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Review

Eco-engineering approaches for ocean negative carbon emission

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ABSTRACT

The goal of achieving carbon neutrality in the next 30–40 years is approaching worldwide consensus and requires coordinated efforts to combat the increasing threat of climate change. Two main sets of actions have been proposed to address this grand goal. One is to reduce anthropogenic CO_2 emissions to the atmosphere, and the other is to increase carbon sinks or negative emissions, i.e., removing CO_2 from the atmosphere. Here we advocate eco-engineering approaches for ocean negative carbon emission (ONCE), aiming to enhance carbon sinks in the marine environment. An international program is being established to promote coordinated efforts in developing ONCE-relevant strategies and methodologies, taking into consideration ecological/biogeochemical processes and mechanisms related to different forms of carbon (inorganic/organic, biotic/abiotic, particulate/dissolved) for sequestration. We focus on marine ecosystem-based approaches and pay special attention to mechanisms that require transformative research, including those elucidating interactions between the biological pump (BP), the microbial carbon pump (MCP), and microbially induced carbonate precipitation (MICP). Eutrophic estuaries, hypoxic and anoxic waters, coral reef ecosystems, as well as aquaculture areas are particularly considered in the context of efforts to increase their capacity as carbon sinks. ONCE approaches are thus expected to be beneficial for both carbon sequestration and alleviation of environmental stresses.

1. Introduction

Mounting evidence supports consensus that the current and projected warming is mainly caused by CO_2 released into the atmosphere by human activities such as fossil fuel burning and land use change [1]. The United Nations Framework Convention on Climate Change Paris Agreement aimed to keep global temperature increase below 2 °C, even below 1.5 °C above the pre-industrial baseline by the end of this century. To meet this requirement, many countries have proposed "carbon neutrality" timetables; however, the roadmap for implementation is unclear. While reducing emissions is absolutely essential, it is no longer sufficient to meet these temperature thresholds [1,2]. The urgent need is to

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achieve “carbon neutrality” by also increasing “negative emissions”, i.e., active CO_2 removal from the atmosphere.

Negative emissions require capturing atmospheric CO_2 and then storing it in a reservoir, either on the land or in the ocean, for a period of time meaningful to climate change mitigation. Many solutions have been proposed in the literature for alleviating global warming using terrestrial ecosystems, including forestation, habitat restoration, soil carbon sequestration, biochar production, bioenergy with carbon capture and storage, enhanced terrestrial weathering, mineral carbonation, direct air capture and CO_2 storage, and the use of low-carbon concrete (e.g., [3]).

The world’s oceans have a gargantuan capacity to serve as a CO_2 buffering system, storing up to 93% of the CO_2 in the Earth system and accounting for approximately 55% of global biological carbon capture through marine organisms [4]. They are estimated to have taken up 30%-37% of fossil fuel emissions since the 1960s (Table 6 of Friedlingstein et al.: Global Carbon Budget 2022 (page 4835) at https://www.globalcarbonproject.org/), thus playing a significant role in mitigating CO_2 accumulation in the atmosphere.

In this review, we aim to explore mechanisms underlying interactions between micro- and macro-processes, such as the biological pump (BP), the microbial carbon pump (MCP), and microbially induced carbon precipitation (MICP). We present scenarios of negative emission practices in an eco-engineering context based on the mechanisms discussed. We do not discuss approaches that are well known to the scientific community (e.g., oceanic iron fertilization and coastal blue carbon) and those that neglect ecological impacts (e.g., direct injection of CO_2 into the ocean interior and burial underneath the seabed). Instead, we identify pathways that would allow us to evaluate the feasibility, magnitude and effectiveness of negative emissions in the marine ecosystem.

We also advocate establishing a framework for marine carbon fingerprints and footprints at micro- and macro-levels because they are of essential importance in delineating the ecological and biogeochemical processes and mechanisms of carbon sequestration in the ocean, thereby providing theoretical foundations for carbon storage estimation and calculation for potential carbon trade.

2. ONCE approaches and mechanisms

2.1. Increasing negative emissions through enhancement of ocean alkalinity

Negative emissions can be enhanced by manipulating two key variables of the marine carbonate system, namely the dissolved inorganic carbon (DIC) pool and the alkalinity pool (Fig. 1). Algae and plants assimilate CO_2 and transform it into biomass (vertical shift in Fig. 1), and if this biomass is removed from the surface ocean, for example, by sinking, the same amount of carbon in the form of CO_2 will be replenished from the atmosphere, until an equilibrium concentration is restored. During the process of reaching a new equilibrium, the formation and removal of biomass take a portion of atmospheric CO_2 and permanently sequester it in the seabed. An increase in alkalinity (horizontal shift in Fig. 1) would allow the water to hold more DIC, which in turn will be supplied by the atmosphere until a new equilibrium with the atmosphere is reached.

