THE ROLE OF NEGATIVE EMISSION TECHNOLOGIES IN ADDRESSING OUR CLIMATE GOALS

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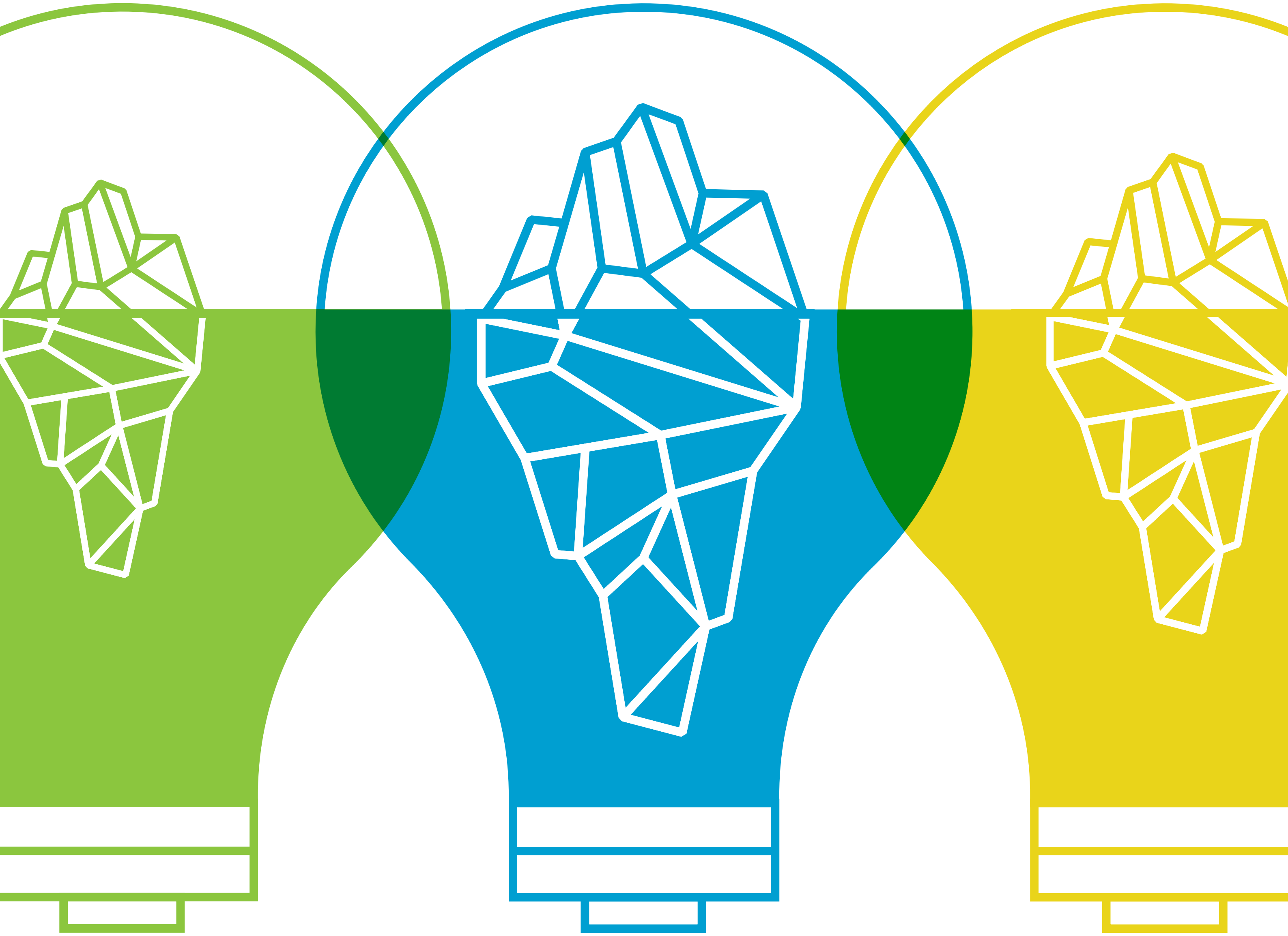

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THE ROLE OF NEGATIVE EMISSION TECHNOLOGIES IN ADDRESSING OUR CLIMATE GOALS

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EDITORIAL
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Editorial: The Role of Negative Emission Technologies in Addressing Our Climate Goals

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Editorial on the Research Topic

The Role of Negative Emission Technologies in Addressing Our Climate Goals

Imagining ourselves in the mid-1700s with perfect foresight of the impending technological revolution, how might our decisions be guided? Would we have done anything different? It is tempting to think that we would skip fossil fuels altogether and that the industrial revolution would have been powered largely by renewables. We may have also avoided obvious environmental failures like the prolific use of asbestos, lead-based paint, and unregulated chlorofluorocarbon use. However, it is likely that we still would have used some fossil fuels. While we may have pushed for quicker deployment of carbon capture and storage, we probably would have afforded ourselves the luxury of unmitigated CO_2 emission into the atmosphere.

If our 1,700 selves had a trillion-ton emission budget, how would we have spent it? We have not entirely squandered that resource. It has raised billions out of poverty, extended our lives and improved living conditions, fuelled exploration and scientific progress, and created opportunities for human connection that have never previously existed in the history of our species. The travesty is that not everyone has benefited from the value of this budget, nor was the transition cost factored into its exhaustion. However, the era of unabated CO_2 emission to the atmosphere must end, and we are now called upon to implement a rapid transition to net-zero greenhouse gas emissions. Most pathways for limiting climate change contain rapid and deep emissions reduction combined with large amounts of CO_2 removal from the atmosphere facilitated by Negative Emission Technologies (NETs).

While NETs have long been part of the climate change conversation, their unique importance has emerged following the 2015 Paris Agreement, and subsequently crystallized by high level reports from the European Academies' Science Advisory Council (2018), The Royal Society Royal Academy of Engineering (2018), National Academy of Sciences Engineering and Medicine (2019), and Intergovernmental Panel on Climate Change (2018). They all acknowledge that NETs are not a replacement for reducing emissions, and that there is an unsettling gap between the assumptions made about the potential of NETs (e.g., in model scenarios) and what is currently known about their feasibility of operating at a global scale. These reports are a call to action for researchers to fill this knowledge gap. In 2019 we launched *Frontiers in Climate: Negative Emission Technologies* as a dedicated home for research in this field, and welcome submissions from any discipline considering the feasibility or implications of NETs. We are delighted to present the collection of articles in our first Research Topic.

Scale is important, not just because it frames the contribution of an approach to solving the problem, but also constrains what might be possible. When an industry operates at a global scale it potentially runs up against hard immovable geophysical limits. Arguably, humanity has surpassed

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several of these limits already (Steffen et al., 2011) and is rapidly approaching the limit of cumulative atmospheric CO_2.

Unquestionably, we have been able to innovate our way out of complicated problems. The story of our species is tethered to that of technological change. This story is also marked by enormous social transformation. Climate change challenges us to innovate and transform further still. However, we postulate that the risk of failing to meet our climate targets depends not only on questions of whether technological or social transformation can occur fast enough, but that ideological conflict between techno-driven or socio-driven approaches may ultimately prevent either from occurring. The perspectives piece from Friedmann makes this point by borrowing from *The Wizard and the Prophet* by Mann (2018).

> "Mann asserts that 'wizards' [those that favor technical solutions] and 'prophets' [those that favor social solutions] represent distinct approaches and tribes, commonly with very different world views and value systems. Understanding this axis of contention is essential to acknowledging the difficulty of the task of climate restoration… Despite their common goals, wizards, and prophets sometimes view each other with contempt."

This "axis of contention" can be a positive reflective force in a world driven by technological change but is essential when confronted by the geophysical limits of global technologies. An early manifestation of this is the potential for new or even hypothetical NETs to act as deterrents to emissions reduction (or "moral hazard"). Within this context McLaren et al. argue for separate targets for emissions reduction and negative emissions. This approach may limit the moral hazard but requires separation of target-setting, incentivization, monitoring, and evaluation regimes.

The perspective of a company developing systems for directly removing CO_2 from the atmosphere is described by Beuttler et al. Switzerland based Climeworks have been operating for a decade, and here they provide an overview of their direct air capture approach, how it was developed, and the current disconnect between markets for CO_2 and what may eventually form part of a global effort to remove billions of tons of CO_2 from the atmosphere. Current policies are not adequate so opportunities that capture CO_2 from air are presently coupled with approaches that help to offset the costs of capture, with most re-releasing CO_2 back into air, whether it be through food (e.g., CO_2 use in greenhouses), carbonated beverages, or synthetic fuels.

Many of the negative emissions approaches create enriched CO_2 gas for storage, and by mid-century about 10 billion tons will need to be stored (e.g., under the "middle of the road" 1.5°C scenario, Intergovernmental Panel on Climate Change, 2018). Kelemen et al. provide a comprehensive review of storage options. This includes sequestration in deep saline aquifers and mineralization. The annual storage potential together with approximate costs are reported. Geological storage of CO_2 has considerable capacity at global scale, and storage in sedimentary basins has been carried out in practice since the early 1970's (e.g., in the Permian Basin).

Over the next 50 years and beyond, our energy transition must reduce fossil fuel use and increase renewables. It is unlikely that fossil fuels will disappear completely, and certainly decarbonizing fossil fuel energy will be an important part of the technology transition. "Enhanced oil recovery" (EOR), the method by which CO_2 is injected into the subsurface to extract oil from depleting reservoirs, has long been acknowledged as releasing more CO_2 than it sequesters. Núñez-López and Moskal challenge us to think again. If optimized for CO_2 storage rather than oil production, an EOR scheme may operate with a net negative carbon balance for a limited period. This potential is further extended if additional process CO_2 is injected lower in the rock formation ("stacked storage"). The finite capacity of an EOR project to be net carbon negative together with constrained scalable potential (Kolster et al., 2017), make carbon storage in EOR a precious resource, which we should take care in using as part of a longer-term transition to decarbonize fossil fuels. Managing the transition to net-zero carbon emissions may include technologies and processes that are incompatible with the goal but otherwise take us several steps closer. For instance, swapping unabated coal for unabated gas lowers emissions from energy generation, but further emissions reduction will be needed to reach our climate targets. Similarly, the feasibility of several NETs (e.g., direct air capture) is sensitive to the emission intensity of the energy supply and are thus inextricably linked to the energy transition.

The oceans play a vital role in the Earth's climate. Marine ecosystems are becoming increasingly stressed by elevated atmospheric CO_2 concentrations. The oceans are becoming more acidic, which makes it harder for some shell forming organisms to grow. Bach et al. explore the possibility of increasing ocean alkalinity through mineral addition. Such proposals plan to remove CO_2 from the atmosphere while also partially ameliorating the impacts of acidification. The potential environmental impact on pelagic marine ecosystems is mapped out here, and they constrain the changes in surface ocean alkalinity, silica, iron, and other metals as a consequence of a range of mineral addition scenarios.

It may surprise many that the two-meter sliver of soil that covers much of our planet contains more carbon than vegetation and the atmosphere combined. Paustian et al. review existing best management practices and frontier technologies for managing and maximizing this resource. They call for a two-stage strategy. First, strong policy that could be enacted immediately to incentivize existing technologies and approaches. Furthermore, continued and leveled-up research and development could be used to create new crop varieties for additional negative emissions mid-century.

Emerging within NET's discourse is the integration of land-based approaches with agricultural reform policy platforms. Within this context, Jacobson and Sanchez review agricultural relevant NETs in the context of the US public administration. They recommend the establishment of a new research agency and technology commercialization program within US Department of Agriculture, improved coordination agencies and foundations and congressional action to establish and fund new NETs programs. The work is valuable insight into how public administration may be shaped by the challenge of balancing residual emissions using NETs.

Considering the global potential of NETs has been an important first step in understanding what options may be available at scale. However, their deployment will undoubtably be on national and regional scales. The perspective article by Fajardy et al. examines the national and regional barriers to a range of approaches including land and water, access to low carbon energy, CO_2 storage, socio-economic issues, and finance.

While the journal is always open to new submissions, future Research Topics will include technology-specific approaches (ocean-based, direct air capture), system-level assessment and rationalization, social sciences, policy and governance. We also welcome suggestions for future Research Topics, and look forward to working with you as editors, reviewers and authors in bridging the NETs knowledge and policy gap.

AUTHOR CONTRIBUTIONS

PR and JW wrote the manuscript.

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POLICY AND PRACTICE REVIEWS
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Opportunities for Carbon Dioxide Removal Within the United States Department of Agriculture

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Farming and ranching communities in the United States sit at the front lines of climate change impacts and responses. In particular, terrestrial atmospheric carbon dioxide removal (CDR) can reduce climate change impacts while increasing resilience to extreme weather. Currently, many CDR technologies and strategies are still under research and development (R&D), and lack sufficient federal support to reach widespread deployment. Here, we provide an assessment of the United States Department of Agriculture's (USDA) existing programs and organizational structure, its capacity to support research and demonstration of CDR, and recommendations for expansion of these capabilities. We summarize USDA's previous and current efforts to incorporate CDR R&D within their research, education, and economics mission, as well as opportunities to refocus and expand existing programs. Potential future actions to expand CDR R&D capabilities include: (1) the establishment of a new extramural research agency and an intramural technology commercialization program within USDA, (2) improved coordination between the Foundation for Food and Agriculture (FFAR) and USDA, (3) improved intra-agency and inter-agency coordination, and (4) congressional action to establish and fund new CDR programs within USDA. USDA can pursue multiple strategies to enhance CDR, driving development, demonstration, and deployment across the United States.

Keywords: carbon dioxide removal, climate policy, research and development, United States Department of Agriculture, Advanced Research Projects Agency

INTRODUCTION

The United States Department of Agriculture (USDA) contains several offices and programs that provide limited support for research, development, and demonstration (RD&D) activities relevant to carbon dioxide removal (CDR) (Sanchez et al., 2018). The bulk of this research and development support is facilitated through Under Secretary of Research, Education, and Economics' (REE) agencies, which include the Agricultural Research Service (ARS), National Institute of Food and Agriculture (NIFA), the National Agricultural Statistics Service (NASS), and the Economic Research Service (ERS). Additionally, offices reporting to other Under Secretaries, such as the Forest Service, the Farm Service Agency (FSA), and the Natural Resources Conservation Service (NRCS), operate programs that perform both research and development functions relevant to CDR. While REE offices focus primarily on applied research, the Forest Service, FSA, and NRCS perform a variety of demonstration and deployment functions. With a collective operating budget of nearly $5 billion annually, collaborative efforts across these agencies have already yielded significant advancements in the research and development of climate change mitigation and adaptive land management strategies and technologies. Below, **Table 1** summarizes the missions, jurisdictions, programmatic focuses, funding levels, and CDR relevant land management strategies of the Research, Education, and Economics offices, as well as the Natural Resources Conservation Offices and the United States Forest Service.

Given USDA's organizational structure, REE mission, and diverse set of expertise and programs, the Department and Congress are well-positioned to refocus and expand existing research programs relevant to carbon dioxide removal. REE's primary objectives of increasing economic opportunity and conserving natural resources through scientific and economic research and education could enable many existing programs to be refocused or modified without congressional action or expanded and augmented through changes in appropriation and authorization legislation to yield significant advancements in CDR RD&D.

We consider three main categories of CDR approaches in this paper: natural, technological and hybrid. Natural solutions include, but are not limited to, improved land management through reduced tillage, increased crop rotation, sowing of cover crops, and increases in forest biomass. CDR-relevant research that is not explicitly focused on CDR, but could provide important lessons for CDR includes carbon cycling in soils, forest management, conservation, and bioenergy crop production. Technological approaches include processes that capture carbon emissions and either reliably sequester them for extended periods or convert them into valuable products or commodities. Hybrid approaches include aspects both of natural and technological CDR, such as bioenergy with carbon capture and sequestration (BECCS). Although we assess how RD&D on all of these approaches could be incorporated within USDA's programs, we focus primarily on natural approaches or "land-use CDR."

This report summarizes USDA's existing program structure and funding mechanisms for research, development, and demonstration (RD&D) with potential applications to carbon dioxide removal, and offers suggestions to refocus or expand these efforts through congressional action, improved coordination, and new research offices. Section I describes the current state and structure of USDA's RD&D and REE initiatives, Section II analyzes past and current RD&D efforts relevant to CDR at USDA and other departments, and Section III provides suggestions on how to improve USDA's capacity for carbon dioxide removal RD&D.

SECTION I: RESEARCH, EDUCATION, AND ECONOMIC AND RELATED FUNCTIONS OF USDA

In the section below, we review the REE functions of USDA. Specifically, we highlight several important distinctions within these offices relevant to CDR RD&D, including: sources and types of funds, funders and performers, and intramural and extramural RD&D. The REE mission within the USDA serves many diverse functions and is detailed below in **Box 1**.

Section I: Preview

- Research, Education, and Economic priorities of USDA and their relevance to CDR
- Structure and process of funding for USDA agencies and offices
- Organizational structure of USDA offices, agencies, and leadership
- Comparative assessment of public and private funding for agricultural R&D
- Summary of intramural and extramural research institutions and agencies supported by USDA
- Summary of development, demonstration, and deployment activities funded and performed by USDA

Additionally, this section assesses how carbon dioxide removal (CDR) could be integrated into the REE functions of the USDA to advance the Chief Scientist's focus areas around renewable energy, natural resources, and environment; plant health and production; agricultural systems and technology; and agricultural economics and rural communities.

Funding for Research and Development

Currently, the two primary pieces of legislation authorizing and appropriating funds for USDA are the 2018 Agriculture Improvement Act—known colloquially as the 2018 Farm Bill—and the Agriculture, Rural Development, Food and Drug Administration, and Related Agencies Appropriations Act. Parallel to other federal departments, USDA receives funds through both the appropriations and authorization processes. Authorization laws establish, continue, or modify mandatory funds, whereas appropriations provide discretionary funding for programs that may or may not be authorized. In Sections I and II, we summarize the structure and funding of USDA agencies, as well as current and prior USDA, interagency, and related efforts to incorporate and enhance RD&D. In Section III, we consider opportunities to alter future appropriation and authorization legislation to provide additional support for RD&D relevant to CDR.

Together, the Farm Bill (**Box 2**) and the Agriculture, Rural Development, Food and Drug Administration, and Related Agencies Appropriations Act provide funds for the REE functions of USDA through intramural agencies, such as the Agricultural Research Service (ARS), the National Agricultural Statistics Service (NASS), and the Economic Research Service (ERS), which depend exclusively on federal funding. These offices carry out their RD&D functions internally through USDA staff and facilities. These two Acts also provide funding for the National Institute of Food and Agriculture (NIFA), an extramural office, which receives some additional funds from state and non-governmental institutions, but re-grants nearly all authorized and appropriated funds to institutions outside USDA to perform solicited RD&D activities, as explained below in **Box 3**. Additionally, the Under Secretary for Research, Education, and Economics, the Office of the Chief Scientist, and the Research, Education, and Economics Extension Office receive operational funding to oversee and orchestrate these offices and their programs, and ensure RD&D efforts are appropriately prioritized across the six components of the REE mission.

TABLE 1 | USDA Under Secretaries and their reporting offices, as well as their programmatic focus and funding.

Under Secretary	Agency or service	Total 2018 funding (in millions)	Science or management jurisdiction	Land use categories	Programmatic focus	Relevant CDR practices	Example programs
Under Secretary of Research, Education, and Economics	Agricultural Research Service (ARS)	$1,388	To research and deploy programs that reduce risks associated with agriculture	Cropland, pastureland, grassland, and aquaculture	Research that reduces economic and ecological risks associated with agricultural production	Agricultural soils Rangeland soils Improved wood utilization Land sparing and intensification (+BECCS/BEBCS) Improved forest management	Soil and Air Program, Sustainable Agricultural Systems Research Program, Grass, Forage, and Rangeland Agroecosystems Program
	Economic Research Service (ERS)	$87	To conduct objective economic research to inform and enhance public and private decision making	Cropland, aquaculture, and pastureland	Research that improves decision making capacity around economic issues associated with agricultural production	Land sparing and intensification Rangeland soils Agricultural soils	Resource and Rural Economics Program, Information Technology Services, and Agricultural Resource Management Survey
	National Institute of Food and Agriculture (NIFA)	$1,564	To provide leadership and funding for initiatives that ensure the long-term viability of agriculture	Cropland, grassland, and pastureland	Funding and programmatic support for research and education programs that enhance the sustainability of agriculture	Agricultural soils Land sparing and intensification Rangeland soils Improved forest management Improved wood utilization (+BECCS/BEBCS) Urban forestry and agriculture	Sustainable Agricultural Systems, Global Change and Climate Programs, and Agriculture and Food Research Initiative
	National Agricultural Statistics Service (NASS)	$191	To provide timely and accurate statistics and data sets on nearly every aspect of agriculture	All land use types	Research to develop objective and unbiased statistics on agriculture needed by people working in and depending upon U.S. agriculture	Land sparing and intensification Rangeland soils Agricultural soils	US Agricultural Census, Crops and Plants, Economics and Prices, and Research, Science and Technology
Under Secretary for Farm Production and Conservation	Natural Resources Conservation Service (NRCS)	$5,202	To provide farmers and ranchers with financial and technical assistance to voluntarily implement conservation practices	Cropland, rangeland, and pastureland	Financial and technical assistance to land managers implementing conservation practices, as well as decision assistance	Land sparing and intensification Rangeland soils Agricultural soils Aquatic ecosystems Improved forest management	Conservation Technical Assistance Program, Landscape Conservation Initiatives, Conservation Stewardship Program, and Environmental Quality Incentives Program
	Farm Service Agency (FSA)	$2,035	To equitably serve all farmers, ranchers, and agricultural partners through the delivery of effective, efficient agricultural programs for all Americans	Cropland, rangeland, and pastureland	Research and extension services with a diverse and multi-talented work force, dedicated to achieving an economically and environmentally sound future for American Agriculture.	Land sparing and intensification Rangeland soils Agricultural soils Improved wood utilization (+BECCS/BEBCS)	Conservation Reserve Program, Emergency Forest Restoration Program, Grassland Reserve Program, Biomass Crop Assistance Program
Under Secretary for Natural Resources and Environment	United States Forest Service (USFS)	$6,649	To sustain the health, diversity, and productivity of the nation's forests and grasslands	Forests and rangelands	Research and extension services that help to preserve the long-term viability of forestry and agriculture	Land sparing and intensification Rangeland soils Improved forest management Improved wood utilization (+BECCS/BEBCS) Urban forestry and agriculture	Forest Inventory and Analysis, Experimental Forests & Ranges, and National Forest System

We segregate land use types into crop (row-crop agriculture), pasture (seeded and heavily managed grazing lands), range (natural ecosystems in which the climax vegetation is primarily grasses), forests (ecosystems in which at least 10% of surface area is covered by trees), and aquaculture (natural or artificial). Authors' analysis, with 2018 Budget Summary data from USDA. BEBCS, Bioenergy with Biochar Systems; BECCS, Bioenergy with Carbon Capture and Storage.

Box 1 | Research, Education, and Economic priorities of the Chief Scientist.

As shown below in **Figure 1** the National Agricultural Statistics Service, the National Institute for Food and Agriculture, the Agricultural Research Service, and the Economic Research Service all report to the Chief Scientist. At USDA the Chief Scientist also serves the role of the Under Secretary for Research, Education, and Economics. The focus of the Chief Scientist is to align the research objectives and programmatic focuses of these four agencies across six focus areas within the REE mission: (1) renewable energy, natural resources, and environment; (2) food safety, nutrition, and health; (3) plant health and production; (4) animal health and production; (5) agricultural systems and technology; and (6) agricultural economics and rural communities.

Box 2 | The “Farm Bill.”

The 2018 Agriculture Improvement Act, known commonly as the “2018 Farm Bill,” is an omnibus bill composed of twelve titles, providing roughly half a trillion dollars in funding for various USDA functions over a period of 5 years. The titles include (1) Commodities, (2) Conservation, (3) Trade, (4) Nutrition, (5) Credit, (6) Rural Development, (7) Research and Extension, (8) Forestry, (9) Energy, (10) Horticulture, (11) Crop Insurance, and (12) Miscellaneous (Johnson and Monke, 2008; Chite, 2014). The bill is renewed roughly every 4 years and has gone by an array of different titles since its inception, but was originally established as the Agriculture Adjustment Act in 1933. While roughly 80% of the funds appropriated through the 2014 Farm Bill were allocated to Title IV: Nutrition, the Bill also provides billions of dollars in financial support to America’s rural constituencies through crop insurance, conservation payments, and loan support (Monke, 2018).

Box 3 | Funding: competitive and formula, intramural and extramural, and mandatory and discretionary.

Within USDA, there are “funders,” or offices without research capacities that re-grant nearly their entire budget to other institutions, and “performers,” or offices that have the capacity to perform R&D activities internally. These performers can be either intramural, meaning they operate within a USDA office, or extramural, meaning the funds are transferred to external institutions or organizations that perform the research on behalf of USDA. These funds can be competitive, meaning all qualified institutions may submit proposals to perform the solicited work, or predetermined as “formula funds” through USDA’s appropriations requests. Finally, funds provided to USDA are either mandatory or discretionary. Discretionary funds are revisited annually through the appropriations process, whereas mandatory funds are established in the Farm Bill and are fixed for the entire period of that bill. While there is significant overlap between mandatory and formula funds, some discretionary funding is disseminated through formula funds. For example, discretionary funding provided to NIFA supports formula funds to Land Grant Universities and Colleges. Conversely, some mandatory funds support competitive grant programs, such as NIFA’s Biomass Research and Development Initiative.

Although Congress has updated research and development priorities through the Farm Bill in recent years, the annual appropriations process provides the most frequent opportunity to amend these priorities.

Structure of Research Offices

As shown in **Figure 1**, all USDA Offices, Assistant Secretaries, and Under Secretaries report directly to the Secretary. Within USDA, the most relevant Under Secretaries for CDR RD&D include the Under Secretaries for Natural Resources and the Environment, Farm Production and Conservation, and Research, Education, and Economics. Below in **Figure 1**, we highlight agencies within the Chief Scientist’s REE authority, as well other agencies with related RD&D objectives, with the potential to support the advancement of CDR technologies and land management strategies.

Intramural and Extramural Funding

In 2018, REE programs received a total of \$3.04 billion with \$1,343.4 million to the ARS, \$1,407.8 million to NIFA, \$191.7 million to NASS, and \$86.8 million to ERS (Monke, 2018). Intramural organizations are primarily supported through annual federal appropriations bills, which provide funding for staff salaries, facilities, and operating expenses (Monke, 2018). Conversely, offices that support extramural research, such as NIFA receive federal funding for a small group of staff that are responsible for distributing the majority of appropriated and authorized federal funds to Land Grant Universities and Colleges, non-profits, and for-profit corporations, through both formula funds and competitive grants.

Formula funds for extramural research, originally established and primarily authorized through the Hatch and the Smith-Lever Acts, provide a base level of support to Land Grant Universities and Colleges and their associated Cooperative Extension Services (Pearson and Atucha, 2015). These institutions are also provided an opportunity to submit proposals for competitive grants which are open to all qualified organizations identified in **Box 4**, and are funded through both discretionary and mandatory funds. While funding for intramural research at USDA agencies is exclusively federal, funding for Land Grant Universities and Colleges and State Agricultural Experiment Stations (SAES) is significantly more diverse. In addition to federal formula funds, Land Grant Institutions and SAES locations also receive support through research grants and contracts from private companies, research grants from trade groups, state governments, philanthropies, and individuals, and revenue and fees from the sale of products, services, and technology licenses.

To demonstrate the scale at which USDA both funds and performs applied research, we compare public and private investment in agricultural research to that of renewable energy. As shown in **Figure 2**, public investment in RD&D for renewable energy sources exceeded that of agriculture for the first time in recent history in 2013. While renewable energy RD&D receives roughly equal support from public and private sources, agricultural research has predominately been supported by the private sector. This is significant with respect to CDR as over half of private investment in RD&D is directed toward crop and seed biotechnology alone (Fuglie et al., 2018). Since the private sector invests almost no funds in research on climate mitigation or natural resource conservation for agriculture, much of this work has been taken up by REE offices, making the USDA an ideal actor to begin work on a variety of important research questions at the intersection of CDR and agriculture (Clancy

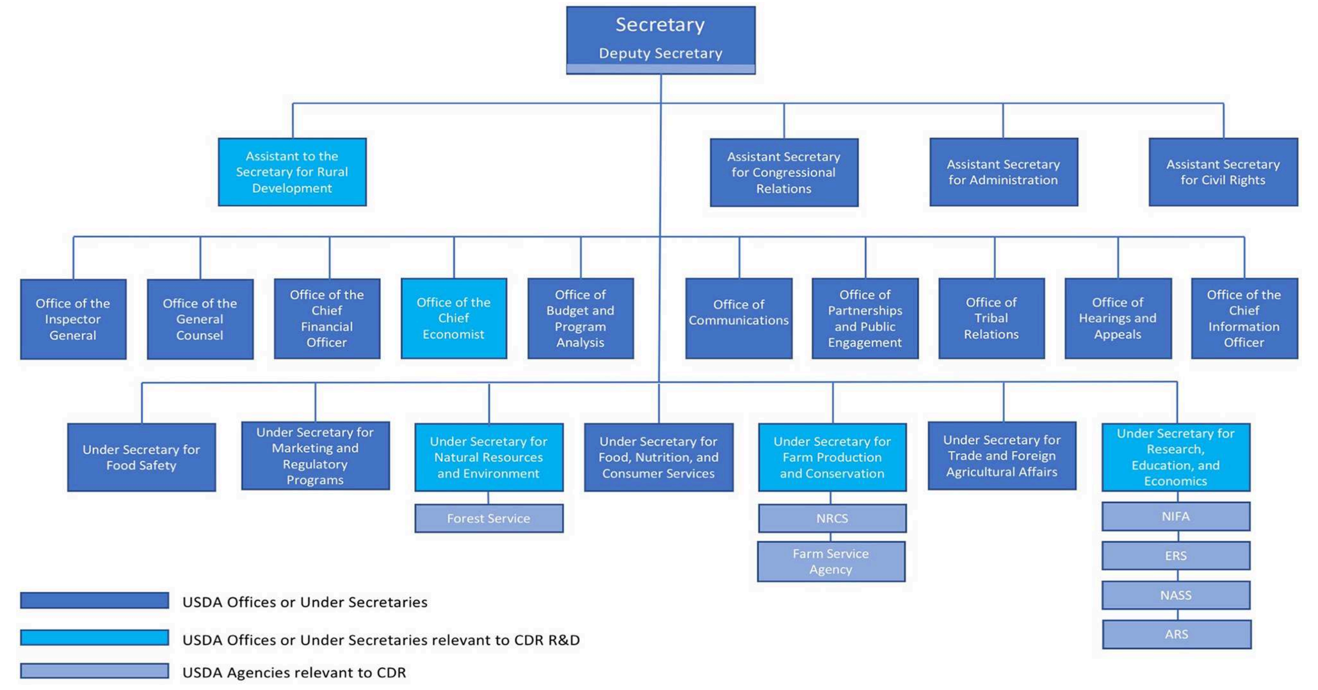

FIGURE 1 | Organizational structure of the United States Department of Agriculture, with focus on Offices and Under Secretaries with opportunities to support carbon dioxide removal (light blue), and the relevant agencies they oversee (gray-blue) (Authors' analysis of USDA Organizational Chart).

Box 4 | Eligibility for NIFA competitive grants.

Eligibility requirements for competitive grants through NIFA differ significantly across programs. The complete list of applicants eligible for at least one program includes (a) State Agricultural Experiment Station; (b) colleges and universities (including junior colleges offering associate degrees or higher); (c) university research foundations; (d) other research institutions and organizations; (e) Federal agencies; (f) national laboratories; (g) private organizations or corporations (including non-profit); (h) individuals who are U.S. citizens, nationals, or permanent residents; and (i) any group consisting of two or more entities identified in (a) through (h). Eligible institutions do not include foreign and international organizations. While not all of USDA's extramural grants have matching requirements, NIFA programs assessed in this report vary from 20 to 100% matching requirements. Notably, both Land Grant Institutions and non-Land Grant Agricultural Universities are exempt from matching requirements.

et al., 2016). Given the Under Secretary of REE's ongoing support for climate and CDR related agricultural RD&D and the private sector's minimal engagement on the topic, most agriculturally relevant CDR RD&D projects have been performed or funded within existing USDA climate and energy programs.

Despite a comparatively small budget with respect to the private sector, USDA is able to perform intramural research in addition to funding extramural research, thus leveraging additional private sector and NGO funds through matching requirements. Matching requirements for NIFA programs relevant to CDR RD&D range from 20 to 100%, and are often dependent on whether the project focuses on research, development, or demonstration. In 2009 the private sector provided 69% of all agricultural research funding, while federal and state funding contributed 21 and 10%, respectively. The utilization of these funds is relatively proportionate, with industry performing 62% of research, USDA performing 27%, and Land Grant Universities and Colleges and SAES performing 11% (Monke, 2018). Accordingly, USDA applies just over half of its REE funding to intramural research, and roughly 7% of private research funds[1] are provided to USDA intramural RD&D initiatives. In this regard, USDA is able to focus intramural efforts on research topics the private sector fails to address, while also enhancing the scale of extramural R&D through matching requirements that necessitate grantees to attract additional state, local, or private funds (Clancy et al., 2016; Fuglie et al., 2018).

Extramural and Intramural Institutions

Extramural institutions relevant to USDA's capacity to perform RD&D on CDR are the State Agricultural Experiment Sites and NIFA Land Grant Universities and Colleges (United States Department of Agriculture, 2016a). SAES sites were established through the Hatch Act in 1887, and require funds to be matched by state and local governments; however, states typically contribute several times the funds provided by USDA (Cash, 2001; Schimmelpfennig and Heisey, 2009). Federal funding for SAES locations is determined by a state's farming and rural population and funded research topics range widely

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[1] These funds consist of (a) research grants and contracts from private companies, (b) research grants from trade groups, philanthropies, and individuals, and (c) revenue and fees from the sale of products, services, and technology licenses.

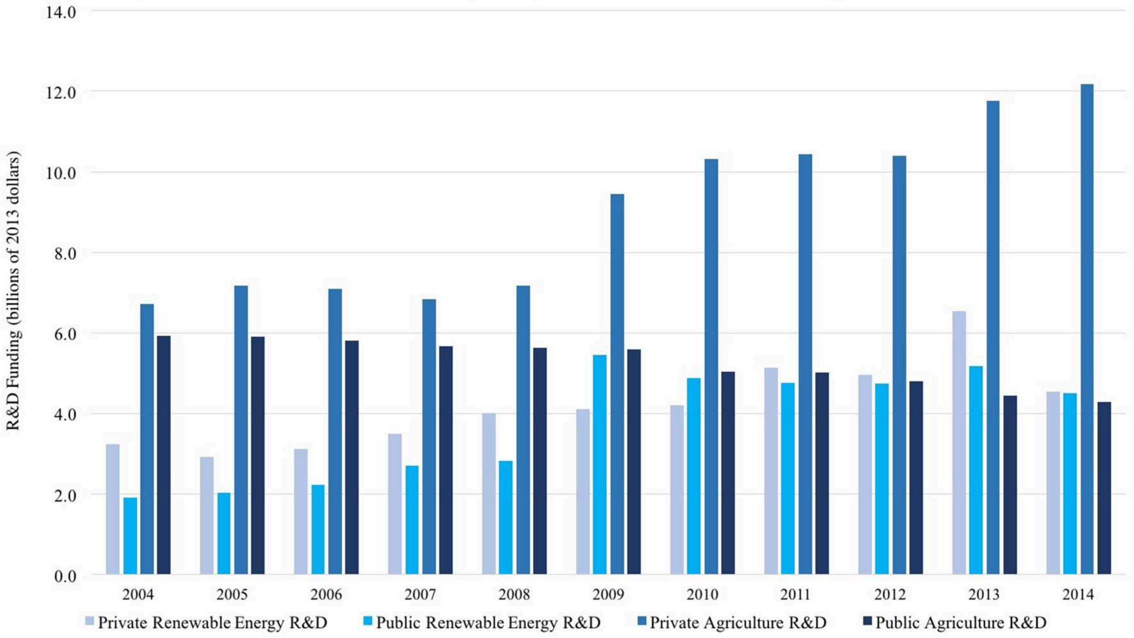

FIGURE 2 | Public and private research funding for agriculture and clean energy from 2004 to 2014 in billions of 2013 USD (Authors' analysis with data from the Economic Research Service and Bloomberg New Energy Finance).

from bioenergy crop genetics to basic carbon cycling science. Accordingly, NIFA's partnership with these localized research sites offers USDA access to and influence over roughly a hundred geographically diverse and cooperatively funded research stations.

With just over a hundred locations across the United States, Land Grant Universities and Colleges are typically paired with SAES and receive over 90% of USDA's extramural funding (King et al., 2012). The Land Grant Universities and Colleges were originally established through the Morrill Acts of 1862 and 1890, which granted federally controlled lands to states for the purpose of endowing and siting educational institutions. Since their inception, these institutions have performed research and outreach to improve the sustainability and efficiency of the agricultural sector, while also helping land manager overcome barriers to productivity.

Within the broader group of Land Grant Institutions, NIFA is also mandated through the 2014 Agriculture Improvement Act to designate Centers for Excellence[2] (COE), defined as institutions that have demonstrated exceptional efficacy at producing cost effective research, leveraging public-private partnerships, and disseminating findings to key stakeholders. Institutions identified as Centers of Excellence are provided priority when applying to funding through NIFA's competitive programs, including CDR-relevant programs, such as the Agriculture and Food Research Initiative, the Biomass Research and Development Initiative, and the Sustainable Agriculture Research and Education Program. Given the competitive nature of the funding and the wide pool of applicants, designation as a NIFA COE will be used as a tie breaker for similarly qualified applicants.

Agricultural Research Service offices, Forest Service offices, and designated USDA Climate Hubs similarly provide vital long-term scientific research in all states, however, they perform this work as intramural offices, funded directly though USDA formula funds. Particularly pertinent to CDR, the ten Forest Service and ARS offices are demarcated as USDA "Climate Hubs," intended specifically to research opportunities for agriculture and forestry operations to adapt to and mitigate the impacts of climate change. Located across the United States, USDA Climate Hubs are intra-agency ("within" agency) institutions, intended to combine expertise from ARS, USFS, and NRCS to provide regionally specific research to aid USDA technical assistance services in developing and implementing climate-informed decision-making frameworks (United States Department of Agriculture, 2016b). The Hubs are guided and overseen by an Executive Committee of senior officials within USDA, and are supported through the intramural funds provided to the associated agencies and extramural grants. These efforts are further bolstered by collaboration with the Department of Interior (DOI) and the National Oceanic and Atmospheric Administration (NOAA), making these Hubs a promising resource to harness the expertise, infrastructure, and ongoing research of multiple departments and USDA offices to research and develop opportunities to integrate CDR into agriculture and land management strategies. Given that many Climate Hubs already perform applied research on carbon sequestration in agricultural soils and vegetation, these locations could enhance and expand these efforts to develop a national network for land-use CDR.

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[2] At the time of publication, NIFA had not yet announced research sites and institutions that would be designated as Centers of Excellence.

Development, Demonstration, and Deployment

Many USDA agencies focus explicitly on or perform demonstration and deployment functions alongside applied research. Offices working on demonstration and deployment initiatives include NRCS, the Forest Service, and the Cooperative Extension Service (**Box 5**).

Similar to funding for REE objectives, funds for demonstration and deployment are appropriated through the Agriculture, Rural Development, Food and Drug Administration, and Related Agencies Appropriations Act and serve to support field office operations and competitive grant programs. Additional support is also provided through the Farm Bill in order to facilitate technical assistance services, direct conservation payments, and cost-sharing programs for land managers. Funds to NRCS and the Forest Service to provide technical assistance for conservation, demonstration activities, and conservation payments to private land owners are provided through the Conservation (II) and Forestry (VI) Titles of the Farm Bill, while demonstration and deployment payments for bioenergy programs are facilitated through the Energy Title (IX) (United States Forest Service, 2017). Each of these programs has recently been, and could further be, tailored and focused to incentivize conservation and cultivation practices that sequester carbon in soils and biomass. The Forest Service and NRCS both carry out limited research activities; however, the focus of their agencies is to provide technical assistance, education, and demonstration resources to private land owners.

Section I: Key findings

- The REE priorities of USDA are well-aligned with CDR RD&D
- Funding for REE agencies is roughly split between intramural and extramural research
- Private funding for agriculture RD&D has significantly exceed public funding in recent years
- USDA funds and operates hundreds of offices, institutions, and research sites across the U.S.
- Beyond research, USDA also funds and performs a variety of development, demonstration and deployment actives

SECTION II: CURRENT AND PRIOR USDA TERRESTRIAL AND HYBRID CDR PROGRAMS

Section II: Preview

- Existing interagency collaborations relevant to CDR RD&D
- Structure and function of federal initiatives outside of USDA relevant to CDR RD&D
- Prior administration efforts to research CDR opportunities in the agricultural sector
- Prior Department of Energy and the White House RD&D efforts relevant to CDR
- New programs or program updates supporting CDR RD&D in the 2018 Farm Bill

Below we summarize recent and current federal efforts to incorporate RD&D relevant to CDR within the programs of USDA and related departments. We focus on the recent efforts of prior administrations, led by the Office of Science and Technology Policy, DOE's Advanced Research Projects Agency-Energy (ARPA-E), and USDA's ARS and NIFA, as well as more recent legislative progress through the 2018 Farm Bill.

Box 5 | Cooperative extension service.

The Cooperative Extension Service (CES), established in 1914 through the Hatch Act, designates an obligation beyond the research and development functions of Land Grant Universities (LGU) to provide non-formal education services to empower farmers, ranchers, and communities to adapt to changing technology, prepare for and respond to emergencies, and protect the environment. As the federal partner of the Cooperative Extension System, NIFA guides and supports the development of educational priorities and curriculum through both formula and competitive funding. NIFA is responsible for leading and directing collaboration among regional offices and LGUs, in order to ensure findings from USDA research programs are disseminated to rural communities. The Service calls for leading academics at LGUs to translate scientific findings into clear language that county-level educators can teach through lessons in practical application. This work is distinct from NRCS's technical assistance function, which is intended to provide farmers and ranchers with individualized on-farm decision assistance specific to regional and practice-specific challenges.

Interagency Collaboration

While USDA collaborations with other departments are currently limited, there are model programs that demonstrate the significant value and potential of interagency initiatives. The Biomass Research and Development Initiative (BRDI), originally established through the Biomass Research and Development Act in 2000, is an interagency collaboration between USDA and DOE, facilitated through NIFA. The program offers 3 years grants from $500,000 to $2,000,000 to eligible institutions for research on the production of biofuels, bioproducts, and biopower, and includes a Technical Advisory Committee that operates as an independent body to provide direction on the focus of the program. The program aims to advance the economic competitiveness of biofuels and biopower and funds projects at all levels of the supply chain (feedstock development, biomass processing, and fuel synthesis), with a focus on technologies at the research stage. Importantly for CDR RD&D, the 2018 Farm Bill revises the program to include bioproducts (chemicals or materials derived from renewable biomass) and specifies that there must be a member on the Technical Advisory Committee with expertise on technological carbon capture and utilization.

Similarly, NIFA's Agriculture and Food Research Initiative includes a partnership with DOE's Office of Biological and Environmental Research through the Plant Feedstock Genomics for Bioenergy Program. Specifically, the program aims to fund projects that research and develop techniques to better understand and improve biomass characteristics, yield, or sustainability, water and nitrogen use efficiency.

While applications are reviewed collaboratively, awards ranging from \$200,000 to \$400,000 are made independently by each department.

Both of these programs highlight the advantageous nature of collaborative initiatives between USDA and DOE, especially on the topics of bioenergy and carbon utilization, due to DOE's longstanding leadership on genome-scale technologies, biomass conversion, and carbon capture, and USDA's expertise on crop improvement. Moreover, ongoing partnerships between NIFA and various DOE programs demonstrate NIFA's capacity to spearhead interagency collaboration through leading extramural research programs for cross-cutting research topics like CDR. Still, interagency efforts between DOE and USDA with relevance to CDR are few and limited to NIFA completive grant programs.

As discussed previously, USDA Climate Hubs perform similar interagency research, development, and education functions, providing premier locations for high-level collaboration between USDA agencies and other departments. Populated by officials and experts from NOAA, DOI, the Cooperative Extension Service (CES), and USDA offices (including ARS, NRCS, and USFS), Climate Hubs serve as geographically diverse locations to coordinate programs across offices to ensure efforts are complementary (**Figure 3**). Operating as an amalgam of the research and outreach efforts performed across USDA offices, Climate Hubs focus on synthesizing the best available science into educational and technical assistance efforts that support land managers in adjusting their production strategies to sequester carbon and adapt to changes in climate.

Terrestrial CDR Research and Development Outside of USDA

Alongside support for many USDA offices and programs, the 2014 Farm Bill established the Foundation for Food and Agriculture (FFAR) as a non-governmental non-profit foundation. The intention of FFAR is to provide increased investment in innovative partnerships and applied RD&D critical to nourishing a growing global population. Relevant to CDR, the Foundation's charter dictates that grants be made to support plant health, production, and plant products, renewable energy, natural resources and the environment, agriculture systems and technology, and agriculture economics and rural communities. With an original endowment of \$200 million, the Foundation operates across three primary programs; Fostering the Future, Challenge Areas, and Strategic Initiatives. While the Fostering the Future initiative aims to educate the agricultural researchers of the future, the Challenge Areas and Strategic Initiatives highlight specific research gaps and barriers in agriculture and fund eligible institutions to develop solutions.

Strategic Initiatives at FFAR relevant to CDR include grants to address the research challenges outlined in NASEM's *Science Breakthroughs to Advance Food and Agricultural Research by 2030* and an initiative to improve the productivity of photosynthesis. A notable success of this initiative is the recent demonstration by the Realizing Increased Photosynthetic Efficiency (RIPE) program, an FFAR grantee, of radically more efficient photosynthesis in engineered crops resulting in up to 40% increase in yield (South et al., 2019). Challenge Areas relevant to CDR include a Next Generation Crops Program and a Soil Health Program. Although the focus of these programs is increased agricultural production and resiliency, their findings could easily be applied to enhance and monitor carbon sequestration in biomass and soils.

In its brief existence, FFAR has yielded impactful advancements in a number of research topics relevant to CDR. However, the current structure and focus areas of FFAR do not effectively support CDR RD&D. For instance,

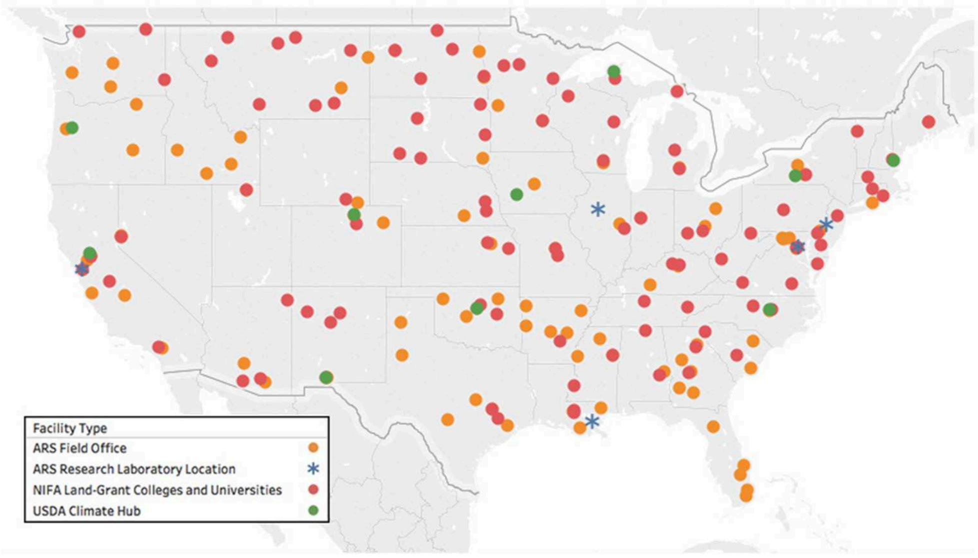

FIGURE 3 | Across the United States, USDA Climate Hubs, Land Grant Universities and Colleges, and Agricultural Research Service offices and laboratories serve as a geographically distributed network of research sites. Collectively, these facilities operate in every state, providing a diverse range of ecosystems, soil types, and climates.

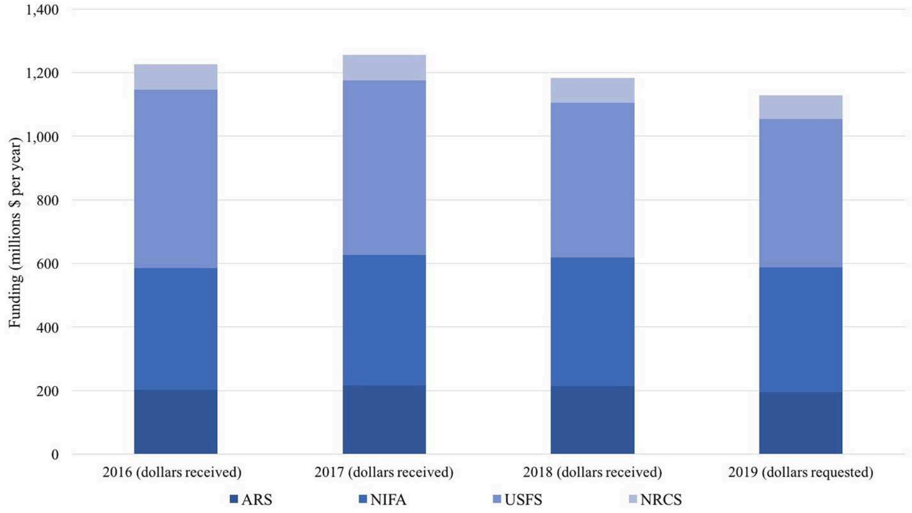

FIGURE 4 | Funding for climate and energy programs within USDA. NIFA funding includes the Agriculture and Food Research Initiative, the Sustainable Agriculture Research and Education Program, the Biomass Research and Developmental Initiative, the Biodiesel Fuel Education Program, and the Renewable Resources Extension Act. ARS funding includes the Environmental Stewardship Program. Forest Service funding includes the Forest and Rangeland R&D Program. NRCS funding includes the Soil Survey Program [Authors' analysis with data from United States Department of Agriculture (2018, 2019) Budget Justification Summaries].

FFAR has one-to-one grant matching requirements for most programs that make it difficult for applicants to qualify for funding. Additionally, as an independent foundation outside of federal jurisdiction, FFAR is not required or well-positioned to coordinate and collaborate with USDA agencies or offices, including the Office of Chief Scientist. As we discuss later in this document, FFAR's support and leadership could be augmented by DOE and USDA offices.

Similar to FFAR's strategy of supporting extramural research on topics too nascent for the private sector or intramural agencies, the DOE Advanced Research Projects Agency-Energy (ARPA-E) funds extramural research to advance high-potential, high-impact energy technologies that are too premature for private-sector investment. Although ARPA-E focuses primarily on energy technology, efforts like the Rhizosphere Observations Optimizing Terrestrial Sequestration (ROOTS) program have clear applications to agriculture and land-use CDR. The ROOTS Program, housed within ARPA-E's Transportation Fuels category, provides grants for advanced phenotyping RD&D occurring at universities and national labs across the country (Advanced Research Projects Agency, 2016). Given the program's explicit goal of developing phenotypes with enhanced capacities to store carbon dioxide in soils, the program's results have clear applications to a variety of agricultural crops and conservation practices intended to sequester carbon dioxide. Specifically, the ROOTS program aims to develop crop varieties with enhanced root structures that sequester 50% more carbon in soils, while also reducing N_2O emissions. Similarly, ARPA-E's Transportation Energy Resources from Renewable Agriculture (TERRA) program is facilitating improvement of advanced biofuel crops, specifically energy sorghum, by developing and integrating remote sensing platforms, data analytics tools, and high-throughput plant breeding technologies.

Prior Administration Efforts: Climate-Smart Agriculture

During recent administrations, USDA has been empowered to develop programs that not only adapt to, but also mitigate the risks associated with climate change. No program is more noteworthy than USDA's initiative on Climate Smart Agriculture and Forestry, developed as a domestic contribution to FAO's international Global Alliance for Climate Smart Agriculture. The initiative, unveiled in 2014 by then-Secretary Tom Vilsack, sought to develop and deploy new technologies and data systems to support land managers in a rapidly changing climate, while also reducing carbon emissions from land use and agricultural production (Sanchez et al., 2018). In 2015, USDA's *Building Blocks for Climate Smart Agriculture and Forestry* set an aggressive objective of reducing agricultural emissions by 120 million metric tons of carbon dioxide per year by 2025 (United States Department of Agriculture, 2010). With regard to CDR, the program focuses on enhanced carbon sequestration in agricultural soils, grassland and pasture stewardship, promotion of hardwood products, and improved forest management.

Funding for programs focused on climate and energy have remained relatively constant (**Figure 4**). Beyond mere road-mapping, USDA had already begun work on many of the research gaps relevant to land-use CDR. In 2009, for instance, ARS began work on series of biochar[3] trials in an effort to evaluate the efficacy of the substance as a soil amendment and water purification substrate under the USDA-ARS Biochar and Pyrolysis Initiative. The Initiative included a 2007–2010 program intended to evaluate opportunities for carbon sequestration in

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[3] Biochar is black carbon produced from biomass sources (i.e., wood chips, plant residues, manure, or other agricultural waste products) for the purpose of transforming the biomass carbon into a more stable form (carbon sequestration).

conventional and novel agricultural systems (funded at $11 million), and another ARS program from 2008 to 2011, to assess and quantify the capacity of biochar to sequester carbon and enhance crop yields (funded at $2.8 million) (United States Government Accountability Office, 2010). This work was expanded in the 2010 budget through an additional $9 million in funds to the Environmental Stewardship Program to perform research on commercially viable technologies to enhance and quantify carbon sequestration in agricultural lands (United States Department of Agriculture, 2015). Collectively, these piecemeal initiatives demonstrate progress, yet greater funding and a refocusing of programmatic structures and objectives within USDA will be required to advance these solutions toward commercial deployment.

