems estimates 1.45-3.44 Pg C/yr (~ 5-13 Gt $CO_2$/yr, 1 Pg = 1 Gt) of technical potential [134], which would involve changing land use practices on something like 70 million km² of agricultural land and degraded land. Other estimates based on biochemical and photosynthesis constraints suggest that the realistic sequestration potential of agroecosystems is less than 1 Pg C/yr (~1-3 Gt $CO_2$/yr) [143, 144]. A likely practical limit on ecosystem-based SOC restoration of at most 10-30% of the total of ~100 Gt of lost carbon (i.e., 40-120 Gt $CO_2$ equivalent) was suggested in [136]. While there is a wide range of variation in estimates, the viable rate of equivalent $CO_2$ sequestration in soil through these kinds of land management practices seems likely to be in the range of hundreds of Mt to a few Gt of $CO_2$ yearly. The effectiveness and permanence of these approaches are also difficult to measure and verify. Even with relatively stable land use patterns, accurately measuring increases in soil carbon is challenging due to a high degree of spatial heterogeneity and relatively small annual changes from management interventions (e.g., < 0.1-0.5% for no-till conversion and biochar application) [101]. However, by comparing soils in similar landscapes at different stages (chronosequence data), estimates of, for example, +2.3%/yr for farm abandonment on Mediterranean lands have been measured [145].

As discussed above, the long-term retention of carbon in soil depends upon environmental factors and continued appropriate land use.

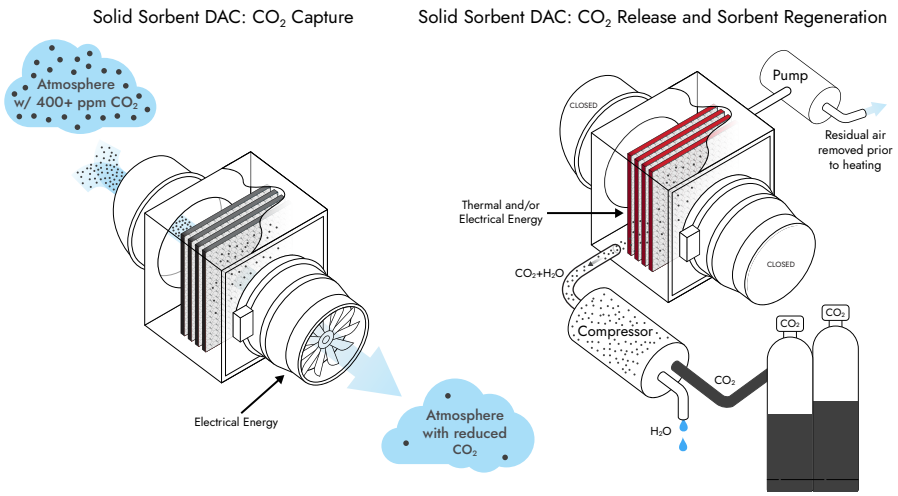
**Carbon retention in terrestrial biomass.** Forests, including the associated living biomass and soil carbon, hold roughly half of the terrestrial carbon on the planet [146], and (primarily tropical) deforestation contributes substantially to net anthropogenic carbon emissions. Well-managed projects to restore forest ecosystems in suitable locations may present one of the most cost-effective methods of large-scale carbon capture in the near future [26, 110] by increasing carbon in terrestrial biomass and soil. This can be done while other larger-scale CDR approaches and technologies covered in this report are gradually brought up to scale. Preserving existing forest ecosystems, particularly in tropical areas, is also essential to reduce the further loss of carbon from terrestrial biomass and soil. An estimate for potential carbon accumulation in above-ground biomass over a 30-year window through the reforestation of ~ 700 Mha (7 M km²) is 1.6 Pg C/yr (~ 5 Gt $CO_2$/yr, with another 50% in potential SOC increase) [147]; a similar estimate (with large error bars) appears in [26]. Like changes in agriculture and grazing practices, the restoration of forest ecosystems can bring many auxiliary benefits including increased biodiversity and ecosystem services to humans. While full ecosystems are highly complex and the goals of precisely reproducing previously stable ecosystems by introducing specific species may be difficult to achieve in many locations, particularly with on-going environmental changes, simply enabling reforestation to occur on degraded lands through, e.g., farmland abandonment, can be effective from the CDR point of view.