The increase in ocean alkalinity is mainly caused by abiotic weathering of natural rocks (mainly silicates), which can convert CO_2 to HCO_3^-, thereby serving as a sink for atmospheric CO_2. It is estimated that natural weathering can absorb about 0.5 Pg of atmospheric CO_2 per year [5], and in earth’s history, vastly enhanced abiotic tropical weathering of the Taconic orogeny might have been a major driver for the Ordovician cooling [6].

Biotic processes can also have tremendous effects on ocean’s alkalinity. For example, the initiation of Neoproterozoic snowball-Earth events has been attributed to enhanced alkalinity generated via microbial sulfate reduction and iron reduction [7].

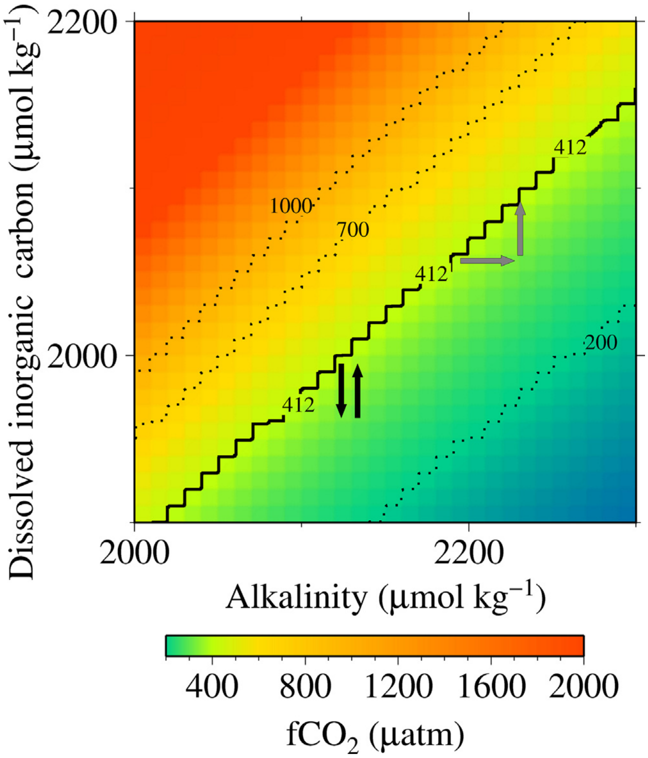


Fig. 1. Dependence of the surface water fugacity of CO_2 (fCO_2) on DIC and alkalinity. The schematic depicts the two fundamental mechanisms to increase the dissolved inorganic carbon, i.e., CO_2 reservoir in marine waters. Arrows indicate the responses to deliberate and natural manipulations. Black arrows represent a decrease in DIC, for example by photosynthetic CO_2 fixation and persistent removal (e.g., burial). The respective CO_2 amount will be replenished by the atmosphere at given atmospheric conditions, the DIC-alkalinity equilibrium conditions will remain unaltered. Grey arrows represent an increase in alkalinity, which allows for the water to hold more DIC at given atmospheric CO_2 conditions. The CO_2 for re-equilibration will be supplied from the atmosphere, the DIC-alkalinity conditions will equilibrate at a higher DIC level.

However, enhancing alkalinity by means of natural processes (either biotic or abiotic) over geological time scales is too slow to meet our carbon neutrality needs given the current speed and magnitude of CO_2 emissions from human activities. Thus, deliberately increasing the alkalinity of seawater to enhance the capacity of ocean carbon sequestration offers a feasible option [8,9].

One way to increase alkalinity is to add alkaline minerals such as olivine to the ocean, the hydrolysis of which leads to the formation of serpentines and release of OH^- [10]. This serpentinization-produced alkalinity transforms CO_2 to the aqueous form of HCO_3^- at high pH, creating favorable conditions for MICP, which can be ecologically engineered for permanent CO_2 sequestration in the ocean floor (see below). It should be noted, however, that collateral effects may arise from artificial mineral-induced ocean alkalinity [11] as the addition of either dissolved or solid (powder) compounds to the ocean may not guarantee a net carbon sink and may cause unforeseen complications in the marine ecosystem. Thus, a comprehensive assessment of ocean alkalinity engineering [12] must be performed before its actual deployment.

2.2. Increasing negative emissions through integrated BP, MCP, and MICP

The BP exports a large amount of particulate organic carbon (POC) below the marine euphotic zone (~15% of the total carbon fixed by photosynthesis), but only less than 1% of the POC can reach the seabed for burial [13]. A unique aspect of the BP is that phytoplankton can accelerate the export of excess dissolved carbon contained in alkalinized waters by the production of organic acids and chelating agents such as transparent exopolymer particles. These compounds facilitate bridging between particles and thereby assist the formation of aggregates and enhance their settling velocity [14]. Experimental data show that the increase in $p\mathrm{CO}_2$ over the next century will facilitate the production of transparent exopolymer particles in the sunlit ocean (e.g., [15]). This part of BP process can be linked to MCP (see below) to allow aggregation of recalcitrant dissolved organic carbon (RDOC) molecules [16] with the sinking transparent exopolymer particles. Similarly, natural dust deposition in the ocean, for example underneath the Saharan dust plume, is thought to act as a ballast for the BP leading to higher efficiencies in organic carbon export [17] though the factors that create high export regimes on a global scale are poorly understood [18]. Soot deposition, such as produced by biomass and fossil fuel burning of human activities, is likely to enhance the BP by stimulating aggregation [19] and is expected to increase in the future [20].