Prior Administration Efforts: Other Programs

In recent years, the U.S. Department of Energy has also provided moderate yet meaningful levels of support for land-use CDR. For instance, in 2010, the DOE's National Energy Technology Lab (NETL) published *Best Practices for Terrestrial Sequestration of Carbon Dioxide* through the Regional Carbon Sequestration Partnership (RCSP), offering a research agenda and roadmap for improving carbon sequestration in agricultural soils, forests, and rangelands (National Energy Technology Laboratory, 2010). The report focuses on developing monitoring, verification, and accounting technologies and protocols for carbon sequestration in soils, forest biomass, and bio-based products. While the report and program were developed through DOE's RCSP, many of the technologies and protocols developed could effectively be implemented and deployed within existing USDA programs and improved through interagency efforts, such as the Biomass Research and Development Initiative (BRDI).

Similarly, DOE's Office of Science supports numerous scientific advances with application to terrestrial CDR, including Earth Systems Modeling and terrestrial ecosystem processes. In particular, Office of Science's Office of Biological and Environmental Research supports science and user facilities to achieve a predictive understanding of biological, earth, and environmental systems. The program seeks to understand the biological, biogeochemical, and physical processes that span from molecular scales to global scales that govern changes in watershed dynamics, climate, and the earth system. Yearly appropriations to the Office of Biological and Environmental Research are ∼$600 million.

In 2014, in response to the *Climate Action Plan–Strategy to Reduce Methane Emissions,* USDA, DOE, and the Environmental Protection Agency collaboratively produced the *Biogas Opportunities Roadmap.* The document outlined existing policies that could be modified to increase support for biogas production, as well as barriers to increased adoption of on-farm biogas systems. While the report aimed to highlight barriers and opportunities for biogas research and development at each of the departments, recent progress through the 2018 Farm Bill's Carbon Utilization and Biogas Education Program has put these learnings into practice. Specifically, the Biogas Education Program aims to educate agricultural producers on the energy, economic, and emissions benefits of implementing on-farm biogas systems.

In 2016, the White House Office of Science and Technology Policy (OSTP) produced a federal framework for soil science, developed in collaboration with more than a dozen other agencies. The release of OSTP's *The State and Future of U.S. Soils* for public comment began a process to established a framework to assesses and overcome three challenge and opportunity categories (land use and land cover change, unsustainable management practices, and climate change) and outlined opportunities for departments to expand or alter existing programs to overcome these challenges (White House Council on Environmental Quality, 2015). These findings helped to guide and motivate ARS to expand soil and climate research at long-term agricultural research sites, NIFA to establish new grant programs for soil health research, and the USFS to develop new models to assess and monitor carbon stocks in soils and biomass under a changing climate (White House, 2016).

Carbon Dioxide Removal in the 2018 Farm Bill

The 2018 Agriculture Improvement Act or "Farm Bill" establishes a variety of new research programs, funding opportunities, and task forces to aid the development and deployment of CDR land use strategies and technologies. Within the omnibus bill, modifications with applications to CDR fall into four main titles; Conservation (Title II), Research, Extension, and Related Matters (Title VII), Forestry (Title VIII), and Energy (Title IX). Within these new provisions, the 2018 Farm Bill supports and incentivizes research on a portfolio of CDR solutions, including land use (soils, forestry, and grazing management), hybrid (bioenergy and biogas/renewable natural gas), and technological (carbon utilization) carbon removal solutions (**Table 2**).

First, the Conservation Title makes extensive improvements to existing programs by incorporating soil carbon sequestration as an explicit criterion of assessment within subtitles, such as the Environmental Quality Incentives Program (EQIP) and the Conservation Stewardship Program (CSP). The Conservation Title, through the Soil Health Demonstration Trial Program, also mandates the quantification of soil carbon sequestration under various conservation and management practices across the country. This program also includes a demonstration component in order to educate land managers on these management and conservation practices, and reinforces the deployment incentives offered through CSP and EQIP with applied education and technical assistance.

These novel soil and conservation efforts are also well-complemented by technology focused programs in the Research and Extension Title (VII). Funded through the 2018 Farm Bill, the Agriculture Advanced Research and Development Authority (AGARDA) Pilot provides $50 million annually for grants to support the development of new technologies that help to enhance the resilience of agricultural systems in response to a changing climate and extreme weather events. The program is a component of the Office of the Chief Scientist, and is intended to be a collaborative effort between the USDA, other federal agencies, and FFAR. Six months from the enactment of

TABLE 2 | 2018 Farm Bill titles and subtitles relevant to CDR and new programs or updates to existing programs that directly support CDR RD&D (Authors' analysis of the 2018 Farm Bill Conference Report).

Title	Subtitles relevant to CDR	New CDR relevant programs or program updates
Title II Conservation	Subtitle B—Conservation Reserve Program Subtitle C—Environmental Quality Incentives Program & Conservation Stewardship Program Subtitle F—Agricultural Conservation Easement Program Subtitle G—Regional Conservation Partnership Program	On-farm conservation innovation trials for soil carbon EQIP and CSP incentives for practices that increase soil carbon Soil Health Demonstration Trial Program Advanced grazing management to increase soil carbon Soil Health and Income Protection Pilot Program
Title VII Research and Extension	Subtitle A—Agricultural Research, Extension, and Teaching Policy Act Subtitle B—Food, Agriculture, Conservation, and Trade Act of 1990 Subtitle D—Food, Conservation, and Energy Act of 2008 Subtitle F—Other Matters	Appropriate Technology Transfer for Rural Areas Program Research Equipment Grants for Land Grant Institutions Agriculture Advanced Research and Development Authority Pilot Sustainable Agriculture Technology Development and Transfer Algae Agriculture Research Program Biomass Research and Development Initiative's inclusion of Carbon Dioxide
Title VIII Forestry	Subtitle A—Cooperative Forestry Assistance Act of 1978 Subtitle B—Forest and Rangeland Renewable Resources Research Act Subtitle C—Global Climate Change Prevention Act of 1990 Subtitle D—Healthy Forests Restoration Act of 2003 Subtitle F—Forest Management	State and Private Forest Landscape Scale Restoration Program Healthy Forests Restoration Act: Carbon Sequestration Amendment Healthy Forest Reserve Program: Carbon Sequestration Amendment
Title IX Energy	Sec. 9002. Bio-based markets program Sec. 9004. Repowering assistance program Sec. 9005. Bioenergy program for advanced biofuels Sec. 9009. Feedstock flexibility Sec. 9010. Biomass Crop Assistance Program Sec. 9011. Carbon utilization and biogas education program	Carbon Utilization and Biogas Education Program

the 2018 Farm Bill, the Chief Scientist will select an AGARDA director that will be responsible for assembling a team to develop project solicitations as well as a strategic plan for the program. This work has the potential to fill many of the technology and knowledge gaps discussed in the Obama administration's *Building Blocks for Climate Smart Agriculture and Forestry* and the Regional Carbon Sequestration Partnership's *Best Practices for Terrestrial Sequestration of Carbon Dioxide.* The Research and Extension Title (VII) also makes important amendments to the Biomass Research and Development Initiative (BRDI), an interagency effort of USDA's NIFA and DOE's EERE. The initiative focuses on developing novel feedstocks as well as bio-products and biofuels, and through the 2018 Farm Bill, now offers funding opportunities for projects focused on the utilization or permanent sequestration of carbon dioxide. Specifically, the text authorizes BRDI to fund projects utilizing carbon dioxide that is byproduct of the production of bio-products or biofuels.

Within the Forestry Title (VII), the State and Private Forest Landscape Scale Restoration Program and the Healthy Forests Restoration and Reserve Programs now take an expansive and explicit focus on carbon sequestration, through both management and conservation practices. The modified Healthy Forests Restoration and Reserve Programs now require that funded forest management plans for land use change, vegetation treatment, or structural management include or evaluate the potential for implemented practices to impact or enhance carbon sequestration.

On the technology front, the Energy Title (IX) also makes important advancements through the Carbon Utilization and Biogas Education Program. The program authorizes $1 million per year for each of two subprograms; a program offering technical assistance and education on carbon dioxide utilization for rural communities with a focus on rural development and economic opportunity and another program offering technical assistance for the development of on-farm biogas systems. These funds will be deployed competitively to appropriate institutions that have demonstrated expertise and experience working on the technical and educational challenges surrounding these issues.

While all of these efforts are productive and praiseworthy, funding for conservation and land management programs within the Farm Bill compose a small percent of total funding, and an even smaller portion is dedicated to programs with potential applications to CDR. In the 2014 Farm Bill, the Conservation Title received 5.8% of funding ($28,165,000,000), the Research Title received 0.2% of funding ($800,000,000), the Energy Title received 0.2% of funding ($625,000,000), and the Rural Development ($218,000,000) and Forestry ($8,000,000) Titles received <0.1% of total funding (Monke, 2018). While the magnitude of these funds is significant, most of these funds have been authorized or appropriated for existing programs with little relevance to CDR. Accordingly, congressional action will likely be needed to develop new programs, or expand existing programs in order to incorporate and support CDR RD&D within USDA.

SECTION III: OPPORTUNITIES TO IMPROVE REE AT USDA

In this section, we provide recommendations to enhance USDA's capacity to perform RD&D on CDR

Section II: Key findings

- DOE and USDA currently lead multiple interagency research initiatives on CDR relevant topics
- FFAR and DOE's ARPA-E both lead initiatives dedicated to enhancing soil carbon sequestration
- Prior administration efforts to establish land-use CDR research have been largely discontinued
- OSTP and DOE established a number of CDR relevant initiatives that could be reinstated
- The 2018 Farm Bill establishes a variety of new RD&D programs and provisions for land-use and engineered CDR

Section III: Preview

- Strategies for USDA to lead inter-agency and intra-agency efforts for CDR RD&D
- A strategy for OSTP to reinitiate, coordinate, and expand its prior research efforts on CDR
- Recommendations to improve the alignment and coordination of FARR and USDA programs relevant to CDR
- Recommendations for an integrated technology incubator program across USDA agencies and Land Grant Universities and Colleges
- Opportunities for congressional action to implement the National Academies of Sciences, Engineering, and Medicine's recommended CDR RD&D research programs
- An outline for a new research agency within USDA using the Advanced Research Projects Agency (ARPA) model

through congressional action or improved inter-agency and intra-agency coordination. We offer recommendations for increased coordination within USDA through OCS, improved collaboration between FFAR and USDA on CDR, the establishment of a new research office and technology commercialization program at USDA, and additional programs that could be authorized through congressional action.

Agency-Led Efforts to Increase CDR RD&D

USDA possesses robust intra-agency ("within" agency) and inter-agency ("between" agencies) science coordination capabilities through its Office of Chief Scientist. Notably, OCS communicates with other science-performing agencies within the federal government, including the Department of Energy. We believe that OCS is a durable framework to promote intra- and interagency coordination on CDR RD&D at USDA.

Nevertheless, several actions could increase CDR RD&D efforts within USDA. First, USDA could empower its Science Council, which advises the Secretary and Chief Scientist, to study terrestrial CDR. For instance, it could establish a team or committee to advise on coordination efforts across USDA offices, including REE offices, Forest Service, NRCS, the Farm Service Agency, and the Office of the Chief Economist (OCE), as well as the Office of Energy and Environmental Policy within OCE. Second, OCS could prioritize inter-agency coordination activities focused on CDR, including diverse agencies like the Environmental Protection Agency, and the U.S. Department of Interior. This could build on the lessons from the Biomass Research and Development Initiative, which coordinates with DOE. Finally, CDR RD&D could benefit from Secretary-level engagement. Each of these actions could coincide with a funding increase or other legislative interventions to support CDR RD&D.

White House-Led Efforts to Increase CDR RD&D

As discussed in Section II, OSTP's *The State and Future of U.S. Soils* helped to guide and motivate ARS, NIFA, and USFS to expand their soil and climate research. While a draft of the report was circulated for public comment in 2016, a finalized version of the report has not been published. OSTP or USDA could revise the report to reflect public comments and publish a final draft of the report, solidifying a research agenda to guide the Office. The impact of OSTP's framework in motivating new research demonstrates the Office's unique capacity to orchestrate interagency RD&D efforts alongside public-private partnerships, largely through the expansion and modification of existing efforts and infrastructure. Previous scholarship has emphasized the role OSTP could play in catalyzing RD&D across CDR technologies (Sanchez et al., 2018). Notably, OSTP may be able to satisfy key criteria for efficient and effective CDR RD&D, including embracing technological diversity and administrative efficiency, fostering agency buy-in, and achieving commercial deployment. Undoubtedly, USDA would play a large role in any OSTP effort to coordinate or enhance CDR RD&D.

Improved Coordination Between FFAR and USDA

As discussed in Section II, a portion of FFAR grants are explicitly intended to address the breakthrough RD&D advancements identified by NASEM in the *Science Breakthroughs to Advance Food and Agricultural Research by 2030* (Breakthroughs 2030) report. The NASEM highlight five main focus areas: (1) microbe productivity in guts and soils, (2) advancements in genetic evaluation and engineering, (3) expanding and enhancing agricultural datasets, (4) developing and improving sensors and biosensors across agriculture, and (5) using transdisciplinary analysis to develop system-wide methods to increase production (National Academies of Sciences, Engineering, and Medicine, 2018a,b). As discussed below, each of these focus areas has the ability to catalyze CDR, either through increased carbon storage in land or land-sparing through increased productivity.

First, the Breakthroughs 2030 report makes recommendations for improved analysis of the role of the microbiome in cycling carbon among other nutrients within soils, and seeks to provide a better understanding of these processes in the broader context of animal and plant interactions. Results from this research could better inform best management practices for carbon sequestration, and biologically enhance the efficiency with which microbial communities cycle and store carbon in soils. Second, with regard to genetic evaluation and engineering, it is clear that advancements in this field could complement and inform the phenotyping efforts for bioenergy feedstocks currently occurring through AFRI and BRDI programs, resulting in more productive

TABLE 3 | To define core research topics, FFAR solicited NASEM to produce the *Science Breakthroughs to Advance Food and Agriculture by 2030* report.

Science breakthroughs to advance food and agricultural research by 2030 as adopted by FFAR	Related CDR research needs identified in negative emissions technologies and reliable sequestration
The potential of microbiomes—in the animal gut, in soil, and everywhere in between—to increase efficiency and overcome obstacles in production	Soil dynamics at depth
Advancements in genetic evaluation and editing, including making the most of CRISPR and other technologies to accelerate the evolution of food production	High carbon input crop phenotypes
Expanding and analyzing the many pools of data involved in growing and producing food	Monitoring of forest stock enhancement projects A National on Farm Monitoring System Data-model platform for predicting and quantifying agricultural soil carbon removal and storage
Developing and improving sensors and biosensors across all agricultural sectors to increase productivity and better target interventions	Soil dynamics at depth Monitoring of forest stock enhancement projects A National on Farm Monitoring System
Examining, through transdisciplinary collaborations, entire systems in food production and finding the keys to adapting and transforming them to overcome challenges and increase production	Experimental network improving agricultural soil carbon processes Biochar studies Scaling carbon sequestering agricultural activities

The focus areas identified in the report are aligned with related research topics also identified by NASEM the same year in their recent Negative Emissions Technologies and Reliable Sequestration report on CDR.

and efficient bioenergy feedstocks. Additionally, results in this focus area could also support the advanced phenotyping efforts occurring through the ROOTS program at ARPA-E, supporting and expediting the creation of crops with advanced carbon sequestering root networks.

While much work is required to enhance and standardize agricultural datasets, the development of sensors and these datasets in unison will be mutually beneficial, and could improve the accuracy and accessibility of carbon stock and flux datasets. Moreover, as demonstrated in **Figure 3**, the immense number of USDA agricultural research sites could easily facilitate data collection on carbon stocks and fluxes at high geographical and temporal resolution.

As discussed in Section I, FFAR is a non-governmental foundation and is not incentivized or required to coordinate with USDA or other federal departments. **Table 3** demonstrates the substantial overlap in the Breakthroughs 2030 NASEM report commissioned by FFAR and the more recent NASEM report on CDR. Given that many of FFAR's current research priorities overlap heavily with research needed to catalyze CDR, improved coordination between USDA and FFAR could allow many of FFAR's existing programs to support significant research on CDR. Through formal or informal alterations to FFAR's mission and charter, FFAR leadership could improve coordination among USDA and FFAR research objectives to resolve jurisdictional conflicts and duplicative efforts to strengthen the impact of both organizations' initiatives relevant to CDR. By aligning the focus of FFAR Challenge Areas and Strategic Initiatives with ongoing USDA research efforts at the executive level, staff at FFAR and USDA would be able to more effectively communicate and complement each other's work, resolving disputes that could arise around duplicity and dominion. Moreover, explicit direction for improved coordination through FFAR and USDA leadership could empower FFAR grantees both in and outside of USDA to more strategically access and align with the resources provided by REE agencies. Finally, informal convening among USDA and FFAR staff could help to develop and strengthen relationships and coordination between staff at both entities.

With greater cooperation between USDA and FFAR, FFAR could explicitly incorporate CDR research within relevant programs, or establish a new program exclusively dedicated to land-use CDR within its Challenge Areas. Additionally, FFAR could decrease or eliminate matching requirements for USDA agencies and institutions exempt from NIFA matching requirements to support projects unlikely to receive additional support from the private sector. Finally, through the USDA's network of technical (ARS, FSA, and USFS), social science (ERS and NASS), and transdisciplinary (NIFA and NRCS) agencies, USDA could offer substantial resources and expertise to FFAR in integrating CDR research within their existing programs. Accordingly, FFAR could also offer USDA greater financial support to pursue research topics related to CDR through its existing programs or a new program dedicated to CDR. Finally, through increased coordination among officials at USDA and FFAR, extramural research initiatives at the department and the foundation could be structured in a complementary and mutually beneficial manner.

Improving Commercialization Support Within USDA Agencies

In order to accelerate the commercialization of the technologies and processes needed to supplement and support the R&D efforts occurring at USDA agencies, additional entrepreneurial and tech-to-market support will likely be necessary. While ARS laboratories and FFAR and NIFA grant programs have effectively delivered impactful discoveries at the research stage, these processes and technologies must rapidly scale and mature beyond the laboratory in order provide benefits to land managers. In addition to the funding provided through USDA extramural grant programs, mentorship, market intelligence, facilities, and professional development trainings could all help to accelerate the transition academic research projects into commercial technologies. In order to rapidly develop the research occurring at ARS laboratories and Land Grant Universities and Colleges into scalable technologies, researchers will need to acquire the skills necessary to secure private investment.

Specifically, proven curriculums from national lab and university technology incubator and accelerator programs could provide excellent models for the creation of a similar program embedded within REE agencies. Broadly, incubators and accelerators are structured programs intended support early stage companies and technologies in order to expedite the commercialization process. While the difference between accelerators and incubators is not well-established, incubators generally operate on an open timeline with less structured curriculum, whereas accelerators have a strict timeline and intensive curriculum (Kushner, 2018). With laboratories located across the US in close proximity to national laboratories, ARS could establish a federal technology incubator program to accelerate the maturation of promising research to commercialized technologies with the capacity for wide-spread deployment.

There is also clear precedent for the establishment of an incubator program within a federal department. DOE's Energy I-Corps, a specialized version of NSF I-Corps curriculum, pairs teams from national labs with industry mentors in order to teach researchers commercialization and business skills through condensed 2-months curriculum. The I-Corps program operates sites at dozens of universities and colleges throughout the US, providing facilities and expertise to researchers and engineers working to commercialize their technologies. An incubator positioned within USDA could offer similar opportunities through the ARS laboratories, research offices, Land Grant Universities and Colleges, and partnerships with national laboratories. Moreover, NIFA Centers of Excellence, having already demonstrated an exceptional capacity for commercialization, education and extension work, could provide valuable curriculum, expertise, and facilities for such a program. Finally, ARS laboratories could offer much needed facilities for entrepreneurs to quickly test and improve their technologies.

Several national laboratories already have incubator or accelerator programs, including Argonne National Laboratory, National Renewable Energy Laboratory, and Lawrence Berkeley National Laboratory. Notably, Cyclotron Road at Lawrence Berkeley Laboratory has demonstrated particularly strong results. Since 2015, the program has provided $15 million in financial support to 41 fellows who have gone on to attract over $80 million in support for their projects. Focused on electronics, clean power, and advanced manufacturing, the program provides entrepreneurial scientists with a 2-years fellowship that includes funding, mentorship, professional development and training, and access to university and national laboratory facilities. The Cyclotron Road model could allow ARS to recruit and mature nascent technologies crucial to measuring, increasing, and enhancing CDR deployment in agricultural, natural, and working lands in the US. A similar program within USDA could leverage a small amount funding to drastically expand the impact of ongoing intramural and extramural research occurring through the department's agencies.

Opportunities for Congressional Action on CDR RD&D

In 2018, the National Academies of Science, Engineering, and Medicine (NASEM) published *Negative Emissions Technologies and Reliable Sequestration: A Research Agenda,* assessing key knowledge gaps in the field of carbon dioxide removal. The report evaluates these gaps and recommends research projects and appropriate funding levels sufficient to address these knowledge and technology gaps. **Table 4** aligns these projects and funding recommendations along the categories of research, development, demonstration and deployment, and makes suggestions for which offices and programs could best house this research, as well as the legislative actions(s) needed to facilitate these programs. Many agencies within USDA and DOE already operate programs similar to those recommended by the National Academies; however, additional funding at the magnitude identified in the report is crucial to unlocking these advancements in a meaningful timeframe.

With an upper bound of $1.35 billion in total additional funding over the next 20 years, USDA and DOE agencies could substantially improve the United States' capacities to research, develop, demonstrate, and deploy land-use and technological carbon dioxide removal. Below, **Table 4** designates appropriate legislative actions in the forms of additional funding, improved direction, and the authorization of new programs. In the case of many NASEM recommendations, existing programs are sufficient in scope and objective to begin research on the recommended topics, however, additional funding through the appropriations process would be needed to integrate the recommended projects. In other cases, existing funding levels and programs are sufficient, but the Appropriations Committee could add language to their report directing the scope, intention, or interagency collaboration of the research projects. Finally, some recommended research projects are entirely outside the scope of existing USDA R&D efforts and entirely new programs would need to be developed to support the recommended projects.

Funding With Offices and Legislative Actions

Some of the efforts recommended by NASEM are actionable through clear direction to agencies through appropriations report language, and in some cases, existing funding is sufficient to pursue these projects. In these instances, report language can be used to explicitly direct collaboration or coordination among agencies. Still, in many other cases, NASEM's recommendations are sufficiently novel that existing programs could not reasonably pursue this research, and the establishment of additional funding or a new program through congressional action will likely be necessary. In most cases, congressional action through the appropriations and authorizations processes will be necessary to establish and fund NASEM's recommended research programs, as existing programs are either not well-suited to

TABLE 4 | National Academies of Sciences, Engineering, and Medicine's recommended research programs and associated funding levels paired with the authors' recommendations for the best agencies to house these initiatives as well as the most effective legislative actions.

	Research focus	Funding	Time frame	Offices	Legislative action
Research	High carbon input crop phenotypes	$40–50M	20 years	DOE (EERE & APRA-E) and USDA (NIFA)	Additional funding and appropriations report language
	Soil dynamics at depth	$3–4M	5 years	USDA (ARS & NIFA)	New program, appropriations report language, and additional funding
	Harvested wood preservation	$2.4M	3 years	USDA (USFS)	Additional funding
	Biochar studies	$3M	5–10 years	USDA (ARS)	Additional funding
Development	Monitoring of forest stock enhancement projects	>$5M	≥3 years	USDA (USFS)	Additional funding
	A National on Farm Monitoring System	$5M	Ongoing	USDA (NRCS & FSA)	Additional funding and appropriations report language
	Data-model platform for predicting and quantifying agricultural soil carbon removal and storage	$5M	5 years	USDA (NASS & ERS)	Additional funding and appropriations report language
Demonstration	Forest demonstration projects: increasing collection, disposal, and preservation of harvested wood; and forest restoration	$4.5M	3 years	USDA (USFS & NRCS)	Appropriations report language
	Experimental network improving agricultural soil carbon processes	$6–9M	≥12	USDA (ARS)	New program and additional funding
	Social sciences research on improving landowner responses to incentives and equity among landowner classes	$1M	3 years	USDA (ERS)	Additional funding
Deployment	Scaling carbon sequestering agricultural activities	$2M	3 years	USDA (NRCS)	Additional funding

Recommendations for report language are included when multiple agencies or offices will require direction to coordinate on a single initiative (National Academies of Sciences, Engineering, and Medicine, 2018a,b with authors' analysis of relevant offices and legislative actions).

house this research or current funding is insufficient. **Table 5** summarizes the recommended increase in appropriations for USDA Agencies or Services based on National Academies of Sciences, Engineering, and Medicine's recommended research programs and associated funding levels paired with the authors' recommendations for the best agencies to house these initiatives.

New Research Offices

Finally, USDA can benefit from enhanced research capabilities that have proven successful in other portions of the federal government. For instance, USDA does not have many authorities granted to certain offices of the DOE and U.S. Department of Defense known as the "Advanced Research Projects Agency" (ARPA) model. These include organizational flexibility on an administrative level and significant authority given to program directors to design programs, select projects, and actively management projects (Azoulay et al., 2019). Some, but not all, of these authorities have been granted to the Advanced Agriculture Research and Development Authority (AGARDA) Pilot program in the 2018 Farm Bill, as described above. Moving forward, this Agency can evolve from the AGARDA Pilot.

Below, we propose an independent research office within USDA to focus on CDR and other climate-related research. We describe the goals, means, role of the director, personnel, and coordination authorities of a new research office, based largely off of legislation establishing ARPA-E within DOE (Gordon, 2007):

- **Goals**: The new office should focus on two primary goals: (1) to overcome the long-term and high-risk technological barriers in the development of agricultural and land management technologies related to climate change and CDR, and (2) to ensure that the United States maintains a technological lead in developing and deploying advanced agricultural and land management technologies that increase economic opportunities.
- **Means**: Much like ARPA-E, this new agency may (1) identify and promote revolutionary advances in fundamental sciences, (2) translate scientific discoveries and cutting-edge inventions into technological innovations, and (3) accelerate transformational technological advances in areas that, due to technical and financial uncertainty, industry is not likely to undertake without Federal assistance.
- **Director**: The Director should report to the Secretary and coordinate with the Chief Scientist to identify relevant scientific priorities and future trends relating to agricultural technologies. The responsibilities of the Director should include: (1) approving new programs, (2) developing funding criteria and assessing the success of programs, (3) administering funds, (4) terminating programs that are not achieving their goals, and (5) ensuring support for a diversity of agricultural practices.
- **Personnel**: Like ARPA-E, the Director should designate term-limited Program Managers. Responsibilities include: (1) establishing research and development goals for the program,

TABLE 5 | Recommended increase in appropriations for USDA Agencies or Services based on National Academies of Sciences, Engineering, and Medicine's recommended research programs and associated funding levels paired with the authors' recommendations for the best agencies to house these initiatives (Authors' analysis of USDA FY2020 Budget Summary).

Under Secretary	Agency or service	Total 2018 funding (in millions)	Relevant CDR practices	Relevant existing programs	Recommended increase in appropriations (in millions)
Under Secretary of Research, Education, and Economics	Agricultural Research Service (ARS)	$1,388	Agricultural soils Rangeland soils Improved wood utilization Land sparing and intensification (+BECCS/BEBCS) Improved forest management	Soil and Air Program, Sustainable Agricultural Systems Research Program, Grass, Forage, and Rangeland Agroecosystems Program	$14.0
	Economic Research Service (ERS)	$87	Land sparing and intensification Rangeland soils Agricultural soils	Resource and Rural Economics Program, Information Technology Services, and Agricultural Resource Management Survey	$3.5
	National Institute of Food and Agriculture (NIFA)	$1,564	Agricultural soils Land sparing and intensification Rangeland soils Improved forest management Improved wood utilization (+BECCS/BEBCS) Urban forestry and agriculture	Sustainable Agricultural Systems, Global Change and Climate Programs, Biomass, Research and Development Initiative, and Agriculture and Food Research Initiative	$27
	National Agricultural Statistics Service (NASS)	$191	Land sparing and intensification Rangeland soils Agricultural soils	US Agricultural Census, Crops and Plants, Economics and Prices, and Research, Science and Technology	$2.5
Under Secretary for Farm Production and Conservation	Natural Resources Conservation Service (NRCS)	$5,202	Land sparing and intensification Rangeland soils Agricultural soils Aquatic ecosystems Improved forest management	Conservation Technical Assistance Program, Landscape Conservation Initiatives, Conservation Stewardship Program, and Environmental Quality Incentives Program	$9.3
	Farm Service Agency (FSA)	$2,035	Land sparing and intensification Rangeland soils Agricultural soils Improved wood utilization (+BECCS/BEBCS)	Conservation Reserve Program, Emergency Forest Restoration Program, Grassland Reserve Program, Biomass Crop Assistance Program	$2.5
Under Secretary for Natural Resources and Environment	United States Forest Service (USFS)	$6,649	Land sparing and intensification Improved forest management Improved wood utilization (+BECCS/BEBCS) Urban forestry	Forest Inventory and Analysis, Experimental Forests & Ranges, and National Forest System	$12.3

Section III: Key findings

- The Office of the Chief Scientist and the Science Council could urge REE agencies to pursue CDR RD&D projects
- OSTP could catalyze and coordinate CDR RD&D across departments
- FFAR and USDA both currently fund RD&D on several topics relevant to CDR but lack coordination. Through improved communication and collaboration, the effectiveness of both entities' research efforts on CDR could be significantly improved
- USDA could leverage its facilities and technical expertise to establish a commercialization program to accelerate the development of early stage CDR technologies
- Current USDA funding is insufficient to pursue the CDR RD&D programs recommended by NASEM. Congressional action through the appropriations process could provide additional funds and direction to establish these programs within USDA agencies.
- Congress could establish a new independent research agency within USDA based on the ARPA model in order to support breakthrough research on climate and CDR

(2) soliciting applications for specific areas of particular promise, (3) building research collaborations for carrying out the program, (4) selecting projects on the basis of merit, (5) preparing technologies for an eventual transfer from lab to market, (6) monitoring the progress of projects supported under the program, (7) recommending program restructure or termination of research partnerships or whole projects.

- **Coordination**: the Agency should work with existing and new advisory committees, along with (1) the President's Council of Advisors on Science and Technology, (2) FFAR, (3) ARPA-E, (3) the Defense Advanced Research Projects Agency, and (4) other professional or scientific organizations with relevant expertise.

The authority and flexibility of this new Agency within USDA would greatly enhance REE efforts relevant to CDR RD&D within USDA.

CONCLUSION

Currently, there are a number of programs within USDA agencies performing and funding research with applications to CDR. Throughout the REE agencies (ERS, NASS, ARS, and NIFA), USFS, and NRCS, there are number of ongoing programs working to quantify, monitor, and enhance carbon storage in agricultural and working lands across the United States. However, we find that the funding for these programs is meaningfully less than the funding recommended by the National Academy of Sciences in their recent report *Negative Emissions Technologies and Reliable Sequestration: A Research Agenda*. Moreover, some of the agricultural and conservation RD&D programs recommended by the NASEM could not be reasonably incorporated within the scope of existing USDA programs.

Given array of ongoing research relevant to CDR occurring through USDA agencies, we conclude there are a number of strategies that could augment the department's capacity to fund and perform the RD&D projects recommended by NASEM. First, we find that there are number of efforts that could be led and coordinated through OCS and the associated Science Council to expand and enhance research on CDR. The OCS could request the Science Council to perform research on CDR and to coordinate this research across REE agencies, as well as the Forest Service and NRCS. The OCS could also coordinate interagency efforts among USDA agencies and external departments, such as DOI, DOE, and EPA.

There are also opportunities for the White House to lead efforts on CDR research at USDA, specifically through OSTP. We find that OSTP is well-positioned to help initiate and orchestrate interagency collaboration on CDR RD&D. OSTP could help agencies across USDA and related department to coordinate and collaborate on a number of key CDR RD&D projects identified by NASEM by using existing resources, such as the *State and Future of US Soils Report.*

Outside of USDA, we find significant overlap in the Strategic Initiatives and Challenge Areas funded by FFAR and the research needs for CDR identified by NASEM. Currently, FFAR's status as an independent foundation does not encourage collaboration between FFAR and USDA, however, through coordination by leadership of each organization, FFAR could more effectively fund USDA agencies and extramural institution to perform research on CDR. Voluntary engagement between executive leadership at USDA and FFAR could avoid duplicative efforts and could improve the effectiveness of both entities.

While greater interagency and intra-agency coordination could improve much of the ongoing RD&D relevant to CDR at USDA, still congressional action will be necessary to establish and fund many of the key CR RD&D projects identified by NASEM. We find that a number of these initiatives will require the establishment and funding of new research programs across USDA in order the make significant progress in actualizing CDR land management strategies. Through the appropriations and authorization processes, Congress can create new programs, provide additional funds to existing programs, and detail requests for collaboration or use funds through report language.

Finally, in the case of some nascent or high-risk CDR research projects, the establishment of a new office within USDA may be necessary. Given the success of the ARPA model, a similar program housed within USDA could support and help to commercialize long-term, high-risk research projects that could not otherwise be pursued by the public or private sectors. Beyond extramural funding, we also consider the potential benefits of an incubator or accelerator program housed within a USDA agency. Drawing inspiration from public programs like I-Corps and public-private partnerships like Cyclotron Road, we argue that a commercialization support program within USDA could help to accelerate the maturation of laboratory stage technologies toward deployment.

Notably, we do not recommend the establishment of a new research office or initiative exclusively for CDR. Instead, we argue CDR RD&D could more effectively be pursued at USDA by incorporating research into relevant existing programs, and adding additional programs within agencies with aligned expertise, missions, and facilities. USDA can pursue multiple strategies for increasing coordination among both USDA agencies other departments, incorporating and funding new research programs within USDA agencies, adding new USDA offices, and improving USDA coordination with FFAR. Taken together, these efforts can catalyze CDR across USDA and the federal government, driving development, demonstration, and deployment across the United States.

AUTHOR CONTRIBUTIONS

DS designed the research. RJ and DS performed the research. RJ wrote the original manuscript and created figures. DS revised the manuscript.

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GLOSSARY

2018 Farm Bill—Agriculture Improvement Act of 2018
2014 Farm Bill—Agricultural Act of 2014
AFRI—Agriculture and Food Research Initiative
ARPA-E—Advanced Research Projects Agency-Energy
ARS—Agricultural Research Service
BRDI—Biomass Research and Development Initiative
CDR—Carbon Dioxide Removal
COE—Center of Excellence
DOE—United State Department of Energy
ERS—Economic Research Service
FAO—Food and Agriculture Organization of the United Nations
FFAR—Foundation for Food and Agricultural Research
FSA—Farm Service Agency
MVA—Monitoring, Verification, and Accounting
NASEM—National Academies of Sciences, Engineering, and Medicine
NASS—National Agricultural Statistics Service
NGO—Non-Governmental Organization
NIFA—National Institute of Food and Agriculture
NIH—National Institutes of Health
NRCS—Natural Resources Conservation Service
NSF—National Science Foundation
OSTP—Office of Science and Technology Policy
R&D—Research and Development
RD&D—Research, Development, and Demonstration
REE—Research, Education, and Economics
USFS—United States Forest Service

frontiers in Climate

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Engineered CO_2 Removal, Climate Restoration, and Humility

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Over the past 200 years, humans have dramatically altered our global environmental envelope accidentally through uncontrolled greenhouse gas emissions. Humans have also developed the technology to both stop emitting greenhouse gases and ultimately to *remove* them from the atmosphere through a combination of natural and engineered pathways. Ultimately, humanity must practice CO_2 removal in addition to maximal reduction in greenhouse gas emissions through conventional mitigation to achieve net-zero greenhouse gas emissions and ultimately net-negative emissions. To accomplish this task will require enormous sums of money and substantial cooperation between groups of people who commonly do not work together: technical experts, financiers, and government officials. In addition to heightened and accelerated ambition, humility is required as well. The task requires frequent and extended achievement in arenas that many scientists and engineers commonly understand only tangentially (e.g., lawmaking, regulatory enforcement, and project finance).

Keywords: CO_2 removal, negative emissions technologies, climate policy, climate finance, public acceptance, CCS, CDR

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> *The map is not the territory.*
> -Alfred Korzybski

We have reached a startling moment in human history. Over the past 200 years, humans have dramatically altered our global environmental envelope accidentally through unbridled greenhouse gas emissions, what Roger Revelle and Hans Seuss called a grand, unplanned geophysical experiment (Revelle and Suess, 1957). Perhaps more audacious still, in the past 10 years we've developed the technology to both stop the experiment and ultimately to *undo* it.

Specifically, we've developed enough technology and learned enough through scientific inquiry to remove greenhouse gases from the air and oceans—carbon dioxide removal or CDR (National Academies of Science Engineering and Medicine, 2018). Engineered approaches to CDR include direct air capture of CO_2 (Ishimoto et al., 2017; Sandalow et al., 2018), accelerated weathering of rocks (Schuiling and Krijgsman, 2006) and bioenergy plus carbon capture and storage or BECCS (Sanchez et al., 2018; Vaughan et al., 2018). Ultimately, in order to remove the dose of combustion-related pollution from the air and oceans, humanity must practice CDR in addition to maximal reduction in greenhouse gas emissions through conventional mitigation, such as efficiency and conservation measures, deployment of near-zero carbon emitting power sources such as solar, wind, or nuclear (Smith and Friedmann, 2017).

Combined, conventional mitigation and CDR can achieve net-zero greenhouse gas emissions and ultimately net-negative emissions. This will almost certainly require both natural approaches like restoring ecosystems and reforesting large areas (Griscom et al., 2017). It will almost certainly involve engineered systems, which have the advantage of dramatic rate improvements compared to nature and dramatic footprint reductions—both of great value on a finite earth.

Recent scholarship has underscored both the magnitude of the task and the rates required to achieve net carbon removal and negative emissions (Fuss et al., 2014). Estimates for the rate of CDR range from 10 to 20 Gt/y by century end, and many gigatons by mid-century (Gasser et al., 2015; Fuss et al., 2018). The IPCC's 1.5°C report (IPCC, 2018) places the integral estimate of CO_2 removal to be from 100 to 1,000 gigatons total by the century's end. This would be additional to full global economy-wide mitigation of 85% emissions reduction by 2050, a number substantially larger than all power sector emissions.

To accomplish this task will require enormous sums of money and substantial cooperation between groups of people who commonly do not work together, namely technical experts, financiers, and government officials, each of whom sees their role and mission very differently. It behooves the scientific, environmental, and policy communities to not be cavalier about this—either the level of difficulty or the cost and how that affects other human endeavors. Two responses seem appropriate simultaneously: ambition and humility.

The harsh arithmetic of climate change demands ambition and extraordinary response, demanding innovation, research, and investment in whole new fields of knowledge (Carbon180, 2018). However, the task is much greater than the technical work alone—it requires frequent and extended achievement in arenas that many scientists and engineers only tangentially understand. Specifically, four arenas stand out: policy design and implementation, markets for products and services, project finance, and social acceptance. These additional dimensions should prompt humility and (ideally) additional ambition, given the scope of the work.

THE LIMITS OF WIZARDRY

In his book, "The Wizard & the Prophet," Mann (2018) describes two approaches to the challenge of restoring balance between man and nature, and two individuals who embodied each approach. Wizards are innovators (like Norm Borlaug, the inventor of dwarf wheat), and are prone to engineering solutions. Prophets are conservationists (like William Vogt, founder of modern environmentalism) and are prone to social solutions (e.g., regulation).

Mann asserts that wizards and prophets represent distinct approaches and tribes, commonly with very different world views and value systems. Understanding this axis of contention is essential to acknowledging the difficulty of the task of climate restoration and widespread deployment of CDR. Each tribe, wizards and prophets, believe that they are right, believe facts support them, and believe the other tribe to be naïve, foolhardy, or reckless. Despite their common goals, wizards, and prophets sometimes view each other with contempt.

Success, however, will likely require extraordinary measures wherein these tribes cooperate. The dimensions to large-scale CO_2 removal and climate restoration are daunting.

- *Net zero by 2050*: A robust finding of many integrated assessment modeling groups is that a 2°C climate stabilization trajectory requires net-zero emissions by mid-century. This extraordinary and unprecedented outcome will require a complete turnover of capital stocks for all power and heavy industry, dramatic improvements in vehicle and end-use efficiency, and immense capital deployment (IEA, 2018). Unfortunately, the long residence time for CO_2 in the atmosphere and the build-up of heat in the oceans makes this outcome insufficient to avoid the worst impacts of climate change (IPCC, 2018).
- *A trillion tons*: The 1.5°C report discussed above estimates that by century end, 100 billion−1 trillion tons of CO_2 must be removed from the air and oceans to stabilize at that target. For the great majority of these scenarios, existing natural carbon sinks lack sufficient rate, and volume to accomplish this task without augmentation or engineered CDR (Smith et al., 2016; National Academies of Science Engineering and Medicine, 2018).
- *Restoring climate*: In the Papal encyclical *Laudato Si*, Pope Francis (2015) argues the moral responsibility for climate stewardship extends beyond abatement and mitigation. To reduce human suffering and minimize global ecosystem damage, humanity must attempt to restore climate as much as practicable, requiring both exertion of technical faculties, and moral sensibilities—a case for accelerated CDR through engineered systems. To be clear, there are many potential states that could be considered "restoring climate" (e.g., just a return to pre-industrial level of atmosphere CO_2 vs. surface albedo reconstruction vs. a restoration of sea level through continental ice volumes reconstruction). Even then, some ecosystems and species are already lost or irretrievably damaged, begging the questions as to what restoration state is sufficient or required.
- *Any failure requires more CDR for success*: At present, the global economy is not on track for any of these outcomes. If mitigation is slowed for any reason (e.g., technical complexity, lack of investment), or if climate impacts accelerate and stimulate positive feedbacks (e.g., rapid polar ice collapse reducing albedo, elevated temperatures increasing wildfire impacts), the climate math demands additional CDR beyond the initial immense scale requirements. This also begs the question regarding the potential role for solar radiation management and its potential Relationship to and interaction with CDR—a topic not discussed here.

Technical success is necessary but insufficient to achieve climate stabilization. Most obviously, technical success (through government, private, and philanthropic investment) would lead to demonstrated cost reductions, making policies easier to enact since the public cost burden and level of disruption are minimized. Cost reduction alone, however, won't drive CDR. To remove CO_2 from the air and oceans at the multi-gigaton scale requires creation of new markets, trillions of dollars of investment, and global deployment (Smith and Friedmann, 2017; Sandalow et al., 2018). Policy, market forces, and wide-spread acceptance are indispensable components to achieving stabilization enabled and augmented by engineered CDR approaches.

THE ROOM WHERE IT HAPPENS

Today, the policy support for engineered CDR approaches is surprisingly robust and evolved. The US has the largest overt support through the 2018 passage of the FUTURE Act, which expanded and amended a small, existing tax credit for capture and storage of CO_2. The amendments, among other changes, explicitly included direct air capture (Energy Futures Initiative, 2018). This policy was the first to provide a government approved value of CO_2 from the air in the form of a transferable tax credit. The State of California has a separate mechanism in the form of the Low Carbon Fuel Standard, first passed into law in 2006. In 2019, it was amended in 2018 to include two provisions relevant for direct-air capture (California Air Resources Board, 2018). The first allowed synthetic fuels made from air-derived CO_2 to qualify as novel fuels for carbon crediting (strictly speaking not CDR). The second allowed for any plant that captured CO_2 from the air and stored it permanently *anywhere in the world* to qualify for carbon crediting. These policies have created new and rapidly expanding markets within the US for CDR.

Market and valuation policies can and have been augmented by additional policy support. For example, the governments of the United Kingdom (UK Natural Environmental Research Council, 2017) and Japan have created a distinct R&D program to support development of CDR technologies. In the US, several state governments are considering creating or amending "buy clean" policy mandates, which give government purchasing agencies mandates or latitude to buy low-carbon products (defined by life-cycle analysis). Some proposed legislation explicitly require states to purchase a fraction of fuels or build materials made with air-captured CO_2. Should they become law, these provisions would create markets, stimulate investment, and provide new grants for innovators, investors, and entrepreneurs.

Despite this recent progress, most nations lack policy mechanisms necessary to deploy engineered solutions to CDR. Central is the absence of a proper market for CDR services, necessary to pay engineers, or the companies that hire them. Key unresolved questions in this undefined market: who pays (e.g., tax payers, rate payers, or consumers) and how (e.g., direct govt. procurement, trading schemes, or feed-in-tariffs on goods and services).

Importantly, this lack of policy is commonly not due to an information deficit—many policy makers have received briefs on CCS, BECCS, CO2U, and even direct air capture and are versed on the viability and importance of the subject. The lack of policy mechanisms reflects in large part an inability of scientists, engineers, and practitioners to frame policy support in a context politicians can use. Overall, engagement is minimal, and communication is often laden with jargon and unduly complex. In some cases, points that scientists and engineers believe are political winners (e.g., showing leadership, creating jobs, maintain competitiveness) are not couched in sensible politics or do not differentiate themselves from similar requests from other constituents (e.g., organized labor, justice advocates, medical researchers). The community of innovators and practitioners must improve their engagement with political decision makers if they want to expand, create, or propagate technology into markets.

OTHER PEOPLE'S MONEY

Those seeking deployment of engineered CDR approaches must recognize that this will ultimately be done through a market. It is unlikely that governments will underwrite the costs completely or mandate public expense. Private capital and public & private companies will deliver solutions to these evolving markets, competing with each other for market share. The business model could be similar to the services provided by waste management and pollution control firms—following mandates and regulations, companies would offer CDR services for a fee.

Thankfully, the appetite for "impact investing," meaning investment vehicles that deliver social benefits in general and environmental benefits in specific, has grown substantially in the last few years (USSIF, 2018). Pension funds, equity firms, hedge funds, and philanthropic investment has increased substantially the amount of money and has expanded the kinds of projects that merit consideration (Global Impact Investing Network, 2016). Overall, these investors still seek substantial capital returns on investment in short time frames (3–5 years) and some investors, like pension funds, carry firm, and well defined fiduciary responsibilities. Many investors require substantial returns (10–30%) in order to merit investment, and they have many competing options for investment. As such, most investment has remained in fields like biomedicine, high technology, or conventional infrastructure. Some clean energy investments have moved only into fairly safe vehicles (e.g., guaranteed renewable power offtakes), while other opportunities such as efficiency or geothermal power have received much less focus (Reicher et al., 2017).

For these reasons and others, substantial challenges remain for CDR to receive large capital flows despite real increased interest in and enthusiasm for impact investment. The largest of these is discussed above—the lack of a market that values CDR services. However, an adequate market signal or carbon price equivalence is not necessarily sufficient. In addition, the technologies and markets are heterogeneous and complex, making it difficult for potential investors to understand the potential technical or market risks. Overcoming these challenges requires patience and dedication, and may require additional policy support to stimulate large-scale investment and capital flows into engineered CDR.

ALL THE RAGE

Even policy support and substantial investment does not necessarily guarantee uptake or propagation. Large-scale CO_2 removal will require public acceptance and support, in large part because of the scale of deployment and magnitude of capital required. Innovations, even ones that dramatically improved people's quality of life, faced substantial public, and governmental opposition (Juma, 2017). The case of CDR is harder, as it provides few immediate, tangible benefits to consumers, or citizens tied to operation. As such, acceptance and right to operate may play an outsized role in deployment.

The technical CDR community ignores the issue of public acceptance at their peril. For example, many teams are seeking pathways to increase soil uptake, BECCS yields and performance, and mineralization though genetic modification of microbial consortia or plant species. Public response to the application of GMOs has been decidedly mixed (Lucht, 2015) and in some cases has led to bans and limiting regulation. Similarly, public response to ocean fertilization experiments and solar radiation management studies has been strong and much of it negative (Abate and Greenlee, 2010; Cummins et al., 2017), greatly complicating future attempts to deploy these approaches.

An important case of social acceptance involves two geological carbon storage projects in Germany: Barendrecht & Schwartzepumpe. These two projects, one power, one industrial, were meant to herald in large-scale deployment of CCS in Europe and create new technologies (e.g., oxyfired coal boiler systems), provide international leadership, maintain jobs, and create green products for export. Both focused on CO_2 storage onshore, which became a focus for local opposition. Poor handling of public engagement at the Barendrecht project strengthened public opposition, and eventually the project was shelved. This led to further political liabilities in Germany, and the ultimate collapse of Schwatrzepumpe (Lockwood, 2017).

While there are many cases where poor public engagement led to failure, there are many stories where positive public engagement led to success (Forbes et al., 2010). In different fields and projects, lessons learned have become strategies for public engagement (Lockwood, 2017). In these cases, neutral technical arbiters (e.g., from universities or government research centers) have a key role in gaining public confidence. However, it is also the case that early engagement, listening and addressing public concerns, and creating processes for public engagement have proven important components of successful strategies.

All engineered CDR approaches are fundamentally new, leading to questions from the public. These could include cost, potential public value, local risk and safety, and ethical concerns. It is important to engage public stakeholders as scientists, practitioners, and engineers with "two ears and one mouth," so as to best address the core questions arising from public discourse.

WHY WE DO WHAT WE DO: CONCLUSIONS

In reviewing the work ahead, it's helpful to remember that the case for CDR is extremely compelling, founded in daunting and incontrovertible math and science. We do what we do first and foremost because it is necessary and because we value our progress, civilization, and the glory of the natural world. This is true regardless of how difficult the path or how vexing the societal circumstances of the undertaking. Cleaning our collective room may be unpleasant but is ultimately necessary and is the work of climate restoration.

In that context, engaging in policy, finance, markets, and society are equally necessary. The work of reducing and reversing greenhouse gas emissions is not like the work of developing a fast microchip or a new medical scanner—dealing with a tragedy of the commons involves engaging the commons.

Toward that end, scientists and engineers involved in engineered CDR should embrace the necessary ambition to make progress and wield it with humility.

They will need to listen carefully to politicians of all stripes to understand their needs and serve them in a way that's consistent with rapid deployment of CDR technologies. This requires perseverance as well as a willingness to postpone an optimal solution for an actionable one.

They will need to study and come to understand the needs of investors and business leaders. This will require trust and patience, and a certain amount of silence.

They will need to meet with public stakeholders who stand with inquiry or in opposition. This will require generosity of time and spirit, and a willingness to be positive at all times.

They will need to improve their skills at communicating with investors, policy makers, the lay public, and media. To do so will require creativity and wiliness to practice and to fail.

Because the challenge is both immense and urgent, it is essential to start today. While there may be opportunities to speed forward on a few key actions, it is most likely that most of the engagements will be slow and laborious. Ultimately, though, that's part of the work needed to succeed, and is demanded of our community. We have few choices—the work is the work.

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The author confirms being the sole contributor of this work and has approved it for publication.

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POLICY BRIEF
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Beyond "Net-Zero": A Case for Separate Targets for Emissions Reduction and Negative Emissions

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Targets and accounting for negative emissions should be explicitly set and managed separately from existing and future targets for emissions reduction. Failure to make such a separation has already hampered climate policy, exaggerating the expected future contribution of negative emissions in climate models, while also obscuring the extent and pace of the investment needed to deliver negative emissions. Separation would help minimize the negative impacts that promises and deployments of negative emissions could have on emissions reduction, arising from effects such as temporal trade-offs, excessive offsetting, and technological lock-in. Benefits for international, national, local, organizational, and sectoral planning would arise from greater clarity over the role and timing of negative emissions alongside accelerated emissions reduction.

Keywords: negative emissions technologies (NETs), carbon trading, offsetting, target-setting, climate policy

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INTRODUCTION

The United Nations Environment Assembly (UNEA) meeting in Nairobi in March 2019 was marked by resistance to a Swiss proposal to study geoengineering governance. Amongst other concerns, the US and Saudi Arabian delegations even explicitly objected to language intended to establish that geoengineering should not be seen as a substitute for accelerated emissions reductions. They argued that negative emission techniques (NETs) will and should be an *alternative*, rather than an *addition,* to emissions reductions (Emerson, 2019). The function of such technologies, the delegations suggested, would be to enable the continued exploitation of fossil fuels.

That debate marked a fundamental shift in understanding of the "mitigation deterrence" effects of carbon geoengineering (often called "moral hazard"). Until now, many researchers have suggested that greater attention to NETs would not significantly delay or deter essential emission reductions. The political stance of the US and Saudi Arabia suggests instead that such mitigation deterrence is already happening, and is actively promoted and defended by some political interests.

Some argue that it shouldn't matter *how* carbon dioxide levels are abated—whether through reductions in fossil fuel combustion, or through NETs. This may hold true if the two approaches do not interact politically in ways that might prevent their effective delivery. Yet we see clear evidence that emissions reductions can be deterred or delayed by efforts and suggestions to use NETs to sustain fossil fuel use. To have any hope of achieving a 1.5°C objective, decarbonization must be accelerated. In this context, substituting negative emissions for emissions reduction could be harmful in itself. Making promises of future negative emissions, instead of reducing emissions now, is even more risky (Fuss et al., 2014; Anderson and Peters, 2016; McLaren and Jarvis, 2018). There is an urgent need to avoid such substitutions. It is crucial to ensure that negative emissions are delivered *in addition to* rapid emissions reduction. This will require careful policy design.

This policy brief outlines a proposal for formal separation of negative emissions targets and accounting from emissions reduction. This proposal is rooted in an analysis of the prospects for effective deployment of negative emissions (or greenhouse gas removal) technologies based on expert interviews and stakeholder deliberation.

A POLICY OF SEPARATION

To avoid substitution, and hence ensure negative emissions deliver the necessary *additional* carbon removal, we suggest that targets and accounting for negative emissions should be explicitly set and managed separately from existing and future targets for emissions reduction. Targets, timetables, accounting methods and incentives could then be clearly and explicitly tailored to the different approaches and technologies involved. This principle should apply to all levels of targets: international, national, local, organizational, and sectoral.