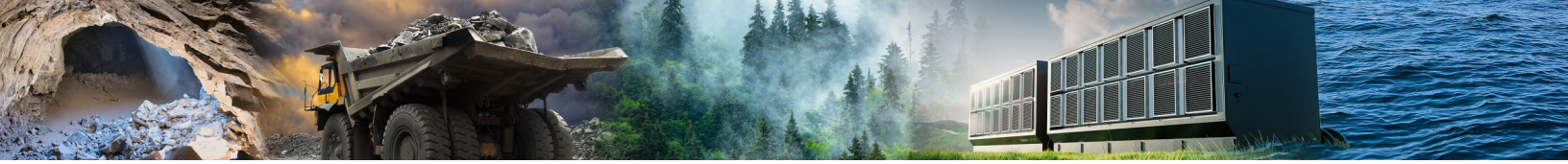
Despite these positive attributes, afforestation/reforestation has been used in many circumstances for questionable carbon offsets (see, e.g., [100]), because it is often difficult to verify that the associated carbon removal satisfies the criteria of being real, permanent, quantifiable, verifiable, etc.; planting trees in unsuitable locations will not lead to lasting carbon removal. Afforestation/reforestation efforts can only be successful in appropriate ecological contexts and often require location-specific active management and ongoing forest preservation. In addition, forest carbon stocks are susceptible to issues such as fires and insect infestation, patterns of which may change significantly as climate change continues [148, 149].

## J. Approaches to Chemical DAC: Details and Further References

This appendix describes some of the technical aspects of different approaches to chemical DAC. More detailed descriptions and analyses can be found in [20].

**Figure 8:** A schematic of a temperature-vacuum swing solid sorbent DAC unit. In the $CO_2$ capture phase (left), air is forced by the capture material. In the extraction phase (right), the chamber is closed off, residual air is removed, and $CO_2$ and water vapor are extracted.





**Solid sorbent systems.** A wide range of technical approaches to solid sorbent DAC systems are under investigation. The furthest developed scheme for solid sorbent DAC uses temperature and/or pressure changes ('thermal / pressure swing') between the adsorption and desorption processes. A generic example is depicted in Fig. 8.

On the left of Fig. 8, air enters the extraction cycle where it is moved through a porous, multi-scale structure that has been chemically treated with a sorbent that will capture $CO_2$ for removal. Adsorber chemistry for the removal of $CO_2$ (a Lewis acid) is base-acid; for example, an amine group might be grafted to the multi-scale structure. Generally, the adsorption process is exothermic, and the sorbent with its supporting structure must conduct away this heat to avoid temperature increases that can reduce sorbent performance. Once the sorbent has reached the desired $CO_2$ saturation, the $CO_2$ must be removed and sequestered and the sorbent regenerated for another capture cycle. Water vapor may also be captured by the sorbent. For some processes this is desirable and increases the $CO_2$ capture rate. There is a concomitant energy penalty when the water is removed from the surface during the regeneration of the sorbent. Materials under investigation for DAC include amine-functionalized silicas, metal-oxide frameworks, porous polymers, zeolites, and porous carbons.

In the desorption process (right side of Fig. 8), the fans are turned off and the sorbent is isolated from fresh airflow. The remaining air inside the unit that houses the sorbent is evacuated, and the sorbent is heated, usually by steam, to a temperature of around 100 °C. This causes the $CO_2$ and any water on the surface to desorb, after which it is pumped to a condenser where $H_2O$ is separated from $CO_2$ as the mixture cools to ambient temperature. Sequestering the $CO_2$ requires further compression and transport to long-term storage or other usage.