Such separation was a central proposal emerging from stakeholder deliberations and expert interviews, involving 80 individuals (including policy makers, business people, academics, and non-governmental organization representatives) from nine countries, conducted between September 2018 and January 2019. The groups discussed politically and technologically diverse scenarios for deployment of negative emissions that spanned favorable and unfavorable contexts for climate policy, and featured several potential mechanisms of mitigation deterrence (Markusson et al., 2018; McLaren and Jarvis, 2018). Stakeholders suggested multiple policy measures to incentivise, accelerate, and underpin the practical delivery of negative emissions. Noticeably, though, all groups also either raised or broadly endorsed measures for the separation of emission reduction and negative emissions targets as a means to help ensure that promotion of carbon removal would not undermine emissions reduction. The discussions suggest that the proposed separation measures could be politically feasible, and also robust both in diverse political settings and for diverse technical options.

The following section briefly reviews how the alternative, treating negative emissions and emissions reductions as entirely fungible, is playing out in current climate politics.

THE ALTERNATIVE: HISTORIC MYOPIA AND CONTINUING CONFUSION

Negative emissions have been included in climate modeling and policy pathways for at least a decade. But until the IPCC's fifth assessment report, and to some extent still, they have been subsumed into net emissions pathways that do not reach net-zero until 2050 or later (Peters and Geden, 2017). In our interviews experts highlighted that this approach unintentionally concealed the role of NETs in model pathways prior to the achievement of net-zero, giving an impression that such technologies were an issue for the post-2050 regime only. As such, failure to separate out negative emissions has fed policy myopia over the need to incentivise NETs early. At the same time this confusing presentation meant that for some years it was unclear that the absolute quantities of negative emissions deployed in the models [especially those arising from bio-energy with carbon capture and storage (BECCS)] were much larger than could be practically or sustainably delivered (Fuss et al., 2014). Furthermore, those same model pathways validated continued delays in mitigation because the later negative emissions—whose costs were heavily discounted—appeared cheaper than accelerating mitigation in the near future (Bednar et al., 2019).

We have tended to overlook the extent to which integrated assessment models replace near-future emissions cuts with *future* negative emissions. But we also overlook the extent to which negative emissions (from enhanced sinks) *have already* substituted for emissions reductions instead of supplementing them. Yet these are not equivalent: carbon captured by sinks is vulnerable to *future leakage*, while emissions reductions have *permanent impacts* on atmospheric concentrations. Furthermore, emissions reductions foregone in the present cannot be substituted in the global cumulative carbon budget by future emissions reductions. So this strategy *increases* future reliance on negative emissions while potentially *consuming* some of the resource (land, energy, or storage capacity) needed to deliver future negative emissions (which will be required not only to reduce atmospheric concentrations of carbon dioxide, but also to counter-balance any ongoing "recalcitrant" emissions). The downsides of policies that emphasize trading carbon in offset schemes as a means to finance forest protection and tree planting, in terms of continued emissions justified by projects that may well have happened anyway (Cames et al., 2016), would become far clearer with a regime that clearly separated negative emissions from emissions reduction.

The failure to separate negative emissions from emissions reductions has impacts at other scales too. The project level accounting of *net emissions* from BECCS has contributed to a common mis-conceptualization of BECCS as an *additional* energy source with negative emissions, rather than as an "energy penalty" on already low-carbon bioenergy to deliver negative emissions. In turn this has boosted BECCS' profile in models and policy, arguably far beyond realistic appraisal. In practice, BECCS development has not been directed toward maximizing negative emissions potential, instead being used to partially offset emissions from production of ethanol-based biofuels (Sanchez et al., 2018). This has helped lock-in a form of biofuel that is sub-optimal (in carbon terms). Moreover, BECCS development has been largely driven by demand for carbon dioxide for enhanced oil recovery, where it acts as an emissions multiplier (Masnadi and Brandt, 2017). All these problems are exacerbated by the opacity of subsuming negative emissions into net carbon calculations at the project level.

Although some of these problems have been recognized, the systemic issue of non-separation has not been addressed. Today we see similar risks emerging in analysis and promises around other carbon removal approaches, including "natural" climate solutions (NCS) and direct air capture (DAC). Economists advocate carbon markets to incentivise such technologies, and modelers produce studies in which huge future removals balance carbon budgets and enable continued delay in eliminating fossil fuel use (Fuss et al., 2014; McLaren and Jarvis, 2018). Some modelers have sought to expose this and produce pathways that minimize reliance on speculative future carbon removal (Grubler et al., 2018; van Vuuren et al., 2018). However, others have introduced alternative novel NETs such as NCS (Griscom et al., 2017) and DAC (Chen and Tavoni, 2013) to balance modeled global carbon budgets (despite the risks of enabling further delay in mitigation).

It can be argued that deployments of NETs as offsets, or in carbon utilization, would help developers improve the technologies and capture economies of scale and of learning, helping make the techniques commercially more viable for future removals. But if we continue to fail to separate and value negative emissions appropriately, such deployments risk locking-in particular socio-technical configurations that sustain or encourage fossil fuel use. Examples include enhanced oil recovery, and the manufacture of synthetic fuel from DAC—which also sustains use of combustion engines and associated technologies. Similarly the inclusion of DAC as a tradable contribution to measures of fuel carbon intensity, for example in the California Air Resources Board system, risks slowing the adoption of e.g., electric vehicles. Negative emissions offsets for air travel or oil production would have the same effect, and we have already seen Heathrow airport using offsets from peat-bog restoration to contribute to its goal of carbon neutrality, and oil major ENI promising expansive afforestation to offset its operational emissions from oil and gas production. Mechanisms and narratives that portray offsetting as possible and desirable overlook risks of socio-technical lock-in, both on the *supply* side, e.g., growing emissions from air-travel, and on the *demand* side, with systems that divert carbon removed from the atmosphere into utilization (and re-release) rather than storage. Our expert interviewees cited several examples of such risks, some of which have been confirmed by more recent events, including the UNEA meeting cited above. In another pertinent example, the chief executive of Carbon Engineering recently told a Senate committee that one advantage of DAC technology was that it could be commissioned independently to offset the emissions of, for example, a coal-fired power station, rather than controlling emissions at source (Senate Committee on Environment and Public Works, 2019, p. 71).

ACTIONABLE RECOMMENDATIONS

Separation has multiple implications for climate policy. Here we offer recommendations in four areas: target definition; offsets and carbon trading; incentives; and modeling and evaluation processes. In each case we outline the implications and advantages of separation, and note potential downsides.

Firstly, then, in *target setting*, the separation approach calls for explicit separate objectives and timetables for emissions reduction and negative emissions. The currently popular policy formulation of achieving "net-zero" by a specified date (used widely by governments and activists) should therefore be unpacked and disaggregated to provide separate goals and timescales for emissions reduction and carbon removal. Using this revised formulation for nationally determined contributions under the Paris Agreement would ensure explicit evaluation of the practicality of each element, and expose any backtracking on emissions reduction.

At the global level, the net concentrations resulting from targets framed as net-zero and from separated targets might be expected to be the same. But the risks of unanticipated shortfalls appear greater with "net-zero," and could escalate at the sub-global level if governments, sectors or businesses conflate emissions cuts and negative emissions. Take agriculture. As a sector it has huge capacity to contribute to net-negative emissions, but also significant recalcitrant emissions. Some of the latter (e.g., those related to meat production) are more politically difficult to reduce than technically (in that dietary change could deliver substantial reductions). Imagine then an agriculture sector, challenged to achieve net-zero, which invests in soil carbon management and perhaps some biochar or enhanced weathering, while continuing to produce large quantities of beef. Its emissions might be somewhat reduced by adoption of renewable energy and other changes in practice and management, and largely offset by its negative emissions from soil management. However, the same sector, pressed first to minimize emissions and supported by promotion of dietary change, could cut its residual emissions dramatically, and additionally free up land for biomass production, perhaps for BECCS. In this scenario the same sector makes a significant net-negative contribution to the national or global goal. It is equally easy to imagine a state with the capability to deliver overall net-negative emissions globally, but taking a politically or economically easier path to net-zero.

Similarly we can picture a transport sector that meets a net-zero goal by buying in offsets from negative emissions to enable further increased aviation, rather than first minimizing emissions by using alternative fuels, and alternative infrastructures to minimize flying. Here constrained capacities for negative emissions get allocated as offsets for emissions that could have been directly cut, increasing the costs of future carbon removals. States which refuse to transform consumption habits (or reduce oil production) but purchase international offsets offer a similar example. This problem is exacerbated by differential responsibility. The states and corporations with the greatest cumulative historical emissions arguably have the responsibility to make larger net-negative contributions. Clarity over the distribution of emissions reductions and negative emissions is essential in making such assessments of climate justice.

Secondly, the formal separation of negative emissions would also require redesign of most *offsetting and carbon trading* systems. Such systems aim to reduce economic costs: to take carbon out of the system where it is cheapest to do so in the current market. But the conflation of negative emissions and emissions reductions can increase overall abatement cost in the long term through lock-in or sub-optimal resource allocation. Moreover, allowing the economic case to determine policy structures reduces climate policy to economic interests and exacerbates injustice. Carbon trading is easily distorted by powerful economic interests, and typically permits luxury emissions while constraining the demands of the poorest (Caney and Hepburn, 2011). Combining emissions cuts and negative emissions in the same markets offers yet another opportunity for such distortions, delaying transformative changes, locking in fossil fuel use, maintaining the political power of fossil-heavy interests, and thus institutionalizing the circumstances in which accelerated emissions cuts continue to be politically and economically expensive. In general, we would recommend not permitting negative emissions to trade in existing carbon markets. A separate market for negative emissions trading might be considered instead. Or if negative emissions were to be included in emissions reductions markets, then the market cap should be reduced in line with the anticipated contribution from negative emissions to ensure that those negative emissions supplement rather than substitute emissions reductions.

Thirdly, separation also implies a different approach to *incentives and portfolio building*. Experience suggests that we cannot simply rely on a high carbon price and offset payments to fund carbon removal (Bolton et al., 2015). But such a mechanism would anyway be unable to generate the inter-temporal financial transfers needed to deliver adequate negative emissions later in the century when the whole world is in net removals territory (Bednar et al., 2019). Separation highlights the need for appropriate financial incentives including and beyond carbon pricing. Avoiding offsetting between removal and emissions cuts would push prices higher in emissions trading markets, stimulating more rapid decarbonization than if offsets were permitted. A counter-argument is that constraints on offsetting might reduce incentives for investments in carbon removal by high-emitting businesses. Other things being equal, this may hinder some negative emissions approaches from reaching commercial viability. For example, investments by oil companies, like those recently made into Carbon Engineering, might appear less likely. But higher carbon prices should drive more abatement of emissions, as well as indirectly supporting carbon removal, and the future market opportunity for removal would become more certain if governments had to deliver credible policies for financing future removals. Enhanced clarity would highlight the need for high support rates in early development of NETs, rather than high carbon prices later in the century. In short, if burdens on business and government are largely displaced onto speculative future agents, the pressure needed to begin transformational change now is lost.

This clarity also helps incentivise portfolios of NETs—another approach seen as essential by our interviewees, yet poorly served by a carbon market vulnerable to price bubbles as finance chases policy (Cretí and Joëts, 2017). Instead, separation enables the introduction of targeted support and risk-reduction for specific NETs or outcomes, perhaps using tools like those designed to increase renewable energy capacity (Mitchell et al., 2006; Ragwitz and Steinhilber, 2014). This would focus attention on the specifics of support needed, and enable the building of effective, context-specific portfolios of NETs varying over both time and space.

Finally, the separation approach implies differences in *evaluation and assessment* methods. Carbon removal potential should be evaluated by independent groups—avoiding vested interests in continued emissions or in carbon removal technology. At a minimum, effective evaluation implies separate processes within bodies like the IPCC, or the UK's Committee on Climate Change, maybe with the equivalent of the "Chinese walls" required in financial organizations to minimize the risks of insider trading. Revised approaches to integrated assessment modeling that are explicit in how they handle and incorporate NETs will also be needed, alongside more careful and reflexive interpretation of model findings to expose the risks of mitigation deterrence.

Separation also clarifies the need for detailed reassessment of baseline assumptions regarding natural climate sinks related to land-use and oceans, given the range of NETs that seek to enhance carbon storage in forests and soils, or through enhanced weathering. If negative emissions were simply absorbed into national net targets, and without such analysis, the risks of double-counting or inappropriate attribution could be significant, yet net emissions approaches leave this gap more easily overlooked.

However, separation might exacerbate some challenges for carbon accounting, especially where techniques involve both emissions and negative emissions (such as habitat restoration and BECCS). Avoiding double counting across two distinct regimes for target setting and monitoring of progress would require careful design, and would not be politically trivial. Nonetheless, based on our stakeholder research, we conclude that such downsides are far outweighed by the potential advantages of a separation approach.

CONCLUSIONS

The arguments presented here validate previous calls for separating "gross-, and net-negative emissions" (Peters and Geden, 2017). They also suggest a need for a clearer formal separation of target-setting, incentivization, monitoring, and evaluation regimes for negative emissions.

"Going beyond net zero" means not only going further, to achieve net-negative emissions, but also reframing the challenge in ways that avoid the shortcomings of "net-zero" discussed here. Clear separation would expose interests and politics—deliberate efforts to substitute negative emissions for emissions reduction could no longer be hidden behind "net-zero" rhetoric; and the justice implications of who generates residual emissions would become clearer. Clarity would reveal both where negative emissions investment and development is inadequate, and where negative emissions (or future promises thereof) could undermine emissions reduction.

At the center of this problem is the myth that a tonne of CO_2 is just a tonne of CO_2 and therefore fungible. But a ton of CO_2 is an object (or indeed a concept) that is always inextricably embedded in technical, social, and political contexts which make different forms distinctive; for example, because of leakage risk, accounting uncertainty, systemic connection to other emissions, or economic or political interest. Climate justice has long distinguished essential from luxury emissions for both normative and substantive reasons (Shue, 1993). Today, for similar reasons, we need to distinguish negative emissions from emissions reductions.

ETHICS STATEMENT

This study was reviewed and approved by Lancaster University's Faculty of Science and Technology Research Ethics Committee. All subjects gave written informed consent in accordance with the Declaration of Helsinki.

AUTHOR CONTRIBUTIONS

All the authors collectively designed and conducted the deliberative workshops, which were facilitated by RW. DM conducted the interviews and drafted the manuscript. All authors contributed to manuscript revision and read and approved the submitted version.

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REVIEW
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Potential of CO_2-EOR for Near-Term Decarbonization

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This paper provides an overview of carbon dioxide enhanced oil recovery (CO_2-EOR) and its ability to reduce greenhouse gas (GHG) emissions (even to the point of negative emissions), the role it needs to play in the challenge of decarbonization, and the need to scale up implementation and deployment in order to meet climate goals. Limitations in current legal and regulatory frameworks for CO_2 injection are explored for both economic and environmental purposes, as well as the economic implications of combining CO_2-EOR with geologic carbon storage. Results from a recent study, which demonstrate that all CO_2-EOR operations produce negative emissions oil during the first several years of production, are analyzed in the context of the urgency of climate change mitigation. Acknowledging that fossil fuels currently provide the energy foundation upon which global societies function, and that a sudden shift in the composition of that foundation can potentially destabilize the global economy and key elements of modern society, we bring CO_2-EOR to the fore as it can supply reduced carbon oil to support the current energy foundation as it steadily transitions toward decarbonization. In order to meet this urgent transition, greater fiscal, and regulatory incentives are needed to begin scaling CO_2-EOR with storage around the globe. A viable and large-scale CO_2-EOR/storage industry depends upon significant capital investments for CO_2 capture and transportation infrastructure. Policy consistency and predictability, combined with targeted subsidies, will help to achieve this goal.

Keywords: CO_2-EOR, negative emissions, decarbonization, CCS, CCUS, carbon capture, storage

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INTRODUCTION

Carbon capture and sequestration (CCS), a technology where carbon emissions are captured at a point source, transported, and injected deep underground into a safe, permitted geologic site for long term storage, was included in the portfolio of climate change mitigation options released in 2005 by the Intergovernmental Panel on Climate Change (IPCC) (IPCC, 2005). The technology continued to be acknowledged and strongly reiterated in successive reports by the International Energy Agency (IEA) (International Energy Agency, 2008a,b, 2011, 2013).

The goal of climate change mitigation is to hold the increase of global average temperature to well-below 2°C above pre-industrial levels (Article 2, United Nations, 2015). Although not explicitly mentioned in the Paris Agreement of November 4, 2016, it is clear that extensive investments in large-scale emission reduction technologies like CCS will be required, at least during a transitional stage (Article 4, United Nations, 2015).

Part of the continued discussion of whether CCS will contribute enough to meet climate targets is the discussion around the role of carbon dioxide enhanced oil recovery (CO_2-EOR). Storing captured CO_2 through CO_2-EOR enters into a category called carbon capture, utilization and storage (CCUS), where the captured CO_2 is utilized for a commercial activity, in this case EOR, as it becomes ultimately stored.

A few large-scale CCUS projects are operational today. The Weyburn-Midale Carbon Dioxide Project located in Midale, Saskatchewan, Canada, started CO_2 injection in October 2000 and continues to produce oil from Weyburn and Midale oil fields at a rate of 14 thousand barrels of oil per day (Jensen, 2019). The CO_2 injected is captured at a lignite-fired synfuels plant in Beulah, North Dakota, U.S., and transported to the fields via a 320 km long transnational pipeline. More recently, a couple of large-scale CCUS projects emerged in the U.S. The Air Products Capture Project injects CO_2 captured at a hydrogen production facility in Port Arthur, Texas, into the West Hastings field for EOR, and the Petra Nova project injects CO_2 captured at NRG's Parish Power Plant, southwest of Houston, into the nearby West Ranch oil field, also for EOR.

Many in the environmental community have argued that CO_2-EOR only serves to prolong the use of fossil fuels and, as it produces carbon emitting oil, is not able to reduce emissions. However, a recent study by Núñez-López et al. (2019), shows that, depending on strategic operational choices, the incremental oil produced from CO_2-EOR can achieve a net carbon negative status throughout most of the life of the operation (i.e., because a large percentage of the injected CO_2 is unavoidably and permanently trapped in the subsurface, as discussed in section Associated Storage of CO_2 through EOR). The study uses a novel dynamic lifecycle analysis (d-LCA) that includes greenhouse gas (GHG) emissions at the CO_2-EOR site and downstream (including crude oil refining and refined product combustion), and is linked to an operational EOR performance model.

Other LCA studies, such as Aycaguer et al. (2001) and Fox (2009) conclude that GHG emissions generated by the combustion of the final product (e.g., gasoline) are offset by the mass of CO_2 storage within the oil reservoir. Most LCA studies for CO_2-EOR, however, are difficult to compare because the boundaries used for GHG emission accounting tend to be drawn differently across studies.

Carbon dioxide enhanced oil recovery (CO_2-EOR) has historically used more captured CO_2 than any other industrial process, and is the only commercially established carbon utilization option that provides large-scale permanent storage for captured CO_2. As opposed to carbon storage generally being seen as a waste disposal activity when done in isolation of market activities, carbon storage paired with EOR can be a profitable activity that also reduces greenhouse gas emissions.

At this point in CCS technology development and deployment, much is still uncertain. Starting CCS projects at EOR sites, where oil profits offset the cost of deployment, is the most intuitive and economically justified action at the current stage of technology development. Many of the risks associated with CCS (carbon capture and storage in deep, brine-filled porous rocks referred to as saline aquifers) are reduced with EOR-associated storage. In CO_2-EOR the (fluid) trapping and confining qualities of the deep subsurface container are well-demonstrated by the existence of the hydrocarbon accumulation. Furthermore, the historical records available to EOR projects contain valuable data that saline storage projects lack, as saline projects have not benefitted from decades of application. This data availability in EOR projects add value and accuracy to testing and monitoring results, which increases confidence in CCS. The only risk that is higher in EOR than in saline CCS is the larger density of well-penetrations, both from legacy operations and from the EOR activity. However, if operators take the necessary precautions, then the benefits are plentiful.

The long-established process of CO_2-EOR has been overseen by existing law, regulations, and standards of the oil and gas industry. Yet, standardizations and framework for monitoring, quantifying and reporting CO_2 retention within the reservoir throughout the EOR process have lagged behind. However, with emerging developments in this area and increased confidence that CO_2-EOR can be used as a legitimate greenhouse gas emission reduction technology, we may see the adoption of laws, regulations, and standards that will lead to the proliferation of this resource to meet larger climate targets.

FUNDAMENTALS OF CO_2-EOR

Oil field development is carried out in two or three recovery stages. During primary recovery, oil is produced by natural drive mechanisms (dissolved gas expansion, gas cap expansion, saline water influx) supported by the reservoir's natural energy. As reservoir fluids are extracted, the reservoir pressure declines, and so do oil production rates. To prolong the duration of primary production, pressure maintenance and fluid lifting techniques are employed. During secondary recovery, a fluid, most commonly water, is injected (a.k.a. waterflooding) not only to maintain reservoir pressure but also to displace oil toward producing (i.e., extraction) wells. On average, only 30–50% of the oil is recovered after secondary recovery and 50–70% of the oil remains in the reservoir (Stalkup, 1984). Extracting the remaining oil requires more advanced and costly technologies; consequently, reservoirs were historically abandoned at this point.

Any technique applied after secondary recovery is considered tertiary recovery (Lake et al., 2014). EOR is often considered a tertiary phase of recovery for this reason, even though it can be applied at any stage of petroleum field development. In an EOR process, the oil is recovered by the injection of a material that is not originally present in the reservoir; in the case of CO_2-EOR, carbon dioxide is the injected material.

Several physical mechanisms enhance oil production when CO_2 is introduced into the reservoir. If the technology is applied after waterflooding, the goal is to produce (extract) the mobile oil that was bypassed by water and the immobile residual oil trapped by capillary force. In the desirable case where the reservoir pressure is above the minimum miscibility pressure (MMP) and the injected CO_2 and residual oil are miscible, the physical forces holding the two phases apart (interfacial tension) effectively disappears. This promotes a mass transfer (extraction/vaporization) of light and intermediate hydrocarbons, which reduces the residual immobile oil saturation. Additionally, the CO_2 rich oil phase expands/swells regaining mobility. Mass transfer is improved at higher pressure, lower reservoir temperature and lighter oil. A reduction of mobile oil saturation can also be achieved through viscosity reduction, and pressure increase (Walsh and Lake, 1989).

Because CO_2-EOR is a displacement process, CO_2 is injected into the deep subsurface rock reservoir through an injection well to displace oil toward a production (extraction) well. CO_2 is produced along with reservoir fluids, separated at the surface, and commonly, reinjected/recycled into the reservoir. The cycle repeats throughout the operation. Although CO_2 is injected in supercritical (dense) phase, it remains significantly less viscous than reservoir fluids and thus highly mobile. When mobility contrast is high, an unstable displacement results in the form of viscous fingering (the uneven advance of CO_2 -resembling fingers in a profile image- toward a producing well), which adversely impacts oil recovery (Juanes and Blunt, 2007).

To reduce the degree of fingering and stabilize the displacement front, water is injected in alternation with CO_2 in a process called Water Alternating Gas (WAG; Caudle and Dyes, 1958). As viscous fingering is a common challenge in CO_2-EOR, over 90% of CO_2-EOR operations around the world have employed WAG techniques (Merchand, 2017). **Figure 1** shows a cross-section illustrating the CO_2-EOR displacement process, the mechanisms that enhance oil recovery, and the water-CO_2 cycles.

COMMERCIAL MOTIVATION FOR CO_2-EOR

The U.S. has an oil resource base on the order of ~600 billion barrels of original oil in place. About one-third of

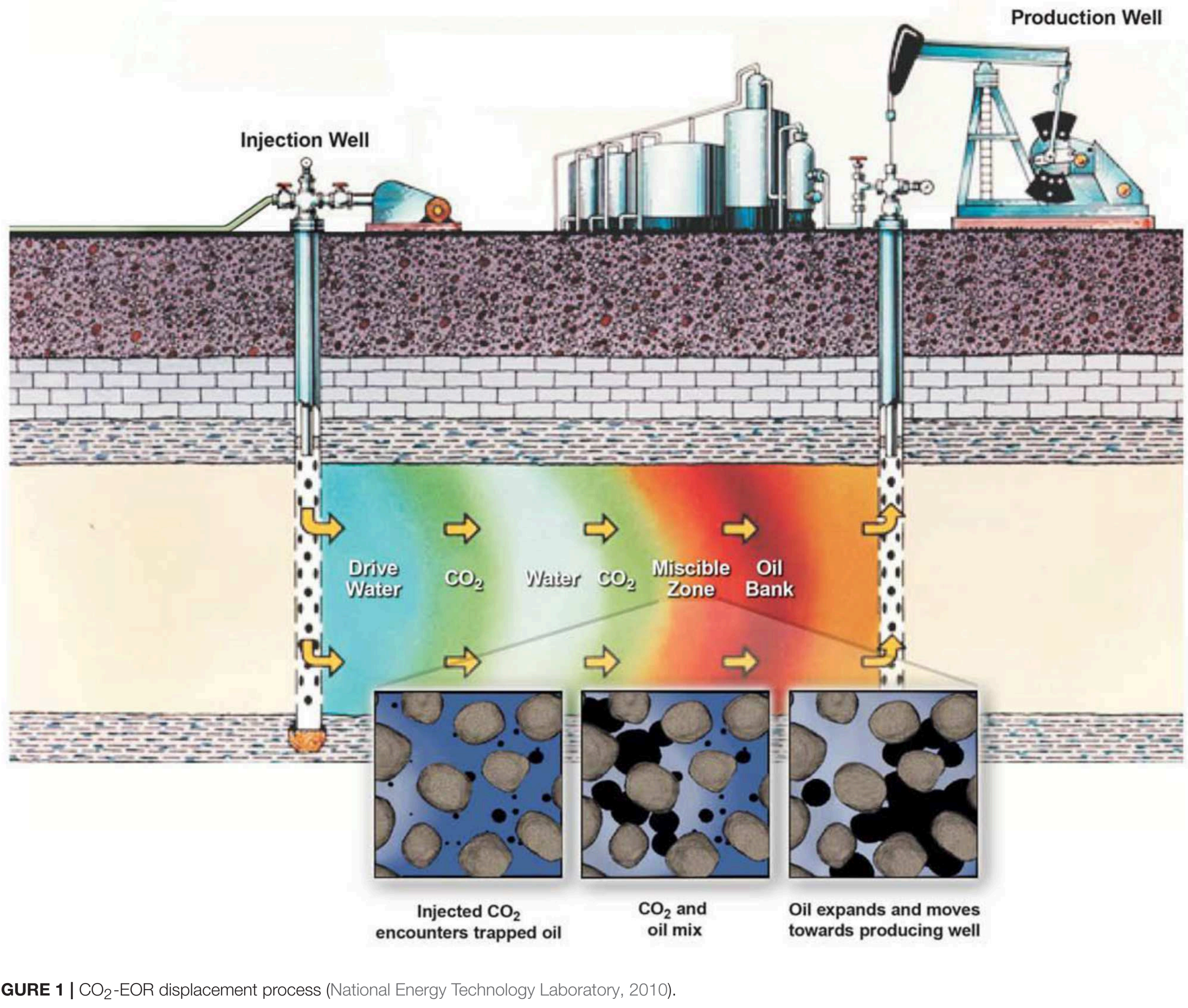

FIGURE 1 | CO_2-EOR displacement process (National Energy Technology Laboratory, 2010).

this resource base, ~200 billion barrels, has been recovered after primary and secondary oil production. This means that a significant target of ~400 billion barrels of oil are still trapped in the subsurface (Kuuskraa et al., 2009). Not all of the stranded oil can be produced or placed into proved oil reserves, and not all is amenable for CO_2-EOR technologies. The same report by Kuuskraa et al. (2009), estimates that from the ~400 billion barrels of oil remaining, ~84.8 billion barrels of oil are technically recoverable through CO_2-EOR applications. However, the CO_2-EOR market is still large and very attractive to developers. Several screening methodologies to identify candidate oil reservoirs for CO_2-EOR exist in the literature, such as Kovscek (2002), Núñez-López et al. (2008), and Bachu (2016). Some methodologies lay out the conditions for miscible displacement (lighter hydrocarbons, higher pressures, lower temperatures), others rely on the estimation of MMP and screen for reservoirs that have pressures above MMP.

Carbon dioxide enhanced oil recovery (CO_2-EOR) has been a successful commercial activity in the U.S. since 1972, when it first started in the Permian Basin, more specifically in SACROC Field, Texas. The Permian Basin, by far the most active CO_2-EOR region in the world, is located in West Texas and southeastern New Mexico. This oil producing region, the third largest in the U.S., has produced over 30 billion barrels of oil, out of which 1.3 billion have been produced with CO_2 (Merchand, 2017). Current EOR activities there produce on average 350,000 barrels of oil per day (BOPD) (the IEA estimates current world CO_2-EOR production at 450,000 BOPD). The CO_2, mostly naturally sourced, is transported to oil fields through a network of 4,500 miles of pipelines. The development of this vast CO_2 supply infrastructure in the region has resulted from the increasing CO_2 demand for EOR. CO_2 supply has not always met the demand, restricting EOR production. The latter is starting to force a change in the low-cost natural CO_2 supply trend, with forecasts including emerging industrial CO_2 capture projects.

ASSOCIATED STORAGE OF CO_2 THROUGH EOR

A large fraction of the CO_2 injected becomes trapped within the formation and cannot be produced back to the surface along with the produced reservoir fluids. In fact, the current operational experience is that 90–95% of the purchased CO_2 remains geologically trapped (Melzer, 2012). The industry refers to this volume as CO_2 retention and is considered a loss that needs to be replaced with new purchased CO_2. In CCUS, the CO_2 lost to the formation through the conventional process of EOR is referred to as associated storage.

The mechanisms that trapped oil within the reservoir also act to trap CO_2. The fact that oil was trapped over geologic time provides confidence that CO_2 will be safely trapped/stored for that long in four different forms: (1) structurally beneath an impermeable barrier, (2) residually as an immobile phase

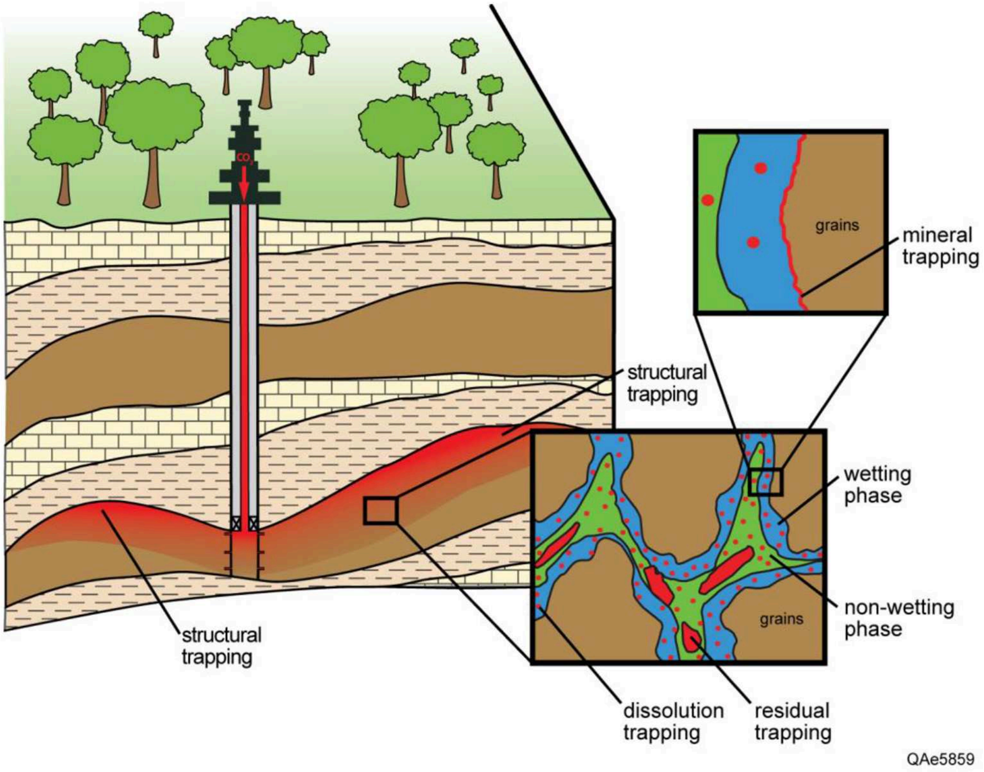

FIGURE 2 | CO_2 trapping mechanisms (Hosseininoosheri et al., 2018).

due to relative permeability and capillary curve hysteresis, (3) dissolved in reservoir brine and oil, and (4) mineralized over time (see **Figure 2**).

As CO_2 is considered a commodity within an economic activity, the primary goal of conventional CO_2-EOR is to produce more oil with less purchased CO_2. If an incentive to store CO_2 exists, through a tax credit or a carbon market, the operator can adjust its practice to achieve a second goal of storing CO_2.

The first adjustment is to switch the source of the CO_2 from natural to captured from industrial emitters. The second adjustment is in regard to operational changes needed to confirm and account for CO_2 storage, and possibly increase the amount of CO_2 use. The IEA developed three models that combine oil production with CO_2 storage: (1) Conventional EOR+: a conventional practice that maximizes oil production and minimizes CO_2 use, but uses additional monitoring and verification practices; (2) Advanced EOR+: the co-exploitation of oil recovery and CO_2 storage with larger amounts of CO_2 use under Conventional EOR+; and (3) Maximum Storage EOR+: a model focused on maximizing long-term storage of CO_2 while achieving the same level of production as Advanced EOR+ (International Energy Agency, 2015).

The use of captured CO_2 for EOR dates back to the origins of the EOR industry in the early 1970s with CO_2 captured from natural gas production (Marston, 2017). However, the use of natural CO_2 (i.e., non-anthropogenic CO_2 from the Earth's subsurface) has always been dominant. As of the mid-2010s, over three-quarters of the approximately 60 million tons of CO_2 (Godec et al., 2013) used in North American EOR operations came from a few naturally-occurring CO_2 reservoirs (Marston, 2018). With recent oil prices, the price paid today for CO_2 injection for CO_2-EOR averages $40-per-ton (Middleton, 2013; Martin et al., 2017). With prices still running about breakeven, industry does not have enough incentive for a major sourcing push from natural reservoirs to anthropogenic sources, which would increase the cost (Massachusetts Institute of Technology, 2010). Oil fields that are farther away from natural CO_2 source reservoirs have more of an incentive to use anthropogenic sources of CO_2 (Kuuskraa et al., 2009). Identifying these hot spot areas for development would further progress a likelihood that more anthropogenic-sourced and greenhouse gas-offsetting EOR projects come online.

CARBON LIFECYCLE ANALYSIS OF CCUS SYSTEMS

Even though large amounts of CO_2 are geologically stored through EOR, the extent to which the technology can reduce greenhouse gas emissions, if any at all, has been assessed by many (Jaramillo et al., 2009; Stewart and Haszeldine, 2014; Cooney, 2015). Carbon lifecycle analysis (LCA) is a systematic process, standardized in ISO 14044:2006, used to assess the environmental impact of a product system throughout the product's lifecycle, from raw materials acquisition through production, use, final treatment, recycle, and disposal. LCA applied to CO_2-EOR answers the question of whether CO_2 emissions resulting from the EOR energy consumption and, more significantly, from the combustion of the incremental oil produced, are offset by the mass of anthropogenic CO_2 stored (**Figure 3**).

The definition of boundaries in LCA studies is very important. Interpretations and comparison of results among different existing LCA studies of CCUS systems is difficult because different boundaries are used. For example, Jaramillo et al. (2009) assessed the net lifecycle emissions of CO_2-EOR in a full CCUS system boundary (cradle-to-grave), from coal mining to product combustion, and concluded that CO_2-EOR projects have historically emitted more CO_2 than they have removed through geologic storage. On the other hand, Aycaguer et al. (2001) assessed CO_2-EOR emissions at the EOR site in a gate-to-gate boundary and concluded that CO_2-EOR effectively results in net CO_2 emission reduction. **Figure 4** shows the general components of CCUS systems and the lifecycle boundaries most commonly used.

Núñez-López, first author of this paper, conducted a study that analyzed the dynamic potential of CO_2-EOR to reduce greenhouse gas emissions using a novel dynamic LCA (*d-LCA*). It was determined that a dynamic approach was needed, given that the rate of crude oil production varies significantly with time. The *d-LCA* linked instant energy demand and associated greenhouse gas (GHG) emissions to instant carbon storage mass, and provided a better understanding of the evolution of the environmental impact (CO_2 emissions) and mitigation (geologic CO_2 storage) associated with an expanded carbon CCUS system, from start to closure of operations.

The *d-LCA* was applied to an ongoing CO_2-EOR operation in Cranfield field, a 3,000 m. deep clastic reservoir in southwestern Mississippi, U.S. A scenario analysis captured oil production, CO_2 storage, and CO_2 utilization curves for four common and recent CO_2 injection strategies used in the USA: (1) Continuous gas injection (CGI), where CO_2 is injected continuously into the oil bearing formation; (2) WAG, where CO_2 slugs and brine slugs are injected in an alternating fashion to improve flood conformance and economics; (3) Water curtain injection (WCI), a continuous gas injection with peripheral water injection (commonly along the oil-water contact); and (4) Hybrid WAG + WCI (see **Figures 5**, **6**).

Results of the gate-to-grave CCUS system, in a CO_2-EOR scheme analogous to IEA's Conventional EOR+ model, indicate that all four CO_2 injection scenarios start operating with a negative carbon footprint and, after years of operation transition into operating with a positive carbon footprint (**Figure 7**). For WAG, the period of negative emissions is longest, from 14 to 18 years depending on the process technology used to separate the CO_2 from other reservoir gases at the surface before re-injection. Gas separation technologies (e.g., Ryan-Holmes, membrane, fractionation, and refrigeration) were included as variables as they are carbon intensive.

The study included a scenario analogous to IEA's Maximum Storage EOR+, where excess CO_2 from the recycling facility is injected into an underlying saline aquifer in a stacked storage fashion for long term CO_2 offtake and storage. This scenario demonstrates significant potential for improving environmental performance while providing a better understanding of how EOR

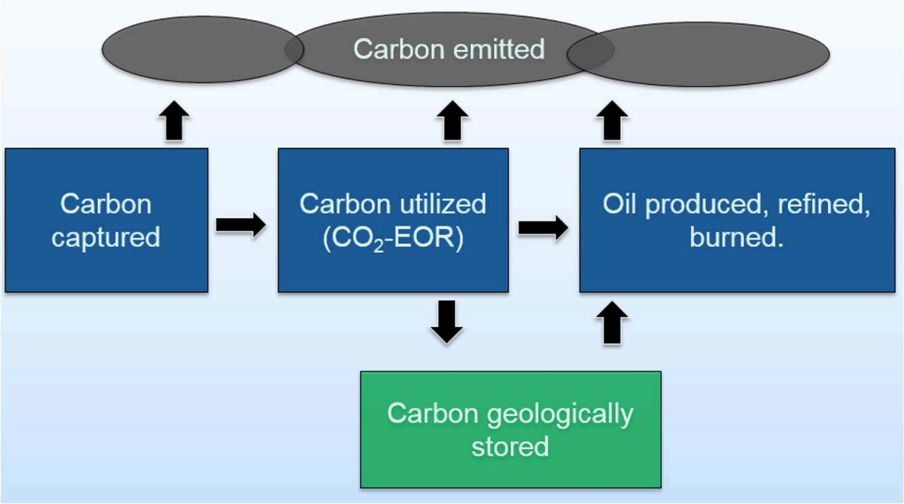

FIGURE 3 | General components of CCUS Systems.

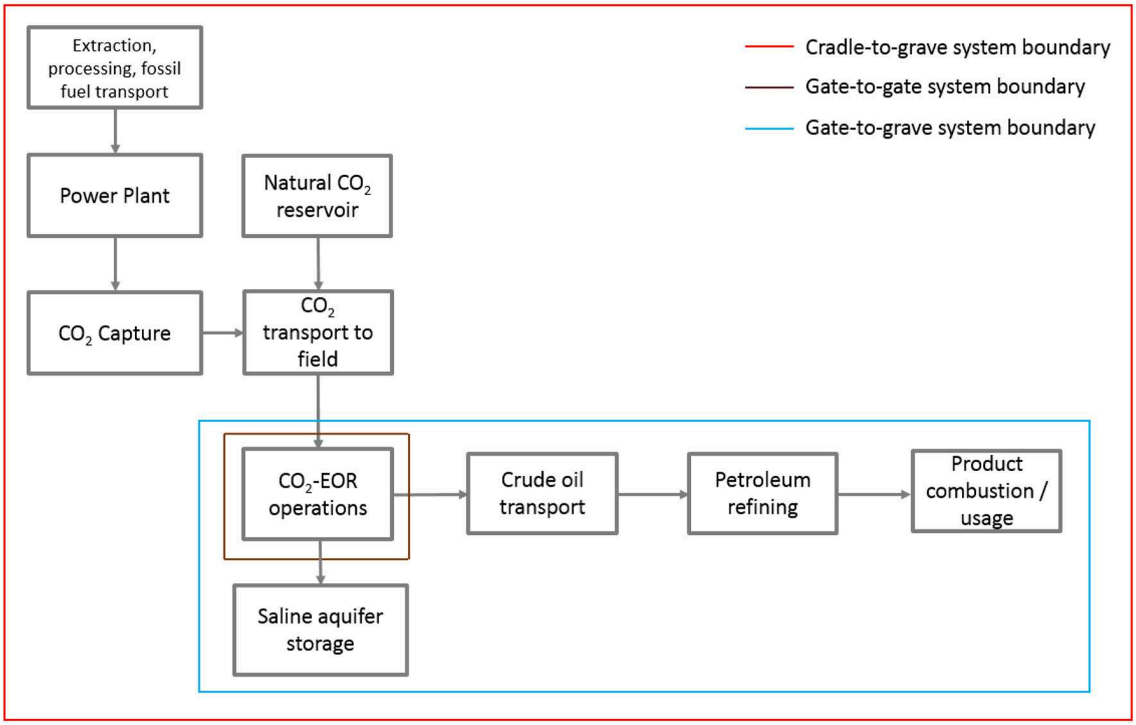

FIGURE 4 | Lifecycle analysis CCUS boundaries. Adapted from Núñez-López et al. (2019).

and saline storage can be co-managed as a CCUS project matures. Results are illustrated in **Figure 8**, where the negative emissions period is extended considerably in all scenarios. In the WAG scenario, oil can be produced with negative emissions from 18 years to potentially the entire life of the EOR project if gases are not separated at the surface.

Results like this, which demonstrate that all CO_2-EOR operations produce negative emissions oil during the first several years of production are critical in the context of the urgency of climate change mitigation. Further details of the study can be found in Núñez-López et al. (2019).

CCUS ECONOMICS

The proposition that CO_2-EOR can be a bridge to deploying CCUS on a larger scale stems from the fact that revenues

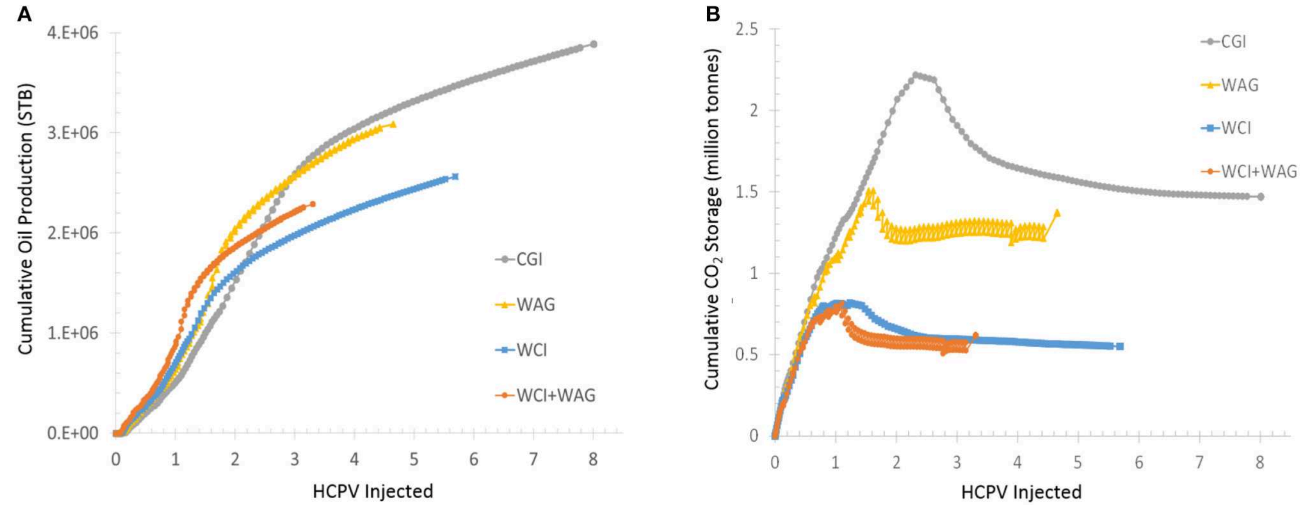

FIGURE 5 | (A) Cumulative oil production as a function of CO_2 volumes injected. **(B)** CO_2 storage as a function of CO_2 volumes injected. HCPV, hydrocarbon pore volume [intergranular pore volume occupied by hydrocarbons]. Núñez-López et al. (2019).

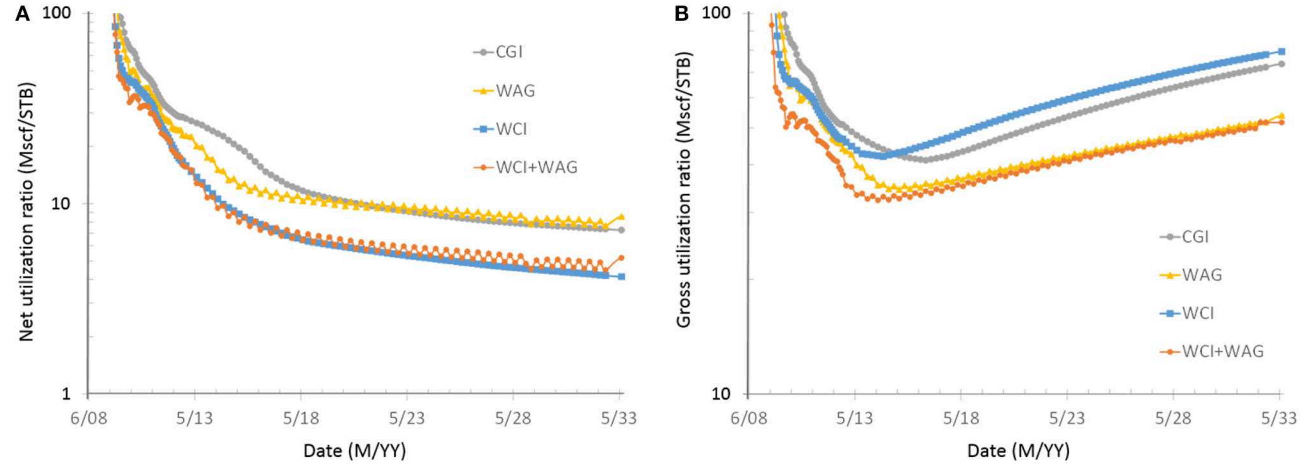

FIGURE 6 | (A) Net utilization ratio (purchased CO_2 needed to produce one barrel of oil). **(B)** Gross CO_2 utilization ratio (purchased plus recycled CO_2 needed to produce one barrel of oil). Mscf/STB, thousand standard cubic feet per standard barrel. Núñez-López et al. (2019).

associated with selling CO_2 to an EOR operator can result in substantial income to offset the cost of employing CCS for emissions-heavy industry. This fact has been cited as an opportunity to speed commercial adoption on a large enough scale to provide a means of climate mitigation (Massachusetts Institute of Technology, 2010). The revenue generated from sales of CO_2 at capture plants can be enough to offset the cost of the capture technology and transportation to an EOR site. According to the Global CCS Institute (2019), for example, this was the case at the Terrell, Enid Fertilizer, and Great Plains CCS facilities.

Determination of a reservoir to be geologically amenable to CO_2-EOR (as discussed in previous sections) does not necessarily imply that CO_2-EOR will be economical. In general, the costs of a CO_2-EOR operation vary depending on three basic extrinsic parameters: oil price, CO_2 cost, and storage tax credit (Ettehadtavakkol et al., 2014). The single largest EOR project cost is the purchase of CO_2. CO_2-related costs, including the capital costs of CO_2 supply, injection, and recycling, can amount to 25–50% of the cost per barrel of EOR oil produced (Kuuskraa et al., 2009).

For this reason, operators design the EOR flood such that the use of CO_2 is optimized and purchases are minimized. Oil reservoirs with higher capital cost requirements and less favorable CO_2 utilization rates (volume of CO_2 needed to produce one barrel of oil) will not achieve economic targets without advanced, highly efficient CO_2-EOR technology, and without tax or other fiscal incentives for storing CO_2 (National Energy Technology Laboratory, 2010). The efficiency of the CO_2 displacement process, which controls critical economic parameters such as oil production rate, CO_2 utilization rate, and CO_2 recycle ratio,

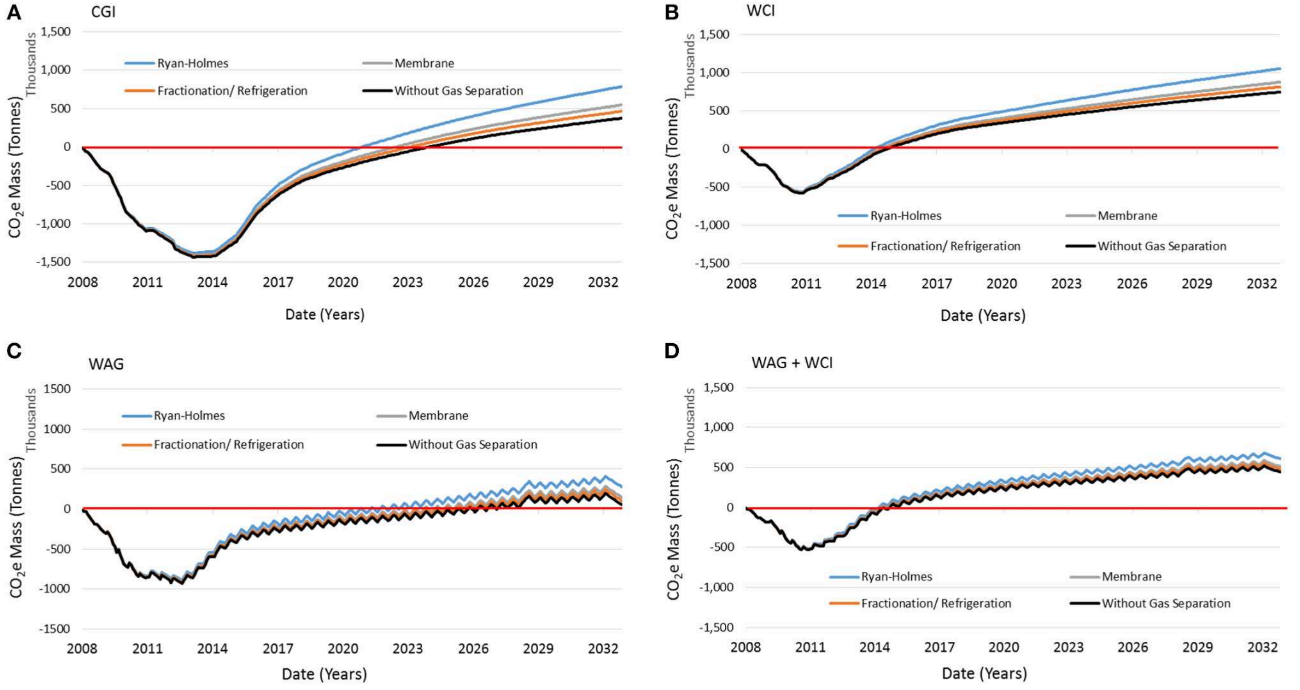

FIGURE 7 | Carbon balance (CO_2e emissions minus CO_2 storage) of the gate-to-grave CCUS system for: **(A)** continuous gas injection, **(B)** water curtain injection, **(C)** water alternating gas, and **(D)** Hybrid WAG + WCI. Núñez-López et al. (2019).

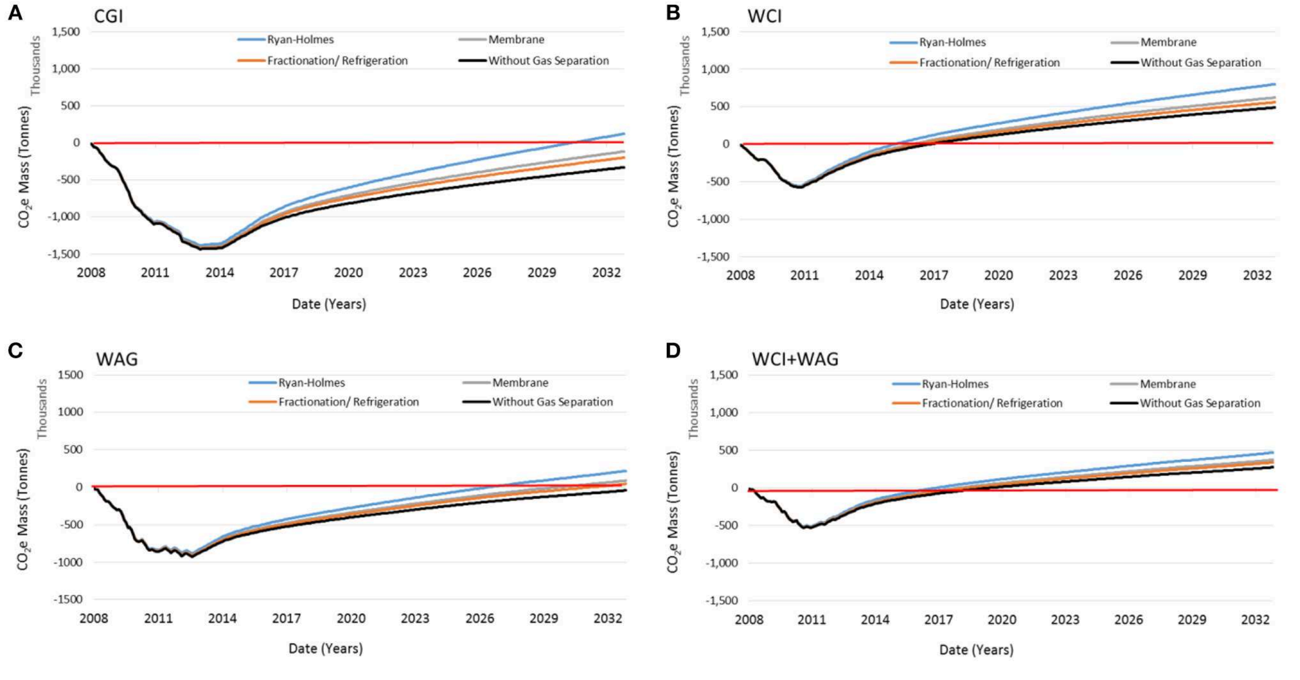

FIGURE 8 | Carbon balance (CO_2e emissions minus CO_2 storage) of the gate-to-grave CCUS system with stacked saline carbon storage for: **(A)** continuous gas injection, **(B)** water curtain injection, **(C)** water alternating gas, and **(D)** Hybrid WAG + WCI. Núñez-López et al. (2019).

is affected by reservoir rock characteristics, oil quality, production history, and other site-specific parameters (Núñez-López et al., 2019).