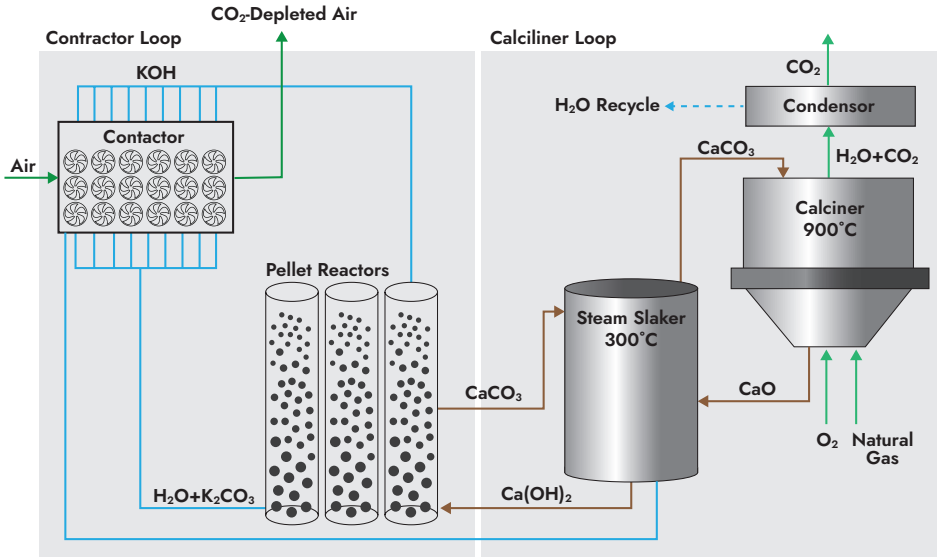
Efficient removal of $CO_2$ requires that the sorbent and supporting structure have sufficient surface area to facilitate $CO_2$ capture, while energy considerations require that $CO_2$ capture systems be designed to minimize the pressure drop needed to push air through the structure. Sorbents should have a high capacity for $CO_2$, energetically favorable thermodynamics, robustness to variations in environmental parameters (humidity, temperature, pollutants), minimal losses during regeneration cycles, and affordability. Energy considerations further favor sorbate structures with a lower heat capacity, and facility size will decrease as sorbate capacity increases and extraction-regeneration times decrease. Table 5.6 [4] provides a summary of the useful parameters governing solid sorbent performance.

**Liquid solvent DAC.** Liquid solvent approaches have been studied extensively in the context of point-source carbon capture, where the $CO_2$ density is higher [6]. A detailed analysis [150] has also been developed for at least one approach to a liquid-based DAC facility, which we review briefly here. In a simplified description, the liquid-based scheme uses two coupled loops to extract $CO_2$. In the contactor loop, air is blown horizontally through the contactor, which consists of a long tube filled with small PVC pellets. A solution of aqueous potassium hydroxide solution (KOH) is sprayed from nozzles above the contactor and coats the pellets as it flows. The KOH reacts with $CO_2$ to form water and potassium carbonate via the reaction $2KOH+CO_2 \rightarrow H_2O+K_2CO_3$.

Regeneration and $CO_2$ removal are accomplished in multiple steps: After flowing through the contactor, the $H_2O$ and $K_2CO_3$ are pumped into a pellet reactor where the $K_2CO_3$ reacts with calcium hydroxide ($Ca(OH)_2$). This process regenerates KOH, which is sent back to the contactor, and produces a precipitate of calcium carbonate ($CaCO_3$). The $CaCO_3$ enters the calciner loop, which is comprised of a slaker and a calciner. This loop separates $CO_2$ for sequestration, and produces the $Ca(OH)_2$ needed in the pellet reactor. (Note that the $H_2O$ flows are not fully described in Fig. 9.) The liquid solvent system involves more steps and generally requires [4] higher temperatures and greater thermal energy input than do solid sorbent systems.

Achieving high efficiency is challenging because it necessitates meticulous management of heat and material losses across multiple chemical processes. These processes proposed for liquid-based schemes, however, derive from well-established industrial applications.

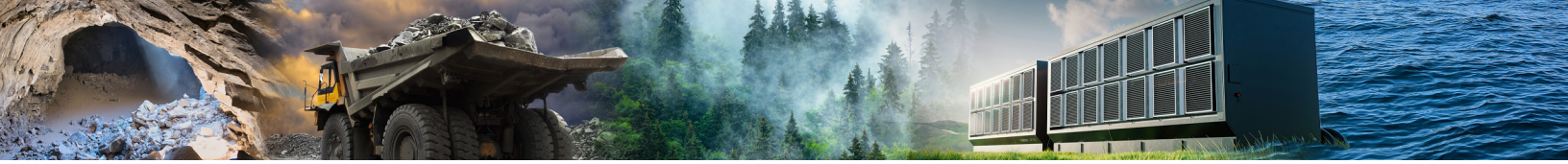
**Figure 9:** Schematic of a liquid solvent DAC following the design of Keith et al. [150]. Green (blue) lines correspond to gaseous (liquid) flows. The dashed line represents $CO_2$-depleted air returning to the atmosphere. On the left, air enters the extraction cycle where it is moved through a honeycomb-like structure chemically treated to facilitate $CO_2$ absorption. Taken from [20].