To transition to a large-scale CO_2-EOR with storage, operators must account for the additional costs of undertaking storage: additional monitoring, measuring, and verification

(MMV), and closure activities, which can all influence project costs. The economic costs of reducing emissions from large point sources must be smaller than the additional costs associated with storage (International Energy Agency, 2015). Though companies employing CO_2-EOR generally do not make the price of CO_2 publically available, the price of CO_2 for EOR is known to be generally linked to the price of oil. A general rule to the price an EOR operator is willing to pay is about 2% of the price of a barrel of West Texas Intermediate (WTI) oil per million cubic feet or 28,316.85 million cubic meters of CO_2 (Kuuskraa et al., 2011; Middleton, 2013). A price of \$40/tCO_2 is prevalent in the literature (Middleton, 2013; Martin et al., 2017).

There are several project design and operator choice decisions that influence the return of the CO_2-EOR with storage project. The forthcoming National Petroleum Council study report on CCUS will likely have a thorough outline of the project-associated costs of a CO_2-EOR project (Tip Meckel, personal communication). Transport and storage costs are outlined in a report from the U.S. Department of Energy (DOE) (National Energy Technology Laboratory, 2013) and the costs associated with verifying CO_2 storage during and after EOR are outlined by Godec et al. (2017). King et al. (2013) studied the cost of vertically integrated systems and found that using anthropogenic CO_2 in EOR projects generally results in a negative profit (aka financial loss), but the State CO_2-EOR Deployment Work Group have reported profit from sourcing anthropogenic CO_2 (State CO_2-EOR Deployment Work Group, 2016). Varied business models and cost data make project cost studies difficult (Jablonowski and Singh, 2010).

U.S. LEGAL AND REGULATORY FRAMEWORK

As early as the 1970s, targeted efforts by the Federal government were made to increase national security by subsidizing tertiary oil recovery to remove financial risk, rather than bring new fields into production, as a way to boost domestic production (Massachusetts Institute of Technology, 2010). Legislation like the 1976 Emergency Petroleum Allocation Act, the President's 1977 National Energy Plan, and Windfall Profits Taxes in the 1980s all supported the advancement of CO_2-EOR (Dooley et al., 2009). Incentives were codified with the U.S. Federal EOR Tax Incentive in 1986 that applied to costs associated with CO_2 flood installation, purchase, and injection, triggering a boom in CO_2-EOR national growth (National Energy Technology Laboratory, 2010).

While geologists and engineers have increasingly gained confidence in the ability of EOR to effectively store carbon dioxide, several risks, logistical, and financial, still exist for the technology in the context of CCS. The role of a legal and regulatory framework will be crucial in the scale-up of CCS for widespread, commercial deployment to reconcile complex relationships.

To date, fluctuating support and the rate of funding and project emergence has slowed deployment. To counter this, political commitments will be necessary to buoy the recent technological advancements. The first regulatory acknowledgment of the associated risks and long-term nature of CCS was in the early amendments to both the London Protocol and the OSPAR Convention (Havercroft et al., 2018). Five main models of the legislation and regulatory frameworks have emerged since then, demonstrating roadmaps for future frameworks (Havercroft et al., 2018), summarized here:

- Stand-alone legislation dealing with CCS as a stand-alone technology
- Stand-alone legislation confined to specific projects
- Adaptation or amendment of existing and familiar petroleum and gas regimes
- Mixed regimes involving a stand-alone regime coupled with significant adaptation to existing legislation
- Adaptation of existing environmental laws to develop a comprehensive CCS regime.

Carbon dioxide enhanced oil recovery (CO_2-EOR) in the U.S. has benefitted from a long history of oil and gas development, particularly in Texas, where the first large-scale CO_2-EOR project was demonstrated in the Permian Basin in the early 1970s at SACROC (Hill et al., 2013). The U.S. legal and regulatory framework for CO_2-EOR (e.g., mineral rights, subsurface data) has largely been addressed since then (Kuuskraa et al., 2011). Because the original intent of CO_2 injection was for tertiary oil production for economic benefit and not permanent storage (Kovscek and Cakici, 2005; Leach et al., 2011; Ettehadtavakkol et al., 2014), the existing U.S. framework for CO_2-EOR is largely based on existing oil and gas regulations. Emerging CCS frameworks have begun to distinguish between CO_2 injection for permanent storage and CO_2 injection for EOR as mutually-exclusive endeavors.

Many active projects benefit from policy-blend initiatives. One example is the Texas Clean Energy Project, a combination of private finance benefitting from a federal grant (e.g., D.O.E. funding), federal tax credits, and state bills (Kapetaki and Scowcroft, 2017). Because CO_2 capture and storage currently has no direct market, additional government incentives have been pursued in recent years. In February 2018, the 45Q tax credit was amended and expanded under the FUTURE Act. The incentive increased to a 10-year ramp up to \$35 and \$50 per ton of CO_2, respectively. The \$35 per ton of CO_2 was also expanded to include CO_2 utilization other than CO_2-EOR (e.g., converting captured CO_2 to fuels) and the cap of 75 million tons was removed (Zapantis et al., 2018). The 45Q expansion amendments follow many recommendations laid out by State CO_2-EOR Deployment Work Group (2016).

In 2015, the EPA issued regulatory guidance that enabled CO_2 injected during EOR to also be classed as stored CO_2 (Global CCS Institute, 2015). CO_2 injection activities associated with EOR are regulated under Class II of EPA's Underground Injection Control (UIC) Program (as opposed to Class VI wells for geologic sequestration of CO_2) and most states have primary enforcement authority—or primacy (Primary Enforcement Authority for the Underground Injection Control Program, n.d.). The EPA has released guidance on how a project can transition into a storage project under Class

VI (Environmental Protection Agency, 2013). Sites aiming to transition will need to plan for increased regulatory requirements like monitoring and verification of storage (Adelman, 2018).

Under the Clean Air Act subparts RR and UU, CO_2-EOR facilities are required by law to report greenhouse gas data to the EPA annually. Subpart RR applies to facilities that inject CO_2 for sequestration and subpart UU applies to facilities that inject CO_2 underground for any reason, including enhanced oil and gas recovery. The first edition of the non-legally-binding ISO standard 27916:2019, titled "Carbon dioxide capture, transportation and geological storage—Carbon dioxide storage using enhanced oil recovery (CO_2-EOR)," was published in January of 2019 to help operators resolve the previous complexities of accounting for long-term storage in CO_2-EOR projects, including how to account for recycling and anthropogenic CO_2 use when mixed with non-anthropogenic sources. The standard accounting does not include lifecycle emissions (International Organization for Standardization, 2019).

ROLE OF CO_2-EOR IN THE LARGER DECARBONIZATION PICTURE

The latest IPCC Special Report (IPCC, 2018) calls for the large-scale transformation of a global energy system that needs to be urgently decarbonized in order to mitigate the impending effects of climate change. Renewable energy and energy efficiency are identified as the most cost-effective pathways to achieve 90% of the emission reduction goals, with CCUS acknowledged in most models as a critical technology in the mitigation portfolio. The IEA Sustainable Development Scenario (SDS) allocates to CCUS 7% of cumulative emission reductions needed by 2040.

The need to reduce GHG emissions is unquestionable, but the subject of decarbonization of energy systems is complex, as developed and developing societies still rely on fossil fuels. Some developing countries justifiably resist carrying out the costs of decarbonization, not having contributed to the global problem as developed nations have. At the same time, developed nations resist bearing those costs alone in a competitive global economy.

According to IEA's Key World Energy Statistics (International Energy Agency, 2018) wind and solar still supply <5% of the world's energy, with fossil fuels continuing to supply a steady 80%. Unquestionably, fossil fuels currently provide the energy foundation upon which global societies function, and any sudden shift in the composition of that foundation can potentially destabilize the global economy and key elements of modern society.

The energy transition will require a balanced action plan in which CCUS plays a critical role, as the only technology with large enough scale to significantly reduce emissions from coal and gas power generation, as well as the only technology through which industries like steel, cement and petrochemicals can be decarbonized (Bui et al., 2018). Without other carbon utilization technologies quickly outpacing CO_2-EOR, negative emission technologies like bioenergy with CCS (BECCS) and direct air capture (DAC) will need geologic carbon storage.

In CCUS, the means for decarbonization is the mass of captured CO_2 that can be stored in oil reservoirs through EOR as well as in saline aquifers underneath the same EOR footprint. Simultaneously, CO_2-EOR can supply reduced carbon oil to support the energy foundation, as well as provide what is most critically needed, time to find long lasting climate solutions.

THE FUTURE OF NEGATIVE EMISSIONS OIL: SCALING UP FOR CLIMATE MITIGATION

An industrial-scale EOR-storage implementation faces several challenges. The revenue of EOR must be matched with accelerating measures such as those seen in other countries that lower the price of commercial-scale implementation, including: tax credits (carrot), carbon pricing (stick), emissions regulation, capital grants, and state ownership of CCS facilities (Global CCS Institute, 2019).

Only nine natural CO_2 reservoirs remain commercially viable in the U.S. Natural reservoirs accounted for 85% of all U.S. CO_2 supply in 2010. Anthropogenic, captured CO_2 from natural gas processing and hydrocarbon conversions are the source for the majority of the remaining CO_2 for EOR (DiPietro et al., 2012). The lack of available CO_2 limits the growth of EOR (Benson et al., 2012). A study by Advanced Resources International (ARI) states that an additional 4–47 billion barrels of domestic resources could be economically recovered using CO_2-EOR and at least 8 billion tons of CO_2 could be sequestered in the U.S. by using EOR (Advanced Resources International Inc, 2011).

Carbon dioxide enhanced oil recovery (CO_2-EOR) offers large CO_2 storage capacity potential and could accommodate a major portion of the CO_2 captured from industrial facilities for the next 30 years (Godec et al., 2013). Assuming that renewable energy generation continues to increase and displace fossil fuel electricity generation, industry-generated CO_2 (steel mills, cement, chemicals) will continue to contribute a larger portion of national emissions. These are often considered "locked-in" emissions because a decarbonized alternative does not exist. The only way to decarbonize those industries is through CCS.

In order to be considered for its climate mitigation value, CO_2-EOR projects must transition to using either anthropogenic CO_2 captured at industrial sources or CO_2 from DAC. Combining DAC with CO_2-EOR/storage on a commercial scale is the goal of a recent project announced by Carbon Engineering and Oxy Low Carbon Ventures, where a DAC facility is being designed to capture 500,000 tons of CO_2 a year directly from the atmosphere for EOR and subsequent geologic storage in a Permian Basin oil field (Global CCS Institute, 2019). This project is being regarded as "the world's largest direct air capture and sequestration facility." However, for scale-up to be sufficient to significantly impact national emissions, capture will have to happen across sectors, starting with the most economical prospects.

Bains et al. (2017) conclude that, based on the purity of the stream (lower cost), the relative contribution to overall industrial process emissions, and the proximity to potential storage reservoirs, the best implementation path forward is to prioritize initial capture efforts on ethanol production facilities, then cement and ammonia industries, and finally natural gas processing and ethylene oxide production plants. The lowest cost opportunities for deploying CCS have been found to be in the Midwest and along the Gulf Coast, where extensive geologic storage reservoirs are co-located with EOR operations (Bains et al., 2017).

The hub nature of scale-up will require intensive upfront time and financial investments as one capture, transport, or storage value chain is completed. Because of this, anthropogenic-supplied CO$_2$ is expected to be delivered in "bulky increments" (Marston, 2018), and early adopters are vulnerable to infrastructure devaluation, stranded assets, and missing markets (Zapantis et al., 2019). Banks and insurance companies offering high risk premiums and insurance can help offset risk (Zapantis et al., 2019). However, Dooley et al. (2010) suggest that this will lead to oligopoly (where a small number of suppliers control supply) when CO$_2$ quantities are scarce at the beginning of scale up, which enables CO$_2$ to have a positive price. As more anthropogenic CO$_2$ sources become available, a set price on carbon will be needed to sustain values.

The scale-up problem that CCS faces is largely the result of an insufficient value on carbon—presenting risks to private sector investments and, by proxy banking institutions, which have the potential to be partially alleviated through policy and regulation (Zapantis et al., 2019). The CO$_2$ pipeline infrastructure scale-up needed between customers and suppliers of anthropogenic CO2 to meet a U.S. climate policy case scenario laid out in Wallace et al. (2015) is unprecedented but comparable to pipeline projections in other sectors. Turning the market focus from the value of oil to climate mitigation would likely require additional funding to help offset the "technology valley of death" that many emerging technologies face (i.e., technologies that are technically proven but unable to bridge the gap to commercial-scale). Because companies are already implementing the technology outside of a stable political and economic framework, the private sector is assuming the economic, technical, construction, and operational risks, but these risks could be partially abated by a policy infrastructure.

Carbon dioxide enhanced oil recovery (CO$_2$-EOR) is the main conduit through which companies planning to or already employing CCS find value in the face of political uncertainty. For simplicity, Marston and Moore (2008) advises to keep CCS frameworks in the hands of the oil and gas regulatory authority since many years will transpire before commercial scale volumes of CO$_2$ from power plants exceed the capacity of the EOR industry.

The Global CCS Institute (2015) concluded that the U.S. framework has not dealt with CCS in a fully-integrated, comprehensive manner at the state or the federal level. Proper division of state and federal responsibilities will be required for commercialization (Ekins et al., 2017). By incorporating what Ekins et al. (2017) call "flexibility instruments" that incorporate legal adaptation in legislation, federal, and local governments can better navigate any unintended consequences of adopted legislation.

The lack of a comprehensive CCS regulatory regime has been cited by numerous experts as a primary obstacle to deployment (Davies et al., 2013). The current mix of federal and state policies is what the State CO$_2$-EOR Deployment Work Group (2016) describes as "too cumbersome for project developers to utilize effectively." The Work Group recommends that Congress establish federal price stabilization contracts, or contracts for differences (CfD) to reduce price volatility between capture facilities and EOR operators (in non-vertically-integrated projects), to make carbon capture eligible for tax-exempt private activity bonds (PABs), and master limited partnerships (MLPs) to provide debt and equity on more favorable terms. There are generalized models of CCS regulatory frameworks that have the potential to support commercial-scale development country-wide (Ekins et al., 2017, p. 83; Jacobs and Craig, 2017).

CONCLUSIONS

Carbon dioxide enhanced oil recovery (CO$_2$-EOR) has potential for decarbonization during the first several years of operation. The timing of EOR net emission reductions (the first years, not the last) is of critical importance given the urgent need to abate climate change. The near-term profitability of this climate mitigation opportunity can accelerate deployment of CCS in general and economically incentivize research in support of this goal. As we get closer to maxing out the global carbon budget, CCS will become an increasingly important carbon removal technology.

Carbon dioxide enhanced oil recovery (CO$_2$-EOR) is the only commercially established carbon utilization option that provides large-scale permanent storage for captured CO$_2$, and CCS is the only technology through which industries like steel, cement, and petrochemicals can be decarbonized. As part of an already established market, carbon storage paired with EOR can be a profitable activity that also reduces greenhouse gas emissions. Until other utilizations for carbon under CCUS become more widely adopted than CO$_2$-EOR, carbon removal technologies like BECCS and DAC projects will require a geological carbon storage counterpart, and the reservoir knowledge gleaned from CO$_2$-EOR projects will prove worthwhile even if oil production and use slows.

Carbon dioxide enhanced oil recovery (CO$_2$-EOR) is at the nexus of energy production and environmental protection, reflecting various tensions in the competing spheres. CO$_2$-EOR has benefitted from decades of technical experience and policy development under an oil and gas regulatory framework while CCS has remained largely under a separate environmental law framework. As CO$_2$-EOR becomes a more appealing and viable entryway to scale up CCS infrastructure, the two legal and regulatory frameworks will have to be reconciled so that the storage achieved during CO$_2$-EOR is supported by robust documentation and procedure.

Though large technology scale-ups take time, they are not unprecedented. The most direct corollary to the type of massive, game-changing buildout that CO$_2$-EOR requires is the development of unconventional natural gas resources. By exploiting the existing conventional oil and natural gas resources, the unconventional natural gas infrastructure developed a critical market mass use in the U.S. within roughly a decade, or what has been called, "seemingly serendipitous development" (Massachusetts Institute of Technology, 2010). With the economy refocusing on a low-carbon future, what was previously ignored as a climate mitigation option may become the basis of a large, multi-pronged market under a carbon capture, storage, and utilization framework. EOR is one way that existing infrastructure can best leverage carbon prices to develop a climate mitigation technology for a shifting energy landscape.

AUTHOR CONTRIBUTIONS

VN-L conceptualized the article and contributed to sections Fundamentals of CO$_2$-EOR, Commercial Motivation for CO$_2$-EOR, Associated Storage of CO$_2$ through EOR, Carbon Lifecycle Analysis of CCUS Systems, and Role of CO$_2$-EOR in the Larger Decarbonization Picture. EM helped in the conceptualization of the article and contributed to sections CCUS Economics, U.S. Legal and Regulatory Framework, and The Future of Negative Emissions Oil: Scaling Up for Climate Mitigation.

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Negative Emissions: Priorities for Research and Policy Design

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The large-scale removal of carbon dioxide from the atmosphere is likely to be important in maintaining temperature rise "well below" 2°C, and vital in achieving the most stringent 1.5°C target. Whilst various literature efforts have estimated the global potential of carbon dioxide removal (CDR) for a range of technologies with different degrees of certainty, regional bottlenecks for their deployment remain largely overlooked. Quantifying these barriers, through national and local case studies, rather than with aggregated approaches, would guide policy and research, as well as investments, toward regions that are likely to play a prominent role in CDR deployment. Five CDR technologies—including afforestation/reforestation, bioenergy with carbon capture and storage, biochar, direct air capture and enhanced weathering—are compared in this work. We discuss main technical, socio-economic and regulatory bottlenecks that have been scarcely investigated at regional level, and provide directions for further research. We identify the availability of accessible land, water, low carbon energy and CO_2 storage as key regional drivers and bottlenecks to most CDR technologies. We discuss the caveats in CO_2 accounting in assessing the performance of each technology, and the need for an international regulatory framework which captures these differences. Finally, we highlight the social, economic and political drivers which are central in unlocking the large scale deployment of CDR technologies, in a cost attractive, socially acceptable and politically achievable way.

Keywords: negative emissions, carbon dioxide removal, climate change mitigation, BECCS, DACCS

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INTRODUCTION

Insufficient climate change mitigation action has led to the need for large-scale carbon dioxide removal (CDR) from the atmosphere to meet the Paris Agreement target (IPCC, 2018). Several *technologies* have been identified as capable of delivering CDR at scale: afforestation/reforestation (AR/RE), bioenergy with carbon capture and storage (BECCS), biochar, direct air carbon capture and storage (DACCS), and enhanced weathering of minerals (EW) (Minx et al., 2018). Other technologies, such as ocean fertilization exist, but were excluded from the study because of their limited potential, particularly when compared to alternative portfolio options (Fuss et al., 2018). The global and regional deployment potentials of these technologies—often in isolation—have been quantified, with different degrees of certainty. A recent review quantifies annual technical and sustainable CDR potentials in

2050 at 0.5–7 $GtCO_2$ for AR/RE, 0.5–5 $GtCO_2$ for BECCS, 0.3–2 $GtCO_2$ for biochar, 0.5–5 $GtCO_2$ for DACCS, and 2–4 $GtCO_2$ for EW (Fuss et al., 2018). These estimates are conservative and account for both physical limits—land, water, CO_2 storage, suitable minerals and energy availabilities—and broader environmental risks—biodiversity loss and albedo effects. Integrated Assessment Models (IAMs) represent the world in aggregated regions, consequently they are incapable of identifying the extent to which these factors represent regional bottlenecks for CDR deployment. Instead, CDR potentials are quantified in a rather coarse manner, by focusing on standalone CDR options (Azar et al., 2010; Klein et al., 2014; Popp et al., 2014), or by investigating two options (generally afforestation and BECCS) at the same time (Humpenöder et al., 2014; Harper et al., 2018). Case studies accounting for a compressive portfolio of CDR technologies and adopting a regional or national approach are currently scarce. The presence of such literature gaps has a 2-fold implication: (1) limited guidance toward implementation of CDRs at national or regional scale (2) difficult identification of key regions for CDR deployment based on site-specific factors.

In the following sections, we highlight regional enabling factors of CDR deployment that have received minimal investigation in the literature, and need further research.

FROM GLOBAL TO REGIONAL BOTTLENECKS OF CDR DEPLOYMENT

Land and Water

The effectiveness of land-based mitigation technologies—AR/RE, BECCS, Biochar and EW—are inherently dependent on land availability, and the quality and accessibility (here considered as proximity to infrastructure) of that land will impact the regional implementation of the CDR options differently. While largely unaffected by land accessibility issues, land availability and quality are critical to AR/RE potential. A key regional limitation for afforestation is also the potential reduction of the earth albedo effect in boreal/northern hemisphere areas (Smith et al., 2016a). Tropical regions are therefore the most attractive places for forest growth practices, because the combined effect of carbon sequestration and albedo change are assumed to lead to a net cooling (Kreidenweis et al., 2016). Whilst some authors have warned against potential side effects, such as food price increase and land use change (Calvin et al., 2014; Kreidenweis et al., 2016) of intense afforestation in such regions, case studies detailing such socio-economic impacts at the local level, are still scarce in the literature (Stoy et al., 2018).

In many regional approaches, the role of water and land for BECCS deployment is relatively nuanced, because of the 2-fold flexibility of biomass feedstock supply. Indeed, BECCS plants can rely on imported biomass feedstock (Fajardy et al., 2018), and opt for biomass feedstock, such as wastes (Pour et al., 2018), residues from forestry and agriculture (Creutzig et al., 2015), pulp and paper industry biomass (Onarheim et al., 2017), and algae biomass (Beal et al., 2018), which do not require dedicated plant growth. Similarly, biochar can be derived form a wide range of feedstock, with properties and utility differing with feedstock type and production condition, including pyrolysis temperature (Kwak et al., 2019). However, large scale deployment of BECCS and biochar will likely require dedicated bioenergy crops, hence the importance of land productivity in regions likely to contribute to bioenergy production. In addition to biomass production, BECCS also requires water at the CO_2 capture level, which is mainly a function of the cooling technology. However, BECCS total water requirement is largely driver by the biomass water footprint (Fajardy and Mac Dowell, 2017). Water requirements for DACCS and EW are typically lower than that of BECCS or biochar by an order of magnitude (Smith et al., 2016a).

Assumptions underlying IAMs typically feature a land productivity increase of a 1 to 3% over time (Fisher et al., 2012; Winchester and Reilly, 2015). Since these assumptions are based on historical trends of conventional (e.g., wheat/corn) crops, the extent of which these increases will be applicable to second generation bioenergy crops, is uncertain. Regional quantifications of actual and potential yields for bioenergy crops is therefore crucial, especially considering that some high yielding and resilient types (e.g., perennial grasses) could remediate set-aside/low productivity land, thus reducing the overall land requirement for BECCS (Smith et al., 2013; Liu et al., 2014). Quantifying the regional availability of such land parcels, and determining to which extent they will be accessible and used by energy producers will enable further evaluations on sustainable bioenergy potential.

Other key drivers for BECCS deployment include land accessibility, for biomass transport and CO_2 infrastructure roll-out, and water requirement at the plant gate (Smith et al., 2016a; Fajardy and Mac Dowell, 2017), which needs to be factored to a smaller extent when deciding on the location of a BECCS plant.

Biochar and minerals for EW can be applied on agricultural lands, in conjunction with conventional farming, which means than there is no direct competition with other uses of the land. Furthermore, potential yield increase in the land from biochar and minerals application could be an added benefit of CDR deployment (Lenton, 2010; Jeffery et al., 2011).

Low Carbon Energy and Energy Systems Integration

A key enabling factor for CDR technologies deployment is the possibility to access low carbon energy sources: whilst BECCS produces energy, its life cycle energy requirement can be high and regions with low carbon fuels and electricity will be favored to produce sustainable biomass (Fajardy et al., 2018). EW, biochar and afforestation also require some form of potentially energy-intensive steps—the total energy requirement typically decreasing in this order—such as material grinding and transport for EW (Renforth, 2012), biochar production via pyrolisis, transport and application for biochar (Smith, 2016), and forest management (e.g., fertilizer application) for afforestation. The availability of low carbon energy becomes even more important when considering DACCS, which process requires ∼9–10 GJ of thermal energy per ton of CO_2. Such energy requirement has three main implications: (1) high running costs of DACCS facilities compared to other CDRs options, (2) carbon efficiency requirement (de Jonge et al., 2019), and (3) implications on the energy system configuration (Daggash et al., 2019).

CDRs options implying energy production (BECCS) or consumption (DACCS), face the challenge of being efficiently integrated into the energy systems, especially considering their anticipated role in climate stabilization scenarios. A recent study calculated that, if emission reductions in line with the 2°C target are to be met by 2100, high rates of end-use energy produced by BECCS and consumed by DACCS (8 and 20%, respectively) would be needed (Creutzig et al., 2019). Given such a prominent role in both energy generation and load, the deployment of these options needs to account for future energy systems configurations, especially considering that the rate of energy systems decarbonization has important consequences for the scale of CDR adoption.

However, studies attempting to understand how CDR technologies will integrate into future energy systems are currently scarce in the literature. Especially crucial are energy systems modeling efforts with high spatiotemporal and technological resolution, to understand: (1) the role of CDR deployment in regional energy demand (DACCS) and energy supply (BECCS) curves, and (2) potential feedback loops between energy market prices and cost/carbon efficiencies of CDRs options.

CO_2 Storage

Accessible CO_2 storage capacity is ultimately the main regional driver of BECCS and DACCS deployment. Few regions (e.g., the USA, UK and Norway) have performed a thorough geological survey to assess its CO_2 storage capacity and quality. When surveys have been carried out, the data is not always publicly available and/or exploitable (e.g., China and Europe) (Dahowski et al., 2009; Geological Survey of Denmark Greenland, 2009; Li et al., 2009). Other regional studies have been performed, but they often consist in qualitative assessments at the country level (e.g., India, Brazil) (Holloway et al., 2008; Ketzer et al., 2014) or quantitative assessment at the basin level (e.g., China). Furthermore, differences in assumptions from one survey to another render them difficult to compare. An open-access and consistent database gathering regional geological storage quality and capacity is mandatory to further explore which role will a region play—if any—in deploying BECCS and DACCS. **Table 1** summarizes the relative importance of regional bottlenecks for each CDR method.

Tracking CO_2 Removal in Space and Time

Common to all CDR technologies is the requirement to track the CO_2 along the value chain in both space and time. The CO_2 negativity—how much CO_2 is actually removed when accounting for life cycle emissions, or the CO_2 efficiency—ratio of CO_2 removed to CO_2 absorbed by the biomass (Renforth, 2012; Fajardy and Mac Dowell, 2017; de Jonge et al., 2019; Tanzer and Ramirez, 2019) including land use change emissions in particular (Searchinger et al., 2008; Harper et al., 2018)—are key metrics that need to be carefully assessed. This is particularly important when comparing different CDR technologies, as illustrated in a recent work comparing BECCS and AR at the global level (Harper et al., 2018), highlighting the relative success of BECCS when accounting for changes on the land total carbon budget. Implementing sustainability criteria and supply chain certification frameworks are central to this endeavor.

Forest certification, a practice that recognizes responsible management via verified compliance with underlying criteria and indicators (CIFOR, 2013), represent a useful tool in biological carbon offsets projects. However, the lack of policy related to certification guidelines and the absence of a universal accreditation framework has resulted in large discrepancies between the claims of different projects (CIFOR, 2013). This, in turn, leads to uncertainty, which might discredit AR as a valid CDR practice and contributes to the growing concern about carbon neutrality of forestry-based biomass sources used in BECCS projects (Searchinger, 2012).

Tracking CO_2 emissions becomes particularly challenging when CDR value chains are potentially international and multipolar, which is specific to BECCS (Fajardy et al., 2018).

Time matters differently for each CDR technology, when it comes to tracking CO_2 removal, hence the need for different metrics to measure their performances. The *rate of CO_2 removal* represents a crucial factor for biological or mineral sinks, i.e., in BECCS, AR, EW, and biochar, and remain a point of uncertainty for EW in particular (Renforth, 2012).

Specific to land-based CDR potentially involving land use change, i.e., BECCS in this case, high initial land use change emissions—also referred to as the carbon debt (Fargione et al., 2008)—as compared to the technology CO_2 removal rate, can lead to *delayed carbon removal*: a CDR system only becomes net negative after several years of operation, if at all (Fajardy and Mac Dowell, 2017).

Natural sinks reach their maximum capacity within decades, as compared to centuries for geological underground storage. Therefore, when CO_2 is stored in biological sinks (trees, land, oceans), the issue of *sink saturation,* i.e., the fact that CO_2 removal is limited in time, is a major limitation. In case of AR, this relates to the fact that trees reach maturity, and therefore the net removal of greenhouse gases from the atmosphere decline to zero (Smith et al., 2016b; Alcalde et al., 2018). Its direct implication is that forest sinks need to be managed and monitored to maintain the optimal CO_2 uptake over time. Finally, monitoring CO_2 sequestered is also key to ensure *permanence* CO_2 removal. Whilst long term geological storage raises questions of liability and insurance (Bui et al., 2018), monitoring CO_2 storage sites represent more of a legal and financial challenge than a technical one. In the case of afforestation however, natural (for e.g., decay) and accidental (for e.g., forest fires, land use change) degradation of the carbon stock, both above ground and below ground, put the permanence of CO_2 removal at risk.

The relative difficulty for each CDR option of tracking CO_2 removal in space and time, could ultimately impact: 1) the complexity of the regulation/certification frameworks required to ensure the performance and sustainability of each option and 2) the economic potential of the technology—by deterring investors with high upfront financial risks and high regulation,

TABLE 1 | The comparative importance of regional bottlenecks to the deployment of different CDR technologies.

	Land			Water	Low carbon energy (fuel and electricity)	Geological CO_2 storage	Misc.
	Availability	**Quality**	**Accessibility**				
AR/RE	++++	+++	+	+++	+ (Management)		Albedo +++
BECCS	++	++	++	++	++ (Supply chain)	++	Wastes/other feedstock ++
Biochar	++	+	+++		+ (Supply chain)		+ Variable impact on yield
DACCS	+		+	+	+++ (Process)	+++	
EW	++		+++		++ (Supply chain)		+ Variable impact on yield

and therefore need to be included in comparative assessments. These observations are summarized in **Table 2**.

TABLE 2 | The comparative importance of life cycle analysis to the performance of different CDR technologies in space and time.

	Space	Time
AR/RE	++ Mapping of sustainable management zones is required	++++ Rate of CO_2 capture Sink saturation Permanence needs management
BECCS	+++ Careful life cycle is challenging especially with biomass international BECCS value chains	++ Potential delayed CO_2 removal if high land use change
Biochar	+ Biomass processing and transport needs to be monitored, usually local	+++ Rate of CO_2 capture Sink saturation
DACCS	+ Carbon footprint of energy needs to be monitored	
EW	+ Rock collection, grinding, and transport need to be monitored, usually local	++ Rate of CO_2 capture, monitoring due to sink evolution over time (from minerals to groundwater and to ocean)

Socio-economic Issues and the Need for a Landscape Approach

Much of the controversy about CDR methods to date is directed at BECCS, emerging as the key technology for cost-effectively limiting global warming to the level set by the Paris Agreement. Studies applying global IAMs indicates a global potential deployment of biomass for BECCS in the range of 1.3–1.6 $GtCO_2$/yr by 2050 (Huppmann et al., 2018; Rogelj et al., 2018). It is well-recognized that such a high deployment of biomass would require a massive transformation of agricultural systems worldwide, with implications for both environmental and social change. Outcomes from IAM studies, have extensively investigated the widespread ecological impacts, such as biodiversity loss, food security along whit access to energy and water (Müller et al., 2008; Popp et al., 2011, 2014) that would occur if BECCS is extrapolated to the required scale. However, case study evidence detailing the societal impacts of bioenergy and BECCS deployment is currently missing in climate actions debates. When the socio-economic dimension is accounted for, this is generally done by considering economic growth or food prices. Other dimensions of human well-beings, such as employment opportunities or change in socio-economic conditions of local communities, remain largely overlooked.

In the attempt to narrow this gap, Creutzig et al. (2013) have analyzed the factors shaping the interaction between bioenergy and livelihood, highlighting how global competition for land use emerging from IAMs scenarios, would directly affect 1.5 billion smallholders. The authors concluded that, because calculation of bioenergy potentials is often presented at aggregated level, impacts of bioenergy schemes on local communities remain systematically underexplored.

The argument of global scale as an inadequate dimension to see potential social impacts at work, has been recently brought up by other authors (Buck, 2018; Lenzi, 2018). While conducting structured interviews and site visits to examine the challenges of the deployment of CDR technologies (Buck, 2018) have identified, in the landscape level, larger than a farm but smaller than a region, the crucial dimension in designing the governance of CDR technologies. A landscape approach, the author argues, would promote a paradigm shift in the way CDR is perceived, as a valuable practice that creates jobs, rather than artifacts being deployed. Hence, adopting a landscape/local prospective, would not only address distributional issues associated with the production model underlying CDR schemes, but also help to resolve some narrative issues around CDR technologies.

The value of the societal dimension is also crucial to quantify potential local co-benefits, such as employment opportunities (Patrizio et al., 2018) and income growth. This is particularly relevant for BECCS and afforestation projects in developing countries, where large areas of land are still unregistered, and surrounding communities must obtain benefits from forest and be actively involved in their management (Greve et al., 2013). Accounting for such elements may influence geographic priorities and serve as a useful initial decision-making tool to maximize the overall benefits of CDR projects.

Financing CDR

The literature on CDR has focused on techno-economic assessments of CDR methods at scale (Fuss et al., 2018).

The economic costs of deploying these technologies have been evaluated, albeit with significant uncertainty owing to the lack of demonstration and commercial-scale projects. It has been repeatedly highlighted that regional carbon prices are insufficient to incentivize CDR deployment. More relevant to note is that, should carbon prices be sufficient, mainstream carbon pricing schemes often only penalize CO_2 emissions and do not remunerate removal. CDR is a public good as it furthers climate change mitigation, so its value needs to be recognized and appropriately remunerated. Failing regulatory/policy changes to carbon pricing mechanisms to incentivize CDR and an increase in carbon prices, a negative emissions credit—a payment for *net* CO_2 removal from the atmosphere—is necessary. Financing mechanisms that can deliver these incentives in a socially-acceptable manner (i.e., without disproportionately affecting some segments of society e.g., fuel tax hikes) have not received enough attention. It has been shown that CDR deployment can allow for the continued utilization of existing CO_2-emitting assets, thereby avoiding stranded assets (Daggash et al., 2019). The revenue generated by some of these assets which would have been otherwise constrained from productive utilization should accrue to the providers of CDR. The market and governance mechanisms that allow for the verification and trading of such negative emissions credits (as discussed earlier), however, are currently non-existent and need to be designed.

Enhanced oil recovery and merchant CO_2 markets have been proposed as routes to CDR commercialization. However, CO_2 utilization processes generally do not contribute to climate change mitigation as they only delay emissions[1]. Commercial-scale CDR which also contributes to mitigation is only possible if a stable revenue stream is available for the service of permanently sequestering atmospheric carbon. Additionally, the volumes of these CO_2 markets are small relative to the scale of CDR needed to meet the Paris Agreement.

Majority of the CDR options interact, to different extents, with the energy system. Since the 1990s, energy markets have been increasingly liberalized, such that investment in energy technologies is largely private sector-driven. Private investment seeks the greatest economic return, therefore incentives (both policy and financial) offered to CDR need to make the technologies not just economically-viable but the most attractive destination for investment.

International Diplomacy

The ability to deliver CDR at scale is contingent on the availability of several bio-geophysical resources—land, CO_2 storage capacity, rocks, ocean, etc.—, all of which are scarce and unevenly distributed geographically. In addition to quantity, the nature and quality of each resource will determine the carbon removal potential of the CDR technique pursued. Economic factors, such as labor costs and taxation rates will influence local CDR costs. Owing to geographic and economic factors therefore, the physical and economic efficiency of CDR technologies for climate change mitigation will be location-dependent. The Paris Agreement—and climate policy, in general—has sought to distribute the burden of climate change mitigation based on "the principle of equity and common but differentiated responsibilities and respective capabilities, in the light of different national circumstances." To achieve both a cost- and politically-optimal distribution of CDR burden, therefore, will require mechanisms that allow for cross-border trade/exchange of resources (e.g., of sustainable biomass, or energy) and emissions (should it prove cheaper for a country to pay for CDR to be done elsewhere). This, in addition to the dominant market-based approach to development of energy infrastructure, means multi-region, multi-stakeholder supply chains and market/governance systems will manifest.

Whilst these socioeconomic and political challenges surrounding CDR have been acknowledged (Honegger and Reiner, 2018; Nemet et al., 2018), there has not been an attempt to quantify what, firstly, a cost optimal distribution of CDR burden globally is. If that involves cross-border biomass trade (in the case of BECCS deployment) as has been suggested (Fajardy et al., 2018; Daggash et al., 2019), then the development of international biomass sustainability certification standards is critical. The development of such frameworks, particularly in countries without pre-existing expertise of sustainability certification, will require cooperation between expected trading partners so as to align verification and monitoring methods. Although several regional emissions trading schemes (ETS) and carbon offsetting mechanisms exist globally, they have failed to incentivize real and verifiable storage of CO_2–and consequently, CDR—by encouraging short-term CO_2 utilization instead (Haszeldine et al., 2018). Adapting carbon offsetting mechanisms to incentivize CDR and include negative emissions in trading permits, and further expansion of emissions trading zones will facilitate the deployment of CDR at scale. Through the nationally-determined contributions (NDCs), commitments to the Paris Agreement are assessed at country-level. If cross-border financing and bio-geophysical resources are used to deliver CDR, a framework needs to be developed to determine to whom the emissions reduction is ascribed. The aforementioned present significant regulatory and political barriers, both locally and internationally, to efficient CDR deployment. However, they have received insufficient investigation, indicated by their absence in the literature. Failure to fully understand these challenges within different socioeconomic and geopolitical contexts, and design appropriate policy solutions to address them will stifle necessary climate change mitigation action.

Knowledge Transfer

IAMs estimate that the largest cumulative contributions of CDR are made by six regions: China, the USA, India, the EU-28, Brazil and Russia (Peters and Geden, 2017). Discussions surrounding CDR, and more broadly, CCS, deployment have centered on these regions, principally the USA and Northwestern Europe. Accordingly, there has been an abundance of research quantifying the bio-geophysical limitations to CDR deployment, often by publicly-funded bodies, such as the countries' geological surveys or energy/environment ministries. High-quality data enables accurate quantification of CDR potential within these

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[1] Conversion of CO_2 into building materials (e.g., concrete) is one of the few CCUS processes that result in the permanence of stored CO_2.

regions—crucial in understanding how the rest of the energy system and society is required to transition to meet the Paris Agreement.

With the exception of India, the centers of population and economic growth in the coming decades are expected outside the aforementioned regions—mostly in Sub-Saharan Africa and South East Asia. Increasing population leads to increased energy demand, and economic development (hence more affluent lifestyles) will lead to higher per capita demand. The rate and scale of energy supply expansion needed to accommodate these demographic explosions means majority of developing countries continue to pursue largely fossil fuel-powered development. Prolonged use of fossil fuels may lead to an increasing reliance on CDR (and CCS) to meet emissions reductions commitments. However, typically, the expertise needed to gather knowledge on CDR/CCS potential is lacking and budgetary constraints limit resource availability to undertake RD&D within those environments. Climate equity—as interpreted in the Paris Agreement—warrants that developed countries provide finance, technology transfer and capacity-building to "assist developing country Parties with respect to both mitigation and adaptation in continuation of their existing obligations under the Convention." This is ongoing *via* special funds under the Global Environment Facility but financing has thus far eluded CDR/CCS projects and focused on alternative mitigation/adaptation solutions. Focus and financing must be re-channeled if the necessary expertise is to be developed, so that it is available when CDR/CCS deployment become necessary. How much and to whom this financing should be given—that is the countries for which CCS/CDR is likely to be a necessity—need to be identified through further research.

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MF, PP, and HD wrote the first draft of the manuscript. All authors contributed to the conception and design of the study. All authors contributed to manuscript revision, read, and approved the submitted version.

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CO_2 Removal With Enhanced Weathering and Ocean Alkalinity Enhancement: Potential Risks and Co-benefits for Marine Pelagic Ecosystems

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Humankind will need to remove hundreds of gigatons of carbon dioxide (CO_2) from the atmosphere by the end of the twenty-first century to keep global warming below 2°C within the constraints of the global carbon budget. However, so far it is unclear if and how this could be achieved. A widely recognized idea is to accelerate weathering reactions of minerals that consume CO_2 when they dissolve. Acceleration could be realized by pulverizing and distributing gigatons of these minerals onto land (termed "enhanced weathering (EW)") or sea (termed "ocean alkalinity enhancement (OAE)") thereby largely increasing their reactive surfaces. However, the desired consumption of atmospheric CO_2 during dissolution would inevitably be accompanied by a release of mineral dissolution products (alkalinity, Si, Ca, Mg, Fe, Ni, and maybe others). Here, we approximate their maximum additions to assess potential consequences for pelagic communities (mainly primary producers) and the biogeochemical fluxes they control. Based on this assessment, we tentatively qualify the potential to induce positive and/or negative side effects to be high for Fe, Ni, Si, intermediate for alkalinity, and low for Ca and Mg. However, perturbation potentials are always higher at perturbation hotspots and would be different for EW than for OAE. Furthermore, ecological/biogeochemical consequences of EW/OAE largely depend on the minerals used. We hypothesize that mainly calcifiers would profit in a scheme where $CaCO_3$ derivatives would be used due to beneficial changes in carbonate chemistry. Figuratively, this may turn the blue ocean into a white(r) ocean. When using silicates, the release of additional Si, Fe and Ni could benefit silicifiers and N_2-fixers (cyanobacteria) and increase ocean productivity ultimately turning the blue ocean into a green(er) ocean. These considerations call for dedicated research to assess risks and co-benefits of mineral dissolution products on marine and other environments. Indeed, both EW and OAE could become important tools to realize CO_2 removal at the planetary scale but associated risks and/or co-benefits should be revealed before deciding on their implementation.

Keywords: plankton, risk assessment, iron, nickel, silicon, silicification, calcification

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FACING REALITY—THE NEED FOR NEGATIVE EMISSIONS TO MEET THE <2°C WARMING GOAL

Anthropogenic emissions of greenhouse gases increase the absorption of heat by the atmosphere thereby inducing global warming (Myhre et al., 2013). It is now widely accepted that anthropogenic warming needs to be limited to not more than 2°C to preserve the Holocene mode of operation of the climate system and thus to prevent intolerable risks for humankind (Schellnhuber et al., 2016). The 2015 Paris Agreement, signed by 194 parties and the European Union, adopted the scientific advice by the Intergovernmental Panel on Climate Change (IPCC) to keep global warming "well below 2°C," and to "pursue efforts to limit the [average] temperature increase to 1.5°C" (United Nations Framework Convention on Climate Change, 2015). This goal can only be achieved when limiting near future CO_2 emissions to ~480–1,130 gigatons (Gt) (Le Quéré et al., 2016; Rogelj et al., 2016). Hence, when continuing at the current emission rate of ~37 Gt CO_2 per year, the 2°C emission target would be missed in ~13–31 years from now (Peters, 2016; Rogelj et al., 2016).

The remaining carbon budget to stay below 2°C calls for immediate and profound emission reductions roughly in the range of 1–10% $year^{-1}$ as anticipated in the techno-economic assessments of the RCP2.6 scenarios (Representative concentration pathways leading to $\leq$2.6 W/m^2 additional radiative forcing) (Rogelj et al., 2016; Rockström et al., 2017). What is widely unknown to the public, however, is that even the very optimistic emission trajectories summarized in RCP2.6 largely fail to remain within the carbon budget constraints for 2°C by decarbonization alone (Peters, 2016). In 108 out of the 116 RCP2.6 scenarios assessed in the last IPCC report, this is only achievable because of the large scale implementation of "negative emission technologies (NETs)" which remove CO_2 from the atmosphere and store it for geological timescales (Rau et al., 2012; Anderson and Peters, 2016; Peters, 2016; Williamson, 2016; Minx et al., 2018). These NETs must be capable of removing something in the range of 600 Gt CO_2 from the atmosphere by the end of the twenty-first century to maintain a likely chance (i.e., >66%) to meet the <2°C goal (van Vuuren et al., 2011; Anderson and Peters, 2016; Fuss et al., 2016; Sanderson et al., 2016). Thus, the inability of societies to initiate dedicated mitigation now, will likely push us toward the future implementation of climate engineering where NETs will likely play a key role.

COULD ENHANCED WEATHERING (EW) AND OCEAN ALKALINITY ENHANCEMENT (OAE) HELP TO SOLVE THE CLIMATE CRISIS?

Every NET has a range of economical and geospatial limitations. As such, a portfolio of approaches is needed to achieve carbon dioxide removal from the atmosphere (CDR) at the necessary scale (Nemet et al., 2018). Various NETs have been proposed so far but only a few have developed beyond the laboratory. There is yet little public funding, generally no established legal framework, and no social acceptance to ramp up any NET to the necessary scale (Oschlies and Klepper, 2017; Braun et al., 2018; Nemet et al., 2018). The public reservations are justified since NETs raise ethical concerns (Lin, 2013; Lawford-Smith and Currie, 2017; Shue, 2017; Lenzi, 2018) and may be associated with side effects for the Earth system (Keller et al., 2014; Fuss et al., 2018; Gattuso et al., 2018). However, since NETs will very likely become key tools for keeping global warming below 2°C it is vital to assess their associated environmental risks before their initiation (Oschlies and Klepper, 2017).

"Enhanced Weathering (EW)" and "Ocean Alkalinity Enhancement (OAE)" are two related NETs which are among the options to realize CDR (**Figure 1**). The idea is to accelerate natural rock weathering e.g., by spreading large amounts

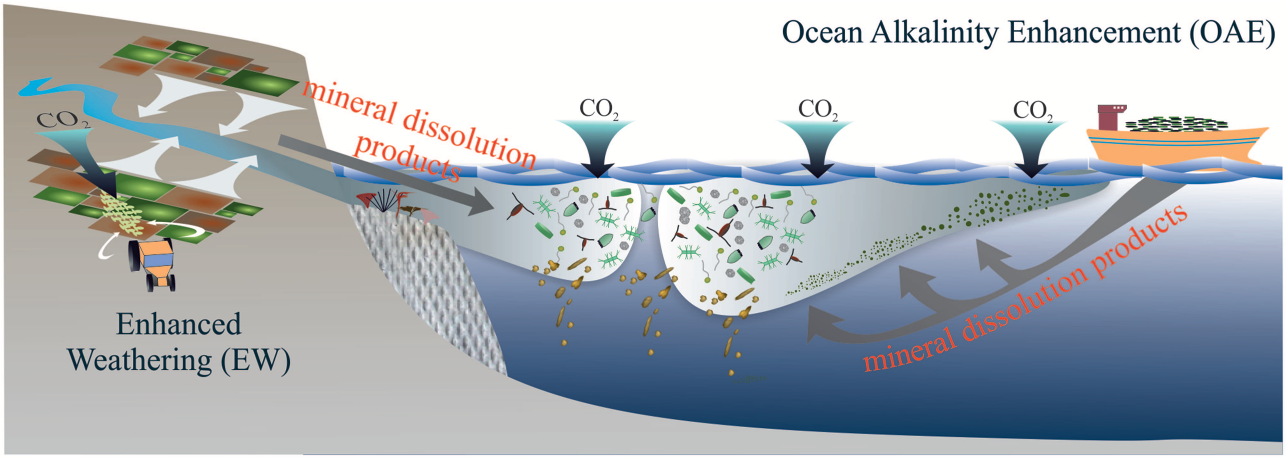

FIGURE 1 | Schematic overview of EW and OAE. EW and OAE will bring mineral dissolution products into marine environments with so far largely unknown consequences for marine ecosystems and biogeochemical cycling therein. During EW, mineral dissolution products enter the ocean mostly via rivers so that perturbations first occur in estuarine and coastal regions. In the case of OAE, cargo ships could transport the minerals to coastal regions or further offshore and the distribution sites would be impacted most strongly.

of pulverized silicate and/or carbonate minerals onto warm and humid land areas (EW) or onto the sea surface (OAE). The increased exposure of these minerals enhances chemical weathering reactions whereby atmospheric CO_2 is consumed (**Box 1**) (Kheshgi, 1995; Schuiling and Krijgsman, 2006). Idealized Earth system model studies have shown that EW and OAE can mitigate climate change significantly when operated at a scale that is appropriate to the challenge (Caldeira and Rau, 2000; Köhler et al., 2010, 2013; Paquay and Zeebe, 2013; González and Ilyina, 2016; Hauck et al., 2016; Taylor et al., 2016; Feng et al., 2017; Lenton et al., 2018).

EW and OAE may be more amenable to implementation (Taylor et al., 2016). First, CDR through rock weathering is a natural process which consumes 1.1 Gt CO_2 $year^{-1}$ already today (Ciais et al., 2013). Second, neither EW nor OAE would require their own land, nutrients, or freshwater (Smith et al., 2016; although dust avoidance during EW may need freshwater, Taylor et al., 2016). They could be applied on open ocean regions or combined with agriculture with the additional benefit of enhancing crop yields and preventing soil erosion (Köhler et al., 2010; Beerling et al., 2018; Dietzen et al., 2018). Thus, in contrast to many other NETs, they are generally not competing with other Sustainable Development Goals like global food and water security but are potentially even beneficial for them (Beerling, 2017; Edwards et al., 2017; Heck et al., 2018). Third, EW/OAE-related alkalinity additions would buffer the CO_2-induced decline in seawater pH (Köhler et al., 2010). Thus, EW/OAE would not only mitigate global warming (and its consequences such as sea level rise) by reducing atmospheric CO_2 but also specifically mitigate ocean acidification which is considered a major threat for marine ecosystems (Doney et al., 2009; Gattuso et al., 2015).

BOX 1 | CDR through alkalinity enhancement in aqueous media.

Total alkalinity (TA) is a complex chemical quantity composed of several ions and molecules (Zeebe and Wolf-Gladrow, 2001). TA is defined as the excess of proton acceptors over proton donors with respect to a certain zero level of protons (Dickson, 1981; Wolf-Gladrow et al., 2007). In terms of chemical concentrations this reads as:

$$TA = \left[HCO_3^-\right] + 2\left[CO_3^{2-}\right] + \left[B(OH)_4^-\right] + \left[OH^-\right] + \left[HPO_4^{2-}\right] + 2\left[PO_4^{3-}\right] + \left[H_3SiO_4^-\right] + \left[NH_3\right] + \left[HS^-\right] - \left[H^+\right] - \left[HSO_4^-\right] - \left[HF\right] - \left[H_3PO_4\right] \quad (1)$$

The dissolution of CO_2 in seawater and the subsequent reaction with H_2O have no effect on TA because the formation of proton donors and acceptors is balanced.

$$CO_2 + H_2O \leftrightarrow H_2CO_3 \leftrightarrow HCO_3^- + H^+ \leftrightarrow CO_3^{2-} + 2H^+ \quad (2)$$

A shift of the carbonate chemistry equilibrium (Equation 2) toward HCO_3^- and CO_3^{2-} would coincide with decreasing CO_2 concentration so that additional CO_2 from the environment could be absorbed and stored permanently. Such a shift toward HCO_3^- and CO_3^{2-} can be induced through the dissolution of minerals like olivine (Mg_2SiO_4) or quicklime (CaO). H^+ is consumed during the dissolution of these minerals and replaced by conservative ions with positive charges (in our cases Mg^{2+} or Ca^{2+}) (Pokrovsky and Schott, 2000; Wolf-Gladrow et al., 2007; Oelkers et al., 2018):

$$\text{Olivine}: \quad Mg_2SiO_4 + 4H^+ = 2Mg^{2+} + H_4SiO_4 \quad (3)$$

$$\text{Quicklime}: \quad CaO + 2H^+ = Ca^{2+} + H_2O \quad (4)$$

The positive charges from Mg^{2+} and Ca^{2+} must be balanced by negative ones due to the constraint of electroneutrality (Wolf-Gladrow et al., 2007). This ultimately forces the shift from CO_2 to HCO_3^- and CO_3^{2-} (**Figure 2**) and is measurable as an increase in TA which is the name-giving feature of "ocean alkalinity enhancement."

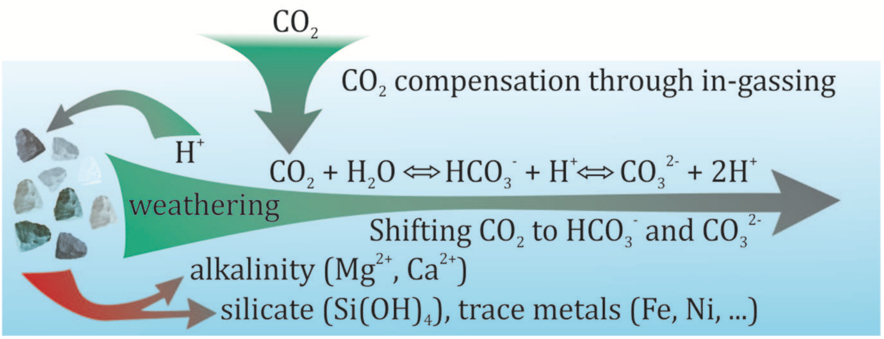

FIGURE 2 | Graphical summary of CDR through chemical weathering in aqueous media (e.g., soil pore water or seawater). When EW/OAE-relevant minerals (e.g., quicklime or olivine) dissolve they consume protons which shifts the carbonate chemistry equilibrium away from CO_2 to HCO_3^- and CO_3^{2-}. Additional CO_2 can subsequently be absorbed by the aqueous media because the shift can cause CO_2 undersaturation relative to the surrounding atmosphere. For EW/OAE it is important that the dissolution occurs as long as the aqueous media is in contact with the atmosphere (e.g., in the surface mixed layer of the ocean) so that the under-saturated medium can be replenished with atmospheric CO_2.

The minerals appropriate for EW/OAE comprise, for example, naturally occurring Mg-rich olivine-type silicates (Mg_2SiO_4) (Schuiling and Krijgsman, 2006). These may absorb 1 Gt of CO_2 for every 1–2 Gt of olivine-rich rocks when accounting for energy expenses due to mining, mineral grinding, and distribution (Moosdorf et al., 2014). Industrially reprocessed carbonates such as quicklime (CaO) are another option (Kheshgi, 1995). To be effective, the energy-related and the chemical CO_2 emissions during the endothermic calcination of $CaCO_3$ in a kiln (i.e., $CaCO_3 \rightarrow CaO + CO_2$) need to be curtailed using carbon capture and storage (CCS) technologies where the generated CO_2 is stored e.g., in underground reservoirs. For quicklime, a net removal of 1 Gt CO_2 from the atmosphere would require ~1.8 Gt $CaCO_3$ source rock to produce ~0.8 Gt CO_2 for CCS and ~1.2 Gt of CaO (Renforth et al., 2013). This estimation includes energy expenses from mining, mineral grinding, and distribution but note that the CO_2 emission along the entire process chain could be reduced considerably when using renewable energies (Meier et al., 2006; Renforth et al., 2013).

The natural reserves of source rocks delivering quicklime, olivine, or other suitable minerals are large enough to sequester thousands of gigatons of CO_2 (Hartmann et al., 2013; Taylor et al., 2016). The additional mining effort needed to remove several gigatons of CO_2 per year roughly equals efforts of the global cement industry which currently extracts ~7 Gt source materials per year (Renforth and Henderson, 2017). Although such amounts are large, they are consistent with the global construction aggregate industry which extracts ~50 Gt year^{-1} (Beerling et al., 2018). The range of technologies that have been proposed for increasing ocean alkalinity may pose significant engineering challenges. However, preliminary cost analyses suggest that they are within the range of other NETs [\$10–500 per ton of CO_2 (Strefler et al., 2018; Pacala et al., 2019)]. Thus, if the world is serious about the <2°C goal and anticipated carbon tax revenues are used to subsidize CDR, then EW/OAE could become economically viable options to realize negative emissions in the twenty-first century.

Under these prospects, it is essential that EW/OAE research expands from feasibility and cost studies to include assessments of risks and co-benefits (Oschlies and Klepper, 2017; Gattuso et al., 2018; Boyd and Vivian, 2019). While this is beginning for terrestrial environments (Beerling, 2017; Beerling et al., 2018), very little is known about impacts on marine systems. Here, several side effects have been conceived (Hartmann et al., 2013; Renforth and Henderson, 2017) but only in four cases were these specifically further investigated *in silico* (Köhler et al., 2013; Hauck et al., 2016) or *in vitro* (Cripps et al., 2013; Gore et al., 2018).

This paper aims to provide a comprehensive overview on potential risks and co-benefits associated with chemical EW/OAE-perturbations for pelagic environments. The overarching goal is to reveal key knowledge gaps thereby providing guidance for necessary future research.

CHEMICAL PERTURBATIONS OF OCEAN ECOSYSTEMS THROUGH EW/OAE

In the case of EW, pulverized minerals would be distributed onto cropland and forests in warm and humid climates (Köhler et al., 2010; Taylor et al., 2016). The dissolution products originating from the chemical weathering reaction would be partially retained in soils and freshwater systems depending on their individual mobilities. However, a certain fraction would ultimately be discharged into the oceans and primarily affect coastal and estuarine systems (**Figure 1**; Laruelle et al., 2009; Dürr et al., 2011; Gaillardet et al., 2014). Indeed, anthropogenic alkalinity enhancement through agricultural liming and other activities are likely already occurring in various coastal and estuarine systems (Guo et al., 2015; Müller et al., 2016; Kapsenberg et al., 2017; Kaushal et al., 2018) but the impact of this on the organisms and ecosystems is unknown.

In the case of OAE, mineral dissolution products would be directly placed into the surface ocean and influence ocean biota in coastal and offshore regions without previous modification by the "terrestrial filter" (**Figure 1**). Logistically, minerals would most likely be added at discrete locations because an even distribution over entire ocean regions seems unfeasible (Köhler et al., 2013). This may lead to the formation of "hotspots" of impact, but this will depend on how minerals are added, the type of material added, and the attenuation/mixing in the surface ocean relative to dissolution rate of the mineral. Most likely, they would be (1) concentrated along ship tracks which disperse the material onto the sea surface (Köhler et al., 2013), (2) in the vicinity of on- and offshore platforms from where the dissolution products are released (Rau et al., 2013), or (3) on beaches or shallow shelf seas where minerals could dissolve on the seafloor and would still be within the mixed layer and therefore in contact with the atmosphere (Hangx and Spiers, 2009; Feng et al., 2017; Meysman and Montserrat, 2017). Accordingly, dissolution products from the weathering reaction will occur in gradients and it will be important to reveal how different ecosystems respond to high and low concentrations of mineral dissolution products.

Previous reviews have listed various suitable minerals for EW/OAE (Hartmann et al., 2013; Renforth and Henderson, 2017). The molecules or elements generated during chemical weathering of these minerals and sustained in solution thereafter comprise for major groups: I) Bicarbonate (HCO_3^-) and carbonate ion (CO_3^{2-}), hereafter summarized as "alkalinity"; (II) Silicic acid ($Si(OH)_4$) hereafter termed silicate; (III) Certain alkaline earth metals like calcium (Ca^{2+}) and magnesium (Mg^{2+}) but potentially also alkali metals like sodium (Na^+) and potassium (K^+); (IV) a variety of "trace metals" associated with the minerals. Among these, iron (Fe^{2+}, or oxidized aqueous species), nickel (Ni^{2+}) will be the major subject in this paper as these elements occur in high concentrations in basic and ultrabasic rocks (e.g., dunite), which are the most widely recognized source rocks for EW/OAE (Schuiling and Krijgsman, 2006; Hartmann et al., 2013). However, depending on the mineral selected, a wider variety of trace constituents should be considered in the future.

RISKS AND CO-BENEFITS OF INCREASED ALKALINITY

Perturbation

The desired effect of EW and OAE is to increase the alkalinity in aqueous media so that additional (ideally atmospheric) CO_2 can be absorbed (**Figure 2**; **Box 1**). Idealized modeling studies suggest that EW/OAE could increase surface seawater alkalinity by about 100 to >2,000 μmol kg^{-1} by the year 2100 although the upper estimates are based on extreme, likely unrealistic, application scales (Köhler et al., 2010, 2013; Ilyina et al., 2013; Paquay and Zeebe, 2013; Keller et al., 2014; González and Ilyina, 2016; Taylor et al., 2016; Feng et al., 2017; Lenton et al., 2018). Alkalinity itself does not affect biology directly because it is a chemical concept, not an ion or molecule that could be sensed by organisms. Nevertheless, the increase in alkalinity substantially modifies pH and the concentrations of various ions and molecules of e.g., the carbonate system which can directly affect biological processes.

In EW applications, new alkalinity will enter the oceans mostly via rivers or streams. These watercourses are usually oversaturated with CO_2 relative to the atmosphere and release globally about 1.8 Gt C $year^{-1}$ mostly from remineralization of terrestrial biomass (Raymond et al., 2013). Hence, the additional alkalinity from EW would likely have exploited its CO_2 uptake capacity before entering the oceans. In the case of OAE, alkalinity is generated in seawater. The perturbation of seawater carbonate chemistry depends on how much alkalinity is added per volume and/or how quickly this volume mixes with surrounding waters. It also critically depends on how fast the perturbed seawater equilibrates with the atmosphere. For example, alkalinity addition would consume CO_2 from the surrounding seawater without immediate replenishment from the atmosphere when mineral dissolution occurs in a water body with slow air-sea gas exchange. In such a scenario carbonate chemistry perturbations are more severe than for fast air-sea gas exchange because high pH (i.e., low H^+) excursions are not immediately mitigated by in-gassing CO_2 (**Figure 3**). To generate negative emissions with OAE, it must be guaranteed that the generated CO_2 deficit in seawater is compensated with atmospheric CO_2. Accordingly, carbonate chemistry perturbations will ultimately have to conform with the "air-equilibrated" scenario shown in **Figure 3**. Nevertheless, reaching this equilibrium may take several years depending on the oceanographic setting (Harvey, 2008; Feng et al., 2017). Thus, more extreme carbonate chemistry perturbations due to "non-equilibrated OAE" at perturbation hotspots may transiently occur (**Figure 3**) and these hotspots will also have a higher potential to affect marine biota.

Current Understanding and Key Unknowns

The strong shift in the carbonate system from CO_2 to HCO_3^-/CO_3^{2-} associated with transient non-equilibrated OAE could potentially drive primary production into CO_2 limitation (Riebesell et al., 1993). Phytoplankton, for example, can acclimate to CO_2 limitation with carbon concentrating mechanisms (CCMs) which utilize HCO_3^- to support C-fixation (Giordano et al., 2005). However, HCO_3^- uptake cannot fully compensate for limited CO_2 since HCO_3^- must ultimately be converted to CO_2 for C-fixation and can leak out of the cells before being incorporated in biomass (Rost et al., 2006). The threshold below which phytoplankton growth rates become notably affected by CO_2 is genotype-specific and depends on growth conditions (e.g., light or temperature) but should generally be roughly below ∼100 μatm (Riebesell et al., 1993; Goldman, 1999; Hansen, 2002; Bach et al., 2011; Sett et al., 2014). Thus, phytoplankton growth could be slowed to a variable degree along gradients of mineral dissolution hotspots. However, dilution with un-perturbed water masses as well as CO_2 in-gassing should quickly alleviate the problem so that new production supported by the available nutrients may shift in space and time but rather not with respect to its overall magnitude as it seems unlikely that the nutrients remain unutilized over the course of a seasonal cycle. Nevertheless, the pulses of low CO_2/high pH will affect species differentially as some will be more sensitive than others (Hansen, 2002; Pedersen and Hansen, 2003).

Perhaps the most fundamental outcome of more than two decades of ocean acidification research is that carbonate chemistry perturbations affect calcifying organisms disproportionately (Gattuso et al., 2015). Thus, it is meaningful to pay special attention to calcification when considering impacts of increased alkalinity on marine organisms and ecosystems. Ocean acidification (black arrow in **Figure 3**) is predicted to have adverse effects on calcifiers (Orr et al., 2005; Kroeker et al., 2010), mainly due to the increased proton (H^+) concentration and decreased $CaCO_3$ saturation state (Ω) (Jokiel, 2011; Bach, 2015; Cyronak et al., 2015; Waldbusser et al., 2015). EW and OAE have the opposite effect on carbonate chemistry conditions. Here, H^+ decreases while Ω increases which could significantly improve the conditions for calcification (**Figure 3**). The Black Sea is a suitable analog for a highly OAE perturbed marginal sea as it has a surface alkalinity of ∼3,300 μmol kg^{-1} already today (**Figure 3**). The major planktonic processes in the Black Sea are similar to other oceans (Sorokin, 2002) but there is a remarkable particularity. The Black Sea harbors some of the most extensive blooms of calcifying phytoplankton (coccolithophores) and thick layers of calcareous ooze cover the sediments (Hay, 1988; Kopelevich et al., 2014). These blooms are likely promoted by the favorable carbonate chemistry conditions since coccolithophores are largely absent in the Baltic Sea—a lower alkalinity marginal sea (Müller et al., 2016) with in other respects comparable characteristics to the Black Sea (Tyrrell et al., 2008).

Important Knowledge Gaps and Testable Hypotheses

We have argued in the previous section that transient shifts in carbonate chemistry conditions should have little impact on the overall productivity of marine ecosystems but could alter the species composition. The latter may be important because even though the perturbation is transient, it could induce knock on effects altering species composition and trophic interactions during the succession following the initial perturbation. Such a restructuring could have more severe biogeochemical consequences than initially anticipated. For

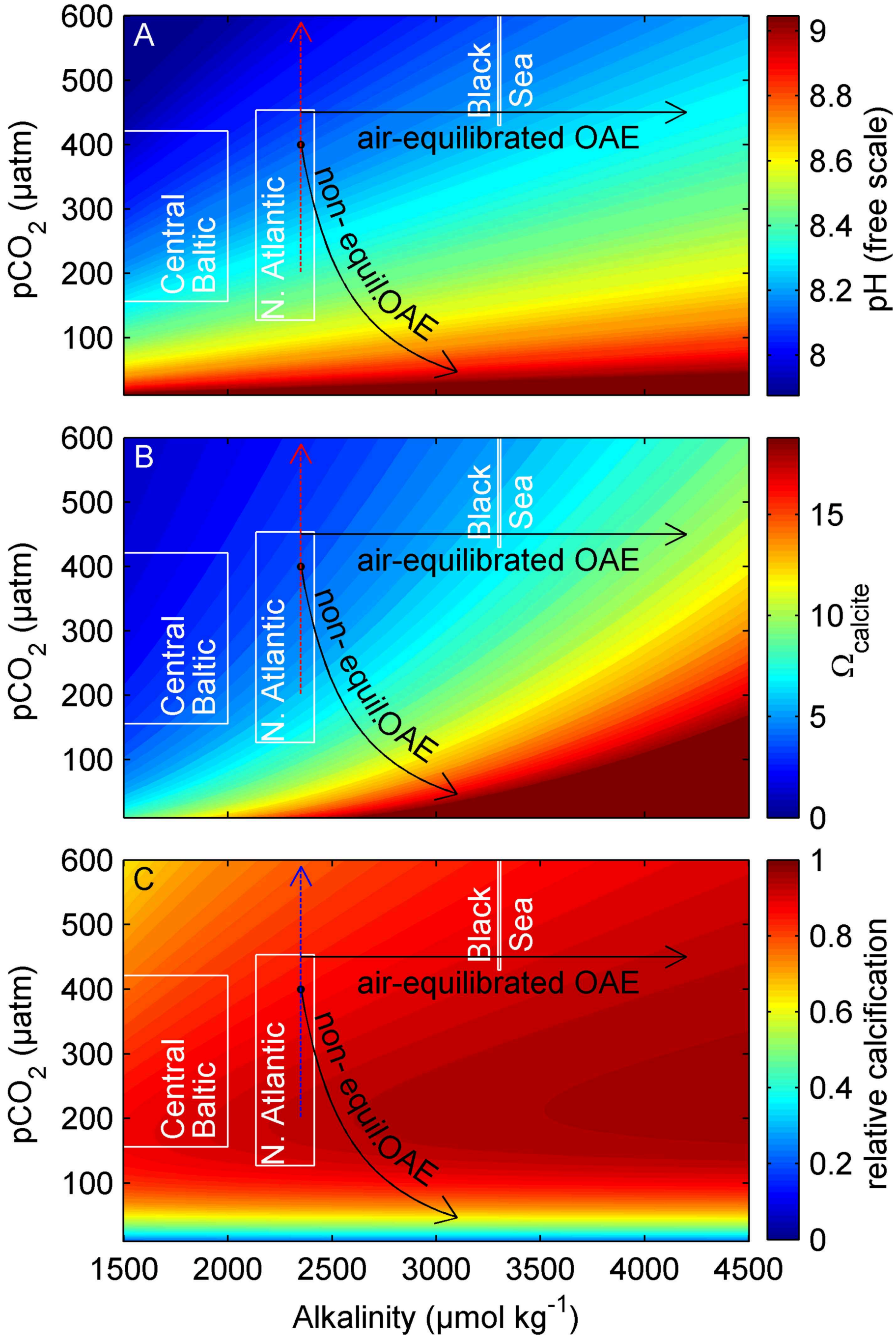

FIGURE 3 | Carbonate chemistry perturbation through EW/OAE. **(A)** Proton concentration, **(B)** Ω of the $CaCO_3$ mineral calcite, and **(C)** relative calcification rates of the coccolithophore *Gephyrocapsa oceanica* as a function of alkalinity and pCO_2. The arrows show the trajectories of ocean acidification (dotted), air-equilibrated OAE, and non-equilibrated OAE. Approximate carbonate chemistry conditions of the Baltic Sea, Black Sea, and North Atlantic (0–100 m depth) are shown in white boxes (data from Goyet et al., 1991; Key et al., 2004; Müller et al., 2016). Calculations were done with CO2SYS (Zeebe and Wolf-Gladrow, 2001) (T =15° C, S = 35, H^+ on free scale, K_1, K_2 from Lueker et al., 2000). Calcification sensitivity of *G. oceanica* was determined in experiments and fitted to the coccolithophore calcification model proposed by Bach et al. (2015).

example, the phytoplankton spring bloom is typically dominated by large phytoplankton species (e.g., diatoms) but in the case of acute OAE perturbations the large species could be replaced by smaller ones which are morphologically better suited to deal with low CO_2 concentrations (Wolf-Gladrow and Riebesell, 1997; Flynn et al., 2012; Chrachri et al., 2018). Boyd and Newton (1995) compared organic carbon export to the deep ocean of the 1989 and 1990 North Atlantic spring blooms where a large diatom species was dominant in 1989 and a smaller one in 1990. They found that, while primary production was similar in both years, particulate organic carbon export from the surface to 3,100 m depth was substantially lower in 1990 when the smaller species dominated (Boyd and Newton, 1995). Thus, it will be important to reveal if the transient but possibly pronounced carbonate chemistry perturbations associated with OAE have the potential to alter succession patterns and associated biogeochemical processes such as carbon export on larger scales.

EW/OAE will very likely have a much more sustained impact on calcifiers because the carbonate chemistry changes beneficial for calcification (i.e., increased Ω, HCO_3^-; lower H^+) will be permanent (**Figure 3**). An expected promotion of calcification through EW/OAE will occur at various scales. At first, low H^+/high Ω conditions will be sensed at the physiological level and facilitated $CaCO_3$ precipitation could allow the organisms to divert more energy into other metabolic processes and growth (Jokiel, 2011; Bach, 2015; Monteiro et al., 2016) (b). This may give calcifiers an energetic advantage and could increase their competitiveness relative to non-calcifiers. Indeed, a recent experiment with natural plankton communities demonstrated that differences in seawater pH can cause seemingly small differences in the growth rates of the dominant calcifier (in this case the coccolithophore *Emiliania huxleyi*) which were sufficient to induce order of magnitude differences in maximum population sizes among treatments (Riebesell et al., 2017). This not only had a considerable influence on the composition of the food web but also strongly altered biogeochemical fluxes such as carbon export or emissions of climate relevant trace gases (Bach et al., 2016; Webb et al., 2016; Riebesell et al., 2017). Accordingly, seemingly small effects of EW/OAE on organism physiology may be amplified in natural environments leading to ecological regime shifts toward calcifying species. Specifying the influence of EW/OAE on pelagic calcification will be important considering the biogeochemical relevance of this process as will be highlighted in the following paragraph.

About 1–2.6 Gt C are precipitated as $CaCO_3$ by marine organisms every year (Lebrato et al., 2010). The vast majority of the $CaCO_3$ is formed by coccolithophores, foraminifera, and pteropods (Sarmiento and Gruber, 2006; Lebrato et al., 2010). The formation of one mole of $CaCO_3$ reduces alkalinity by 2 moles (Zeebe and Wolf-Gladrow, 2001) so that calcification counteracts the desired effect of EW and OAE. For example, net calcification in the surface ocean was increased by 10 % (in mol) through OAE, then 20 % more alkalinity would be consumed and the effectiveness of OAE would be strongly reduced. Furthermore, the EW/OAE-induced increase of Ω could decrease $CaCO_3$ dissolution in sinking aggregates which are $CaCO_3$ under-saturated microenvironments in the surface ocean (Milliman et al., 1999). As a consequence, more alkalinity bound into sinking biogenic $CaCO_3$, would be exported to the deep ocean where its capacity to store atmospheric CO_2 is not exploited. These relatively easily quantifiable negative feedbacks are complicated by the influence of pelagic calcification on the efficiency of the biological pump (Armstrong et al., 2002; Francois et al., 2002; Klaas and Archer, 2002). If EW/OAE stimulates calcification, then more $CaCO_3$ ballast material becomes available to accelerate sinking velocities of particulate organic carbon (Honjo et al., 2008; Bach et al., 2016). The accelerated sinking could amplify the efficiency of the biological carbon pump because organic carbon that is built with CO_2 originating from the surface ocean would on average sink deeper into the ocean and lock the respired CO_2 in the deep ocean for a longer time (Kwon et al., 2009). This amplified deep ocean CO_2 sequestration of photosynthetically fixed carbon through the higher availability of ballast $CaCO_3$ could allow CO_2 to be taken up more efficiently by the oceans and therefore constitute a positive feedback on the CDR potential of EW/OAE. Thus, the hypothetical proliferation of calcification under EW/OAE applications could have a positive or negative feedback on EW/OAE efficacy, depending on if the "more ballast feedback mechanism" or the "less alkalinity feedback mechanism" become dominant [see (Riebesell et al., 2009) who discussed the influence of these two feedbacks in the context of ocean acidification]. An assessment of the "real" EW/OAE efficacy therefore needs to account for the response of calcifiers to EW/OAE and the biogeochemical feedbacks they drive.

RISKS AND CO-BENEFITS OF INCREASED SILICATE CONCENTRATIONS

Perturbation

EW and OAE can be realized with different minerals (Hartmann et al., 2013; Renforth and Henderson, 2017). Olivine-rich silicate rocks like dunite are considered as most effective because they occur naturally, are relatively fast dissolving, and do not require energy-intense chemical processing before their dissolution (Schuiling and Krijgsman, 2006; Renforth and Henderson, 2017). The flipside is that the desired generation of alkalinity coincides with the release of other chemicals such as dissolved silicate (DSi). Dissolving one mole of olivine leads to an increase in total alkalinity by 4 moles and in silicic acid (H_4SiO_4) by one mole (Köhler et al., 2013; **Box 1**).

Estimating how much DSi enters pelagic ecosystems is difficult for EW but possible for OAE. The difficulty for EW comes from the unknown fraction of DSi that is retained in soils, as well as on land and in freshwater ecosystems. This retention will most likely be significant although a considerable amount will undoubtedly be discharged into in coastal environments (Köhler et al., 2010; Dürr et al., 2011). OAE inputs are easier to assess because OAE only makes sense when all, or at least the vast majority, of the mineral added to the oceans dissolves in water layers which are in contact with the atmosphere within a few years after dissolution. Therefore, it can be assumed that most of the Si contained in silicates such as olivine would dissolve and enrich the DSi pool in the surface ocean. Assessments under more realistic constraints (e.g., accounting for mineral grinding and distribution) suggest that the maximum scale of mineral addition through OAE lies somewhere at around 10 Gt year^{-1} which is roughly 70 Tmol DSi year^{-1} if dunite was used (**Box 2**). Thus, OAE would be a source of DSi to the surface ocean that 5–14 fold higher than all natural sources combined (**Box 2**).

Current Understanding

DSi is an essential nutrient for silicifying organisms and by far the most important silicifiers in the ocean are diatoms (Tréguer and De La Rocha, 2013). These are a group of globally distributed phytoplankton that utilize DSi to form a shell (frustule) made of biogenic silica (opal) (Sarthou et al., 2005). Diatoms were estimated to contribute ∼25% to primary production on Earth (Nelson et al., 1995; Field et al., 1998). They usually initiate the seasonal phytoplankton succession and form blooms until they become nutrient-limited and other taxa with alternative nutrient acquisition strategies take over (Sommer et al., 2012).

DSi is often the nutrient that constrains diatom proliferation because it is highly deficient relative to other macronutrients throughout most subsurface water masses that supply nutrients to the euphotic zone through upwelling (Sarmiento et al., 2004). In fact it is the very "success" of diatoms throughout the Cenozoic that is thought to have driven the decrease in global ocean Si concentration (Conley et al., 2017). The deficiency in modern subsurface waters likely has two reasons. First, the opaline frustule dissolves relatively slowly so that DSi is remineralized on average at greater depths than other nutrients (Dugdale et al., 1995). Second, much of the subsurface water is formed by the subduction of Southern Ocean surface water (Sloyan and Rintoul, 2001). Before this water mass is subducted and spreads northwards, diatoms strip out and export DSi more than other nutrients thereby trapping DSi in the Southern Ocean (Sarmiento et al., 2004; Primeau et al., 2013; Holzer et al., 2014). These mechanisms explain why global diatom productivity in the present oceans is lower than it could be without prevailing shortage of DSi (Ragueneau et al., 2000; Sarmiento et al., 2004).

Important Knowledge Gaps and Testable Hypotheses

Applications of silicate-based EW and OAE could relieve the present day DSi deficiency (Köhler et al., 2013). The DSi concentrations where diatom DSi uptake rates reach half of the theoretical maximum ($K_{1/2}$) are species specific and range between 0.2 and 22 μmol kg^{-1} (Sarthou et al., 2005). High DSi affinity (i.e., low $K_{1/2}$) typically coincides with lower maximum uptake rates (V_{max}). Accordingly, increased DSi in the surface ocean would favor diatoms with high V_{max} and $K_{1/2}$ and therefore presumably fast-growing species (Sarthou et al., 2005). Furthermore, it could allow diatoms to reduce energy investments for DSi acquisition and enable them to construct thicker frustules (Martin-Jézéquel et al., 2000; Sarthou et al., 2005). Energy savings could accelerate diatom growth rates because available energy can be diverted into the acquisition of other nutrients while reinforced frustules would reduce grazing mortality (Hamm et al., 2003; Wilken et al., 2011; Friedrichs et al., 2013; Liu et al., 2016). Indeed, diatoms typically out-compete other phytoplankton when DSi is plentiful and other essential resources are sufficiently available (Armbrust, 2009). Thus, under silicate-based EW/OAE, diatoms could consume a larger fraction of the other limiting nutrients like N and P and ultimately become more dominant than they are already today. However, it will be important to assess if all diatoms profit equally from DSi enrichment due to the large range of DSi affinities among species (Sarthou et al., 2005). In theory, more heavily silicified diatom species should profit over proportionally from silicate-based EW/OAE as these have inherently higher DSi requirements to satisfy. Thus, it will be important to unravel if silicate-based

BOX 2 | Additions of key elements in a global OAE scheme.

In this BOX we approximate how much Si, Ca, Mg, Fe, and Ni could be added to the surface ocean under an intense and immediately (i.e., 2020) initiated application of OAE. All approximations should therefore be seen as an upper threshold. Please note that we refrain from approximating element inputs via EW due to the unconstrained degree of element retention by the terrestrial filter. Our estimations are based on idealized scenarios calculated by Köhler et al. (2013), Keller et al. (2014), and Lenton et al. (2018) assuming an even addition of 10 Gt of Mg_2SiO_4 (Köhler et al., 2013) or $Ca(OH)_2$ (Keller et al., 2014; Lenton et al., 2018) to the surface ocean excluding ice covered areas ($Ca(OH)_2$ "slaked lime" is $CaO + H_2O$). For both minerals this results in a TA addition of around 250 Tmol year^{-1} because olivine adds double the amount of TA when dissolving but also has roughly double the molecular weight of $Ca(OH)_2$. Lenton et al. (2018) simulated OAE for the twenty-first century also under the low emission scenario RCP2.6 and our estimation follows their scenario because they show that it only makes sense to implement OAE on a global scale when it is accompanied with rapid decarbonization.

The amounts of Si, Ca, Mg, Fe, and Ni added alongside TA strongly depend on the purities of minerals used for OAE. For our estimations we used elemental compositions of dunite provided by the Geochemistry of Rocks of the Oceans and Continents (GEOROCS) database (http://georoc.mpch-mainz.gwdg.de/georoc/). Values given in **Table 1** are means ±SD of 609 Si, 604 Ca, 665 Mg, 496 Fe, and 145 Ni dunite samples all of which are globally distributed. For CaO, we used approximate compositions of $CaCO_3$ precursors from America and central Europe (Gabe and Rodella, 1999; Galván-Ruiz et al., 2009; Šiler et al., 2018).

Table 1 shows that the perturbation potential under an even distribution of minerals scales with element background concentrations in seawater. When considering a relatively even distribution we would consider the perturbation potentials to be high for Si, Fe Ni, intermediate for TA, and low for Ca and Mg. However, as mentioned in the main text, an even distribution of minerals over the entire surface ocean seems unfeasible due to logistic constraints (Köhler et al., 2013). Thus, even those elements marked with a lower potential under these idealized conditions may have significant influence on pelagic ecosystems at perturbation hotspots where they occur at high concentrations.

It is important to emphasize that all values given in **Table 1** should be seen as rough approximations. Large uncertainties are, for example, due to large variabilities in chemical source rock composure or uncertainties in estimates of natural inputs of TA and the other elements into the oceans. Furthermore, the added amounts do not account for bioavailability of elements. For example, Si in quicklime could be non-soluble quartz so that inferences on bioavailability must consider solubilities of individual phases. Likewise, Fe has generally a low solubility in seawater (Boyd and Ellwood, 2010) so that a very large fraction of the added Fe would presumably not become bioavailable. Nevertheless, in the particular case of Fe, the amounts that would be added in this 10 Gt dunite-based OAE scenario are so large relative to the surface seawater concentration that they could offset Fe limitation in large parts of the ocean even under the assumption that only 0.1% of dunite Fe becomes bioavailable (Hauck et al., 2016).

(Continued)

BOX 2 | Continued

TABLE 1 | Estimated additions of key components in an OAE scenario with 10 Gt rock year^{-1} (see **BOX 2** for details).

Component	Residence time in the oceans[a] (years)	Dissolved surface ocean concentration[b] (μmol kg^{-1})	Natural input of dissolved element into the oceans[c] (Tmol year^{-1})	Source material for OAE	Element content in source rocks (mol kg rock^{-1})[d]	Amount added via OAE[e] (Tmol year^{-1})	N-fold the natural inputs	Amount added from 2020–2090 (Tmol)	Surface ocean concentration increase by 2090[f] (μmol kg^{-1})
TA	100,000	2,000–2,500	30			250	8	17,500	25–400
Si	10,000	0–80	5–14	Dunite	6.7 ±0.6	67	5–14	4,692	
				Carbonate	0.70	7	0.5–1.4	490	
Ca	1,000,000	8,800–11,700		Dunite	0.3 ±0.6	3.2		225	0.3–5
				Carbonate	15	150		10,500	15–240
Mg	11,600,000	45,500–60,600	6	Dunite	10 ±2.4	101	16	7,045	10–160
				Carbonate	0.4	4	0.7	280	0.4–6
Fe	40	mostly <0.001		Dunite	1.4 ±0.5	14		971	
				Carbonate	0.18	2		126	
Ni	4,000–10,000	0.002–0.012	1.5	Dunite	0.04 ±0.01	0.4	0.3	30	
				Carbonate	0.00003	0.0003	~0.0002	0.02	

[a] *Residence time for TA was estimated by dividing the oceanic inventory (~3 × 10^{18} mol) by riverine inputs (next column); Si from Tréguer and De La Rocha (2013); Ni from Gall et al. (2013); other values from Sarmiento and Gruber (2006).*

[b] *TA from Takahashi et al. (2014); Si from the World Ocean Atlas (https://www.nodc.noaa.gov/OC5/indprod.html); Ca and Mg from Zeebe and Wolf-Gladrow (2001) for a salinity range from 30 to 40 assuming conservative behavior; Fe from Schlitzer et al. (2018); Ni from Gall et al. (2013). Please note that the given ranges cover most of the surface ocean but deviations can occur regionally (e.g., Fe can be considerably higher in coastal regions or when affected by dust or hydrothermal plumes).*

[c] *TA from Amiotte Suchet et al. (2003) with only riverine alkalinity (HCO_3^-) influx considered; Si from Tréguer and De La Rocha (2013); Mg from by Tipper et al. (2006); Ni from Gall et al. (2013). Please note that values for Ca and Fe are not provided due to large uncertainties (Jickells et al., 2005; Fantle and Tipper, 2014). For example, inputs of DFe are hard to quantify as it is difficult to constrain solubilities of particulate Fe sources (Jickells et al., 2005; Boyd and Ellwood, 2010).*

[d] *Please keep in mind that the element contents within source rocks for EW/OAE vary widely. Given the large globally distributed deposits of silicate and carbonate rocks, it may be possible to extract material to optimize the chemical composition or minimize the environmental impact. Such a procedure would require careful regulation and verification.*

[e] *The assumption is that 10 Gt of rock coincide with an addition of 250 Tmol TA year^{-1}. The other elements are scaled to this mass with the molar contents per kg rock^{-1} given in the previous column. Please note that the addition of 250 Tmol TA year^{-1} for 10 Gt of rock is an (idealized) upper estimate based on the assumption that pure Mg_2SiO_4 or $Ca(OH)_2$ dissolve. In reality annual TA additions through 10 Gt rock would likely be lower because such chemical purities will not be reached.*

[f] *Concentration increase for TA from Lenton et al. (2018) which vary regionally as indicated by the given range. The upper estimate is based on a scenario where TA was added evenly over tropical surface ocean. Please note that an even addition is probably unfeasible so that higher concentrations would very likely occur in a more realistic scenario with localized additions. Concentration increases are also provided for Ca and Mg as these should, like TA, behave conservatively.*

EW/OAE has the potential to shift diatom communities toward more heavily silicified species.

A particularly interesting aspect in the context of community shifts is the competition between calcifiers and silicifiers. We hypothesized above that calcifiers profit from alkalinity enhancement and would become more dominant under EW/OAE. However, a prerequisite for the success of calcifiers would be that growth of the usually more competitive silicifiers is kept in check by DSi limitation (Egge and Jacobsen, 1997; Tyrrell and Merico, 2004). Under silicate-based EW/OAE, this limitation is relieved so that the fertilization of calcifiers through alkalinity enhancement could be overcompensated by DSi fertilization on silicifiers. Humborg et al. (1997) provide an excellent case study where this effect may actually have been observed. The river Danube contributes ~70 % of the river inputs into the Black Sea but was dammed in the early 1970s. The damming reduced DSi inputs into the Black Sea and promoted coccolithophores at the expense of diatoms (Humborg et al., 1997). The reverse case (i.e., increase of DSi) would be true for silicate-based EW/OAE. Here, calcifiers may not be able to benefit from more favorable carbonate chemistry because silicifiers benefit at the same time. Addressing this is very important because the balance between calcification and silicification has tremendous influence on the efficiency of EW/OAE. For example, if DSi inputs reduce net calcification and the subsequent export of $CaCO_3$ into the deep ocean by 10 % (in mol), then 20% more alkalinity remains in the surface ocean to absorb atmospheric CO_2. Such a reduction of marine alkalinity sink would enhance the CO_2 uptake capacity of the oceans and therefore increase the effectiveness of EW/OAE. Indeed, paleo-oceanographic studies suggest that increased DSi leakage from the Southern Ocean during glacial periods may have led to a shift from calcification to silicification globally (Brzezinski et al., 2002). This could have reduced the marine alkalinity sink and enhanced the oceanic CO_2 uptake capacity to such an extent that it can explain parts of the glacial-interglacial difference in atmospheric pCO_2 (Matsumoto et al., 2002). It may therefore be an argument to use silicate minerals for EW/OAE as this may reduce the biotic alkalinity sink and thereby indirectly increase the EW/OAE efficacy.

A shift from calcification to silicification under silicate-based EW/OAE would also alter quality and quantity of ballast material in water column. $CaCO_3$ has a higher density (2.7 g cm^{-3}) than BSi (2.1 g cm^{-3}) and is therefore considered to be more effectively accelerating the sinking of organic matter into the deep ocean (Klaas and Archer, 2002). However, $CaCO_3$ ballast is currently considered to be only regionally important in the oceans whereas the influence of BSi ballast is thought to be more widespread (Wilson et al., 2012; Le Moigne et al., 2014; Tréguer et al., 2018). Accordingly, a further increase in diatom biomass and potential shifts toward more heavily silicified diatoms at elevated DSi inputs (see above) may amplify the ballast potential of BSi and therefore carbon export in silicate-based EW/OAE schemes (Köhler et al., 2013). Thus, we speculate that the proliferation of silicifiers, even if this comes at the expanse of less pelagic calcifiers, could lead to a net increase of CO_2 sequestration into the deep ocean and therefore constitute a positive feedback on EW/OAE efficacy. If true, this could be another argument for silicate minerals.

RISKS AND CO-BENEFITS OF INCREASED (EARTH) ALKALINE METAL CONCENTRATIONS

Perturbation

It is expected that the most likely minerals to be used for EW/OAE are Ca or Mg oxides (e.g., CaO) and/or Ca or Mg silicates (e.g., Mg_2SiO_4). Ca and Mg are important ions for all organisms and in particular for calcifiers: Ca as a main ingredient and Mg as an inhibitor of precipitation which is excluded from the mineralisation space (Davis et al., 2000). Due to the high background concentrations of these alkali Earth metals in seawater, the relative change in their concentrations through addition to the surface ocean will be small (**Box 2**). However, it is likely that there will be considerable local perturbations through EW/OAE, and therefore Ca and Mg, at the input location for these schemes, on a regional scale. Biotic impacts through Ca or Mg may be pronounced at these perturbation hotspots before mixing through ocean currents disperses these chemicals to lower concentrations.

Current Understanding

High intracellular calcium is incompatible with life. At high intracellular concentrations, Ca^{2+} causes aggregation of proteins and nucleic acids, affects the integrity of lipid membranes, and can precipitate with cytosolic phosphate removing a major nutrient from bioavailability within the cell (Case et al., 2007). All forms of life, from their origin, need an effective system for Ca homeostasis, which keeps intracellular Ca^{2+} at concentrations low enough to prevent apatite precipitation, at levels of ~0.1 μM, and ~10,000–20,000 times lower than that in seawater (Carafoli, 1987). Although maintenance of such a large Ca^{2+} concentration gradient can be used effectively for Ca signaling as a core component of cellular regulation, it comes at a metabolic cost. Thus, any perturbation of the environmental Ca^{2+} concentration could impose an additional metabolic cost to all forms of life from bacteria to the highest eukaryotes, meaning that marine food webs could be affected by high Ca^{2+} at perturbation hotspots.

Magnesium is the second most abundant cellular cation after potassium. In comparison to Ca, high intracellular concentrations of total and free magnesium ion (Mg^{2+}) appear to be essential to regulate numerous cellular functions and enzymes, including ion channels, metabolic cycles, and signaling pathways up with intracellular concentrations ranging from 17-20 mmol kg^{-1}, just 2.5 times lower than ambient seawater (Romani, 2011). Understanding how cells regulate Mg^{2+} homeostasis remains incomplete but again must occur at some, but lesser than Ca, energetic cost to the cell (Romani, 2011).

Important Knowledge Gaps and Testable Hypotheses

From a biomineralization perspective, it is possible that calcification will benefit from additional Ca^{2+} in a quicklime

OAE scheme (Stanley et al., 2005). The influence of Mg in an olivine scheme is more complicated. Mg is known to be a strong inhibitor of inorganic calcite precipitation through incorporation into the $CaCO_3$ lattice (Davis et al., 2000). Mg incorporation raises the dissolution rate of the advancing crystal edge, which subsequently increases the mineral solubility, resulting in corresponding reduced net calcification. Aragonite is not inhibited by Mg incorporation into the $CaCO_3$ lattice in the same way that calcite is. Incorporation of Mg into aragonite's crystal structure has been shown experimentally to have no effect on growth rate of aragonite crystals (Berner, 1975; Gutjahr et al., 1996). Changes in the concentration of Mg in seawater relative to Ca do appear to be reflected in the plankton fossil record. Here, high Mg/Ca ratios through low absolute Ca concentrations, as in the modern ocean, promoted aragonite, and high Mg-calcite mineralizers. Conversely, low Mg-calcite mineralizers such as the coccolithophores, were more prevalent during the Mesozoic, which was characterized by oceans with low Mg/Ca and high absolute Ca^{2+} concentrations (Stanley and Hardie, 1998; Ries, 2010). Therefore, studies in palaeoceanography may be useful to elucidate future changes we could expect from EW/OAE scenarios by acting as suitable analogs.

There is also the possibility that higher Ca^{2+} and Mg^{2+} concentrations may interfere with the uptake of nutrients by phytoplankton (and/or bacteria). For example, Ca and Mg have been shown to influence Fe uptake in the freshwater cyanobacteria *Microcystis aeruginosa* (Fujii et al., 2015). It is therefore possible that Ca and Mg have similar effects on marine phytoplankton which may have implications from primary production at EW/OAE perturbation hotspots.

Given that foraminifera mineralize first by the engulfing of seawater by a membrane, the low Mg/Ca of their calcite compared to inorganic predictions suggests an active role for pumping of Mg away from the region of mineralization (Segev and Erez, 2006). Two species of benthic foraminifera incorporate more Mg into shells as Mg/Ca increases, suggestive that control on the Mg content of the mineralizing space is not strictly regulated. The greatest increase in precipitation of $CaCO_3$ was obtained at Mg/Ca of 1, and not at present-day seawater ratio ($Mg/Ca = 5$). Thus, it is possible that any decrease (increase) in the Mg/Ca ratio of the ocean will both increase (decrease) the degree of calcification of foraminifera, in addition to making the carbonate less (more) soluble by lowering its Mg/Ca ratio.

Pteropods are holoplanktonic gastropod molluscs which are the major producers of aragonite in the surface ocean today (Andersson et al., 2008). The response of pteropods to changing Mg/Ca is not well-constrained due to the difficulty of keeping pteropods alive in culture (Howes et al., 2014). The origination of the pteropods and rise in prevalence during high Mg/Ca in the Cenozoic may suggest that pteropods are favored by the emerging aragonite sea conditions (Burridge et al., 2017). Accordingly, they could be influenced oppositely to coccolithophores and foraminifera by Mg/Ca alterations associated with EW/OAE.

As a first step, it will be essential to constrain the impact of elevated Ca and/or Mg on the order of magnitude predicted in **Table 1**. To our knowledge, no work has specifically been completed on the response of plankton other than coccolithophores or foraminifera to increased Mg, Ca or altered Mg/Ca. This work may also be important to complete for the major group of silicifiers, the diatoms, due to the competition between silicifiers and calcifiers mentioned before. If CaO were added to the ocean, thereby decreasing Mg/Ca, it is reasonable to hypothesize that coccolithophores as competitors of diatoms would be further strengthened and take a larger share of the available nutrient pool. There is huge opportunity for the utilization of palaeoceanographic studies to help with these predictions, due to the rich fossil and sediment record providing evidence of secular changes in Mg/Ca ratios over geological time showing that silicifiers have risen to open ocean dominance in high Mg/Ca oceans (Falkowski et al., 2004).

RISKS AND CO-BENEFITS OF INCREASED TRACE METAL CONCENTRATIONS

Perturbation

All minerals suitable for EW/OAE will contain elements considered as "trace metals" (Hartmann et al., 2013; Schlitzer et al., 2018). Trace metals occur at relatively low concentrations in seawater so that perturbations at the scale anticipated for EW/OAE are likely sufficient to have a fertilizing and/or toxic effect on organisms (Hartmann et al., 2013; Moore et al., 2013; Hauck et al., 2016). The diversity of trace metals added to the oceans via mineral dissolution would be large and highly dependent on the source mineral used for EW/OAE. For example, alkaline industrial residuals such as steel slags are considered to be useful for EW/OAE but they can contain trace metals such as Cr, Mo, Ni, Pb which could negatively influence the environment above critical threshold concentrations (Mayes et al., 2008; Renforth, 2019). Likewise, naturally occurring minerals like olivine or source $CaCO_3$ for the production of CaO are associated with various elements, depending on their source region. It is beyond the scope of this paper to discuss the potential impacts of all trace elements contained in source material appropriately. We therefore focus on two particularly important ones—iron (Fe) and nickel (Ni). Fe is in focus because it is one of the key nutrients influencing pelagic ecosystems in the surface ocean (Moore et al., 2013; Tagliabue et al., 2017) and is abundant in minerals considered for EW/OAE (Hartmann et al., 2013). Ni is currently not considered as a key nutrient in the surface ocean (Moore et al., 2013) but its abundance in dunite, one of the major candidate rocks for EW/OAE, is very high so that highlighting this element is very reasonable (Meysman and Montserrat, 2017; Beerling et al., 2018).

Fe is rather immobile in soils (Gaillardet et al., 2014) so that Fe enrichment of rivers through EW should be relatively small. However, differences exist among rivers with those that contain large amounts of dissolved organic ligands ("blackwater rivers") likely transporting more dissolved Fe to the oceans (Krachler et al., 2015; Vieira et al., submitted) which is an important aspect to consider when selecting EW application sites on land. Once riverine Fe reaches the ocean, much of it will be rapidly scavenged, for example by salt-induced flocculation or by settling particles so that Fe perturbations through EW would be restricted mostly to coastal regions (Boyd and Ellwood, 2010) even though physical conditions at some estuaries (e.g., Congo) could transport material further offshore (Vieira et al., submitted). Ni is not as abundant as Fe in olivine but is more mobile in soils and more soluble in seawater (Gaillardet et al., 2014; Montserrat et al., 2017) so that Ni inputs through EW could be higher in coastal systems and potentially further off-shore. OAE short-circuits the terrestrial filter so that the trace metal contamination of the ocean realm would be significantly larger and directly depend on the dissolution rates of mineral powder and the solubilities of the dissolution products (Montserrat et al., 2017).

Current Understanding (Fe)

Fe is an essential micronutrient needed as a co-factor in many enzymes (Tagliabue et al., 2017). The concentration of dissolved Fe (DFe) is mostly <1 nmol L^{-1} in the surface ocean where productivity is in large parts Fe-limited (Moore et al., 2013; Schlitzer et al., 2018). Therefore, even a relatively small Fe addition of a few tons to Fe-limited surface ocean regions can induce a massive phytoplankton bloom visible from space (Boyd et al., 2007). The ideal olivine mineral for EW/OAE would be pure forsterite (Mg_2SiO_4; **Box 1**) but in nature such purity rarely exists. Here, olivines typically occur as a mixture of forsterite and fayalite (Fe_2SiO_4) (Taylor et al., 2016). A commonly found forsterite/fayalite mixing ratio is ∼9:1 and this rock type, termed "dunite," exists in sufficiently large quantities to sequester maybe some hundreds of Gt CO_2 from the atmosphere (Taylor et al., 2016). Fe enrichment scenarios tested with an Earth System model suggested that this iron fertilization of ocean primary production would increase OAE efficacy roughly by one third (Hauck et al., 2016). However, more recent thermodynamic modeling indicated that fayalite dissolution reduces alkalinity through secondary reactions (Griffioen, 2017), although this could not be confirmed in a dissolution experiment (Montserrat et al., 2017). Thus, the overall influence of Fe perturbations on the CDR potential of EW/OAE is very difficult to determine with our current level of understanding. Fe enrichment could, to a smaller extent, also occur under applications of artificially processed minerals. Quicklime for example is made by calcining natural $CaCO_3$ rich source rocks with purities of <98 % typically achieved at industrial scale (Renforth and Kruger, 2013; Renforth et al., 2013). The majority of the remaining mass is composed of silicate and iron oxides. Therefore, Fe or Si addition when using calcined limestone will certainly be lower than from silicates such as dunite (**Box 2**).

Important Knowledge Gaps and Testable Hypotheses (Fe)

Iron is often not the most limiting nutrient for phytoplankton in coastal environments as these usually receive DFe supply from sediment fluxes or terrestrial sources (Boyd and Ellwood, 2010). Increased DFe river discharge through EW may therefore have a limited effect on primary production near coasts. In the case of OAE the situation could be different. Counterintuitively, the addition of mineral particles could scavenge bioavailable DFe thereby reducing primary production. This may apply for mineral powder additions in DFe replete regions where DFe scavenging (adsorption) by volcanic ash particles has been shown to outweigh the Fe release through dissolution thereby leading to a net removal of DFe (Rogan et al., 2016). However, when particles are added to a DFe deficient system the dissolution term outweighs the scavenging term leading to a net increase in DFe (Rogan et al., 2016). Thus, OAE applied in DFe-limited areas should enhance primary production unless the ecosystem is acutely co-limited by other nutrients such as nitrate. Indeed, DFe and N co-limitation is widespread, especially on the edges of sub-tropical gyres (Browning et al., 2017). OAE would strongly relieve DFe but not N limitation thereby favoring N_2 fixing cyanobacteria (e.g., *Trichodesmium*) which are largely controlled by DFe availability in oligotrophic systems (Mills et al., 2004). It will therefore be essential to test if OAE could stimulate N_2 fixation rates of cyanobacteria in these regions that may ultimately increase primary production and the CDR efficacy of OAE.

Altogether, the DFe related effects on primary production would most likely coincide with pronounced shifts in the composition of the plankton community. For example, a specific stimulation of cyanobacteria may initiate a reorganization of the energy flow along trophic pathways, ultimately affecting the marine food web in its entirety. Such indirect effects of EW/OAE may even be stronger than the first order chemical perturbation and therefore need to be taken into account (Strauss, 1991; Walsh, 2013). Ultimately, all the positive and negative side-effects that have been anticipated for ocean fertilization with iron (e.g., increased ocean productivity, deep-ocean hypoxia; Oschlies et al., 2010) would also have to be considered in the context of EW/OAE when minerals with considerable Fe content are used.

Current Understanding (Ni)

Ni is rare in the Earth's crust but over-proportionally abundant in forsterite-rich olivine (Simkin and Smith, 1970). Ni is more mobile in soils than Fe especially at low pH and dissolves relatively quickly so that it is currently considered as the most important contaminant under silicate-based EW/OAE (Nieminen et al., 2007; Hartmann et al., 2013; Meysman and Montserrat, 2017; Montserrat et al., 2017; Beerling et al., 2018). It is an essential co-factor in variety of enzymes but Ni nutrient requirements and sensitivities to excess concentrations vary greatly among organisms (**Figure 4**). Eukaryotic microalgae have lower Ni requirements (mostly <2 nmol kg^{-1}) and are more sensitive to high concentrations than prokaryotes such as cyanobacteria (**Figure 4**). Ni has a higher solubility than Fe and occurs in the surface ocean mostly as Ni^{2+} in concentrations of about 2–12 nmol kg^{-1} so that bulk primary production is usually not limited by it (Konhauser et al., 2009; Gall et al., 2013; Moore et al., 2013). This also explains the general absence of a nutrient-like vertical concentration profile of Ni with the exception of the North Pacific (Glass and Dupont, 2017) where occasional Ni limitation of cyanobacteria communities have been observed (Dupont et al., 2010).

Important Knowledge Gaps and Testable Hypotheses (Ni)

The influence of dissolved Ni (DNi) on pelagic systems has not been a particularly strong focus in Biological Oceanography so

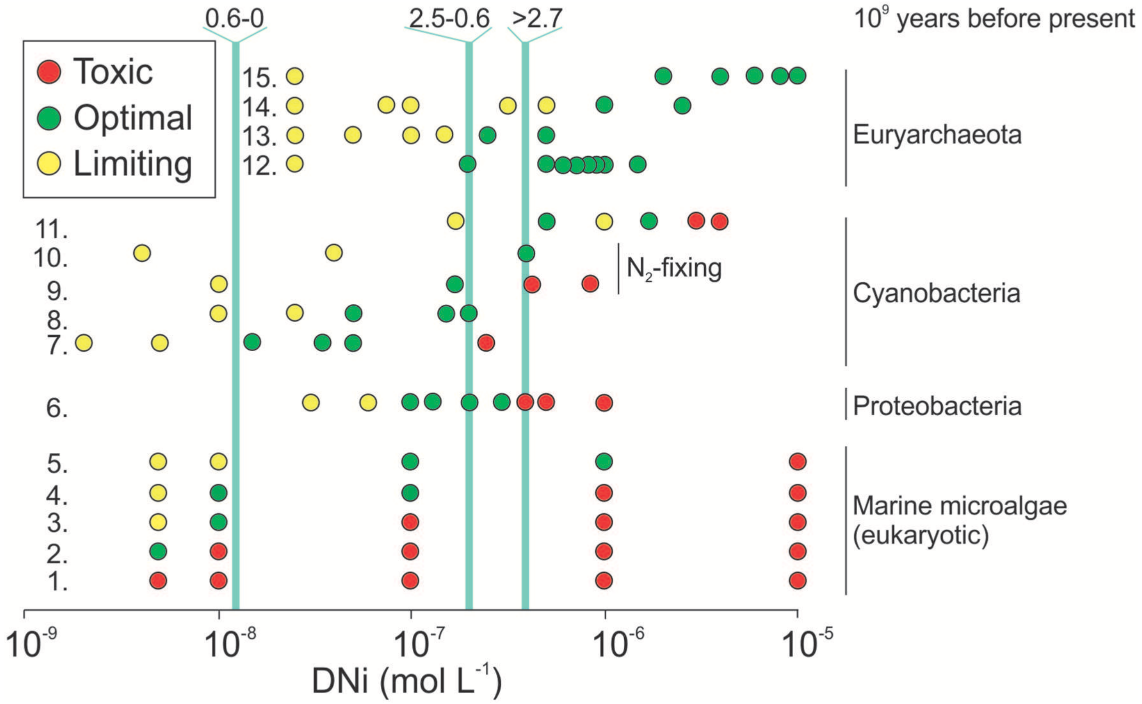

FIGURE 4 | Effects of DNi on growth of (marine) microbes. Marine microalgae were grown in urea. The phyla of each microalgae is given in brackets behind each species name based on www.AlgaeBase.org (Guiry et al., 2014): 1. *Pavlova lutheri* (Haptophyta), *Chaetoceros gracilis* (Bacillariophyta "diatom"), *Olisthodiscus luteus* (Ochrophyta); 2. *Prymnesium parvum* (Haptophyta); 3. *Rhodomonas* sp. (Cryptophyta), *Achnanthes brevipes* (Bacillariophyta "diatom"), *Amphidinium carterae* (Miozoa "dinoflagellate"), *Skeletonema costatum* (Bacillariophyta "diatom"), *Hymenomonas elongata* (Haptophyta), *Porphyridium cruentum* (Rhodophyta); 4. *Thalassiosira* spp. (Bacillariophyta "diatom"); 5. *Cyclotella cryptica* (Bacillariophyta "diatom"); Proteobacteria: 6. *Cupriavidus necator*; Cyanobacteria: 7. *Synechococcus* spp.; 8. Oscillatoria sp.; 9. *Nostoc muscorum* (freshwater); 10. *Trichodesmium erythraeum*; 11. *Arthrospira maxima*; Euryarchaeota: 12. *Methanococcus voltae*; 13. *Methanococcoides methylutens*; 14. *Methanobacterium thermoautotrophicum*; 15. *Methanothrix soehngenii*; Light blue lines are reconstructed paleo seawater DNi concentrations in 10^9 years before present. The figure was adopted from Glass and Dupont (2017) who compiled the data from numerous references. Reproduced by permission of The Royal Society of Chemistry.

far but it may become important in the context of EW/OAE. Here, DNi could either function as nutrient or toxin with pronounced impacts on primary production and plankton community composition.