**Electrochemical and other mechanisms for DAC** A number of alternatives to the temperature/pressure swing approach are being actively pursued [151, 152, 22]. In a 'moisture-swing' process, $CO_2$ is removed at low humidity and the sorbent is regenerated by exposure to high humidity. In this scheme, water, rather than heat, provides the free energy needed to remove the $CO_2$ from the sorbent. It has been argued that this approach comes closer to the thermodynamic bound for energy [25]. However, abundant water supplies are not available everywhere, limiting the siting possibilities or requiring additional infrastructure/energy expenditures.

Newer technologies for DAC which utilize only electrical power or a hybrid of heat and electrical power can avoid or lessen thermodynamic losses that are inevitable when heat is used to desorb $CO_2$. The temperature/electrical [153] swing process employs a combination of thermal energy and Joule heating to realize the desired temperature swings in the process. Microwave-accelerated [154] processes use microwaves to reduce the heat input needed to remove $CO_2$ from the adsorber. The magnetic induction [155] swing process uses an alternating current magnetic field which inductively couples to embedded magnetic nanoparticles embedded in a metal-oxide framework. The nanoparticles, in turn, heat the adsorber through collisions, and the heated material releases captured $CO_2$.

Electrochemical swing [156, 157, 158] technologies that reduce the need for heat or mechanical (vacuum/pressure) changes have the potential to increase the efficiency of $CO_2$ removal. Electrochemical DAC systems can utilize a pH-swing process, where the solubility of $CO_2$ in a working fluid is changed by varying the fluid's pH. A variety of methods for electrochemical DAC are under study [158, 157], including electrolysis, bipolar membrane electrodialysis, reversible redox reactions, and capacitive deionization. Elec-



trochemical DAC offers potential advantages over other methods. Power can be supplied by solar arrays, and in some realizations they can be part of a process to produce hydrogen fuel.

## K. Some Further Ideas for CDR Approaches

In general, most CDR approaches will fit into some of the primary paradigms described in the main text. We briefly list a few further ideas here for CDR to illustrate some of the range of possibilities; these are all currently at a rather speculative stage.

**Cryogenic separation of $CO_2$.** Cryogenic separation of carbon dioxide from air is already in use at industrial scale [159]. Condensation of carbon dioxide ice ('dry ice') occurs on the surface of Mars, and temperatures in the interior of Antarctica occasionally drop low enough for carbon dioxide to solidify. This has been proposed as a possible way to capture $CO_2$ directly from the atmosphere [160]; at least one company has proposed doing so on balloons [161].

**Plasmons.** Natural photosynthesis converts about 1% of the energy in sunlight to the reduction of carbon dioxide to form carbohydrates. Artificial photosynthesis driven by surface plasmons (electron waves that propagate on the surface of a conductor) offers a possible route to more efficiently use sunlight [162, 163, 164], converting atmospheric $CO_2$ to either hydrocarbon fuels or carbon-containing material suitable for storage.

**Geothermal water with high alkalinity.** Geothermally heated water from carbon-depleted aquifers could be brought to the surface to generate electrical power and absorb atmospheric $CO_2$ before being injected into ultramafic rock for in situ mineralization [39]. It may be a challenge to sufficiently scale up this approach.

## L. Anthropogenic $CO_2$ and Climate Warming

On average, when Earth's climate is in an approximate equilibrium, roughly 70% of the net incoming solar radiation that hits Earth is absorbed. The land and oceans balance the incoming radiation flux by emitting outgoing thermal radiation in the infrared. Water vapor, $CO_2$, and other greenhouse gases in the atmosphere absorb and re-emit this outgoing thermal radiation, adding to the downward flux of radiation (radiative forcing), thereby maintaining Earth's surface temperature at a higher value than it would have been in the absence of greenhouse gases. An increase in atmospheric $CO_2$ levels drives additional downward radiative forcing, which leads to planetary warming until the extra downward radiation flux is balanced by increased thermal radiation from a higher equilibrium temperature. [67]

While it is not clear that a quasi-equilibrium approximation will accurately describe changes in Earth's climate from anthropogenic $CO_2$, such a model is widely used to provide a rough estimate of expected temperature increase for a given change in atmospheric $CO_2$ levels. Based on a synthesis of multiple approaches, including the best available current models of the full climate system and evidence from historical and paleoclimate data, along similar lines to [165], the Intergovernmental Panel on Climate Change (IPCC) AR6 report estimates that the rise in Earth's mean surface temperature from a doubling of $CO_2$ from preindustrial levels (ECS = equilibrium climate sensitivity) would be roughly $\Delta T_{2\times} = 3\,^{\circ}C$, with uncertainties giving a likely range of 2.5-4 °C [131]. From the IPCC estimate of climate sensitivity and the logarithmic dependency of radiative forcing on $CO_2$ levels, in the quasi-equilibrium approximation the

[67] This story is complicated by a number of factors. Multiple positive and negative feedbacks, such as clouds and albedo changes due to variations in snow and ice coverage, modify the simple direct effect of $CO_2$. Earth's climate systems are complex, nonlinear, multiscale, and multifluid. The net effect of atmospheric $CO_2$ changes takes decades to be fully realized. Numerous other natural and anthropogenic forcing factors are relevant.

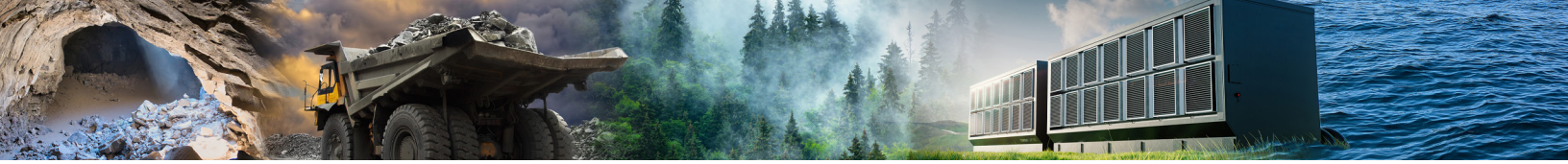
expected rise in mean surface temperature from preindustrial times due to anthropogenic $CO_2$ emissions already present in the atmosphere would be expected to be roughly 1.75 °C (≅ 3 °C × ln (420/280)/ ln 2). Because equilibration takes place on a decades-long time scale, this temperature shift has only partially taken effect, and $CO_2$ already in the atmosphere will continue to drive increased warming through the remainder of this century, in the absence of other factors. Most anthropogenic atmospheric $CO_2$ emissions (~ 80%) have occurred since 1950, when levels were around 310 ppm. The measured mean surface temperature rise of over 0.8 °C since 1950 [166, 167] is largely attributed by the IPCC and others to human activity, with atmospheric $CO_2$ a primary driver of this warming. As an order-of-magnitude reality check, this observed warming since 1950 is roughly compatible with the IPCC ECS estimate and a rough estimate that something like half of the expected warming from $CO_2$ in the atmosphere has taken place since then.

A similar computation using the ECS of 3 °C shows that the atmospheric $CO_2$ levels associated with roughly a 2 °C warming would be roughly 445 ppm, which would be reached by around 2035 if emissions were to continue at the current rate and CDR is not implemented at scale. It is worth noting, however, that these and similar estimates of temperature variation with $CO_2$ concentration depend upon the ECS estimate, and accurate evaluation of the ECS is very challenging. For example, other estimates based on the same data [168] suggest an ECS value of 2.2 °C. This would reduce the expected temperature change from $CO_2$ already in the atmosphere to 1.3 °C, with warming reaching 2 °C only after another 40 years of emissions at current rates. By contrast another recent study [169] finds an ECS value of 4.8 ± 1.2 °C. (See Fig. 1 [170] for an overview of different estimates of climate sensitivity.) If the ECS takes the upper value in the IPCC's likely range, 4 °C, then the warming expected from $CO_2$ already in the atmosphere would be over 2.3 °C.

The level of temperature rise considered tolerable is a question that depends upon many factors, including impacts on ecosystems, agriculture, and human living conditions, many of which are not well understood scientifically or have high uncertainties, as well as tradeoffs between addressing climate and other critical issues.

The IPCC and other entities have analyzed a variety of scenarios, based on different assumptions about many controlling factors including human activities and varying levels of future emissions, as well as the future behavior of the ocean and land carbon sinks. In scenarios where atmospheric carbon levels are managed to avoid a surface temperature rise of more than 1.5 °C or 2 °C and $CO_2$ emissions are not drastically and immediately reduced, substantial carbon dioxide removal (CDR) implementation is incorporated (including from LULUCF), at the level of 1-20 Gt $CO_2$/yr in the latter part of this century. Specifically, in the IPCC AR6 (WGI) report [12] (page 622), the median of $CO_2$ removal needed by 2100 in various scenarios is quoted as 730 Gt $CO_2$, with a range of 1-20 Gt $CO_2$/yr yearly after 2050; very aggressive emission reduction scenarios can reduce the necessary rate of carbon removal toward the lower values. Similar and related analyses appear in [1]. The NASEM 2019 report [4] concludes that CDR will likely need to remove ~ 10 Gt $CO_2$/yr globally by 2050 and ~ 20 Gt $CO_2$/yr by 2100. The analysis of [10] suggests that somewhat lower amounts of CDR, although still at the level of multiple gigatons of $CO_2$ per year, may be feasible in scenarios keeping temperature rise below 2 °C.