Recent dissolution experiments with olivine have shown that an increase of alkalinity of about 100 μmol L^{-1} coincided with an increase of $\sim$3 μmol L^{-1} in dissolved Ni (DNi) due to non-stoichiometric dissolution (Montserrat et al., 2017). These DNi concentrations are $\sim$3 orders of magnitude higher than ambient concentrations and would likely be harmful for many organisms (**Figure 4**). Thus, olivine-based EW/OAE could strongly affect marine plankton communities near perturbation hotspots. As for Fe, impacts of DNi will likely differ regionally and depend on the constitution and evolutionary history of plankton communities. For example, communities near estuaries may be adapted to higher DNi concentrations, especially when discharging rivers come from basaltic, olivine rich catchment areas (Nieminen et al., 2007). However, this should be the exception rather than the rule so that we would expect rather detrimental effects on biodiversity and productivity when too much DNi is released locally.

DNi is generally not considered to be an element that is bio-accumulated strongly by organisms (bio-accumulation = enrichment within organism tissues) (Nieminen et al., 2007). Without bio-accumulation, there is also little potential for Ni bio-magnification along the food chain (i.e., Ni enrichment with increasing trophic level). However, as mentioned earlier, Ni is likely one of the most important trace metal contaminants under olivine-based EW/OAE but certainly not the only one. Other potential trace metals (e.g., Cr, Cu, or Cd) (Simkin and Smith, 1970; Beerling et al., 2018) may become bio-accumulated and bio-magnified, particularly in phytoplankton evolutionarily associated to the "red lineage" (Wilson et al., 2019). Furthermore, a suite of anthropogenic materials (e.g., slag from the production of steel, or mining residuals) could be used for EW (Renforth, 2019). These can contain a range of elemental compositions, and a material by material assessment is required for wider-scale impact. Thus, it may be possible that certain trace metals introduced into the environment via EW/OAE could bio-accumulate and bio-magnify which may cause health problems, particularly for end-members in the food chain like humans (Järup, 2003). It is therefore important to identify minerals associated with such risks and not use them for EW/OAE. Indeed, trace metal toxicity may become a key decisive factor for the acceptability of EW/OAE applications and strongly influence the choice of rocks that could ultimately be used.

The toxicity-related hypothesis outlined above refers to EW/OAE scenarios where very high DNi concentrations occur. However, after dilution of the perturbation signal in the oceans through space and time DNi may dilute from harmful to fertilizing concentrations (**Figure 4**). Ni is an essential co-factor for enzymes involved in urea cycling and utilization (Nieminen et al., 2007; Glass and Dupont, 2017) and has been shown to limit growth of cyanobacteria- and picoeukaryota-dominated phytoplankton communities (Dupont et al., 2010). Therefore, EW/OAE-related DNi inputs could stimulate primary production in certain oligotrophic settings. Indeed, the growth rate of the predominant N_2-fixer *Trichodesmium* has been shown to profit strongly from DNi amendments (Ho, 2013) such that ecological regime shifts toward more cyanobacteria in subtropical gyres would be a plausible consequence of olivine-based EW/OAE. If true, moderate increases of DNi may under certain circumstances locally enhance marine productivity and therefore EW/OAE efficacy through a stimulation of N_2-fixation.

SYNTHESIZING THE INFLUENCE OF DIFFERENT PERTURBATION SCHEMES IN A COMMON FRAMEWORK

In the sections above we highlighted potential risks and co-benefits associated with individual mineral dissolution products. In the following we try to synthesize the various factors in a common framework to establish potential scenarios under sustained EW/OAE.

EW and OAE at the scale needed for relevant CDR could shift the present day ecological/biogeochemical equilibrium ("blue ocean") into a new steady state depending on the applied source mineral (**Figure 5A**). Quicklime or other industrially reprocessed minerals should contain relatively fewer contaminants (**Box 2**) so that EW/OAE would mostly constitute an increase of alkalinity and Ca^{2+}. Here, calcifiers should profit from the favorable carbonate chemistry while silicifiers and N_2-fixers are affected to a minor extent (perhaps slightly negatively because calcifiers acquire more of the available nutrients). In such a scenario, the blue ocean could turn into the "white(r) ocean" where calcifiers become more important (**Figure 5B**).

When natural minerals such as olivine are used then the desired alkalinity increase coincides with a substantial DSi and trace metal enrichment (**Box 2**). In this scenario, global primary production would likely increase mostly due to Fe fertilization in Fe-limited open ocean regions (Köhler et al., 2013; Hauck et al., 2016). Silicifiers could profit from the additional DSi and strengthen their global dominance. Coccolithophores could be out-competed by silicifiers. N_2-fixers could profit from Fe and Ni enrichment and reinforce primary production further until other nutrients (e.g., P) become limiting. This transformation under olivine-based EW/OAE could result in a more productive "green(er) ocean" (**Figure 5C**).

If ecological shifts toward a white(r) or green(er) ocean were shown to be realistic consequences of different EW/OAE schemes, they may influence: 1) if humankind is willing to accept changes of the affected ocean environments including their associated risks and co-benefits; 2) the choice of minerals to be used and amounts to be added in order to find the right balance between CDR efficacy and acceptability of EW/OAE.

OUTLOOK

The risks and co-benefits discussed in this paper are based upon the presently available information on the most prominent dissolution products of the most widely considered source minerals (quicklime and silicate minerals such as olivine). Future research might reveal other minerals to be more suitable for EW/OAE and these minerals may be associated with other risks and co-benefits. Furthermore, the timing of mineral additions may become an important factor to consider in OAE schemes since side-effects may play out differently when additions occur in different seasons. Finally, we acknowledge that there are most certainly other risks/co-benefits that have been neglected in our discussion and the list will only become more complete once the hands-on research on this topic gained momentum.

Testing the "white or green ocean hypothesis" will require a broad and mechanistic understanding how dissolution products affect species and, more importantly, entire ecosystems. It needs the full scale of experimental approaches, including physiological studies, mesocosms, and modeling. Importantly, lessons learned from previous research efforts suggest that *in situ* surveys should be initiated early in a developing field to cover the ecological complexity of natural plankton communities (Riebesell and Gattuso, 2015). This could be realized by studying natural analogs such the Black Sea as an example for a high alkalinity marginal sea. Likewise, sites of intense natural weathering of basaltic rocks along volcanic islands may provide insights on more regional impacts (Hartmann et al., 2009). Studies of marine paleo-environments may also improve our understanding of EW/OAE, since there are episodes in the geological past where seawater chemistry had similarities to what could be expected under EW/OAE in the future [e.g., higher DNi (Glass and Dupont, 2017) or higher alkalinity (Tyrrell and Zeebe, 2004)].

Ultimately, it will be important to plan and eventually execute (sub-) mesoscale *in situ* OAE experiments since only experiments at the scale envisioned for OAE perturbations can provide a more comprehensive picture of ecosystem-wide impacts (Carpenter, 1996). Indeed, previous mesoscale iron enrichment studies have provided insights on the efficacy and potential side-effects of this NET that would probably not been revealed with conventional methodology (Boyd and Bressac, 2016). However, comprehensive risk assessments for environment including ethical considerations in an open public discourse would have to be made to assess whether such experimentation would be morally acceptable (Lawford-Smith and Currie, 2017; Oschlies and Klepper, 2017; Pidgeon and Spence, 2017; GESAMP, 2019).

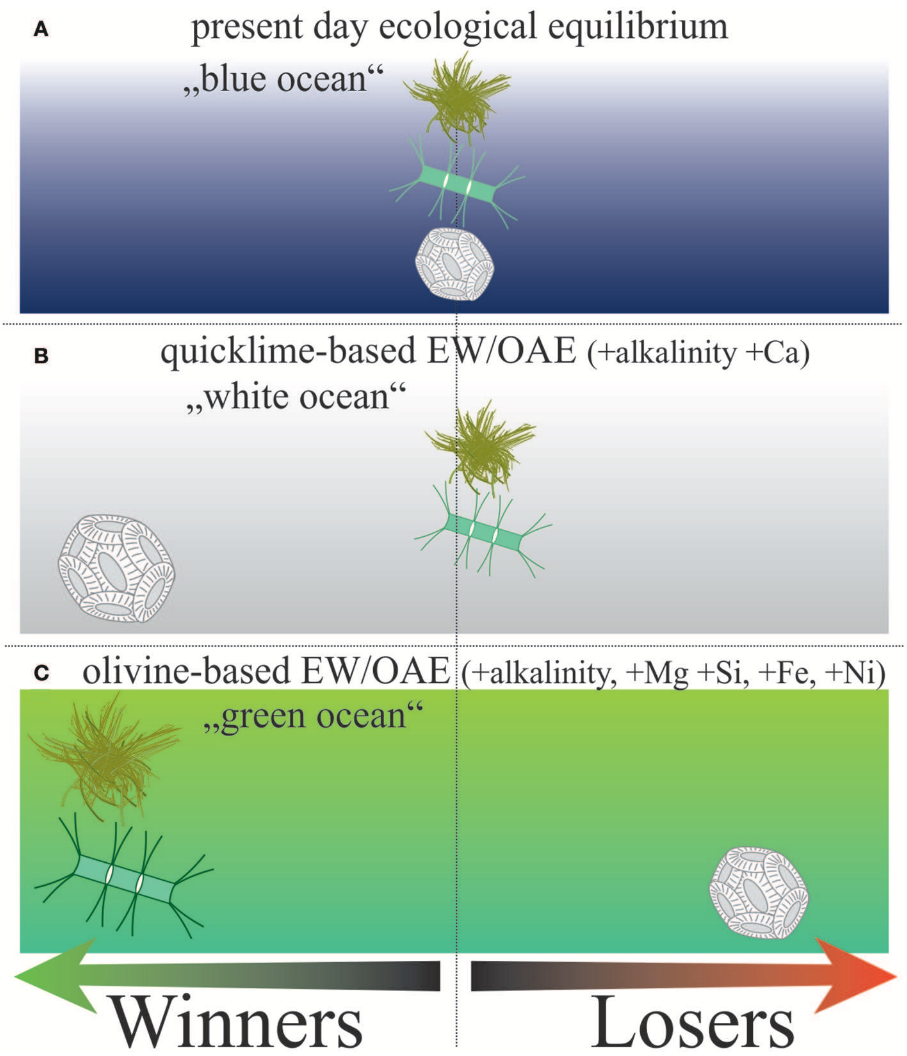

FIGURE 5 | The "white or green ocean hypothesis." This schematic drawing synthesizes the potential changes in the present day ecological equilibrium **(A)** under either quicklime-based **(B)** or olivine-based EW/OAE **(C)**. ✸ = cyanobacteria; ◍ = coccolithophores; ✳ = diatoms.

Concerning legal aspects, there is currently some unclarity with respect to the governance for NETs (Boyd and Vivian, 2019). OAE may fall into either national regulation if activities were limited to the inland waters, or the global "London Convention (LC)/ London Protocol (LP)," treaties that regulate marine pollution from the dumping of wastes or other matter at sea [The International Maritime Organization (IMO) listed 87 members to the LC and 51 members to the LP where the latter was agreed to further modernize the LC and eventually replace it (July, 2019; www.imo.org)]. The LC/LP sets high hurdles on legitimate scientific research involving additions of minerals to the open oceans (e.g., ocean iron fertilization and potentially also open ocean alkalinity enhancement) but does not prohibit it (GESAMP, 2019). EW is not within the scope of the LC/LP because land-based sources of dissolution products would not be considered within their definition of "dumping" materials in the oceans. The assessment of risks and co-benefits of EW/OAE should therefore be paralleled by the development of a precise regulatory framework that is informed by the independent scientific community (Oschlies and Klepper, 2017; Gattuso et al., 2018; Boyd and Vivian, 2019).

Assessing EW/OAE impacts on ocean ecosystem is a new and understudied research branch in Biological Oceanography but the methodology and the scientific expertise is already largely established. For example, the widespread interest in the marine carbon cycle and ocean acidification has catalyzed the proliferation of global networks of stationary or robotic carbonate chemistry observation platforms which could be used to assess the success of OAE to store CO_2 in seawater (e.g., http://www.goa-on.org or http://biogeochemical-argo.org). This constitutes an invaluable basis since only certified long-term storage (ideally $>>1{,}000$ years) can be used for carbon management. Furthermore, biology-centered ocean acidification research formed a well-trained research community that can easily design and perform experiments to investigate EW/OAE-related carbonate chemistry impacts on marine biota. Marine Si cycling and trace metal ecotoxicology are equally established research fields and the available knowledge of highly skilled scientists will become invaluable to determine risks and co-benefits of EW/OAE for the oceans. The key challenge is to bring together expertise from already established fields in Biological Oceanography in order to make the necessary progress within the limited timeframe given by the remaining carbon budget to stay below 2°C.

DATA AVAILABILITY STATEMENT

Publicly available datasets were analyzed in this study. All references where the data is available is given in the corresponding text passages.

AUTHOR CONTRIBUTIONS

LB conceptualized the research, compiled information in **Table 1**, composed the figures, and drafted the manuscript except for section risks and co-benefits of increased (Earth) alkaline metal concentrations. SG and RR drafted risks and co-benefits of increased (Earth) alkaline metal concentrations. All authors revised and improved the manuscript.

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Soil C Sequestration as a Biological Negative Emission Strategy

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Soil carbon (C) sequestration is one of three main approaches to carbon dioxide removal and storage through management of terrestrial ecosystems. Soil C sequestration relies of the adoption of improved management practices that increase the amount of carbon stored as soil organic matter, primarily in cropland and grazing lands. These C sequestering practices act by increasing the rate of input of plant-derived residues to soils and/or by reducing the rates of turnover of organic C stocks already in the soil. In addition to carbon dioxide removal potential, increases in soil organic matter/soil C content are highly beneficial from the standpoint of soil health and soil fertility. Practices to increase soil C stocks include well-known, proven techniques, or "best management practices" (BMP) for building soil carbon. A second category includes what we refer to as frontier technologies for which significant technological and/or economic barriers exist today, but for which further R&D and/or economic incentives might offer the potential for greater sequestration over the longer term. We reviewed published estimates of global soil carbon sequestration potential, representing the biophysical potential for managed cropland and/or grassland systems to store additional carbon assuming widespread (near complete) adoption of BMPs. The majority of studies suggests that 4–5 $GtCO_2$/y as an upper limit for global biophysical potential with near complete adoption of BMPs. In the longer-term, if frontier technologies are successfully deployed, the global estimate might grow to 8 $GtCO_2$/y. There is a strong scientific basis for managing agricultural soils to act as a significant carbon (C) sink over the next several decades. A two-stage strategy, to first incentivize adoption of well-developed, conventional soil C sequestering practices, while investing in R&D on new frontier technologies that could come on-line in the next 2–3 decades, could maximize benefits. Implementation of such policies will require robust, scientifically-sound measurement, reporting, and verification (MRV) systems to track that policy goals are being met and that claimed increases in soil C stocks are real.

Keywords: biophysical potential, carbon sequestration, best management practice, carbon dioxide removal and storage, agricultural soils

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INTRODUCTION

Together with other terrestrial ecosystem-based strategies for CO_2 removal [i.e., afforestation, bioenergy with carbon capture and storage (BECCS)], soil carbon sequestration relies on plant photosynthesis to carry out the initial step of carbon "removal" from the atmosphere. However, rather than increasing the storage of carbon contained in plant biomass, soil C sequestration relies on management practices that increase the amount of carbon stored as soil organic matter, primarily in cropland and grazing lands. Part of the attraction of soil C sequestration as a biological negative emission (BNE) strategy is that carbon stocks are most depleted on lands currently under agricultural management and thus this approach to CO_2 removal doesn't require land use conversions (e.g., to forests) and competition for land resources. In addition, increases in soil organic matter/soil C content are highly beneficial from the standpoint of soil health and soil fertility, which provides additional incentives for adopting soil carbon sequestering practices. In this paper, we review and summarize data and understanding, from field to global scale, of the capacity for soil carbon sequestration to play a role in helping draw down atmospheric CO_2 concentration (NASEM, 2019).

Globally, soils contain about 1,500 Gt of organic carbon (C)[1] to 1 m depth and 2,400 GtC to 2 m depth (Batjes, 1996). Thus, the total size of the soil carbon reservoir exceeds the total mass of carbon in vegetation and atmosphere combined. About 45% of global soils are under some form of agricultural use (i.e., cropland and grazing land). In most soils, organic matter[2] makes up a small fraction (∼1–10%) of the total soil mass which is dominated by mineral matter (i.e., sand, silt, and clay particles); these are so-called "mineral soils." The vast majority of agricultural lands are on mineral soils. In contrast, "organic soils" (e.g., peat soils), as the name implies, have very high organic matter content. Organic soils form where anaerobic conditions restrict decomposition, such that partially decayed plant material accumulates, making up the matrix of the soil.

Most agricultural soils (both mineral and organic) are depleted in C relative to the native ecosystems from which they were derived, due to reduced net primary production and export of harvested biomass—which reduce C inputs to soil; nutrient depletion, intensive soil disturbance, and soil erosion are other contributing factors to soil C depletion (Paustian et al., 1997). Most cropland mineral soils have lost 30–50% of the C stocks in top soil layers (0–30 cm) relative to their native condition (Davidson and Ackerman, 1993). In contrast, grassland soils managed for grazing may or may not have suffered similar C losses relative to their native state, depending on how they have been managed. Grasslands that have been overgrazed and poorly managed are likely significantly depleted in soil C, whereas well-managed grasslands may have C stocks equal to or exceeding their original native condition (Conant et al., 2016).

The organic carbon content of soils is governed by the balance between the rate of C added to the soil from plant residues (including roots) and organic amendments (e.g., manure, compost), and the rate of C lost from the soils, which is mainly as CO_2 from decomposition processes (i.e., heterotrophic soil respiration). Other organic C can be lost as CH_4 from anaerobic (e.g., flooded) systems as well as from leaching of dissolved organic C, but these are minor loss processes in most ecosystems. Soil erosion can greatly affect C stocks at a particular location, but at larger scales erosion may not represent a loss process *per se* but rather a redistribution of soil C. Effects of erosion on the global C balance is a subject of continued research but soil erosion may actually result in a small net C sink, because burial of C-rich sediment reduces its decomposition rate and, with erosional exposure, low C subsurface soil layers can have a higher capacity to store additional C (van Oost et al., 2007).

In native ecosystems the rate of detrital C inputs is a function of the type (e.g., annual vs. perennial, woody vs. herbaceous) and productivity of the vegetation, largely governed by climate but also nutrient availability and other growth determining factors. Decomposition rates are controlled by a variety of factors including soil temperature and moisture, drainage (impacting soil O_2 status) and pH. Soil physical characteristics such as texture and clay mineralogy also impact the longevity and persistence (i.e., mean residence time) of soil C, by affecting organic matter stabilization processes, i.e., the extent to which organic matter is "protected" from decomposition through mineral-organic matter associations (Schmidt et al., 2011).

In managed ecosystems such as cropland and grazing land both the rate of C input as well as the rate of soil C loss via decomposition are impacted by the soil and crop management practices applied. In general, soil C stocks can be increased by: (a) increasing the rate of C addition to the soil, which removes CO_2 from the atmosphere, and/or (b) reducing the relative rate of loss (as CO_2) via decomposition, which reduces emissions to the atmosphere that would otherwise occur.

However, three key points need to be made regarding the pattern of gains or losses of soil C. The first is that with increased C inputs and/or decreased decomposition rates, soil C stocks tend toward a new equilibrium state and thus after a few decades C gains attenuate, becoming increasingly small over time (Paustian, 2014). Secondly, because the soil C balance is governed by biotic processes, changes in management that lead to C gains are potentially reversible, i.e., if management reverts back to its previous condition, much or all of the gained C can be lost. Thus, practices that led to increased soil C need to be maintained long term. Third, mineral soils (i.e., non-peat soils) have an upper limit or "saturation level" of soil C (Six et al., 2002). While this maximum soil C concentration is much higher than is found in most managed soils, it does mean that soils that already have very high organic matter levels (e.g., >5% C by mass) have a low propensity for further C gains.

An additional consideration that has been raised regarding constraints on aggressive targets for soil C sequestration, is the need for additional inputs of nitrogen (van Groenigen et al., 2017). In most mineral soils, soil organic matter has a relatively narrow C:N stoichiometry, typically ranging from 8 to 20, with a

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[1] In this paper, C refers to carbon and CO_{2eq} refers to CO_2-equivalents. One tone of C is equivalent to 3.67 tones of CO_{2eq}.

[2] About 50% of the mass of soil organic matter is carbon.

C:N of 10–12 as a general "rule-of-thumb" for agricultural soils. Thus, to maintain this balance, if soil organic matter stocks were to increase by say 4 billion tons CO_{2eq}/y (1.1 GtC/y), then about 100 million tons per year of N would need to be incorporated into the added soil organic matter. van Groenigen et al. (2017) point out that this is equivalent to about 75% of the current global synthetic N fertilizer production. While this is a valid point, many of practices being promoted for increasing soil C include using more legumes (e.g., N-fixing cover crops, legume hay/pastures in rotation with annual crops) that could help meet demands for additional N inputs into soil organic matter. Moreover, many cropland soils in North America, Europe, China, India, and SE Asia currently lose a significant amount of added N (from fertilizer, manure, N-fixation) as gaseous losses and leached nitrate, and thus improved practices that could "mop up" some of this N and incorporate it into soil organic matter would yield multiple environmental benefits. Undoubtedly, improved management of N inputs, both to sustain crop productivity and soil organic matter increases and to minimize N_2O emissions (the most potent biogenic GHG on a per mass basis) and other losses of pollution-causing reactive nitrogen to the environment, will be an important part of strategies for negative emissions from soils.

MANAGEMENT PRACTICES TO INCREASE SOIL C STORAGE AND NET CO_2 REMOVALS

In evaluating management interventions to increase soil C stocks, the recent National Academies report divides soil carbon sequestering management practices into two broad categories (NASEM, 2019). The first category includes known, proven conservation management systems that can increase soil C on lands with existing crops and management techniques. These are practices that are typically not (yet) in dominant use, but are being practiced by more conservation-minded farmers and have the potential to become much more widely adopted. Such management techniques can be referred to as BMPs ("Best Management Practices") for increasing soil carbon storage. With proper incentives, such BMPs can be quickly adopted to provide near-term soil C stock increases. The second category of practices are referred to as "frontier technologies" which represent systems or practices for which significant technological and/or economic barriers exist. Thus, they represent technologies and practices that are still largely experimental, with little or no occurrence in production agricultural systems and thus are not yet mature enough to deploy at scale. However, with further R&D and sufficient economic incentives these frontier technologies may offer the potential for greater soil C increases over the longer term.

Conventional Conservation Practices (BMPs) to Sequester Soil C

Conservation practices that can contribute to an increase in carbon stocks in soils are well-known from numerous field experiments and comparative field observations. **Table 1** lists several classes of practices, classified according to their main mode of action in either increasing C inputs to soils and/or reducing C losses from soils.

TABLE 1 | Examples of agricultural management actions that can increase organic carbon storage and promote a net removal of CO_2 from the atmosphere and the main mode of action on the soil C balance (from Paustian, 2014).

Management practice	Increased C inputs	Reduced C losses
Improved crop rotations and increased crop residues	✓	
Cover crops	✓	
Conversion to perennial grasses and legumes	✓	✓
Manure and compost addition	✓	
No-tillage and other conservation tillage		✓
Rewetting organic (i.e., peat and muck) soils		✓
Improved grazing land management	✓	

Improved Crop Rotations and Cover Cropping

Farmers may adopt a number of cropping choices that increase inputs of C into soils: planting of high-residue crops, seasonal cover crops/green manure, continuous cropping (reduced fallow frequency), and planting of permanent or rotated perennial grasses (CAST, 2004). For example, a recent global review of cover crops reported a mean annual sequestration rate of 0.32 tC/ha/y, with several studies reporting rates higher than 1 tC/ha/y (Poeplau and Don, 2015). In many dry climates, farmers fallow croplands every other year to conserve soil moisture and stabilize grain yields. Intensifying and diversifying crop rotations in such systems can increases average annual C inputs, leading to higher soil C stocks than high fallow frequency systems (e.g., West and Post, 2002; Sherrod et al., 2005; O'Dea et al., 2015). In moister environments, adding 2–3 years of perennial hay/forage crops to row crop rotations increases C inputs from fine roots and boosts SOC stocks (e.g., Dick et al., 1998).

Manure and Compost Addition

Organic matter additions such as compost and manures can increase soil C contents, both by virtue of the added C in the amendment itself and through improving soil physical attributes and nutrient availability, such that plant productivity and residue C inputs increase as well (Paustian et al., 1997). One difficulty in assessing the overall impact of organic amendments on net CO_2 removals is that the amendments typically originate from an "off-site" location and thus don't directly reflect on-farm CO_2 uptake from the atmosphere as with other practices described in this section. Hence a full life cycle assessment (LCA) approach, in which the boundaries of the assessment extend outside the farm to include the source of the amendment, is needed for an accurate accounting of C accrual and net GHG reductions. An example is given by work in California on compost addition to rangeland, in which Silver and coworkers (Ryals and Silver, 2013; Ryals et al., 2015) found substantial increases in soil C storage following modest compost additions (a one-time ∼1.3 cm thick surface dressing), in part attributed to improved infiltration and water retention, increased grass productivity and hence greater grass root and residue inputs to soil. Without counting C in the compost addition, they estimated an increase in C storage of 0.5 tC/ha (1.8 tCO_{2eq}/ha) and 3.3 tC/ha (12.1 CO_{2eq}/ha) at two contrasting rangeland sites, respectively, 3 years after compost addition. Further, where the compost was sourced from organic waste in which the business-as-usual case involved land filling and thus potential large emissions of methane, DeLonge et al. (2013) estimated an average net GHG mitigation of 23 tCO_{2eq}/ha, over the 3 year study duration, considering the full LCA including landfill waste emissions vs. compost production, transport, application, and subsequent soil improvement impacts. Considering the large amount of organic waste generated by urban centers and impacts of landfilling on GHG emissions and the potential benefits of organic amendments to soil, use of compost is a potentially attractive option that merits additional R&D to assess the full range of environmental costs and benefits.

Tillage

Tillage is used by farmers to manage crop residues and prepare a seed bed for crops, and is the main source of soil disturbance in annual croplands. Advances in tillage technology and agronomic practice have allowed farmers in recent decades to reduce tillage frequency and intensity, sometimes ceasing tillage altogether with a practice known as "no-till" (NT). The main impetus for many farmers to reduce tillage is to mitigate soil erosion. Studies have reported highly significant reductions in soil erosion under NT, often as high as 90% (Langdale et al., 1979; Ghidey and Alberts, 1998; Williams and Wuest, 2011). Tillage also acts to speed the breakdown of stable soil aggregates that can "protect" organic matter from decomposition (Six et al., 2002). Under NT, aggregation and aggregate stability is significantly enhanced, which is believed to be the main mechanism promoting increased C storage under NT (Six and Paustian, 2014). Many field studies and reviews have shown increases in soil organic carbon (SOC) following adoption of reduced till and NT, with variations due to soil texture and climate (Denef et al., 2011). For example, Ogle et al. (2005) estimated increases under NT of approximately 0.25 tC/ha/y and 0.29 tC/ha/y on sandy and non-sandy soils, respectively. In a global analysis, Six et al. (2004) reported increases in dry climates of 0.1 tC/ha/y and 0.22 tC/ha/y in humid climates. Sainju (2016) recently assessed the net impact of NT to the atmosphere, and found NT systems to have 66% lower Global Warming Potential (GWP) and 71% lower greenhouse gas intensity (GHG emissions per unit of yield) than conventionally tilled systems. However, there are instances in which no-tillage does not increase soil C relative to conventional tillage (Angers and Eriksen-Hamel, 2008), primarily in soils with already high surface C concentrations and often cooler (and wetter) areas where crop productivity and C inputs may be lower under NT, e.g., because of delayed germination (Ogle et al., 2012).

In humid and subhumid croplands, particularly for soils with moderate to poor drainage and with high C concentrations in surface layers relative to subsurface horizon, a one-time deep inversion tillage may be highly effective at promoting a significant increase in soil C stocks, over a multi-decadal period. This practice entails the burial of C-rich surface horizons to a depth of 60–80 cm depth and the transfer of low-C subsoil material to the surface. Burial of C-rich surface soil can significantly slow its decomposition (and promote deeper root penetration) while "conventional" C sequestering practices—e.g., high residue crops, cover crops, and no-till—applied to the newly exposed subsoil material, could rapidly build new C stocks in surface soil layers. For example, Alcantara et al. (2016) sampled 10 sites in Germany that had been subjected to a single deep tillage operation between 1965 and 1978 (done to alleviate compaction of subsurface layers) and found that the deep-tilled sites contained on average 42 t/ha greater SOC stocks (to 1.5 m depth) than similar soils that were not deep-tilled. Crop yields were similar on the fields that received the deep tillage treatment and on untreated fields. The implied average rate of soil C increase following the deep tillage operation was 0.96 tC/ha/y (3.5 tCO_{2eq}/ha/y), over a 45 year period.

Conversion to Perennial Grasses and Legumes

Where croplands are converted to perennial vegetation (grasses, trees), we observe both an increase in C inputs and a reduction in soil disturbance (Denef et al., 2011). Lands retired from cropland cultivation are often referred to as "set-aside." In the U.S., the Conservation Reserve Program (CRP) pays farmers to retire marginal and highly erodible croplands, with peak cumulative enrollments of just over 35 million acres (USDA FSA, 2012). The EPA National Greenhouse Gas Inventory report credits CRP land as a key contributor to agricultural soil carbon sinks in the U.S. (USEPA, 2017). A synthesis by Conant et al. (2016) estimated C stock increases of 39% after conversion of annual cropland to permanent vegetation, with an average rate of almost 0.9 tC/ha/y. Initial rates of SOC accumulation can be high under set-aside, and long-term field studies have noted that accumulations can continue for several decades, approaching levels of native SOC stock (Baer et al., 2010; Munson et al., 2012).

Rewetting Organic Soils

The soils and practices discussed to this point relate to "mineral soils," soils in which the bulk of the soil mass is made up of mineral matter, i.e., sand, silt and clay, and where organic matter normally constitutes only a few percent of the total mass. In contrast, organic soils (referred to as "histosols" in formal soil classification systems), include peat and muck-derived soils for which the total mass consists mainly of organic matter. These soils are formed under waterlogged conditions (hence very low O_2 concentrations) which strongly inhibit decomposition processes, leading to the buildup of deep layers of partially decomposed plant material. In contrast to mineral soils, organic soils are not subject to saturation in the same way—that is, organic matter can continue to accumulate, with the soil "depth" increasing, as long as the conditions inhibiting decomposition remain. When organic soils are exploited for agriculture they are typically drained, limed, and fertilized. They can be very productive for annual cropping, but conversion to agriculture gives rise to extremely high rates of CO_2 emissions, as much as 40–80 tCO_2/ha/y (as well as substantial N_2O emissions; IPCC, 2006) as the soil mass is being oxidized, which can continue as long as organic layers remain exposed to aerobic (i.e., ambient O_2

concentrations) conditions. Consequently, where organic soils can be taken out of production and hydrological conditions restored (referred to as "rewetting"), the very high CO_2 and N_2O emissions can be abated and the soil C accumulation can resume (Wilson et al., 2016). When wetland conditions are restored, CH_4 emissions can increase but, overall, restoring cultivated organic soils provides very large per hectare net emission reductions. However, the area of cultivated organic soils is very small in comparison to that of mineral soils so that that overall mitigation potential is relatively modest (Paustian et al., 2016a).

Improved Grazing Land Management

In the US, non-forested grazing lands are typically differentiated into two main categories: pastures and rangelands. Generally speaking, rangelands refer to grasslands dominated by native species, often occurring in drier environments, and where conventional management interventions are largely restricted to manipulating grazing intensity and duration. In contrast, pastures are often made up of non-indigenous and/or non-native species, are often derived from other land covers and support more intensive and more diverse management options (e.g., fertilization, irrigation, plant species introduction, as well as grazing management).

With the exception of some managed pastures, grazing lands are generally never tilled. Therefore, increasing SOC stocks under perennial grasses relies mainly on enhancing C inputs from plant roots and residues. Ranchers may achieve this by managing plant biomass removal from grazing or increasing forage production through improved species, irrigation and fertilization, yielding increases in SOC stocks of as much as 10% (Conant et al., 2016). Other analyses of grazing land BMPs (including adjusting animal stocking rates and managing plant species) found SOC stock increases of 0.07–0.3 tC/ha/y on rangelands and 0.3–1.4 tC/ha/y on managed pastures (Morgan et al., 2010). Looking at individual practices, Conant et al. (2016) estimated average positive stock changes for improved grazing (0.28 tC/ha/y), sowing legumes (0.66 tC/ha/y) and fertilization (0.57 tC/ha/y).

For improving productivity and soil condition on grazing lands, there is heightened interest in intensive grazing practices employing high animal stocking rates for short durations, from a few hours to a few days, on an area of pasture, with frequent movement of animals and relatively long "rest periods" for the vegetation between grazing events. Various terms including rotation grazing, mob grazing, or adaptive multi-paddock (AMP) grazing are used to label such management systems although terminology is far from standardized. Some studies suggest very dramatic effects from AMP grazing systems in terms of improved productivity and soil physical properties and increased soil carbon stocks. Teague et al. (2011) reported rates of soil C accumulation of about 3 tC/ha/y in AMP systems compared to heavy, continuous grazed systems and Machmuller et al. (2015) reported even higher C accrual rates of up to 8 tC/ha/y on annually cropped soils converted to intensive rotational grazing systems. However, others have questioned whether AMP/rotational grazing systems are superior to well-managed continuous grazing systems (Briske et al., 2008) and there is an ongoing debate within the scientific community. A confounding issue is that adaptive grazing systems, by definition, are dynamic in response to varying weather and other environmental conditions that affect grassland productivity. Thus it is difficult to set up traditional replicated field experiments to compare different grazing systems at the landscape scale (Teague et al., 2013). In any case, additional research and better understanding of grazing impacts on SOC stocks is needed determine optimal management conditions for increasing soil C stocks and minimizing N_2O and CH_4 emissions from livestock in these systems.

"Frontier Technologies" to Sequester Soil C

Several "non-conventional" management practices offer considerable promise for producing negative emissions but require further research to develop the necessary technology and/or better constrain estimates of costs and life-cycle emissions under large-scale deployment. Technologies that we consider here include application of biochar to cropland soils, deployment of perennial grain crops, and adoption of annual crops that have been bred to produce deeper and larger root systems for enhanced C inputs.

Biochar Additions

Biochar is a carbon-rich solid produced from biomass using a thermochemical conversion process known as pyrolysis. A range of temperatures can be used in pyrolysis, with lower temperatures/longer residence times favoring solid biochar formation and higher temperatures/shorter residence times producing a greater proportion of gases and liquid bio-oil and less char (Tripathi et al., 2016). Tradeoffs therefore arise between energy production, which generally favors maximal production of volatiles and bio-oil, and soil applications which favors maximal production of biochar. Biochar also occurs in the soils of many fire-prone ecosystems (where it is typically referred to as pyrogenic carbon), including grasslands, savannas and woodlands, and can make up as much as 35% of the total organic C in these systems (Skjemstad et al., 2002; Glaser and Amelung, 2003; Bird et al., 2015). Hence biochar/pyrogenic carbon is a natural constituent of many soils and soil function is not generally impaired (and may be enhanced) with the addition of large quantities (e.g., 100 t/ha or more) of biochar. Thus, many soils have a potential large storage capacity for added biochar.

Biochar amendments can impact soil C storage and net CO_2 removals from the atmosphere in three different ways. For biochars produced as a coproduct of biofuel pyrolysis processes, when added to soils, most of the biochar mass (80–95%) is highly resistant to microbial decay, with a mean residence time of 100s of years or more (Santos et al., 2012; Wang et al., 2016). Hence, the biochar itself represents a carbon stock that once added to soil tends to persist for a long time. Secondly, biochar additions can also interact with the native organic matter already present in soils, and either stimulate or reduce the rate of decomposition of the native soil organic matter. These interactions could involve a number of factors including impacts on soil water holding capacity and soil moisture, changes in pH or nutrient availability and direct impacts of biochar additions on microbial community activity and composition. Both positive and negative effects on native SOM decomposition following biochar addition have been found (e.g., Song et al., 2016; Wang et al., 2016), but in most cases these effects on the long-term soil C balance are small (Wang et al., 2016). Finally, biochar additions can influence plant productivity and hence C inputs to soil in the form of plant residues. Impacts of biochar addition on plant productivity can vary widely depending on the characteristics of the biochar and soil/plant characteristics. Results from meta-analyses suggest that biochar additions generally have neutral or positive effects on plant growth, with small increases on average (typically $<10\%$) in temperate cropping systems and larger increases (e.g., 10–25%) in tropical systems, particularly on acid, nutrient-poor soils (Liu et al., 2016).

Aside from impacts on soil C storage, a number of studies suggest that biochar amendments may decrease soil N_2O emissions, which would further contribute to greenhouse gas mitigation. A recent meta-analysis by Verhoeven et al. (2017) reported average reductions of N_2O emissions of 9–12% while an earlier global assessment (Cayuela et al., 2014) suggested greater average reductions of almost 50%, compared to non-biochar amended soils. Differences in these meta-analyses are due to different selection criteria for the studies included and the weighting factors used. Regardless, there is an emerging consensus that, on average, biochar applications help to reduce N_2O emissions. The exact mechanisms involved are uncertain since many of the controls on nitrification and denitrification processes (by which N_2O emissions occur), for example pH, mineral N concentrations, soil moisture, and O_2 concentrations, can be impacted by the presence of biochar.

In summary, the main effect of biochar amendment on the GHG balance is associated with the long term storage of the biochar when added to soil. Because the production and transport of the biochar (and bioenergy coproducts) entail a number of different GHG emission sources, the actual mitigation attained (*vis a vis* the atmosphere) depends on the full biochar life cycle and emissions of the biomass feedstock production and harvesting, biochar production process, and field application. This net life cycle C offset value may vary considerably with system design and location, and better knowledge of biochar system LCAs is needed to support broad-scale deployment. One of the few global assessments of biochar amendments as a CO_2 mitigation strategy, by Woolf et al. (2010), suggested a climate change mitigation potential of 1.8 Gt C per year. Due to the complexity of biochar-bioenergy-agricultural systems, the viability of large-scale biochar production and soil application will be spatially variable and process dependent. One cost-benefit analysis found that (without a C price), the net present value of biochar application to soils was positive in a sub-Saharan African context but negative in a Northwestern European context, due to a combination of greater production costs and more modest yield benefits in the latter scenario (Dickinson et al., 2015).

Deployment of Perennial Grain Crops

There have been breeding efforts underway over the past three decades to develop cereal grains (and other annual crops) with a perennial growth habit. The perennial grasses selected for breeding stocks, such as intermediate wheatgrass, are notable in having deep and extensive root systems with a higher proportion of dry matter allocation belowground than conventional annual crops. Hence C inputs to soil are much greater than annual crops and thus will support greater SOC stocks. Perennial crops would also greatly reduce the need for tillage and its negative effects on SOC stocks and soil erosion. Larger and deeper root systems could also reduce nitrate leaching losses to waterways and possibly N_2O emissions to the atmosphere (Glover et al., 2010; Pimentel et al., 2012; Abalos et al., 2016; Crews and Rumsey, 2017).

Because of the relatively recent focus on developing agronomically-viable perennial grains, there are few long-term experiments that are of sufficient duration to document increases in SOC from adoption of perennial grain crops. Culman et al. (2013) found that intermediate wheatgrass increased the amount of labile soil C after 4 years compared to annual winter wheat in SW Michigan, but there was no significant increase in total SOC. However, results from other long-term studies and chronosequences involving perennial grass (e.g., hay, pasture) systems probably provide a reasonable proxy for what would be expected for the longer term response of soils under perennial grains. Some rates of SOC change observed following conversion of annual cropland to a variety of managed perennial grasslands systems are given in **Table 2**.

From **Table 2** it is reasonable to assume that perennial grains could sequester, on average, about 1 tC/ha/y (about 3.6 tCO_2/ha/y) over a number of years, on land converted from continuous annual crop production in the central US grain belt.

At present there are several barriers to adoption of perennial grains on significant areas of land currently allocated to conventional annual crops. Chief among these barriers are low yields and hence questionable economic viability if brought to scale. Yields for intermediate wheatgrass (presently the most commercially viable perennial grain) are typically $<1{,}000$ kg/ha, which is 5–10 times less than annual wheat yields at the same locations (Culman et al., 2016). Between-year variability is also high—in a 4 year study in Southwestern Michigan, Culman et al. (2016) reported average yields ranging from 119 kg/ha/y

TABLE 2 | Observed rates of SOC change under various managed perennial systems.

Cropping system	Mean ΔSOC (t C/ha/y)	Range (t C/ha/y)	Source
Restored prairie	0.77	0.62–0.91	Tilman et al., 2006
Hayed grassland	0.47	None given	Culman et al., 2010
Conversion of annual crops to pasture	0.87¶		Conant et al., 2016
Meta-analysis of perennial bioenergy crops			
Switchgrass	3.10	−5.4 to 13.0	Qin et al., 2016
Miscanthus	1.97	−4.7 to 8.2	
Poplar	0.56	−3.4 to 6.0	

¶*Mean value from a global meta-analysis of 93 studies.*
Results are annualized rates of change from multi-year studies.

(in 2012) to 1,493 kg/ha/y (in 2011), with a mean over the 4 years of 485 kg/ha/y. In a 4-year trial of more than 75 lines of perennial wheatgrass in Australia, several had first-year yields that approached a profitability threshold (without considering any value for potential carbon mitigation benefits), but yields for the following three seasons declined to negligible levels (Larkin et al., 2014). Other issues include problems with grain shattering, lodging, small seed size, and sparse knowledge on optimal agronomics. Such challenges are not unexpected given the few years of active breeding efforts so far, and thus further selection, breeding and field experimentation are likely to improve yields and agronomics[3]. However, there are likely persistent tradeoffs involving resource allocation by perennial plants between dry matter belowground to roots and aboveground to grain (Smaje, 2015; Vico et al., 2016) that will set limits on grain production capacity.

There are also clear tradeoffs in the case of replacing higher yielding annual crops with lower yielding perennials in terms of land use impacts at regional to global scales. If food/feed supply is decreased as a result of adopting lower yielding perennials, there would be pressure to replace that lost production through conversion of new land to agriculture elsewhere, leading to potentially large increases in GHG emissions due to land use conversion. This phenomenon, termed indirect land use change, has been extensively analyzed in the case of substituting energy crops for food crops (e.g., Searchinger et al., 2008; Creutzig et al., 2015) and tradeoffs between crop yields and land use choices are central to arguments underpinning sustainable land use intensification (e.g., Foley et al., 2011; Garnett et al., 2013). However, the potential for mixed grain and forage production and targeting the use of marginal lands that are poorly suited for annual grain production offer opportunities for successful initial commercialization of perennial grain crops (Bell et al., 2008; Culman et al., 2016). In summary, perennial grains show promise for broadening the array of ecosystem services provided by agriculture, including building SOC, but considerable work remains to produce cultivars with reliable regrowth and adequate grain yields, among other important agronomic traits (Cox et al., 2010; Crews et al., 2016).

Annual Crops Bred to Develop Deeper and Larger Root Systems

Another future option, somewhat similar to the deployment of perennial cereals, would be to modify, through targeted breeding and plant selection, existing annual crop plants to produce more roots, deeper in the soil profile. Thus, while the crops would still have an annual life cycle, both C inputs to soil would be increased and deeper root distributions, where decomposition rates are slower compared to surface horizons, would act to increase soil C storage. In a concept paper, Kell (2012) laid out a rationale for the potential to direct plant breeding efforts toward developing varieties for our major grain crops, e.g., corn, sorghum, wheat, and barley, that would have much greater allocation of C to roots and also deeper root distribution compared to current annual crop varieties.

In an analysis to support a new program launched by DOE's ARPA-E, Paustian et al. (2016b) performed a "bounding analysis" to estimate what level of soil C increase and total greenhouse gas mitigation (including N_2O emissions) might be possible based on specifying feasible increases in total root mass and changing root depth distributions toward those found in perennial grasses. They estimated that widespread adoption of annual crop phenotypes designed to have deeper and larger root systems could yield soil C stock increases of 0.5 Gt CO_2/ha/y on current US cropland.

ESTIMATES OF THE BIOPHYSICAL POTENTIAL FOR CO_2 REMOVAL AND SEQUESTRATION IN SOILS

As described in the preceding section, there are a wide variety of management practices that can be adopted on agricultural lands to remove CO_2 from the atmosphere and convert it into soil organic matter. The question then is "how much?"—how much carbon can actually be added to and maintained in soils and is it large enough to matter?

Over the past 20 years there have been several estimates of the soil C sequestration potential globally and for the US. In nearly all cases these represent the *biophysical potential* for managed cropland and/or grassland systems to store additional carbon assuming widespread (near complete) adoption of the sequestering practices. As such, these represent upper-bound estimates of the C sequestration potential. Economic or policy-related constraints are generally not considered as they require a detailed coupled ecosystem and economic modeling approach. In terms of methods, most estimates, particularly at global scale, are based on highly aggregated data on total area by land-use type, stratified into broadly defined climate types, and then applying estimates of representative per ha soil C sequestration rates for different management practices or suites of practices, based on measurements from long-term field experiments.

Despite somewhat different scope (land types included) and assumptions (practices considered), there is fairly close alignment among global estimates (**Figure 1**), suggesting a technical soil C sequestration potential of 2–5 Gt CO_2 per year, for what were characterized in the section above as existing best conservation management practices. Estimates toward the lower end of this range consider either less land area (e.g., cropland only) and/or a more restricted set of practices. It is not surprising that these various estimates are in reasonably close alignment since the two main determining factors, land area by land use type and observed rates of soil C sequestration from long-term field trials, are fairly tightly constrained. Thus, there seems to be good support for an estimate of as much as 4–5 Gt CO_2 per year for widespread adoption of a broad suite of BMPs for soil C sequestration on global grassland and cropland. These rates of C storage could be sustained for a limited time period, on the order

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[3] Glover et al. (2010) estimated that commercially viable perennial grains could be available by 2030.

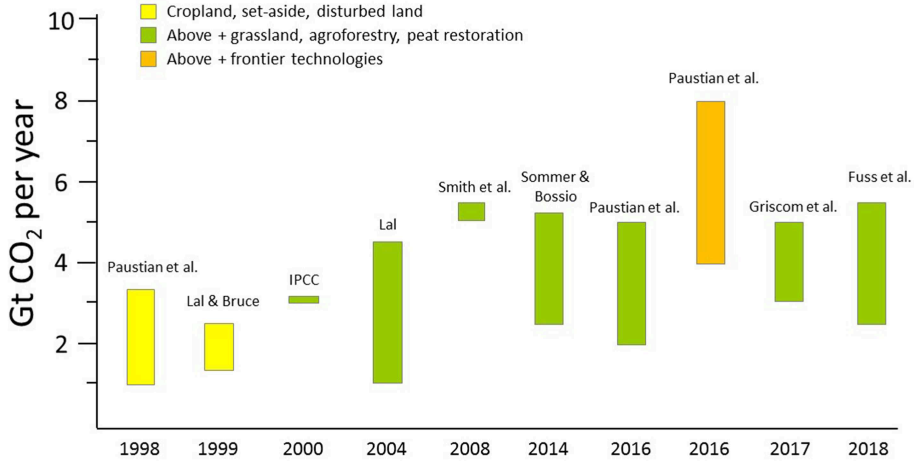

FIGURE 1 | Published estimates of global biophysical soil carbon sequestration potential, assuming (near) full adoption of C sequestering practices. Estimates vary in terms of land use types and mix of practices included; e.g., the pre-2000 estimates (Paustian et al., 1998; Lal and Bruce, 1999) focused on cropland and set-aside to grassland of marginal crop land, while later estimates (Lal, 2004; Smith et al., 2008; Sommer and Bossio, 2014; Paustian et al., 2016a; Griscom et al., 2017; Fuss et al., 2018) included a fuller range of options on all agricultural lands. The IPCC (2000) estimate provided a point estimate whereas other publications provide a range of per annum rates. The two estimates shown for Paustian et al. (2016a) denote with and without frontier technologies.

of 2–3 decades before decreasing, as soil C levels approach a new equilibrium.

The estimate by Paustian et al. (2016a) that goes as high as 8 Gt CO_2 per year, includes ∼3 Gt CO_2/y from what we've referred to as "frontier technologies," in this case biochar amendments and high root C input crop phenotypes, in addition to the conventional conservation technologies included in other global estimates. However, estimates of technical potentials for these frontier technologies are much more uncertain, either because empirical data on their performance in the field (e.g., in long-term field studies) is much scarcer, or in the case of novel crop types (e.g., perennial grains, enhanced root phenotype annual crops), the technologies themselves are still in an early developmental stage.

In conjunction with the negotiations for the Paris climate accords, the French government announced an initiative dubbed "4 per mille" which advocates for a massive effort to increase global soil C stocks as a core greenhouse gas mitigation strategy. As articulated by INRA, the French National Institute for Agricultural Research (INRA, 2017), if global soil C stocks in the top 40 cm (860 GtC) could be increased on average by 0.4% (i.e., 4 per mille) per year that is equivalent to about 3.4 GtC/y or 12.6 $GtCO_2$/y. That level of net CO_2 uptake would offset most of the current annual increase in atmospheric CO_2 (15.8 $GtCO_2$/y), assuming that the current ocean and terrestrial C sinks remained intact. There is considerable debate about whether this level of soil C sequestration is indeed possible, and whether all soils or mainly agricultural soils should be targeted (e.g., Chambers et al., 2016; Minasny et al., 2017). In any case, as an aspirational goal, the 4 per mille concept has certainly spurred debate and "raised the profile" of soils as a potentially key mitigation strategy.

As points of comparison, current global GHG emissions are about 40 $GtCO_{2e}$/year (with about 83% of that from fossil fuel combustion), and meeting the goals of the Paris agreement may require negative emissions of about 15 Gt CO_2/year by the end of the century (NASEM, 2019).

CONCLUDING REMARKS

There is strong scientific evidence for agricultural soils to act as a significant carbon (C) sink over the next several decades and thereby to contribute to meeting the objectives of the Paris Climate Accord. There are a wide variety of C sequestering practices that can be applied and the best solutions vary according to climate, soil, and farming practices. Many practices (e.g., improvements in crop rotations, use of cover crops, tillage changes, N fertilizer management) are already developed and their efficacy is relatively well-understood. Wide-scale adoption of such measures could take place quite rapidly. Other potential practices, requiring development of new crop varieties and broad-scale use of soil amendments such as biochar, require additional research and development to overcome technological hurdles and/or improve economic feasibility.

This suggests a "two-stage" strategy. Strong policy could be enacted immediately to begin an international effort to increase soil carbon sequestration, based on existing technologies. Key ingredients are efficient policies that incentivize farmers to adopt improved (C sequestering) practices, by compensating them for additional costs and/or added risk. Expanded education and outreach can also help to overcome knowledge or "know-how" barriers. Meanwhile, continued R&D, with increased investments could be devoted to further developing new crop varieties, both perennial grains (and "perennialization" of other crops such as oil seeds) and breeding for annual crops with larger and deeper root systems. This could lead to viability of these new crops for use by about 2030 and beyond, when the need for negative emission strategies will be growing.

Implementation of these policies will require a robust, scientifically-sound measurement, reporting, and verification (MRV) system to track that policy goals are being met and that claimed increases in soil C stocks are real. Much of the infrastructure for an effective MRV system for soil C sequestration could be assembled relatively quickly and with modest research and development investments (NASEM, 2019; Paustian et al., 2019). Existing ground-based data from long-term field experiments (e.g., Harden et al., 2018) together with national networks for on-farm soil monitoring (van Wesemael et al., 2011) can support the continued improvement and deployment of process-based predictive models. Expanded use of remote sensing can help to monitor management practices (e.g., Hively et al., 2018) and constrain local-scale estimates of CO_2 assimilation and C input to soils by crops (e.g., Guan et al., 2017). This extensive and broad-based melding of ground-based experiments and monitoring, dynamic predictive models, remote sensing and farmer-based knowledge of management practices can form the basis for quantification tools that can inform policy and program implementation, at field- (Paustian et al., 2018) to national-scales (Ogle et al., 2014).

In summary—by leveraging existing scientific knowledge and infrastructure, together with modest investment to further advance the knowledge base and develop new technologies, many countries could move to implement negative emission strategies in the agricultural sector and at the same time improve the health and resilience of their soils. This would stimulate and encourage global-scale initiatives (e.g., Schleussner et al., 2016; INRA, 2017), to help achieve the goal of limiting average global temperature increases to $<2^\circ$C.