One simple way to think about these numbers (see Reality Check box) is that, assuming an equilibrium climate sensitivity in the range of 2.2-3 °C, the anthropogenic $CO_2$ already in the atmosphere has already 'locked in' an eventual temperature change of ~ 1.5 °C. To keep the temperature increase below that goal, this simple model implies that most or all of any further net $CO_2$ emissions must eventually be recaptured. Because it will likely take at best several decades to reduce the current rate of emission below 10 Gt $CO_2$/yr, this suggests a necessary rate of removal of multiple gigatons of $CO_2$ per year over comparable or longer time scales to achieve such climate goals.



### Reality Check: Temperature Change and CDR (Approximate Analysis)

The scenarios described by the IPCC and others for emissions and warming trajectories involve a complicated set of assumptions. It is easy to see, however, from the rough numbers involved that the IPCC estimations of climate sensitivity imply that Gt-scale CDR would be needed to keep warming from anthropogenic $CO_2$ below 1.5 or 2 °C. In particular, the eventual warming that will occur from $CO_2$ already in the atmosphere is around 1.75 °C, assuming that the central IPCC estimate of ECS = 3 °C for a doubling of $CO_2$ is correct. With the more optimistic ECS = 2.2 °C, the 'locked-in' warming would be around 1.3 °C and the increase of 1.75 °C would be reached in 26 years. Thus, to keep warming at 1.5 °C, most or all future net increases in atmospheric $CO_2$ (i.e., emissions minus sinks) would need to be canceled by CDR. Because such net emissions are currently over 15 Gt $CO_2$/yr, and are unlikely to decrease below 10 Gt $CO_2$/yr very quickly, CDR at the multiple gigatons per year scale would be needed to achieve that target.

There are a number of other significant greenhouse gases besides $CO_2$. Water vapor ($H_2O$) is the dominant greenhouse gas, and changes in atmospheric water vapor content and the associated lapse rate (rate of change of temperature with altitude) represent important climate feedbacks. Methane, which also contains carbon, has an even stronger effect on radiative forcing per unit mass than $CO_2$; however its lifetime in the atmosphere is much shorter (~ 10 years for methane rather than thousands of years for $CO_2$). Other greenhouse gases include nitrous oxide and other molecules with anthropogenic sources through agriculture or other activities. The focus in this report is on $CO_2$, which is a key greenhouse gas for several reasons. First, its long lifetime makes $CO_2$ the primary factor that will control Earth's climate over the time scale of centuries. Second, $CO_2$ is currently the primary driver of planetary warming. Third, the primary emissions from fossil fuel combustion are $CO_2$ molecules that represent additional carbon brought into the atmosphere-terrestrial carbon cycle. [68]

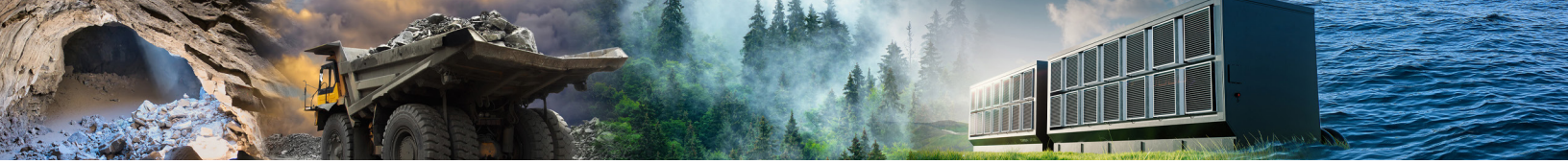
This report is about methods to remove carbon in the form of $CO_2$ from the atmosphere to keep the system closer to the approximate equilibrium that it has been in over recent millennia. Given a fixed amount of carbon in the atmosphere-terrestrial system, many other factors influence changes in radiative forcing and climate over shorter time scales. Human activities, including grazing, agriculture, and forest management practices, move carbon between terrestrial ecosystems and the atmosphere, and change the form of carbon (e.g., methane release from farming). Some of these activities fit into the realm of biological CDR, as discussed in §4.2. There are also many other shorter-lived greenhouse gases such as nitrous oxide that do not specifically contain carbon. In considering how to manage atmospheric carbon levels to achieve specific climate-related goals, it may be useful to conceptually separate efforts like chemical DAC that can permanently remove carbon from the atmosphere-terrestrial system from other activities that move the carbon around within that system. It should be noted that other effects beyond fossil fuel use and CDR may impact the total atmosphere-terrestrial carbon budget, particularly in a context where climate is out of equilibrium; for example, a warming climate can lead to release of methane from clathrates in formerly frozen tundra leading to a secondary source of increased carbon in the terrestrial-atmosphere system (and a potentially nonlinear climate feedback). [69] All of these issues must be taken into account when determining the right balance of resources to put into CDR versus management of other relevant activities.

[68] Fossil fuels are carbon-containing resources in Earth's crust that were formed from organic material over millions of years. Burning fossil fuels therefore releases into the atmosphere-terrestrial carbon system an abundance of carbon-based molecules that have been locked away in geological repositories, thus changing the net quantity of carbon in the atmosphere-terrestrial system. While over time scales of thousands of years, carbon from the atmosphere and terrestrial system gradually equilibrates with the much larger deep ocean carbon repository, over the time scales currently relevant for humanity — decades or centuries — carbon, once released from fossil fuels, increases the atmosphere-terrestrial carbon account, generally in the form of increased $CO_2$.
[69] Note that the IPCC estimates that it is unlikely that release of methane from permafrost clathrates will have a significant impact on the expected emissions trajectory this century [131].

To put the effects of anthropogenic $CO_2$ and climate warming in context, it is helpful to consider atmospheric carbon changes that have occurred over longer time scales. While the current atmospheric $CO_2$ levels are believed to be the highest reached in several million years, Earth's atmosphere has endured enormous fluctuations in carbon content over longer time periods. Over the longest 'tectonic' time scale of tens of millions of years, atmospheric $CO_2$ levels have been affected by large-scale changes in the arrangement of Earth's continents and oceans. Fifty-five million years ago in the Eocene, $CO_2$ levels were estimated, based on various proxies such as benthic $^{18}O$ levels, to have been at least five times greater than current values. At that time there were no polar ice caps, the temperature was higher by 5-10 °C (with larger differential at higher latitudes), and sea levels were hundreds of meters higher than they are currently. Life was abundant and mammals began to proliferate. Over the interceding tens of millions of years, $CO_2$ levels and global temperatures have dropped fairly steadily (with some temporary increases in between). The precise mechanisms for the reduction in $CO_2$ levels and temperature over this tectonic time scale are not fully understood, but it is believed that chemical weathering of rock exposed in the formation of the Tibetan plateau, a decrease in the rate of sea floor spreading, and changes in ocean circu-lation (such as the formation of the Antarctic circumpolar current) may all have played substantial roles in driving these changes. Over the last 3 million years or so, superimposed on a gradual overall cooling, Earth's climate has been dominated by regular glacial/interglacial periods with oscillations of 40,000-100,000 years, driven by changes in Earth's orbital parameters described by the now widely accepted Milankovitch theory.

While the presence of high atmospheric $CO_2$ levels and temperatures earlier in Earth's history at a time when life was abundant provides reassurance that Earth's climate and ecosystems can in principle thrive in such environments, it should be kept in mind that these changes occurred over many millions of years, giving organisms and ecosystems time to adapt to the changes. Even if anthropogenic carbon has a more modest effect of running $CO_2$ conditions back 5-10 million years, this will cause changes in climate over the next century at a rate much faster than most geologic changes, and more quickly than species and ecosystems adapt through evolution. A recent report [171] estimates that one million animal and plant species are threatened with extinction [70], and expects that rapid climate change will exacerbate the rate of ecosystem and biodiversity loss in coming decades. Human society, which has developed in the relatively stable climate of the last 10,000 years, also may face substantial challenges in the face of changing climate impact on weather, sea level, and ecosystem services. These changes are arising at the same time as many other critical issues, such as a large and still growing population where many lack access to adequate energy, water, and other resources, and dangers of increasing conflicts and the threat of nuclear war. Addressing and balancing these multifaceted and interconnected problems presents a complex task that will require coordinated efforts and innovative solutions.