AUTHOR CONTRIBUTIONS

KP and EL formulated the idea for the paper. KP led the development and writing of the paper. JK, EM, and AS helped compile and analyze data and contributed to the writing of the paper.

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REVIEW
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An Overview of the Status and Challenges of CO_2 Storage in Minerals and Geological Formations

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Since the Industrial Revolution, anthropogenic carbon dioxide (CO_2) emissions have grown exponentially, accumulating in the atmosphere and leading to global warming. According to the IPCC (IPCC Special Report, 2018), atmospheric warming should be <2°C to avoid the most serious consequences associated with climate change. This goal may be achieved in part by reducing CO_2 emissions, together with capturing and sequestering CO_2 from point sources. The most mature storage technique is sequestration in deep saline aquifers. In addition, CO_2 can be mineralized and sequestered in solid form by various techniques, i.e., *ex-situ, surficial* and *in situ* mineralization. *Ex situ* and *surficial* approaches may produce valuable products while mitigating environmental hazards. *In-situ* mineralization uses ultramafic and mafic geological formations for permanent, solid storage. In addition, the IPCC portfolio that limits warming to <2°C by 2100 includes avoiding CO_2 emissions and removal of CO_2 from air. Regardless of the specific mix of approaches, it will be essential to permanently sequester about 10 billion tons of CO_2 per year by mid-century, and roughly twice that amount each year by 2100. Maximizing the potential of technologies for CO_2 removal from air and CO_2 storage will help to meet global climate goals. The research agenda published by National Academies of Sciences Engineering Medicine (2019) calls for roughly \$1 billion over a 10–20 years time period to advance the deployment of CO_2 sequestration in deep sedimentary reservoirs at the $GtCO_2$/yr scale and develop CO_2 mineralization at the $MtCO_2$/yr scale. This would lead to a deeper understanding of the reservoir characteristics from the nano- to kilometer scale, some of which may include the distribution of the reaction products, the reaction rate of the minerals, the permeability evolution, the pressure build-up in the reservoir, the large-scale impact of chemicophysical processes leading to clogging or cracking, the effects of potential geochemical contamination, etc. This overview presents the advantages, drawbacks, costs, and CO_2 storage potential of each technique, the current and future projects in this domain, and potential sequestration options in geologic formations around the world.

Keywords: geologic storage, mineral carbonation, mineralization, carbon sequestration, negative emissions, carbon capture and storage

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INTRODUCTION

The cumulative release of anthropogenic carbon dioxide (CO_2) into the atmosphere has been estimated at 2,035 ± 205 $GtCO_2$ from 1870 to 2015 (Le Quéré et al., 2015). Today, emissions are about 40 $GtCO_2$/yr (IPCC Special Report, 2018). Each year, roughly half of these emissions are removed naturally by uptake in the ocean and the terrestrial biosphere, while the remainder accumulates in the atmosphere and contributes to global warming. To avoid the worst impacts of global warming and subsequent climate change, the Paris agreement recommended limiting average warming of the atmosphere to $<2^\circ$C, and preferably $<1.5^\circ$C (UNFCCC, 2015).

Recently the IPCC published a report stating that, to reach the 1.5°C goal, negative emission technologies have to remove tens of Gt CO_2 per year (IPCC Special Report, 2018), in addition to reducing emissions and capturing CO_2 from point sources. Options for CO_2 removal from air (CDR) include an increase of carbon storage in soils and biomass, but also Direct Air Capture using synthetic sorbents (DACSS) and carbon mineralization via enhanced weathering (DACEW). The likely scenario is that most of these options, operating in parallel, will be necessary to achieve the required level of global CDR, approximately 10 Gt/yr by 2050, and 20 Gt/yr by 2100 (United Nations Environment Programme (UNEP), 2017; National Academies of Sciences Engineering Medicine, 2019). While utilization of some captured CO_2 may be attractive, to produce fuels with net-zero emissions for example, most of the captured CO_2, from point sources and from air, must be permanently stored. Starting now, carbon capture and storage (CCS) methods must sequester about 125 Gt of CO_2 by 2100 (National Academies of Sciences Engineering Medicine, 2019). Two possible methods for long-term CO_2 storage are underground sequestration in sedimentary formations and carbon mineralization (National Academies of Sciences Engineering Medicine, 2019).

Underground CO_2 sequestration in sedimentary formations is the most mature technique for storage of CO_2 captured from point sources, and perhaps by DAC, as commercial-size CO_2 injection projects are already operating today. This approach has been carried out over two decades globally, currently storing roughly 3.7–4.2 $MtCO_2$/year, for a cumulative total of 30.4 $MtCO_2$ at the end of 2017 (Global CCS Institute, 2019; National Academies of Sciences Engineering Medicine, 2019). Additionally, roughly 28 $MtCO_2$/year of captured CO_2 are injected into the ground for CO_2-enhanced oil recovery (CO_2-EOR) (Global CCS Institute, 2019). Approximately 85% of CO_2-EOR projects use CO_2 sourced from natural CO_2 gas fields (DiPietro et al., 2012). Less common use of CO_2 captured from smokestacks or removed from air leads to a lower carbon footprint of produced oil. More specifically, in the U.S., about 64 $MtCO_2$/yr are used for CO_2-EOR, from which 21 $MtCO_2$/yr is from anthropogenic sources (U.S. Environmental Protection Agency (EPA), 2016). Conventional CO_2-EOR today is optimized to produce as much oil as possible with the least amount of CO_2 injection. In practice, about 0.3–0.6 tons of CO_2 is injected for every barrel of oil produced—which compares to the 0.4 tons of CO_2 emitted when the oil is burned (McGlade, 2019). During oil production, some of the CO_2 is produced along with the oil. In many such cases, it is separated from the oil and reinjected underground. Larger amounts of CO_2 could be sequestered in depleted oil reservoirs if advanced EOR+ or maximum storage EOR+, both which optimize the amount of CO_2 sequestered, were used in place of conventional CO_2-EOR. The combined underground storage capacity in saline aquifers and hydrocarbon reservoirs is estimated to range between 5,000 and 25,000 Gt CO_2 (de Coninck and Benson, 2014), which is larger than necessary to limit warming to $<1.5^\circ$C through 2100 (Rogelj et al., 2018).

Another storage option is carbon mineralization of mafic and ultramafic rocks. This has been implemented in the Carbfix Project in Iceland. There, more than 10,000 tons of CO_2 captured from geothermal steam is injected into subsurface pore space in basaltic lavas, where it is mineralized within a year (Gislason et al., 2010; Aradóttir et al., 2011; Matter et al., 2011, 2016; Snæbjörnsdóttir et al., 2017; Gunnarsson et al., 2018). This approach is at a lower stage of technological readiness and is under continued investigation on the lab- and pilot-scales. Globally, carbon mineralization in these rock types has the potential of sequestering up to 60,000,000 $GtCO_2$ if the resource is economically-accessible and ultimately fully carbonated.

CARBON MINERALIZATION

Processes and Methods of Carbon Mineralization

Carbon mineralization was proposed 30 years ago as a strategy for CO_2 removal from the atmosphere (Seifritz, 1990; Lackner et al., 1995). It is a long-term and non-toxic method of storing CO_2 in solid form, that can also help in mitigating health and environmental hazards in specific contexts (National Academies of Sciences Engineering Medicine, 2019). This strategy of underground sequestration increases the uptake of CO_2 in the reservoir through the interaction with rocks bearing magnesium or calcium ions (Gunter et al., 1993). Additionally, since it is converted to a stable carbonate form, this represents the safest storage mechanism with regard to minimizing leakage (Zhang and DePaolo, 2017).

Carbon mineralization involves the formation of solid carbonate minerals (calcite, magnesite, dolomite, and a variety of hydrated Mg-carbonate minerals such as nesquehonite) through reaction of CO_2 (gas, liquid, dissolved in water, or supercritical) with rocks rich in calcium or magnesium. The best sources of Mg and Ca are mafic and ultramafic rocks (mantle peridotite, basaltic lava, and ultramafic plutons), mine tailings (mafic and ultramafic), and industrial byproducts (cement kiln dust, steel slag, and fly ash). The source of CO_2 varies depending on the context and local availability, including ambient air ($\sim$0.04% CO_2), flue gas from the power sector ($\sim$5–12% CO_2), seawater or Mg-rich peridotite groundwater saturated in air ($\sim$0.01% CO_2), fluids enriched in or saturated with CO_2, or high purity CO_2 gas and fluid (+99.5%). Direct Air Capture via Enhanced Weathering (DACEW) uses CO_2 directly from air or surface water for the purpose of carbon mineralization, and can be combined with long-term solid storage (Kelemen et al., under review).

Various methods exist for carbon mineralization: (1) *ex-situ*, where the alkalinity source is transported to a site of CO_2 capture, ground to a small particles, and combined with CO_2 in a high temperature and pressure reaction vessel, (2) *surficial*, where dilute or concentrated CO_2 is reacted with the alkalinity source on-site at the surface (e.g., mine tailings, smelter slag), and (3) *in-situ*, where CO_2-bearing fluids circulate through subsurface porosity in geological formations. The rate and cost of carbon mineralization depends on available CO_2 dissolved in solution, available alkalinity in the solution, and optimal reaction conditions (Kelemen et al., under review). Variations in pH are particularly important, as low pH favors mineral dissolution whereas high pH accelerates carbonate precipitation (Park and Fan, 2004; Pokrovsky and Schott, 2004).

Research results are difficult to compare because of the wide range of reagents and conditions used. Few comparisons of carbon mineralization kinetics exist as a function of temperature, CO_2 partial pressure, and other variables (Matter and Kelemen, 2009; Kelemen et al., 2011; Gadikota et al., in preparation). Several processes have been considered. For instance, in two-step processes, the first step involves the dissolution of the alkalinity source at low pH, with precipitation of carbonates during a second step at high pH. In most cases, the dissolution of the alkalinity source is the rate-limiting step. Alternatively, dissolution of silicates and carbon mineralization may be combined into a single-step process. Experiments with olivine as the alkalinity source used $NaHCO_3$ as a catalyst that buffers the pH (O'Connor et al., 2005; Chizmeshya et al., 2007; Gadikota et al., 2014). In the natural reactive flow of water through ultramafic rocks, a pH-swing occurs spontaneously making both dissolution of the alkalinity source and precipitation of carbonate possible (Bruni et al., 2002; Paukert et al., 2012; Canovas et al., 2017; Paukert Vankeuren et al., 2019; Leong and Shock, under review). Carbon mineralization rates of serpentine may be increased by heat treatment, which removes hydroxyl groups from the mineral and increases the rate of dissolution and carbon mineralization (e.g., O'Connor et al., 2005; Li et al., 2009; Larachi et al., 2012).

Kinetics

The mineral wollastonite ($CaSiO_3$) has the highest reaction rate with CO_2, in the range of $8.0{\cdot}10^{-9}$-$2.0{\cdot}10^{-7}$ $mol{\cdot}m^{-2} \cdot s^{-1}$ at 25°C and in the range of $1.6{\cdot}10^{-5}$-$5.0{\cdot}10^{-4.}$ $mol{\cdot}m^{-2} \cdot s^{-1}$ at 180°C (**Figure 1**) (Palandri and Kharaka, 2004; O'Connor et al., 2005). However, the availability of wollastonite is low, with global reserves estimated at only 100M tons. Its high Ca content makes it a good analog for industrial waste byproducts (i.e., cement kiln dust, steel slag, and fly ash), that may have initial reaction rates similar to wollastonite, but they generally contain less Ca, and/or contain Ca-Al silicates that are slow to react [e.g., (Huijgen et al., 2007; Pan et al., 2012; Sanna et al., 2014; Gadikota et al., 2015; National Academies of Sciences Engineering Medicine, 2019)].

Olivine (Mg_2SiO_4) reacts with CO_2-bearing aqueous fluids almost as fast as wollastonite (**Figure 1**) (Palandri and Kharaka, 2004; National Academies of Sciences Engineering Medicine, 2019). Additionally, as the major mineral of peridotite, which constitutes the Earth's upper mantle, olivine is highly abundant, making it a good candidate for carbon mineralization. Addition of $NaHCO_3$ in single-step carbon mineralization experiments buffers the pH and helps for both dissolution and carbon mineralization reactions (O'Connor et al., 2005; Chizmeshya et al., 2007; Gadikota et al., 2015).

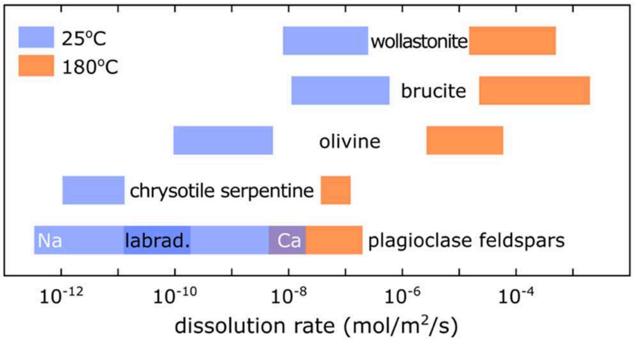

FIGURE 1 | Dissolution rates of magnesium- and calcium-rich minerals at 25°C (blue) and 180°C (orange). The plagioclase feldspars are a solid solution between a sodium-end-member (albite $NaAlSi_3O_8$) and a calcium-end-member (anorthite $CaAl_2Si_2O_8$) indicated as Na and Ca. The most common plagioclase feldspar in basalts is labradorite (labrad.). At high temperature, there is less variation of dissolution rates as a function of composition for plagioclase feldspars [Figure modified from National Academies of Sciences Engineering Medicine (2019), after data from Palandri and Kharaka (2004) and Thom et al. (2013)].

Plagioclase feldspars, one of the major constituents of basalt and gabbro (the major constituents of the oceanic crust), have lower reaction rates than wollastonite and olivine. At low temperature (25°C) the calcium-rich end-member (anorthite $CaAl_2Si_2O_6$) dissolves faster than the sodium-rich end-member (albite $NaAlSi_3O_6$). In basalts, the plagioclase feldspar is usually labradorite (50–70% anorthite), which has reaction rates in the range of $1.3{\cdot}10^{-11}$-$2.0{\cdot}10^{-10}$ $mol{\cdot}m^{-2} \cdot s^{-1}$ at 25°C, and $6.3{\cdot}10^{-9}$-$6.3{\cdot}10^{-8}$ $mol{\cdot}m^{-2} \cdot s^{-1}$ at 180°C (**Figure 1**) (Palandri and Kharaka, 2004; Gadikota et al., in preparation; National Academies of Sciences Engineering Medicine, 2019). To the extent that steel slag and smelting waste contain Ca-Al silicate compounds similar to anorthite, formation of calcite ($CaCO_3$) may be retarded by slow diffusion of Al.

The dissolution rate of serpentine ($Mg_3Si_2O_5(OH)_4$) is similar to the lower bound for plagioclase feldspars (**Figure 1**) (Thom et al., 2013; National Academies of Sciences Engineering Medicine, 2019). Serpentinite is formed by the alteration of peridotite, which is often partial. This results in a mineral association of serpentine, olivine, and brucite ($Mg(OH)_2$), with olivine and brucite being among the most reactive minerals for carbon mineralization. Also, fibrous "asbestiform" chrysotile serpentine, composed of nano-scale cylinders, allows for rapid carbon mineralization due to its high surface-to-volume ratio. Consuming asbestiform serpentine also mitigates health hazards due to asbestos contained in mine tailings (National Academies of Sciences Engineering Medicine, 2019).

The low crystallinity of amorphous basaltic glass and some industrial byproducts may make them more reactive than crystalline minerals and rocks with the equivalent composition (Oelkers and Gislason, 2001; Gislason and Oelkers, 2003).

Reaction between the solid source of alkalinity and dissolved or supercritical CO_2 can coat grains of solid reactants, thereby reducing the reactive surface area and limiting the extent of reaction (known as "passivation"). However, in most studies, there is no negative effect of passivation observed for olivine, for which the rate carbon mineralization is constant, in part due to reaction-driven cracking and formation of etch pits as the reaction proceeds (Gadikota et al., in preparation). This is not the case for many other reactants, for which the reaction rate decreases over time (perhaps due to passivation). This could be an issue for extensive carbonation of mafic intrusions and basalts rich in plagioclase feldspars and pyroxenes. Understanding the key parameters that favor reaction-driven cracking rather than surface passivation is essential for forecasting the extent of carbon mineralization reactions (National Academies of Sciences Engineering Medicine, 2019).

Ex-situ Carbon Mineralization

The most studied mineral for *ex-situ* carbon mineralization is olivine, as it is abundant, has a high reaction rate, and exhibits little to no passivation. The optimum temperature for carbon mineralization of olivine is ∼185°C (O'Connor et al., 2005; Gadikota et al., 2014). Serpentine-rich mine tailings are an intriguing option as the carbonation of asbestiform chrysotile would mitigate health and environmental hazards, and products of rapid heat-treatment of serpentine-rich mine tailings could be used as a feedstock for carbon mineralization using concentrated sources of CO_2 (McKelvy et al., 2004; Maroto-Valer et al., 2005; O'Connor et al., 2005; Li et al., 2009; Larachi et al., 2010, 2012; Balucan et al., 2011; Fedoročková et al., 2012; Balucan and Dlugogorski, 2013; Werner et al., 2013, 2014; Dlugogorski and Balucan, 2014; Ghoorah et al., 2014; Hariharan et al., 2014, 2016; Pasquier et al., 2014; Sanna et al., 2014; Hariharan and Mazzotti, 2017). However, these *ex-situ* methods are more expensive than the projected cost of direct air capture of CO_2, and significantly more expensive than CO_2 storage in subsurface pore space (see section Costs and Reservoir Capacities).

Another interesting source of alkalinity is waste byproducts (i.e., cement kiln dust, steel slag, smelter slag, and fly ash) (Huijgen et al., 2007; Pan et al., 2012; Sanna et al., 2014; Gadikota et al., 2015) because these are directly available for carbon mineralization. Using these materials for carbon mineralization also mitigates hazards due to chemical contamination associated with alkaline industrial wastes. However, it should be kept in mind that these contain variable, sometimes low concentrations of Ca and Mg, and that Ca may be "locked up" in slow-reacting Ca–Al silicate species. *Ex-situ* carbon mineralization using industrial waste, wollastonite, and olivine (excluding mine tailings) has the potential to carbonate many $GtCO_2$/yr (Kelemen et al., 2011; Renforth et al., 2011; Sanna et al., 2014; Gadikota and Park, 2015; National Academies of Sciences Engineering Medicine, 2019). Nevertheless, costs remain high, and producing this volume of carbonated material raises additional questions regarding handling, transportation, and disposal.

The cost of *ex-situ* carbon mineralization is about 10 times higher than CO_2 injection and sequestration into subsurface reservoirs (National Academies of Sciences Engineering Medicine, 2019). *Ex-situ* carbon mineralization could instead be used to create valuable products (Huijgen et al., 2007; Pan et al., 2012; Sanna et al., 2014; Gadikota et al., 2015), particularly for construction applications. If it were economically viable to use CO_2-added aggregates in concrete, instead of currently-used materials, this could reduce the net emissions of the concrete industry. Replacing 10% of building material by material carbonated with anthropogenic CO_2 could reduce CO_2 emissions by up to 1.6 Gt/yr (Sridhar and Hill, 2011). Details about the mitigation capacity and the cost of various methods are provided in section Costs and Reservoir Capacities of this study.

Surficial Carbon Mineralization

Surficial carbon mineralization uses mafic and ultramafic mine tailings. At surface conditions most minerals in mafic and ultramafic rocks have relatively slow reaction rates, with the exception of brucite and asbestiform chrysotile (Power et al., 2011, 2013a; Harrison et al., 2013; Wilson et al., 2014). Mine tailings have a high surface area-to-volume ratio compared to subsurface geologic formations, resulting in increased reactivity. They are also low-cost sources provided that they are wastes of mafic and ultramafic rocks quarried or mined for other resources such as nickel, chromium, platinum-group elements, and diamonds. Mine tailings represent a small mitigation opportunity that would consume <36 $MtCO_2$/yr and require potential transportation and large disposal areas (National Academies of Sciences Engineering Medicine, 2019). Still, mine tailings have a high uptake potential of carbon and mines that carbonate their tailings could offset on-site CO_2-eq emissions (Harrison et al., 2013; Mervine et al., 2018).

Figure 2 summarizes data on the rate of carbon mineralization of mafic and ultramafic mine tailings in contact with air, and in contact with more CO_2 rich gases and fluids. Carbon mineralization in mine tailings can be enhanced by several methods, such as sparging air, air-saturated water, or CO_2-rich gas through the tailings (Assima et al., 2013, 2014; Harrison et al., 2013), stirring the tailings to have a turnover of reactive surfaces in contact with air (Kelemen et al., under review), or adding microorganisms that accelerate the formation of carbonates in disposal ponds (Power et al., 2011, 2013b). These methods can significantly increase the CO_2 uptake rate of the tailings (Assima et al., 2013; Harrison et al., 2013). For instance, sparging CO_2-rich gas through ultramafic tailings could accelerate the CO_2 uptake rate up to a million times (Harrison et al., 2013; National Academies of Sciences Engineering Medicine, 2019). Though surficial mineral carbon mineralization methods could prove to be more expensive than the combination of Direct Air Capture using Synthetic Sorbents (DACSS) combined with geological sequestration, this method could provide local solutions where mine tailings are close to a local source of concentrated CO_2. And, as noted above, mine tailings containing asbestos present health hazards that would

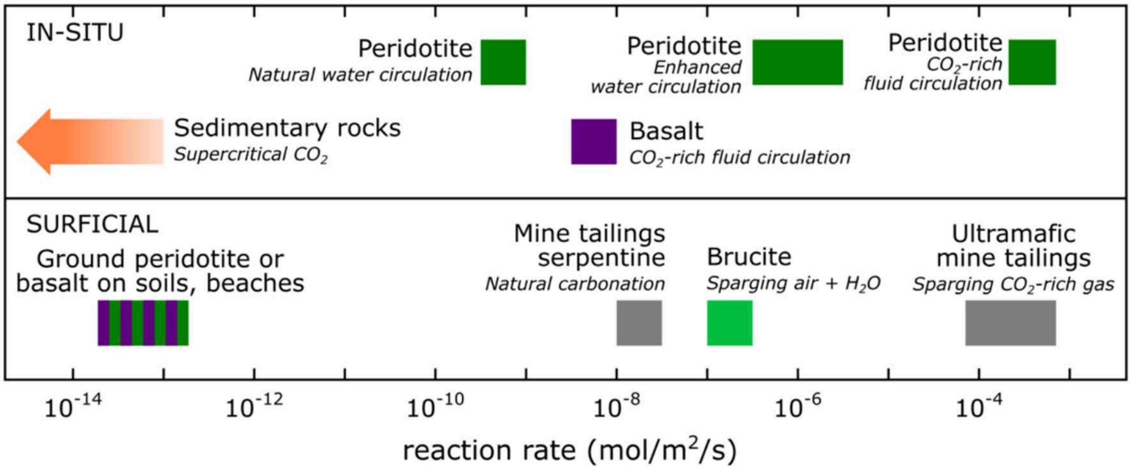

FIGURE 2 | Reaction rate in mol·m^{-2} · s^{-1} of *in-situ* and surficial mineralization of CO_2 using mine tailings or rocks as the source of alkalinity. Mine tailings are in gray, peridotite in dark green, basalt in purple, brucite in light green, and sedimentary rocks in orange. From National Academies of Sciences Engineering Medicine (2019) and references cited therein.

be mitigated by carbon mineralization (National Academies of Sciences Engineering Medicine, 2019).

Another option for surficial carbon mineralization is mining of mafic and ultramafic rocks for the purpose of CO_2 capture and storage, which might cost \$55–500/tCO_2 depending on the reactivity of specific tailings, the duration of their exposure to air, and the cost of "stirring" the tailings to prevent passivation (Kelemen et al., under review). This range of costs overlaps with the range of cost estimates for DACSS, combined with storage in geologic formations (National Academies of Sciences Engineering Medicine, 2019).

Recently, looping, or re-use, of Mg-rich mine tailings has been proposed (Kelemen et al., under review; McQueen et al., under review). The simplest version of this process would begin with magnesite ($MgCO_3$). Heating of magnesite ("calcining") produces a nearly pure stream of CO_2 gas plus a solid residue of caustic magnesia (MgO). Subsequent weathering of caustic magnesia will rapidly remove CO_2 from air, with nearly complete conversion to Mg-carbonates expected in about a year. Mg-carbonates can then be calcined again, and so on. Initial feedstock could also be ultramafic tailings, with the first step of calcining forming "heat-treated serpentine" and producing $H_2O + CO_2$ gas. After a few cycles of weathering, the solid residue of calcining is likely to be composed of caustic magnesia plus more or less crystalline SiO_2 (opal, chalcedony, and quartz). The caustic magnesia can be used for many more cycles of weathering to remove CO_2 from air. We believe that this process has costs and area requirements similar to, or less than, DACSS.

Grinding ultramafic and mafic rocks to sizes smaller than mine tailing sizes and spreading them over agricultural soil, forest soil, or along beaches is another possibility for CO_2 removal (**Figure 2**) (e.g., Schuiling and Krijgsman, 2006; Renforth et al., 2015; Edwards et al., 2017; Kantola et al., 2017; Beerling et al., 2018). This process may be enhanced by microbial processes occurring in soils (Power et al., 2009, 2013b) and may be competitive with direct air capture using chemicals combined with geologic sequestration of CO_2. However, mafic and ultramafic rocks contain nickel and chromium that, once oxidized, might present a significant health hazard if they accumulate in soils.

IN-SITU MINERAL SEQUESTRATION

Storage in Subsurface Pore Space

When CO_2-bearing fluids or supercritical CO_2 are injected at depth into geologic formations, the sequestration of CO_2 in the subsurface porosity relies on the impermeability of the caprock of the reservoir. Suitable reservoirs are saline aquifers, depleted oil, and gas reservoirs (may or may not be associated with enhanced oil recovery, i.e., CO_2-EOR), possibly hydraulically fractured shale formations, and thick formations of basalt and peridotite (Tao and Clarens, 2013; National Academies of Sciences Engineering Medicine, 2019). Ideal properties of saline formations for CO_2 storage include high permeability to enable high rates of CO_2 injection without large pressure buildup, thick formations with many interbedded low permeability barriers to use the pore space efficiently (Wen and Benson, 2019), and having salinity in excess of 10,000 ppm of total dissolved solids to comply with groundwater protection regulations. It is important to limit pore pressure buildup in order to avoid fracturing the caprock and inducing seismic events (National Academies of Sciences Engineering Medicine, 2019).

Secondary trapping mechanisms aim to improve CO_2 storage and include solubility trapping by dissolution of the CO_2 into aqueous pore fluid (Emami-Meybodi et al., 2015), residual gas trapping by capillary forces (Krevor et al., 2015), and mineralization by chemical interactions between the CO_2, pore fluid, and rock (**Figures 2**, **3**) (Zhang and DePaolo, 2017). The extent of secondary trapping mechanisms is highly site-specific, and depends on the geology, structure, and hydrology

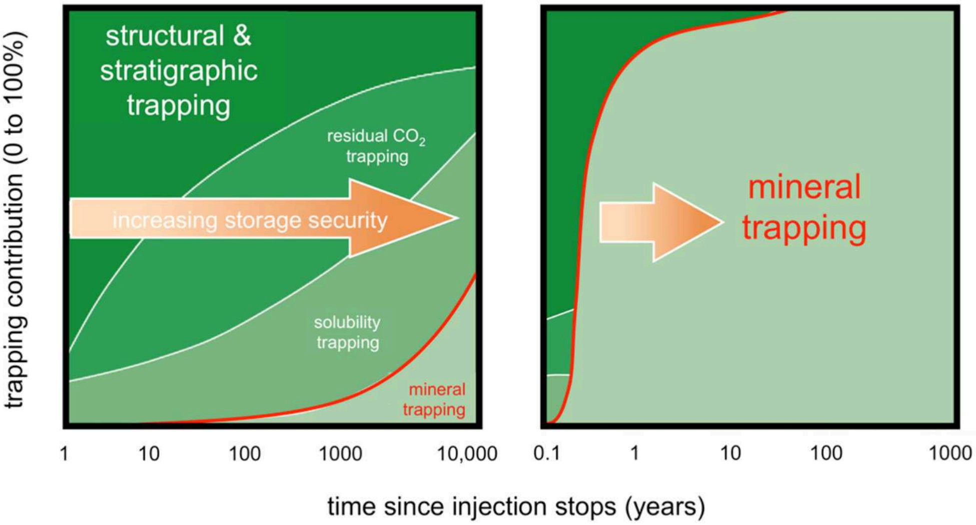

FIGURE 3 | Evolution of the extent of CO_2 trapping mechanisms with time. The extent of each trapping mechanism is highly site specific and depends on several parameters including the type of rock: carbonatitic and siliciclastic rocks (left panel), or mafic and ultramafic rocks that have the ability to react much faster with CO_2 to form carbonates (right panel) [from (National Academies of Sciences Engineering Medicine, 2019), Figure 6.7 and (Kelemen et al., under review), Figure 8, modified from (Benson et al., 2005); also see Figure 9 in (Snæbjörnsdóttir et al., 2017)].

of each reservoir. For instance, increasing pore fluid salinity decreases CO_2 solubility (Gunter et al., 1993). The purity of the CO_2 also affects the storage capacity of the reservoir (Talman, 2015). Secondary trapping mechanisms in and of themselves may be sufficient for the mitigation of the risks of CO_2 leakage (National Academies of Sciences Engineering Medicine, 2019), for example in basalt and peridotite formations where carbon mineralization rates are highest and 90% of the CO_2 may be mineralized in a few months to decades (**Figure 3**, right panel) (National Academies of Sciences Engineering Medicine, 2019). The rate and capacity of mineral trapping are low in many sedimentary reservoirs due to the dominant quartz composition which has very limited reactivity with CO_2 (**Figure 3**, left panel). In sedimentary settings, CO_2 may remain mobile for centuries and trapping relies primarily on the impermeability of the caprock. This is observed for closed structural traps, in which fluids do not move far and both solubility and capillary trapping remain low. Conversely, large saline aquifers are hydrodynamic traps where CO_2 moves rapidly through the pore space and interacts with a larger volume of the reservoir, increasing the extent of all secondary mechanisms (National Academies of Sciences Engineering Medicine, 2019).

Carbon Mineralization in Basalts for CO_2 Storage

The high porosity and permeability of basalt increases its reactivity with CO_2 making it an ideal medium for CO_2 injection and storage. Two pilot projects—Wallula in Washington state, USA and CarbFix in Iceland—demonstrate this high potential for CO_2 storage in basalt. For both projects the composition, structure, and hydrology of the reservoir were studied, to assess the viability of the targeted reservoir for the injection and sequestration of CO_2 in the rock pore space (National Academies of Sciences Engineering Medicine, 2019). The Wallula pilot plant successfully injected 977 tons of pure CO_2 between 838 and 886 meters in depth over the course of 25 days during the summer of 2013 for the purpose of CO_2 storage (McGrail et al., 2014, 2017a,b).

The aim of the CarbFix project is to dispose of H_2S as well as CO_2 derived from condensation of natural steam. In Phase I, ∼200 tCO_2 were injected at a depth of 500 m with a 100 m flow path through the basalt. New techniques were developed in both Phase I (Sigfusson et al., 2015) and Phase II (Gunnarsson et al., 2018) to inject water and CO_2 separately with proportions insuring the complete dissolution of CO_2 in water at depth. These techniques are very important, because they eliminate the need for an impermeable caprock and thereby greatly increases the number and extent of potential basalt storage reservoirs. Tracers including SF_6 and ^{14}C, were used to assess the fate of CO_2 in the basalt-hosted aquifer. Rapid carbon mineralization removed dissolved CO_2 from pore water within months. Phase II is more ambitious and continues to inject 10–20 $ktCO_2/yr$ and a roughly comparable mass of H_2S at 1,500 m in depth, forecasting a 2,000 m flow path. Over 3 years, the carbon mineralization rate has been about $5 \cdot 10^{-9}$ $mol \cdot m^{-2} \cdot s^{-1}$ (a value that can be compared with the summary of experimental data in **Figure 2**). This offers the potential to recycle pore water for continued co-injection with CO_2, and addresses concerns about the extent of water consumption for solution trapping methods. Moreover, Climeworks has installed direct air capture unit on site, to investigate the potential for the project to be carbon negative (Gislason et al., 2010; Aradóttir et al., 2011; Matter et al., 2011, 2016; Snæbjörnsdóttir et al., 2017; Gunnarsson et al., 2018).

These pilot projects have demonstrated the potential for CO_2 storage in basalt reservoirs (McGrail et al., 2014; Gíslason et al., 2018) and monitoring is ongoing for a better understanding of the process (Alfredsson et al., 2013; Matter et al., 2014, 2016). Basalts contain alteration minerals and glass that could be more reactive than the original, igneous minerals, which are generally more abundant (Oelkers and Gislason, 2001; Gislason and Oelkers, 2003; Wolff-Boenisch et al., 2006; Flaathen et al., 2010; Alfredsson et al., 2013). It is thus important to know the role and proportion of alteration minerals and glass at specific sites of carbon mineralization. Other concerns are potential formation of a passivation layer that may decrease the reactivity of the rock over time (Power et al., 2013b). Also, crystallization of reaction products in pore space may lead to clogging of pore space (Alfredsson et al., 2013) and subsequently, a decrease in permeability. Monitoring will quantify the evolution in space and time of a dissolution zone in acid waters close to the injection well, and a precipitation zone that will occur further away and pushed outwards as new carbonates are formed.

Storage of CO_2 in basalts is estimated to cost \$20–\$30/tCO_2, as compared to <\$20, perhaps <\$10, for storage in pore space in sedimentary reservoirs. Basalt storage is likely to be a preferred choice in volcanic provinces, and possibly additional gigantic near-shore reservoirs in the extensive basalt layer that comprises the upper few kilometers of oceanic crust, worldwide (Goldberg et al., 2008, 2010; Goldberg and Slagle, 2009). Because they cool very rapidly during submarine eruptions, basalts in oceanic crust may have higher proportions of basaltic glass that react much faster than crystalline basalt. Overall, abundant basalts, some close to populated areas, can potentially store thousands of gigatons of CO_2.

Carbon Mineralization in Peridotites

Peridotite is the major constituent of Earth's upper mantle and is primarily composed of olivine, which reacts spontaneously with air or fluids containing CO_2. The biggest massif of exposed peridotite is the Samail ophiolite in Oman and the United Arab Emirates. There are almost equally large massifs in New Caledonia and Papua New Guinea. The contiguous 48 US States contain relatively small bodies of peridotite that cumulatively have about the same mass as the Samail ophiolite (Krevor et al., 2009). The primary minerals in tectonically exposed mantle peridotite (mainly olivine, pyroxenes and spinel) transform partially or completely into mixtures of serpentine, brucite, Fe-oxides and -oxyhydroxides. Extensive carbon mineralization in the Samail ophiolite has been observed, forming carbonate veins in fractures, and travertine terraces around alkaline springs. The rate of natural carbon mineralization in the Samail ophiolite is on the order of 1,000 tCO_2/km^3/yr (**Figure 2**) (Kelemen and Matter, 2008; Kelemen et al., 2011; Streit et al., 2012; Mervine et al., 2014). The presence of listvenites—fully carbonated peridotites composed entirely of magnesite and dolomite ($MgCO_3$, $CaMg(CO_3)_2$) + quartz (SiO_2) + hematite and goethite (Fe_2O_3, $FeO(OH)$) + relict spinel and/or chromian mica—show that full carbonation of peridotites is possible at natural conditions (about 100°C) (e.g., Falk and Kelemen, 2015; Kelemen et al., 2018).

Peridotite reservoirs have the potential to mineralize and sequester 10^5-10^8 GtCO_2 and are potentially the least expensive route for combined CO_2 capture from air and storage. Because olivine, serpentine and brucite are far from CO_2, H_2, and O_2 exchange equilibrium with air and surface water, peridotites are also an immense reservoir of chemical potential energy that could yield heat and work from weathering reactions. Nevertheless, compared to basalts, peridotites are less permeable, have less abundant outcrops, and are farther from population centers and sources of anthropogenic CO_2.

As in any *in situ* carbon mineralization system, the extent of carbon mineralization could become limited through negative feedbacks, i.e., passivation of reactive surfaces and clogging of pore space causing a decrease in permeability. While basalt formations may be more vulnerable to passivation than peridotite, pore space clogging is less important in basalts because of their higher initial porosity and permeability (National Academies of Sciences Engineering Medicine, 2019). However, in peridotite formations, natural alkaline springs have persisted in the same locations for tens to hundreds of thousands of years without running out of reactive surface areas or destroying permeability (Kelley et al., 2001; Früh-Green et al., 2003; Ludwig et al., 2006, 2011; Kelemen and Matter, 2008; Kelemen et al., 2011, 2019). In some cases, every magnesium and calcium atom in peridotite has combined with CO_2 to form listvenite (previous paragraph). This may be the result of positive feedback, in which reaction-driven volume changes cause increases in differential stress in the rock and generate fractures, maintaining or increasing the permeability of the rock and exposing fresh reactive surfaces (e.g., Jamtveit et al., 2008; Kelemen and Matter, 2008; Rudge et al., 2010; Kelemen and Hirth, 2012; Evans et al., 2018). Reaction-driven cracking is probably maximized when the rate of reaction is highest. During some experiments on carbon mineralization in peridotite, a reduction in permeability has been observed (e.g., Andreani et al., 2009; Hövelmann et al., 2012; Godard et al., 2013; van Noort et al., 2017), emphasizing the necessity to better understand of the parameters and mechanisms driving positive (cracking) or negative (clogging) feedbacks. The analog experiments of Zheng et al. (2018) illustrate the efficacy of reaction-driven cracking under the right conditions. In addition to their application to *in situ* carbon mineralization, a greater understanding of these mechanisms could be applied toward geothermal power generation, *in-situ* mining, extraction of oil and gas from tight reservoirs, and conversely, ensuring clogging to avoid failure of caprock and wellbore cement overlying CO_2 storage reservoirs.

Geologic examples demonstrate that complete, *in situ* carbon mineralization of peridotite is possible at elevated temperature and relatively high partial pressures of CO_2 (e.g., Falk and Kelemen, 2015; de Obeso et al., 2017). Thus, at about 185°C and more than ~70 bars P_{CO2}, carbon mineralization may be implemented for solid storage of CO_2 at a cost on the order of \$10 per ton [(National Academies of Sciences Engineering Medicine, 2019), Figure 6.17]. Modeling has shown that injecting CO_2 at depth in peridotite formations could increase the mineralization rate by 16,000× over natural systems, and the weight of CO_2 sequestered per kilogram of peridotite could be 350× higher

(Paukert et al., 2012). However, complete carbon mineralization is unlikely, and the cost of injecting fluid, per ton of CO_2, becomes very high at temperatures lower than 50°C and/or CO_2 partial pressure $< \sim 1$ bar.

As noted above, peridotite undergoing low temperature alteration and weathering produces C-depleted, Ca-rich alkaline waters, via precipitation of Mg-carbonates and dissolution of Ca-silicates along the subsurface reaction path. When these alkaline waters emerge at springs on the surface, they combine with atmospheric CO_2 to form calcite ($CaCO_3$) travertine deposits (e.g., Barnes and O'Neil, 1969; Bruni et al., 2002; Paukert et al., 2012). Thus, accelerated production of water from peridotite-hosted, alkaline aquifers represents a low-cost opportunity for CO_2 removal from air coupled to solid storage. The size and recharge rate of such aquifers are unknown. Note that only very limited amounts of pumping could be employed. In near-surface peridotite catchments, natural water saturated in air contains ~40–400 ppm CO_2 (calculated with all dissolved C as CO_2, using C concentrations from Kelemen and Matter, 2008, Figure S2). Similarly, we calculate that alkaline water at the surface will consume about 400 ppm CO_2 to form calcite (Vankeuren, personal communication). At these concentrations, every penny spent pumping a ton of water becomes \$20–\$200 per ton of CO_2. Thus, to produce alkaline water, it is best to seek areas with artesian springs, relatively high subsurface permeability, and/or a significant thermal gradient promoting thermal convection of water in pore space. Recharge of alkaline, peridotite-hosted aquifers could be accelerated by recirculation of produced water into peridotite, with costs of \$50–\$100/tCO_2 in formations with permeability $\sim 10^{-12}$-10^{-13} m^2 (1–0.1 Darcy), with each well having the capacity to capture and store a few thousand tCO_2/yr (Kelemen et al., 2016; Kelemen et al., under review). Unfortunately, costs are strongly correlated with permeability, so that in low permeability formations (e.g., $\leq 10^{-14}$ m^2), the costs would exceed \$1,000/tCO_2. On the other hand, costs are negatively correlated with the CO_2 concentration in the circulated fluid. Initial investigations into relatively low-cost enrichment of CO_2 from 0.04% in air to, e.g., 5% using direct air capture to increase the CO_2 purity, followed by *in situ* carbon mineralization (Kelemen et al., under review) are ongoing.

In summary, near-surface peridotite formations have the capacity to store over 100,000 GtCO_2 at relatively affordable costs that could be similar to CarbFix or injection in subsurface sedimentary reservoirs. Drilling to depths where temperature exceeds 100°C, and pumping of high P_{CO2} fluids, could yield efficient, permanent, solid storage of CO_2 captured elsewhere. At lower temperature, production of alkaline water from peridotite-hosted aquifers could provide a route to CO_2 removal from air, combined with solid storage that is cost-competitive with direct air capture using chemicals. A combination of peridotite carbonation and geothermal power generation could be a win-win situation, because the extraction and reinjection of geothermal fluid drives fluid flow through peridotite, necessary for extensive carbon mineralization, an exothermic reaction that could preserve the high temperature in the fluid reservoir, maintaining thermal buoyancy, and power generation.

Geological Sequestration in Saline Aquifers and Oil and Gas Depleted Reservoirs

Supercritical CO_2 (density around 600 kg/m^3) may be injected and stored in deep sedimentary formations. Ideal reservoirs for CO_2 sequestration are thick reservoirs located below 1 km in depth with high porosity and high permeability ensuring a high storage capacity (i.e., 50–100 MtCO_2 /project) at commercially meaningful injection rates (MtCO_2/yr). Suitable reservoir rocks have high porosity and permeability such as sandstone, limestone, dolomite, basalt, or mixtures thereof, whereas a suitable caprock is made of shale, anhydrite, or low permeability carbonate rocks. Long-term CO_2 sequestration and reservoir safety rely on the impermeability of the caprock and the extent of the secondary trapping mechanisms (section Storage in Subsurface Pore Space) (National Academies of Sciences Engineering Medicine, 2019).

Injections of CO_2 into subsurface reservoirs began in the 1970s for enhanced oil recovery. Today CO_2-EOR has the potential to sequester 30 GtCO_2 if the CO_2 used in oil recovery [assuming an industry standard of 3 bbl oil per tCO_2 sequestered] is left in the ground after oil production (Kuuskraa, 2013; International Energy Agency, 2015). Advanced CO_2-EOR methods, designed to co-optimize both oil recovery and CO_2 sequestration, could sequester over 90 GtCO_2 (National Academies of Sciences Engineering Medicine, 2019), and this could increase oil production compared to standard CO_2-EOR methods (Benson and Deutch, 2018).

The Sleipner Saline Aquifer Storage Project is the first commercial-scale CO_2 storage project. It commenced in 1996 and has injected CO_2 into an offshore saline aquifer formation, comprised of permeable sandstone beneath a low permeability shale caprock for permanent disposal and climate change mitigation (Baklid et al., 1996). This project shows that CO_2 storage in subsurface sedimentary reservoirs at a rate of 1 Mt/yr is possible and safe. In addition, the project helped increase our understanding of CO_2 plume migration. The project led to the avoidance of a tax of roughly 320 NOK/tCO_2 (36 USD at 9 NOK/USD) (Torp and Brown, 2005). Four commercial-scale projects globally are currently collectively sequestering 4.2 MtCO_2/yr and a fifth pending project in Western Australia is aiming to sequester 3.4–4 MtCO_2/yr. These projects store anthropogenic CO_2 from natural gas processing, chemical production, and power generation (**Table 1**) (Global CCS Institute, 2019; National Academies of Sciences Engineering Medicine, 2019).

Sedimentary basins around the world have the potential to store 8,200 GtCO_2 in a conservative estimate and between 20,000 and 35,000 GtCO_2 in high estimates (Benson et al., 2005, 2012; U.S. Geological Survey (USGS), 2013; de Coninck and Benson, 2014; U.S. Department of Energy (DOE), 2015; Global CCS Institute, 2019). A reasonable CO_2 footprint for sequestration in sedimentary reservoirs lies between 0.5 and 5.0 tCO_2/m^2, and depends on the architecture of the reservoir and the caprock, the petrophysical properties of the rocks, the pressure and the temperature in the reservoir, and the

TABLE 1 | Global CO_2 sequestration projects for climate change mitigation (Rutqvist et al., 2010; Vasco et al., 2010; Eiken et al., 2011; Shi et al., 2013; McGrail et al., 2014; Gíslason et al., 2018; Marieni et al., 2018; Global CCS Institute, 2019; National Academies of Sciences Engineering Medicine, 2019).

Project	CO_2 source	Date	CO_2 injection rate (Mt/yr)	Observations
CO_2 SEQUESTRATION IN SEDIMENTARY FORMATIONS				
Sleipner *Offshore Norway*	Natural gas processing	1996–present	1	1st project injecting supercritical CO_2 in a saline aquifer for long-term storage
In Salah *Algeria*	Natural gas processing	2004–2010	0.7	• Large pressure build-up in the reservoir • Unexpected geomechanical deformation
Snøhvit *Offshore Norway*	Natural gas processing	2008–present	1	Fast decrease in CO_2 injectivity, remedied by injecting into a different interval
Decatur *Illinois,United States*	Chemical production	2011–2014 2017–present	0.3 1	
Quest *Alberta, Canada*	Power generation	2015–present	1.2	
Gorgon *Barrow Island, Australia*	Natural gas processing	Under construction	3.4–4	
CO_2 SEQUESTRATION IN BASALT FORMATIONS				
CarbFix *Iceland*	Geothermal power generation Direct air capture	2012–2016	200 tCO_2	Ending reason: upscaling of the project
		2014–present	6,500 tCO_2/yr	• Alternated injections of CO_2 and water, so that CO_2 entirely dissolves in water at depth • Co-mineralization of carbon and sulfur
Wallula *Washington State, United States*		2009–2013	977 tCO_2	Injection of supercritical CO_2

extent of secondary trapping mechanisms (National Academies of Sciences Engineering Medicine, 2019).

True realization of a given capacity depends on a number of factors. More specifically, the uncertainties surrounding the pore space that is usable for CO_2 storage include: the pressure buildup in the reservoir during injection; the quality of the seal, including the presence old boreholes that penetrate the seal; other uses of the formation; and other restrictions on above-ground or surface resources (Ehlig-Economides and Economides, 2010; Zoback and Gorelick, 2012; Bachu, 2015). The concept of dynamic capacity has been developed to address the issues surrounding pressure buildup and the maximum rate of CO_2 injection. This parameter depends on other reservoir users and if active pressure management is implemented (e.g., brine removal) (Buscheck et al., 2012). Actual CO_2 sequestration capacities could thus be in the lower range of assessed reservoir capacities (National Academies of Sciences Engineering Medicine, 2019). A more salient issue is the proximity of quality storage reservoirs to sources of CO_2 emissions. While long-distance transport of CO_2 is possible, it can add considerably to the cost of CCUS.

In general, even the conservative estimates for CO_2 sequestration capacity far exceed the cumulative anthropogenic CO_2 emissions since the Industrial Revolution (2,035 ± 205 $GtCO_2$) (Le Quéré et al., 2015). Worldwide potential for sequestration of supercritical CO_2 in sedimentary rock pore space is estimated to be between 8,200 $GtCO_2$ and 34,700 $GtCO_2$. Most of the possibilities are located in Asia (2,964–2,984 $GtCO_2$), North America (2,600–21,770 $GtCO_2$), and South America (~2,030 $GtCO_2$) (Global CCS Institute, 2019). Given this geographic dispersion, highly concentrated CO_2 sources and storage opportunities are often co-located, which minimizes CO_2 transportation costs and associated emissions. If the strategy is to use the reservoirs with the highest capacities or the highest injection rates, CO_2 may be provided by long distance transport or direct air capture depending on the relative costs of each (National Academies of Sciences Engineering Medicine, 2019).

Increasing the Security of *in-situ* CO_2 Storage in Subsurface Pore Space

Sequestration in sedimentary formations is the most mature technology for long-term storage of CO_2 in subsurface pore space. The requirements for secure reservoirs are described in sections Storage in Subsurface Pore Space and Geological Sequestration in Saline Aquifers and Oil and Gas Depleted Reservoirs. Assurance of long-term storage security can be enhanced by secondary trapping mechanisms that immobilize the CO_2, including dissolution, residual gas trapping, and mineralization. Prior to CO_2 injection, targeted reservoirs are modeled using multiscale and multiphysics numerical simulations coupled to field observations in order to assess the ability of the reservoir to securely sequester CO_2. During and after the injection it is necessary to monitor the evolution of CO_2, the reservoir, and the caprock (Jenkins et al., 2015), in order to detect leakage from the reservoir. It is thus necessary to have suitable conditions for monitoring. In addition, underground sequestration of CO_2 aims to avoid any competition with other vital resources, such as freshwater and land use (National Academies of Sciences Engineering Medicine, 2019).

Modeling and Simulating for Safety Assessment of Storage in Subsurface Pore Space

Numerical modeling of the target reservoir helps to design and optimize CO_2 injection and confirms the performance of the formation. The goal is to predict the evolution of the plume, its center and spread, as well as the CO_2 phase distribution. Modeling outputs include CO_2 plume migration, pressure buildup, geomechanical effects, and geochemical reactions (National Academies of Sciences Engineering Medicine, 2019). These phenomena are coupled and can be described by a set of nonlinear differential equations that model fluid thermodynamics and physical processes. Solving these set of equations requires using finite differences, finite volume, or finite element simulations (U.S. Department of Energy (DOE), 2017d). Mature codes using paralleled temporal and spatial discretization and related equations to model the fluid thermodynamics and physical processes provide similar results (Pruess et al., 2004; Class et al., 2009).

Accurate modeling of CO_2 evolution in subsurface reservoirs faces several challenges: (1) the complexity of physical and chemical processes occurring, (2) spatial and temporal scales ranging over more than 12 orders of magnitude (from nm to km, and from ms to 1000s years, respectively), and (3) incomplete high-resolution knowledge of the subsurface geology that necessitates probabilistic modeling. Simultaneously addressing these challenges in a single simulation is too computationally intensive. Hence, modelers use reduced physics models (Nordbotten et al., 2012), upscaling, and empirical parametrization provided by lab experiments. However, in order to make the experiments feasible, experimentalists use small samples over short timescales, high flow rates, and other parameters that may not be representative of natural subsurface reservoirs like relative permeability. These parameters must be extrapolated in order to understand the natural reservoir dynamics. Modeling is carried out over specific time and space scales and relies on computationally-intensive simulations using advanced fluid mechanics in realistic pore networks (Raeini et al., 2018). Efforts are underway to properly bridge the gaps between the various scales (Abu-Al-Saud et al., 2017). Additional work is required for improved rate constant data of the various chemical processes (Zhang and DePaolo, 2017) and the parametrization of convective dissolution of CO_2 in saline formations (Riaz et al., 2006). Additionally, a high-resolution description of the reservoir is only available at the wellbores. A description of the rest of the reservoir relies on indirect methods using geophysical imaging such as seismic surveys and probabilistic methods. The discrepancies on parameters and assumptions made from the various models result in differences in plume evolution and CO_2 phases distribution, illustrating the need for improved modeling tools (Nordbotten et al., 2012).

In practice, subsurface reservoir modeling uses an iterative method that aims to match observations and simulations by refining and calibrating the numerical model with direct and indirect observations of the reservoir characteristics. This method is used today and is shown to work well for geological sequestration projects (National Academies of Sciences Engineering Medicine, 2019). A combination of rapid growth in computational power and algorithm evolution are promising for the emergence of more accurate forecasting of CO_2 evolution in subsurface reservoirs, ultimately leading to the scale-up of geologic sequestration on the order of $GtCO_2$/yr (National Academies of Sciences Engineering Medicine, 2019).

Monitoring for Risk Prevention of Storage in Subsurface Pore Space

Major risks associated with the storage of CO_2 in underground pore space include leakage from wellbores or non-sealed fractures in the caprock, pressure buildup in the reservoir that could result in caprock hydraulic fracturing, and contamination of drinking water. Prevention of these risks relies on the monitoring of CO_2 plume migration, pressure in and above the reservoir, induced seismicity, extent of secondary trapping mechanisms, and the chemistry of freshwater aquifers close to the CO_2 reservoir (National Academies of Sciences Engineering Medicine, 2019). Injection wells for CO_2 sequestration are Class VI and in the U.S., guidelines exist that prescribe monitoring requirements throughout the lifecycle of a CO_2 storage project (European Parliament the Council of the European Parliament, 2009; U.S. Environmental Protection Agency (EPA), 2010).

Pressure-variation monitoring is one of the best indicators of CO_2 movement. Even if the horizontal displacement of the brine is small, the injection of CO_2 can create a pressure buildup >100 km away from the injection zone (Birkholzer et al., 2009). Pressure measurements are performed with pressure sensors placed in injection and monitoring wells and in aquifers above the storage formation. Pressure buildup assists in tracking CO_2 plume migration (Strandli and Benson, 2013; Strandli et al., 2014), but pressure changes in aquifers above the reservoir indicate leakage in the overlying aquifer (Meckel et al., 2013; Kim and Hosseini, 2014; Cameron et al., 2016). Interferometric synthetic aperture radar (InSAR) measurements of ground surface deformation have also proved to be efficient for estimation of large pressure buildup in the reservoir in regions with few seasonal variations (e.g., In Salah, Algeria) (Vasco et al., 2010). However, the most favored technique for CO_2 plume monitoring is seismic imaging (Hovorka et al., 2006; Pevzner et al., 2011; Ivanova et al., 2012; Ajo-Franklin et al., 2013; White, 2013; Jenkins et al., 2015). Tracking the CO_2 plume can be used to calibrate and validate reservoir models, detect leakage of CO_2 into overlying aquifers, and indicate the "CO_2 footprint" of the plume. It could also provide information about the interactions between several injection sites in the same formation (U.S. Environmental Protection Agency (EPA), 2013; National Academies of Sciences Engineering Medicine, 2019).