[70] According to the report, the direct drivers of change in natural systems, in order of significance, are: human land and sea use, direct exploitation of organisms, climate change, pollution, and invasive species.



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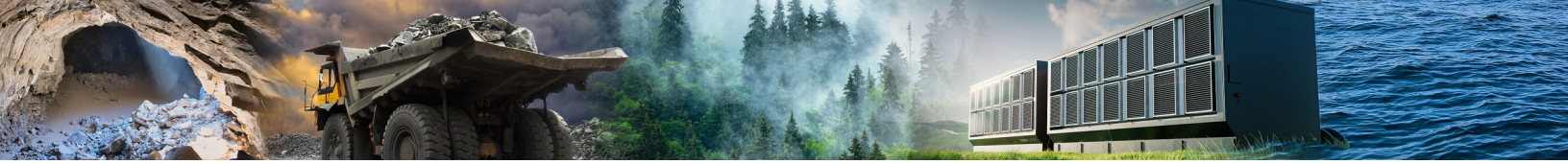


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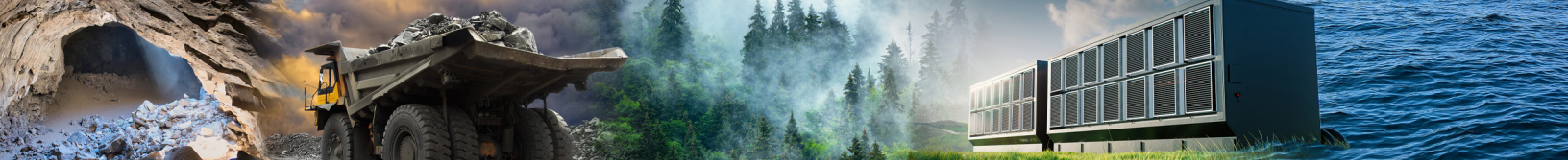


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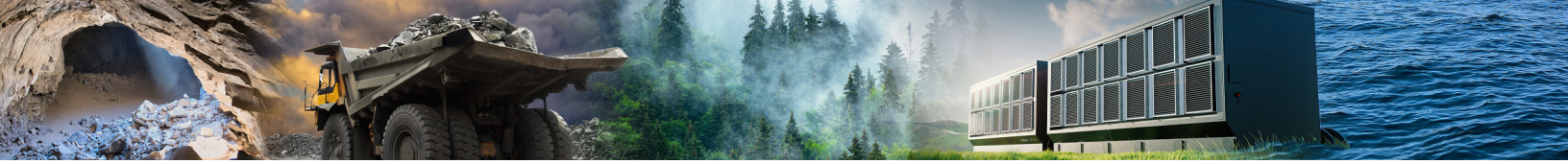


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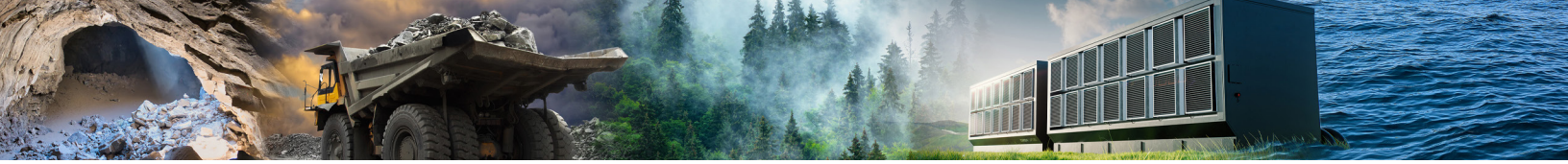


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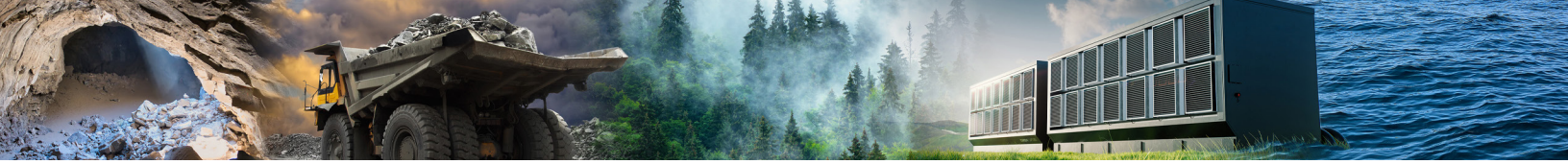


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