Injection of CO_2 in subsurface reservoirs may create induced seismicity, likewise for injection of oilfield brine disposal or hydraulic fracturing for enhanced oil and gas recovery (Ellsworth, 2013; Walsh and Zoback, 2015; Langenbruch and Zoback, 2016). Induced earthquakes are mostly of magnitude 2 or below and are not felt at the surface and require sensitive geophone arrays to be recorded, identified, and located. Still, microseismicity monitoring at Decatur (USA) and In Salah (Algeria) sequestration sites have shown thousands of induced earthquakes per year (U.S. Department of Energy (DOE), 2017c). Most studies consider that induced earthquakes have a low risk of triggering fault displacements and endangering the security of the reservoir, but some authors have argued that even small- to moderate-magnitude earthquakes could damage the seal and compromise the integrity of sequestration reservoirs (Zoback and Gorelick, 2012). Experience to date has shown that the vast majority of micro-seismic events are not in the seal, instead, occurring in crystalline basement rocks far below the storage reservoir which may be due to a pre-existing geological structure—posing no risks to the storage integrity of the reservoir (Kaven et al., 2014).

The monitoring of CO_2 reservoirs also includes checking for CO_2 leaks at the surface or dissolved in groundwater. At the surface, CO_2 is commonly detected using infrared detectors or techniques developed for studying the carbon cycle in the terrestrial environment (Fessenden et al., 2010; Krevor et al., 2010; Lewicki et al., 2010; Male et al., 2010; Rouse et al., 2010). In groundwater, the presence or an increase of CO_2 can be deduced from reaction products (Hovorka et al., 2006; Jenkins et al., 2012; Romanak et al., 2012; Yang et al., 2013; Anderson et al., 2017), from analyses of pressurized samples representative of the subsurface conditions (Freifeld et al., 2005), and from tracers that help track waters containing dissolved CO_2 (Kharaka et al., 2009; Ringrose et al., 2009; Würdemann et al., 2010; Matter et al., 2011). Probabilistic modeling of risks associated with leakage suggest that 98% of the CO_2 will be retained in 10,000 years (Choi et al., 2013; Alcalde et al., 2018; Rogelj et al., 2018). An increase in CO_2 in aquifers may mobilize lead and arsenic contained in rocks (Zheng et al., 2009) creating an environmental hazard if drinking water sources are affected. However, numerical simulations have shown that the migration of fluids between reservoirs is unlikely through low-permeability sealing units (Birkholzer et al., 2009).

CARBON MINERALIZATION IMPLEMENTATION

Costs and Reservoir Capacities

In order to meet the 2°C target of the Paris agreement, 125 $GtCO_2$ has to be removed from air and permanently stored by 2100 (National Academies of Sciences Engineering Medicine, 2019). The cheapest storage methods with large capacity cost roughly \$7–\$30 per ton of CO_2 sequestered (storage costs only) and range from the injection of supercritical CO_2 into subsurface sedimentary formations to *in-situ* carbon mineralization via injection of CO_2-enriched fluids into on-land basalt and on-land peridotite formations (**Figure 4**). Storage of a total of 125 Gt CO_2 would result in a capital input of approximately 1–4 trillion dollars in total and 10–50 billion dollars per year until 2100. For comparison, the total cost of the energy transition, i.e., decarbonizing energy through shifting to an energy infrastructure comprised primarily of renewables, required for <1.5°C of warming was estimated to be 1.6–3.8 trillion dollars per year until 2050 (McCollum et al., 2018; Rogelj et al., 2018).

Recently the IPCC published a report stating that, to reach the 1.5°C goal, negative emissions technologies must be implemented at the scale of tens of Gt by mid- century (IPCC Special Report, 2018). In other words, in addition to reducing emissions and capturing CO_2 from point sources, CO_2 removal from air (CDR) is required. The options for CDR include an increase of carbon storage in soils and biomass, but also Direct Air Capture using Synthetic Sorbents (DACSS) and carbon mineralization via enhanced weathering (DACEW). It is likely that all of these options, operating in parallel, will be necessary to achieve the required level of global CDR (United Nations Environment Programme (UNEP), 2017; National Academies of Sciences Engineering Medicine, 2019). **Figure 4** summarizes the estimated cost vs. the annual capacity for several *in-situ*, *surficial*, and *ex-situ* techniques of CO_2 storage, and for a smaller number of techniques combining CO_2 removal from air with solid storage.

With regard to storage, the least expensive options with the greatest capacity involve injection of CO_2 into subsurface pore space in sedimentary rock, basalt, or peridotite formations on land. Sequestration in on-land basalt is competitive with storage in sedimentary reservoirs, but costs rise for storage in seafloor basalts. Various low-cost methods (e.g., sparging of concentrated CO_2, bioleaching, stirring, etc.) have been developed for CO_2 storage in mine tailings at a cost <\$30/tCO_2. In **Figure 4**, unless otherwise specified, costs for sedimentary reservoirs, on-land and seafloor basalt, and on-land peridotite include only the costs for storage. The total cost of capture *and* storage of CO_2 would include all costs incurred by the CO_2 capture method used (e.g., DACSS, DACEW, point-source, etc.), compression, and transport (if appropriate) to the sequestration site.

One of the lower cost options proposed for combined CO_2 removal from air and storage involves *in situ* circulation of water in equilibrium with air through peridotite, either in tailings or in the subsurface. Such methods may be optimized where they are co-located with geothermal power production. This technique does not require any CO_2 capture equipment, though recirculation of water is required to maximize uptake, since the CO_2 in air-saturated water is very dilute. With or without co-generation of geothermal power, configurations in which fluid circulation is driven by thermal convection or gravity could be competitive with DACSS coupled with storage in sedimentary reservoirs. However, where configurations require pumping water into low-permeability formations, costs rise to over \$1,000/tCO_2.

Another option for combined CO_2 removal from air and solid storage is carbon mineralization in ultramafic mine tailings and alkaline industrial wastes, which spontaneously react with CO_2 from air to form carbonate minerals during surficial weathering. In the case of *in situ* carbon mineralization in peridotite, mining and crushing peridotite for the purpose of CO_2 removal from air may be cost competitive with DACSS. This is particularly true if tailings are weathered, calcined, and weathered again. In this case, CO_2 produced during the calcining step could be stored, or sold for utilization. The storage potential of ultramafic

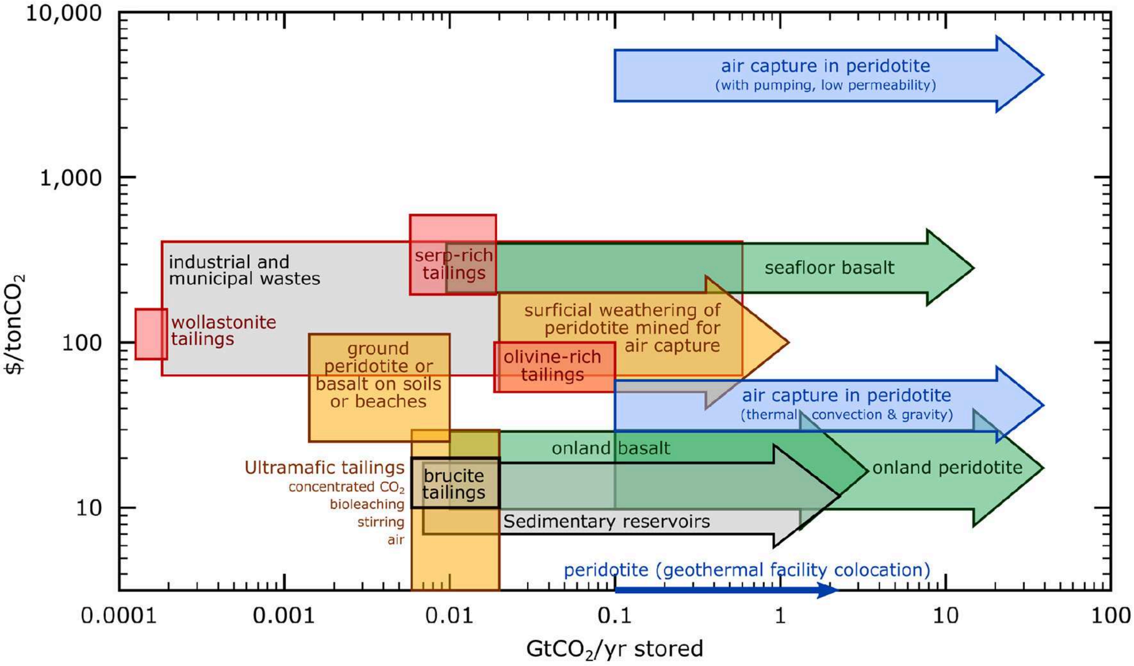

FIGURE 4 | Summary of the cost of CO_2 stored (US\$/tCO_2) vs. storage potential of CO_2 per year (GtCO_2/yr). Red boxes illustrate costs and rates for *ex-situ* CO_2 mineralization using heat and concentrated CO_2. Yellow boxes are for surficial CO_2 mineralization of mine tailings, of ground peridotite added to soils or beaches, and of peridotite mined and ground for the purpose of CO_2 removal from air with solid storage. Green arrows are for *in-situ* carbon storage by injection of CO_2-enriched fluids into mafic and ultramafic formations (e.g., CarbFix). Blue arrows are for *in-situ* carbon sequestration by circulating water saturated in air into peridotite formations, for CO_2 removal from air with solid storage. Gray arrow is for *in-situ* carbon sequestration by injecting supercritical CO_2 into subsurface sedimentary formations. Figure modified from (National Academies of Sciences Engineering Medicine, 2019), Figure 6.19, with data from references therein (Tables 6.1 and 6.2). Details on the capacity of industrial and municipal wastes are provided in **Table 2**.

tailings (about 0.2 GtCO_2/yr) and industrial and municipal wastes (0.1–1.3 GtCO_2/yr) depend on rates of mining and waste production, and are small compared to geological reservoirs. However, it may be that rocks can be mined and ground for the purpose of DACEW, again with costs and area requirements similar to, or less than, optimistic future cost estimates for DACSS. Moreover, *surficial* carbon mineralization can have other advantages: it can help industries lower their carbon footprint, and it may mitigate health and environmental hazards (e.g., from asbestos). Work has been done to create commodities from industrial waste (e.g., fabrication of CH_4 or more complex hydrocarbons, and so on) with nearly-net-zero emissions. A few such applications (e.g., adding CO_2 to aggregate for concrete) could provide CO_2 storage as well as value-added products (Shao, 2014; Monkman et al., 2018).

The total capacities of reservoirs proposed for *in-situ* CO_2 sequestration are $8.0 \cdot 10^3$-$3.5 \cdot 10^4$ GtCO_2 for sedimentary formations, $1.0 \cdot 10^3$-$2.5 \cdot 10^5$ GtCO_2 for on-land basalts, $8.0 \cdot 10^2$-$1.0 \cdot 10^5$ GtCO_2 for submarine basalts, $6.0 \cdot 10^4$-$6.0 \cdot 10^5$ GtCO_2 for on-land peridotite aquifers (National Academies of Sciences Engineering Medicine, 2019). There is perhaps another $6.0 \cdot 10^7$ GtCO_2 of capacity in seafloor fractured peridotite aquifers. Thus, storage capacities are not a limitation for CO_2 mitigation, given that current emissions are about 40 GtCO_2/yr (Le Quéré et al., 2015; IPCC Special Report, 2018). An important limitation for CO_2 sequestration in subsurface pore space is the maximum injectivity achievable in a given reservoir, which is 0.001–60 MtCO_2/yr in the U.S. in sedimentary reservoirs (Baik et al., 2018). Such rates are compatible with the climate goal requirements of storing 125 GtCO_2 by 2100 (National Academies of Sciences Engineering Medicine, 2019).

Ex-situ carbon mineralization is one of the most expensive techniques reviewed here, with costs generally estimated at hundreds of dollars per ton (**Figure 4** and **Table 2**) depending on the reactant and the carbon mineralization technique. It is generally more expensive to carbonate serpentine-rich and olivine-rich tailings *ex-situ* than to carbonate basalt and peridotite *in-situ*, due to the price of heating and transportation of the tailings. Still, less costly opportunities may be found where a source of concentrated CO_2 and a source of waste heat are close to a mine of mafic or ultramafic rock, or an industry producing waste byproducts with high alkalinity.

A large variety of sequestration options are available globally (**Figure 5**). The most effective for climate change mitigation is sequestration in geological formations, but locally—and where no geological formation is close to a source of concentrated CO_2–other solutions such as *surficial* or *ex-situ* carbon mineralization may be cheaper or present other advantages. Large geological formations far from anthropogenic CO_2 sources could be used to store CO_2 concentrated by direct air capture, or via enhanced weathering processes that combine CO_2 removal from air with solid storage.

TABLE 2 | Details of industrial and municipal wastes regarding the quantities produced per year, the potential CO_2 uptake per year, and the cost of the method (National Academies of Sciences Engineering Medicine, 2019).

Industrial and municipal wastes	Production (Gt_{waste}/yr)	Conditions of carbon mineralization	Uptake ($MtCO_2$/yr)	Cost ($)
Steel and blast furnace slag	0.17–0.5	- High temperature and pressure - Reaction with purified CO_2 or CO_2 saturated water ± Various pretreatment steps	2–100	75–100
Cement waste	0.42–2.1		1–300	Probably steel slag cost divided by ratio of weight fraction CO_2 for steel slag to this commodity
Construction and demolition waste	1.4–5.8		100–600	
Municipal solid wastes	1.3		4–160	
Coal ash	0.4–0.6		0.2–90	
Red mud, wastes from Al extraction	0.12		0.01–6	~150
Total CO_2 uptake			~100–1,250	

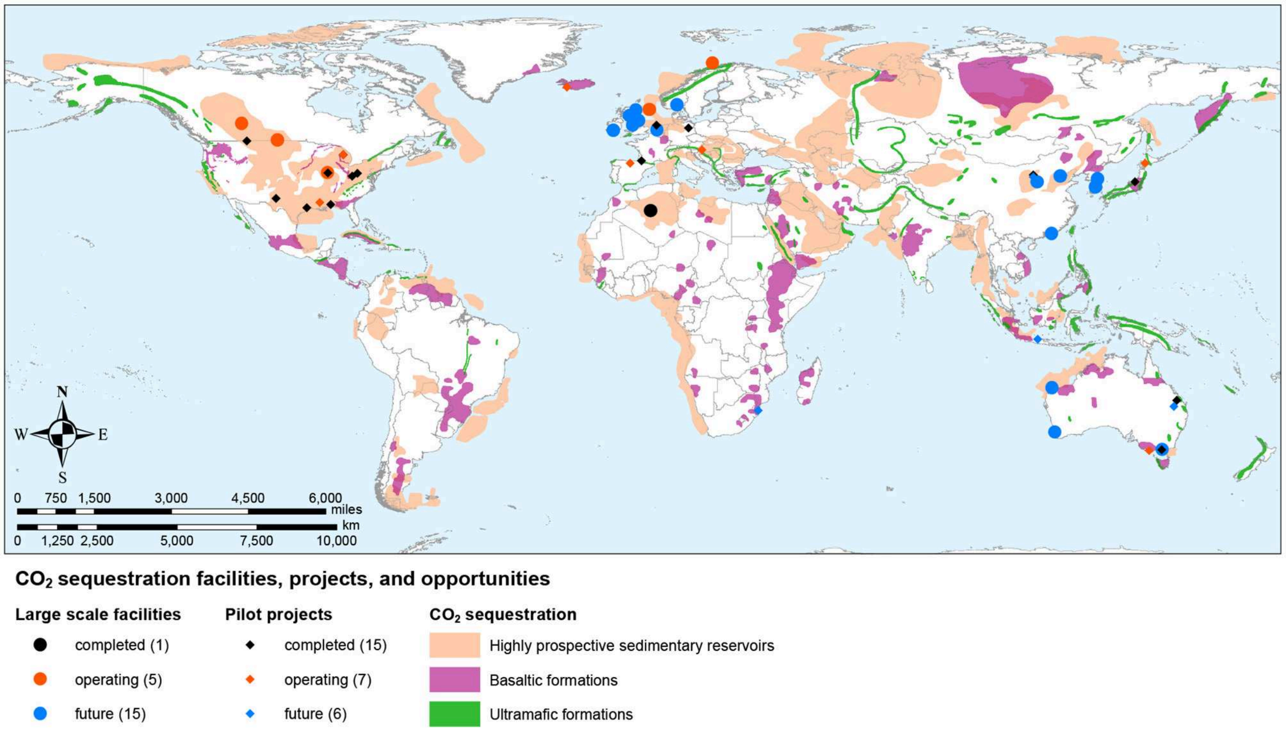

FIGURE 5 | Map of CO_2 sequestration facilities, pilot projects, and long-term storage potential in geologic formations (Kelemen, 1998; Bradshaw and Dance, 2005; Oelkers et al., 2008; Krevor et al., 2009; Global CCS Institute, 2019; National Academies of Sciences Engineering Medicine, 2019).

Current and Future Scientific Challenges

Ex-situ and *surficial* carbon mineralization are currently under development, and *surficial* carbon mineralization is ready for kt to Mt CO_2 field experiments. Research gaps remain in the area of carbon mineralization process improvement. One of the top priorities is to fill the gaps between datasets in order to compare different materials (rocks and industrial byproducts) at the same temperature, pressure, $p(CO_2)$, grain size, surface area, and fluid composition using the same units for comparison. Currently, research on *ex-situ* carbon mineralization focuses on the creation of usable products to offset high costs of this method relative to surficial and *in situ* carbon mineralization (for CDR and storage), and storage of supercritical CO_2 in subsurface pore space.

Surficial carbon mineralization methods would benefit from the optimization of heat-pretreatment, grinding processes, mining, and the development of new techniques (e.g., microwave treatment and/or microbial acceleration). Current production of mine tailings (and alkaline industrial wastes) limits the capacity of these methods, but mining of peridotite at the Gt/year scale for the purpose of CO_2 removal from air via carbon mineralization combined with solid storage (DACCM), may be cost competitive with the use of direct air capture with synthetic sorbents (DACSS) coupled to storage in subsurface pore space, particularly if the tailings are calcined and recycled. Some mine tailings sites are close to surface water and groundwater resources, requiring evaluation of the potential effect of geochemical contamination. More research is also needed to evaluate the impact of carbon mineralization on terrestrial, coastal, and marine environments. Current efforts to reduce the hazards of asbestos could be combined with carbon mineralization in tailings.

The CarbFix and Wallula pilot-scale CO_2 storage projects have shown that *in-situ* carbon mineralization for storage in basalt formations is feasible and safe, and that an impermeable caprock is not always necessary when CO_2 is dissolved in pore water at depth. Additional sites should be developed, ideally with high geothermal gradients to drive fluid flow and rapid reaction, readily available water to co-inject with CO_2, and high proportions of basaltic glass for accelerated carbon mineralization. Research is needed on potential contamination of nearby aquifers, and on the time evolution of the carbonate-forming reaction front, to get feedback about reaction rates, the three-dimensional distribution of reaction products, and permeability evolution. Medium-size pilot projects to address these questions are estimated to cost tens of millions of dollars per year for several years (National Academies of Sciences Engineering Medicine, 2019).

In-situ carbon mineralization in peridotite formations should be implemented on a pilot-scale, with particular emphasis on the maintenance or enhancement of permeability, perhaps via reaction-driven cracking as described in section Carbon Mineralization in Peridotites. It is essential to understand the chemicophysical parameters that lead to cracking vs. clogging, which can then be applied to favor cracking in reservoirs and clogging in caprocks. Currently, the size, injectivity, permeability, geomechanics, and microstructure of peridotite reservoirs are relatively unknown. Understanding nano-scale rock behavior is essential to understanding the macro-evolution of the reservoir, and more research is needed in this field. In order to assess the possibility of CO_2 storage in peridotite formations, two to three small-to medium-scale pilot projects are needed. This will lead to a greater understanding of the behavior of hierarchical fracture networks, which are difficult to simulate in laboratories. These pilot projects would cost about $10M—20M/yr based on Phase I of the CarbFix project and would test the efficiency of CO_2 mineralization when circulating surface water or CO_2-enriched water into peridotite (National Academies of Sciences Engineering Medicine, 2019).

Sequestration of CO_2 in sedimentary formations already stores about 4.2 $MtCO_2$/year. Current challenges are associated with upscaling more than 100 times in order to reach the $GtCO_2$/year scale and beyond. This requires field experimentation and testing, and sequestration of CO_2 in regions lacking major emission sources. Currently, it would cost $5M per well for supercritical CO_2 injection in sedimentary formations. Major research concerns are related to site selection, reservoir performance (e.g., increasing the speed of secondary trapping mechanisms, co-optimizing CO_2-EOR and sequestration, improving models), improving monitoring and reducing its costs, assessing risks (e.g., induced seismicity, groundwater contamination) and managing risks in compromised sequestration systems, as well as improving public confidence in secondary trapping mechanisms. Development of negative emission technologies also relies on the engagement of the community and on the favorable opinion of the general public that should be informed about the needs, opportunities, risks, and benefits of CO_2 sequestration in geologic formations.

Regulations, Financing, Partnerships, and Public Acceptance

Regulation already exists in the U.S. regarding fluid injection in subsurface pore space, due to the routine injection of wastewater into deep saline aquifers. Best practice manuals have been developed by the Department of Energy (DOE) for safe injection and sequestration of CO_2 in sedimentary formations (U.S. Department of Energy (DOE), 2010a,b, 2017a,b,c,d,e). Still, some problems remain to be resolved, e.g., the financial responsibility for long-term liability, pore space ownership, and regulatory impediments. Indeed, CO_2 injected into pore space in depleted oil reservoirs and deep saline aquifers can remain mobile during hundreds of years. Even if secondary mechanisms increase reservoir safety over time, regulations are needed to determine who is responsible for monitoring and remediation after injection projects shut down. Sedimentary units extend over tens or hundreds of km^2, and thus are shared by hundreds of owners, potentially restricting the scope of CO_2 sequestration projects. These two issues are easier to solve in countries where underground resources are owned by the government, which can act as a deciding body for project realization and be responsible for long-term management. Currently, the requirements for monitoring are significant and costly for operators. More flexibility adapted to each situation would still ensure safety and reduce the costs, making more projects affordable thereby facilitating the required deployment at the gigaton scale.

In the U.S., most research on CO_2 subsurface injection and carbon mineralization at the laboratory- and pilot-scale would be financed mainly by the DOE and the National Science Foundation (NSF), with support from the Environment Protection Agency (EPA), the US Geological Survey (USGS) and the Bureau of Land Management (BLM). The NSF would fund more fundamental research in hydrology, geochemistry, geophysics, biochemistry, and social sciences, whereas the DOE usually funds applied research on topics such as site characterization, modeling, optimization of trapping mechanisms, co-optimized carbon capture and oil production, monitoring, induced seismicity, *ex-situ* and surficial carbon mineralization, mitigation of environmental hazards, and pilot-plant projects in basalt and ultramafic formations. The EPA oversees regulatory aspects regarding site selection, operation, and monitoring, and aims to assess, minimize, and monitor risks of ground water contamination and mitigate environmental hazards. The USGS is in charge of mapping the geology of the U.S. and has already assessed sites suitable for CO_2 sequestration in sedimentary reservoirs and could help with site selection in mafic and ultramafic formations. Scaling up CO_2 sequestration in sedimentary reservoirs to $GtCO_2$/yr, would require using federal lands, and BLM could help investigate possibilities for CO_2 sequestration in these locations. The 10-year research agenda established in the 2019 National Academies of Sciences report calls for \$250/yr to realize the $MtCO_2$ to $GtCO_2$ upscaling for sedimentary reservoirs and for \$700M to expand pilot-scale studies in mafic and ultramafic formations (National Academies of Sciences Engineering Medicine, 2019).

Successful and rapid development of CO_2 sequestration and mineralization projects depends on data sharing, community coordination and funding. Research topics being shared between several institutions and partnerships would facilitate research and development. Partnerships are also essential between universities or government agencies and industry. Such partnerships already exist for some projects on carbon mineralization in mine tailings, and for the CarbFix project. Oil and gas industries have expertise in drilling, injection of fluids, and monitoring reservoir evolution. The oil industry currently injects megatons of CO_2 per year for CO_2-EOR, and their input in CO_2 sequestration projects would be valued, even if the quantity required to be injected for CO_2 sequestration is much larger. Some academic and government labs have expertise in laboratory analyses, computational modeling, monitoring, and practical experience with sequestration, that could result in a three-way transfer of knowledge. The federal tax credit 45Q is designed to support CO_2 injection in subsurface pore space. The credit increases progressively to \$35/tCO_2 in the case of EOR and up to \$50/tCO_2 for storage in deep saline aquifers.

Sequestration of CO_2 in the amount needed for climate change mitigation requires scaling CO_2 sequestration to the industrial scale. Public acceptance is essential. Thus, it is important to involve local communities and the general public in project realization. Education, and the demonstration of safe storage of CO_2 with successful pilot projects, would help in gaining public support.

CONCLUSIONS

Sequestration of CO_2 has to increase to the $GtCO_2$/yr scale to play a significant role in mitigation of climate change. Many options have to be considered to ensure fast development of some or all of the methods described in this paper. The research agenda outlined here, and described in more detail in National Academies of Sciences Engineering Medicine (2019), includes priorities ranging from fundamental research on kinetics, rock mechanics, numerical simulation, and database creation to small and medium-scale pilot projects for surficial and *in situ* carbon mineralization to assess the efficiency of reservoirs and to reduce associated risks. Supercritical CO_2 injection is the most mature storage method. Research is needed to improve actual techniques and reduce risks concomitant with injection and leakage. The 10-year research agenda requires \$250M to lead this field from the current, $MtCO_2$/yr scale to the $GtCO_2$/yr scale required for climate change mitigation. Carbon mineralization methods are at an earlier stage of development but include a set of reactants and techniques that are promising in terms of CO_2 uptake and safety of CO_2 storage. The research agenda aims to expand pilot-scale studies, with a total of \$700 M needed to reach this goal. These approaches complement each other as carbon mineralization is the ultimate step for permanent CO_2 storage, while injection of CO_2 into sedimentary reservoirs at the Gt/year scale leads the way in terms of technique, regulation, and public acceptance.

AUTHOR CONTRIBUTIONS

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

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The Role of Direct Air Capture in Mitigation of Anthropogenic Greenhouse Gas Emissions

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In recent years Direct Air Capture (DAC) has established itself as a promising approach to atmospheric Carbon Dioxide Removal (CDR) also referred to as Negative Emissions. However, due to the amounts likely needed to be removed CDR technologies like DAC will only become climate relevant if they rapidly reach gigaton scale, around the middle of this century. Here we give a brief insight into DAC and in particular, the modular low temperature DAC technology developed by Climeworks of Switzerland. We discuss potential co benefits, in particular in relation to the Sustainable Development Goals (SDGs) of the United Nations and conclude by suggesting some policy approaches on how a climate relevant scale could be achieved in time.

Keywords: direct air capture, DACS, negative emissions, NETs, mitigation, climate policy, climate change

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INTRODUCTION

In the 2015 landmark Paris Agreement, the international community committed to limit global warming by the year 2100 to "well below 2°C" and "to pursue efforts to keep warming below 1.5°C" compared to pre-industrial levels (United Nations, 2015).

To achieve that, first of all a worldwide rapid decline toward zero emissions is urgently needed. However, currently annual greenhouse emissions continue to rise (Quere et al., 2018), rendering these goals increasingly hard to achieve in time and near impossible with emissions reductions alone (Strefler et al., 2018). Therefore, atmospheric Carbon Dioxide Removal (CDR) or negative emissions technologies have increasingly been included in the vast majority of mitigation pathways and are now considered necessary to keep below 2°C and even more so for 1.5°C.

More concretely, *all* IPCC emissions pathways that are compatible with 1.5°C without overshoot[1] and 87% of pathways compatible with 2°C, rely on the assumption of large-scale atmospheric CDR, reaching zero emissions at some point in the 21st century followed by a period of net-negative emissions—meaning that CDR rates exceed residual emissions (IPCC, 2014, 2018; European Academies Science Advisory Council, 2018). In most scenarios the rate of CDR reaches several billions of tons (1Gt = 1 billion tons) per year over several decades as shown in **Figure 1**. Such large scale remains indicative, as global mitigation efforts and their success varies depending on shared socio-economic pathways, e.g., outlined by Rogelj et al. (2018) and technological feasibility of country-level net zero emission targets, e.g., outlined by Kaya et al. (2019). In all cases, CDR within a gigaton order of magnitude remain relevant and show a substantial impact on achieving climate targets, e.g., Realmonte et al. (2019). It is however most important to highlight that CDR is complementing rather than replacing mitigation.

Prominent approaches to CDR discussed in the scientific literature include afforestation and change of land use, enhanced weathering, bioenergy and carbon capture and storage or

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[1] Notably, "overshoot" is actually just a different formulation for building a carbon removal debt.

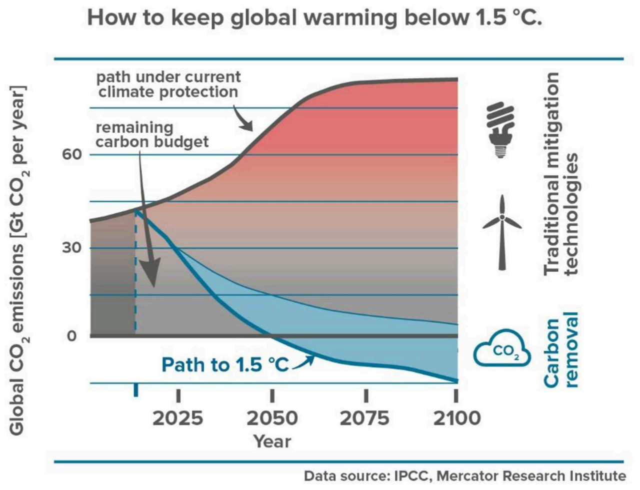

FIGURE 1 | Schematic illustration of NET deployment timing and scale[2].

BECCS, as well as direct air capture and storage of CO_2 or DACS (Smith et al., 2015).

DAC refers to a range of technological solutions that are able to extract CO_2 from ambient air at any location on the planet This is possible because in ambient air, CO_2 is nearly evenly distributed around the globe at average concentrations of, at present 405.5 parts per million and rising[3] (World Meteorological Organization, 2019).

Currently there are a handful of companies active in the DAC field, all currently designing and utilizing different DAC technologies, to focus on different markets.

In Canada, Carbon Engineering (CE) uses liquid alkali metal oxide sorbents regenerated by heat at around 800°C. CE currently uses natural gas to power its machines, co-capturing CO_2 from the flue gas stream of the burnt natural gas in addition to atmospheric capture (Keith et al., 2018).

A further key DAC player is Global Thermostat (GT) in the US. The GT process uses a solid amine-based sorbent material, regenerated at around 80–100°C (Ping et al., 2018).

Switzerland-based Climeworks AG uses a DAC design based on an adsorption/desorption process on alkaline-functionalized adsorbents, which is described in more detail in the following. CO_2 adsorption is performed at ambient conditions and CO_2 desorption is performed through a temperature-vacuum-swing (TVS) process (Wurzbacher et al., 2016). During this process, the pressure in the system is reduced and the temperature is increased to 80 to 120°C, thereby releasing the CO_2. After a cooling phase, the whole process begins anew, as illustrated in **Figure 2** below. The process results in the production of gaseous CO_2 at 1 bar with a purity level of >99.8%. Depending on the ambient conditions, in particular relative humidity, the current Climeworks process can also extract H_2O from the air as a byproduct[4].

Climeworks adopted a modular design to reduce manufacturing and operating costs, support scalability and diversity in deployment, ease of transportability and enable automated mass manufacturing. CO_2 adsorption and desorption is performed within the same devices, referred to as "CO_2 Collectors" or "Collectors." Collectors are engineered to fit efficiently into a steel frame, with 6 Collectors fitting into a standard 40-foot shipping container. The present nominal annual CO_2 Collector capacity is 50 tons of CO_2. This will increase as the technology is optimized. The modular Collectors are designed to operate together as a unit and can be scaled with the addition of new modules to expand capacity. They have been designed for commercial mass production that uses conventional metal fabrication technology.

Another important characteristic of the Climeworks DAC process is that a large share (around 80%) of the energy demand can be met by low-temperature heat in the range of 80–120°C. Heat at these temperatures is available from a variety of sources including industrial low-grade waste heat (Ammar et al., 2012). Long-term energy requirement projections based on current technology assumptions for the DAC process are expected at around 2,000 kWh per ton of CO_2 (400 kWh electrical and 1,600 kWh thermal). Climeworks plants can and will be solely powered by renewable energy sources and/or waste heat. The plants do not require a nearby fossil power plant in order to work. Costs per ton of CO_2 permanently removed from the atmosphere and safely stored via mineralization (CDR via DACS) are calculated to come down to around \$100 per ton of CO_2[5] within a decade, based on the detailed development roadmap for cost reduction and the learnings derived from the total of 14 DAC plants built and set into operation by the company across Europe so far.

In 2009 Climeworks was founded and first working laboratory prototypes were developed. In 2011, Climeworks built its first demonstration prototype. In 2015, a Climeworks system was commissioned for the world's first synthesis of renewable methane from atmospheric CO_2.

In 2017, Climeworks commissioned the world's first commercial DAC plant in Hinwil, Switzerland (**Figure 3**). The plant nominally captures 900 tons of CO_2 per year from the air and delivers the CO_2 to a nearby greenhouse.

2017 marked another milestone: together with their partners and project leader Reykjavik Energy, Climeworks built the world's first negative emissions plant in Icleand using DACS technology, known as the CarbFix2 project. The first CarbFix project was initiated by Reykjavik Energy. The aim of the project is to

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[2] Graph by Climeworks, based on a Graph by Mercator Research Institute on Gloabl Commons and Climate Change (2019) and Data from IPCC (2018). 2.6 RCP. Blue shading represents the amount of negative emissions likely needed in this scenario.

[3] There is regional and seasonal variation, of several ppm, which affects DAC only marginally. The variability in near surface CO_2 concentrations is visualized on the cover of the Royal Society report on Greenhouse Gas Removal: https://royalsociety.org/-/media/policy/projects/greenhouse-gas-removal/royal-society-greenhouse-gas-removal-report-2018.pdf

[4] With current technology in moderate climate conditions roughly at an average ratio of 1 ton of H_2O per ton of CO_2.

[5] For its cost calculations, Climeworks currently assumes 12 years plant life time and 20 years in the long term. Lifetimes in industries of comparable equipment are typically at the order of 30 years.

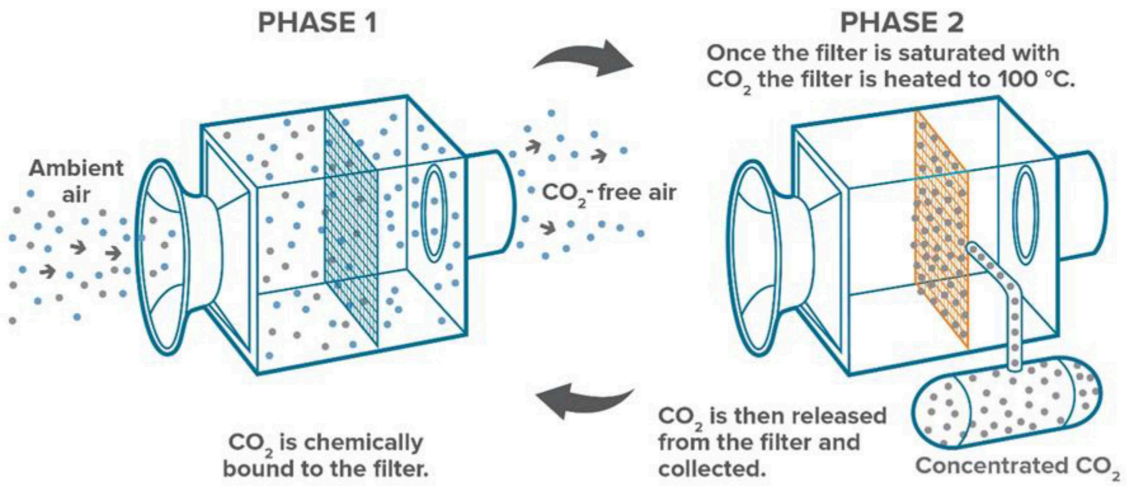

FIGURE 2 | Schematic illustration of Climeworks direct air capture process.

FIGURE 3 | The world's first commercial plant for direct air capture of CO_2 located in Hinwil, Switzerland.

develop safe, simple and economical methods and technology for permanent CO_2 mineral storage in basalts. The first pilot injections took place at Hellisheidi in South West Iceland in 2012; 175 tons of pure CO_2 were dissolved and injected at about 500 meters depth and about 35°C (Matter et al., 2016). Following the success of the pilot injections, the project was scaled up to industrial-scale in June 2014 as a part of the CarbFix-SulFix project, with injection of 65% CO_2–35% H_2S gas mixture at about 800 m depth and about 230°C. The injection is now an integral part of the operations at the Hellisheidi power plant (Sigfusson et al., 2018).

The CarbFix2 project builds on the success of the CarbFix project (as illustrated in **Figure 4** below), with the goal of moving the demonstrated CarbFix technology from the demonstration phase, to a general and economically viable complete CDR process chain that is replicable in other parts of the world. To this end, CarbFix2 extended the original CarbFix approach from the demonstration stage in one location to implementation under more diverse conditions. A main goal of the CarbFix2 project is to combine the storage approach with DAC technology, as developed by Climeworks, and thus create an integrated CDR solution with potential for negative emissions as well as global multiplication (Gutknecht et al., 2018). An integrated and safe solution toward permanent CDR has been created through the combination of the CarbFix and Climeworks technologies (Friedl, 2017). To date, the project has run for almost 2 years and demonstrates that negative emissions via DAC with rapid mineralization is possible.

In December 2018, Climeworks announced its partnership with Coca-Cola HBC Switzerland and that Climeworks will be

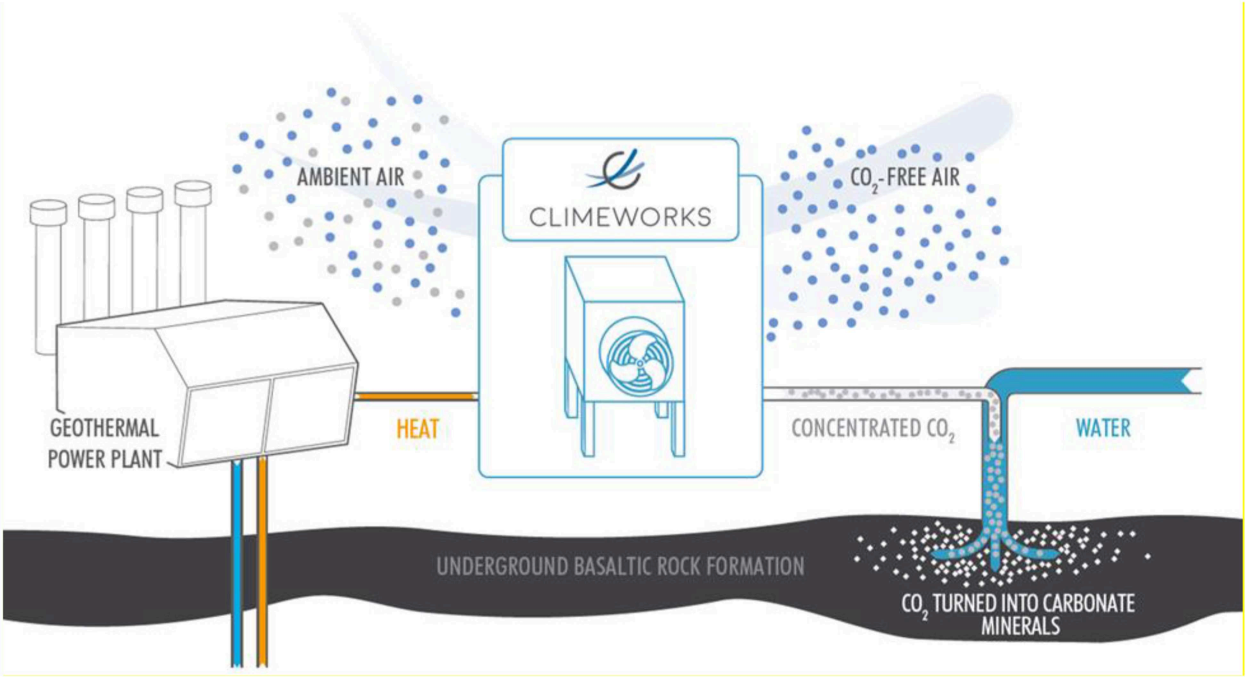

FIGURE 4 | Schematic illustration of the Climeworks-CarbFix injection at Hellisheidi, Iceland.

supplying atmospheric CO_2 to Coca-Cola HBC Switzerland's mineral water brand Valser from April 2019 on[6]. For 2020, Climeworks plans to build its so far largest DAC plant, scaling its DACS operations in Iceland by offering to remove emissions from the atmosphere to individuals[7]. Overall, the 14 DAC plants built by Climeworks so far will be complemented during 2020 with three more plants currently in planning or production. The company employs the currently largest team of DAC experts in the world at 70 full time equivalent positions[8].

When facing the need for negative emissions at gigaton scale, considerations have to be given to potentially limiting factors such as land use and resource constraints. Different CDR approaches have different requirements in terms of surface area and type of land they need to operate. Whilst afforestation and BECCS need large quantities arable land to grow trees and biomass that binds carbon, and will thus likely put pressure on ecosystems and food production systems if implemented at gigaton scale (Smith et al., 2015; IPCC, 2018), DACS can operate anywhere where renewable energy (such as PV, wind, or geothermal) can be found or installed. Further advantages of DAC(S) are that it does not require arable land, and has a much smaller physical footprint than bio-based approaches. According to Climeworks' estimations, around 2,000 km^2 of non-arable land would be needed to remove 1 gigaton of CO_2 net from the atmosphere, including the required renewable energy production. This calculation is based on the assumption that solar photovoltaic (PV) is the sole energy source, where heat is produced using heat pumps with a coefficient of performance (COP) of 3.5 and energy consumption is based on the long-term requirements as shown above. For the PV system, the following assumptions are taken: the PV ground cover ratio is 100% (to be adapted individually to local sites), the PV module efficiency is 25% and the annual solar radiation of suitable solar PV sites is assumed at 1.9 TWh per km^2, a value that can be achieved in parts of the US for example[9]. The footprint of the actual DAC plants would cover just 62 km^2. It is important to point out that depending on the location these footprints may vary and their calculation should receive further analysis. However, even if for example doubled in size, the DAC footprint would still be several orders of magnitude smaller than that of other CDR solutions.

CO_2 from the atmosphere can also be used for the production of a range of synthetic materials and fuels by using renewable energy (Bhanage and Arai, 2014). This has the advantage that "hard-to-electrify" sectors such as aviation, or long-distance heavy transportation, can be indirectly electrified via synthetic hydrocarbon fuels such as synthetic methane or Fischer-Tropsch fuels (Olah et al., 2009; Verdegaal et al., 2015), as well as a range of other hydrocarbon based products (e.g., polymers) can be made that previously relied on fossil resources. Together with its partners, Climeworks has demonstrated the viability of combining these technologies—usually referred to as Power-to-X (or PtX)—in Troia, Italy[10]. Here, CO_2 from the air is provided for conversion into renewable methane as part of the Horizon 2020-funded STORE&GO research project. The primary objective of

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[6] https://ch.coca-colahellenic.com/en/news-and-media/news/valser-to-get-its-fizz-from-the-air-with-climeworks/

[7] https://climeworks.shop

[8] As of August 2019.

[9] https://solargis.com/maps-and-gis-data/download/world

[10] www.storeandgo.info

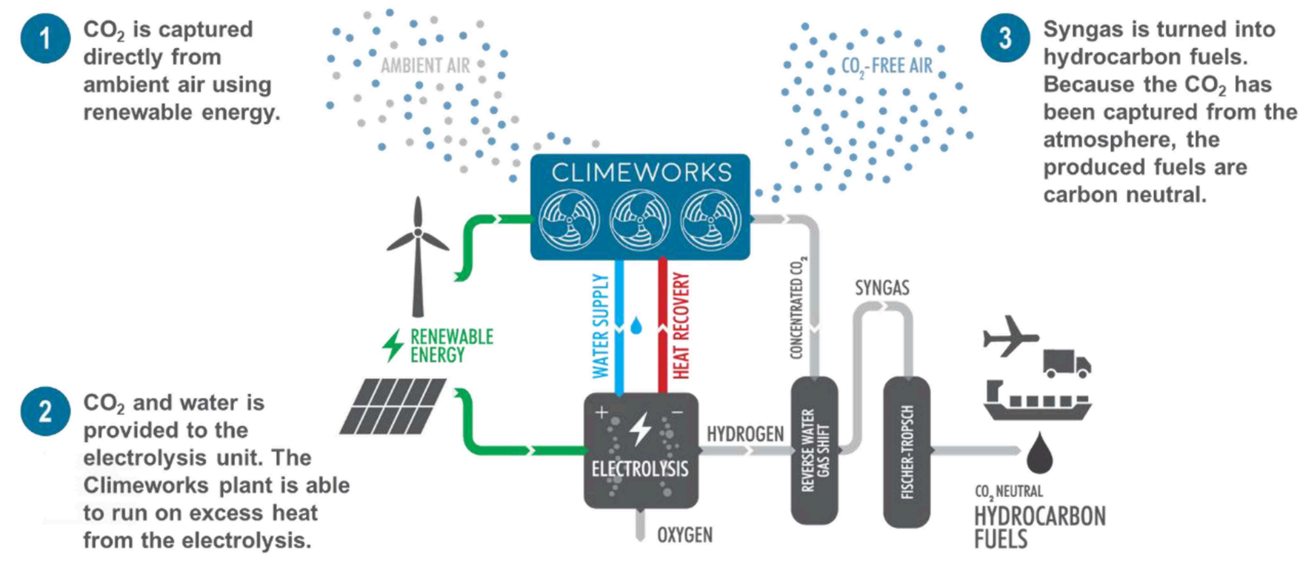

FIGURE 5 | Schematic illustration of the process chain for the production of renewable hydrocarbon fuels (in this case Fischer-Tropsch fuels), including DAC.

the STORE&GO project is to demonstrate the viability of large-volume energy storage through Power-to-Gas technology in real-life applications.

Besides energy storage, another advantage of air-captured CO_2 as a feedstock for synthetic fuels is that DAC allows for *location-independent* sourcing of CO_2. This means transport costs as well as emissions from transport are greatly reduced for any application. Being location-independent in particular allows for synthetic fuel production to be performed at locations with most favorable renewable energy prices. A cost advantage of just 1ct/KWh could offset or even overcompensate any potential extra costs of CO_2 derived from DAC compared to, for example, CO_2 that is captured from industrial point source emissions. Also, if atmospheric CO_2 is used, the fuels can be near-CO_2 neutral as the CO_2 that is emitted to the atmosphere when the fuels are burnt have been previously captured from that atmosphere (as illustrated in **Figure 5** above). In this way, DAC technology can contribute to the development of a circular economy independent from fossil hydrocarbons and to conventional mitigation as well. There are further co-benefits of DAC, especially in relation to the UN sustainable development goals. There are positive effects on SDGs 13, 9, 6 as the byproduct H_2O could be used as fresh water in general and, after corresponding purification, as drinking water in particular. A DAC expansion would bring new green jobs and thereby contribute to SDG 8, whilst it can also drive the scale-up of renewable energy, thereby driving SDG 7. DAC as a feedstock for PtX has positive effects on SDG 12 and 11 by allowing cities to produce their own fuels and materials, including synthetic building materials made from atmospheric CO_2. Lastly because synthetic fuels release far less air pollutants when burned (Olah et al., 2009) DAC could also have a positive effect on health and well-being (SDG 3).

Besides their removal and mitigation potential and contribution to SDGs, DAC and other NETs might offer more intangible but likewise valuable benefits—namely their contributions to the outlook on, and narrative around, the current climate crisis.

Over the last four decades, it has become apparent how difficult it is to agree on, and implement, appropriate policies that result in the mitigation pathways so urgently needed. In part, because in the absence of technologies that could replace fossil resources, such policies would have to be based on abstinence thereby resulting in having to relinquish a large proportion of lifestyles that especially those in industrialized countries have become accustomed to. Being able to decarbonize, for example, air travel with DAC and synthetic fuels at least offers the possibility of continuing to fly in a carbon neutral way.

Also notable is the current narrative around the reduction of carbon emissions to zero, in which lies implicitly the assumption that any human activity is a burden on the planet, as it is bound to have some emissions. In this context NETs open up the possibility of a much more positive self-image around a balance between residual (unavoidable) carbon emissions and natural carbon sinks or CDR, that humankind can actively manage and be the responsible stewards of.

Under current economic conditions that largely have not priced in the externalities of greenhouse gas emissions, CO_2 for niche markets, fuels and materials is a way for DAC companies to operate already today.

Current markets will likely allow DAC and NETs companies to grow to considerable size. However, these markets will almost certainly not be large enough to become "climate relevant," i.e., to produce negative emissions at the scales that are forecast/needed in time. To reach these scales, these externalities need to be priced in or regulated, which in turn means designing appropriate policies to ensure the scale-up needed.

Various climate scenarios predict negative emissions at gigaton scale by mid-century. What does this mean for CDR scale-up pathways? An example: to reach a mean pathway of around 6 gigatons of CDR by 2050 as calculated in a recent comprehensive review of the relevant literature (Nemet et al., 2018), from 2019 onwards, CDR would require an annual growth rate of over 55%. Delaying scale-up to 2025 would already require a sustained growth of 80% per year, whilst scale-up starting in 2030 when most CDR policies are currently recommended to set in, would require roughly a *yearly doubling* of CDR capacity. Scales like these are hard to achieve and from a risk perspective, it would therefore be vital to start scaling earlier.

To achieve such a feat, the dialogue between science, NET providers and policy makers urgently needs strengthening. Effective policy measures need to be developed and adopted, both for R&D on CDR and for its scale-up. How should these look like?

From the perspective of a NET/DAC company, first the technologies need to be made eligible in *existing* mitigation policies wherever possible. However, *new* policies also need to be designed so current NETs can make use of them.

A good example is the recently passed 45Q tax credit in the US, where DAC is eligible for CDR[11]. However, the policy still has a number of limitations. Firstly, the minimum installed capacity for eligibility is 100,000 tons of CDR per year. Building plants of this size would very likely exceed current market capitalization of all leading DAC companies and put the ones that decide to try at considerable risk of failure. In addition, the revenue stream the 45Q would create (at up to USD 50 per ton of CO_2), would not be sufficient to cover the costs of atmospheric CDR with current technologies (Keith et al., 2018)[12].

In general, any policy that puts a price on carbon usually plans to start low and raise the price subsequently in order to not disrupt current economic processes too much. However, technologies follow the opposite pathway. These are expensive to start with and have low capacities. Importantly, though, costs will fall rapidly as soon as the new technologies are being implemented. Future CDR policies thus need to accommodate this fact by allowing for *lower removal thresholds at higher prices*. As the installed capacity increases, the prices may fall corresponding to the (assumed rapid) technological development. In other words, the overall CDR volume that requires policies with higher prices, is relatively small and the absolute amount of money, "the world needs to spend on high-price CDR" is negligible compared to the required gigaton-scale carbon removal.

Also, polices like the recently passed European Renewable Energy Directive 2 (RED2) allow for atmospheric CO_2 as a feedstock for synthetic fuels (renewable fuels of non-biological origin) as described above. However, the regulation also allows for CO_2 captured from flue gas from (fossil) point sources. CO_2 from flue gas can be typically sourced at much lower cost, as the CO_2 concentration in the gas stream is higher. In the marketplace, such regulations therefore place an incentive on CCS CO_2 rather than CO_2 captured from air, again not triggering much needed CDR scale-up effects.

Lastly, unless the CO_2 in the flue gas stream comes from biomass, CO_2 from fossil point sources does not have the same CO_2 reduction effect, as fossil CO_2 in essence is reused *once more* in the synthetic fuel, before it is released back into the atmosphere. It is therefore important that this is reflected in appropriate life cycle analysis (LCA) methodologies (e.g., cradle to grave instead of cradle to gate) and regulations (von der Assen et al., 2013; Cuéllar-Franca and Azapagic, 2015; von der Assen, 2015).

CONCLUSION

DAC is a promising set of technologies that can deliver negative emissions and contribute to mitigation at scale. There are different technological approaches to DAC resulting in different resource requirements. It is important that these differences are taken into account, especially for large-scale predictions of NETs potentials. In general, DAC's main benefit is that the technology is space efficient and can be built on non-arable land, therefore not adding further pressure on either ecosystems or food systems. The technology can be used as a sustainable CO_2 feedstock for PtX technologies (synthetic fuels and materials). Such applications can help the scaling-up of DAC in the absence of a functioning price mechanism or policy for CDR. Negative emissions technologies like DAC can have a positive Impact on UN sustainable development goals and can create a positive impetus for narratives around climate change. DAC's biggest challenge is achieving climate-relevant scale, which—as any other NET—it cannot do without a sufficiently high price on carbon or other relevant policy measures. Because of the scales likely needed and the time it takes to develop these, climate policy *urgently* needs to develop and implement suitable mechanisms to trigger sufficient mitigation and scaling of NETs alike. As any other NET, DAC is not a substitute for mitigation but rather needed *in addition* to ambitious mitigation efforts. Like any NET, DAC is also not a silver bullet though likely to be a main contributor to a future NETs portfolio.

AUTHOR CONTRIBUTIONS

This paper was written by CB with inputs from JW and LC. Any opinions and own calculations presented in this article state the views of the main author only and are not necessarily shared by his affiliations.

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[11] https://www.betterenergy.org/blog/primer-section-45q-tax-credit-for-carbon-capture-projects/

[12] As well as Climeworks's own calculations.

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