MCP was proposed as a new theoretical framework emphasizing the contribution of marine microbes to RDOC production via their orchestrated metabolic activities: (1) the modification and secretion of RDOC during microbial cell growth and metabolism, (2) the release of RDOC during the lysis of host cells by viruses, and (3) the release of RDOC by predators such as protozoa feeding on microbial cells [21,22]. Different from the sinking process driven by BP, the MCP is a microbially-mediated DOC transformation process, in which the marine microbes can readily metabolize the labile DOC, but not the RDOC. The remineralization efficiency of RDOC is only about 0.1% that of labile DOC [23], leaving the majority of DOC stored in the ocean in the form of RDOC [21,22]. This MCP-driven RDOC accumulation in the ocean interior has thus played a significant role in marine carbon cycling and potentially combating global climate change [24].

MICP can further process the sinking transparent exopolymer particles and other organic matter including RDOC to drive carbonate precipitation in the seafloor sediment. Sulfate-reducing bacteria, halophiles, methanogens, and some planktonic microbes and associated viruses living in the water column, have been experimentally demonstrated to be capable of mediating carbonate mineral formation [25,26]. This process had been observed to serve as an important mechanism for massive carbonate precipitation from the seawater during the Proterozoic and Permian-Triassic dolomite formation [27,28]. The dolomites ($\mathrm{CaMg(CO_3)_2}$) first precipitated from seawater as nonstoichiometric and disordered protodolomite through microbial mediation. MICP also played a fundamental role in the formation of ancient stromatolites, which occurred abundantly in the Precambrian and some critical periods of the Phanerozoic [29]. Stromatolites consist of alternating layers of benthic algae/cyanobacteria and calcium carbonate and have a high potential for permanent $\mathrm{CO_2}$ sequestration in shallow marine sediments.

Another important setting for MICP is ocean bottom cold seeps along continental margins where anaerobic oxidation of methane by archaea was coupled to bacterial sulfate reduction [30]. This microbial process can either increase the alkalinity in sediments or provide more free-carboxyl groups that can favor the dehydration of $\mathrm{Mg[H_2O]_6}^{2+}$, and effectively bind cations to facilitate the equal incorporation of Mg^{2+} and Ca^{2+} into the precursive crystal of dolomite [26]. Massive carbonate deposits have been observed in modern cold seeps in the Gulf of Mexico and the South China Sea, as well as in ancient geological formations [31,32].

The continuum of BP-MCP-MICP activities, however, requires full understanding of their coupling mechanisms at the molecular and genetic levels so that their synergy can be best harnessed for effective negative emissions in a given ecosystem.

2.3. Increasing negative emissions by means of artificial upwelling

Phytoplankton carbon fixation takes place in the euphotic zone and often leads to depletion of nutrients in surface ocean. While deep waters are nutrient-replete due to re-mineralization of sinking particles and reduced biological utilization, warming-intensified stratification of the water column blocks the vertical supply of nutrients from the deep ocean to the surface layer by preventing water mixing, limiting carbon fixation by phytoplankton. This problem can be resolved by artificial upwelling. The idea is supported by an investigation of the 2018 Kilauea eruption in Hawaii [33]. Despite the deposition of potentially lethal lava material into the ocean, a phytoplankton bloom occurred within several days of the eruption. Analysis of this situation revealed that the sinking lava heated deeper waters sufficiently to cause an upwelling of these nutrient-rich waters and triggered a phytoplankton bloom [33].

Artificial upwelling theoretically can be manipulated by providing just the amount of nutrients needed for normal photosynthesis (i.e., no algal blooms). The system can be operated during daytime only to bring oxygen-saturated surface water down to lower depths by compensational mixing, and high-DIC and nutrient-rich water from lower depths up to the euphotic zone to fuel carbon fixation. Unlike iron and nutrient fertilization, artificial upwelling can be tuned to adjust nutrient supply and demand within the ecosystem [34]. It is an ecological regulation inside the ecosystem rather than perturbation of new nutrients from outside, which is expected to enhance production of organic matter in the euphotic zone and mitigate formation of hypoxia and nutrient “bombs” in bottom water. Although artificial upwelling is still in its early development, a coastal ocean observatory network and the deployment of sophisticated devices such as the biogeochemical argo floats may eventually allow us to use artificial upwelling to balance nutrient stoichiometry (e.g., Redfield ratio) in the future.

Nevertheless, research into artificial upwelling systems is confronted with challenges. One of them is that the equipment must operate continuously in the complicated and variable marine environment of fluid mechanics [35]. The artificial upwelling system developed in China uses self-sufficient energy and injects compressed air to accelerate the migration of deep ocean water to the euphotic zone [36,37]. Several trials have been carried out, showing that the device can be employed to enhance primary productivity.

The application of artificial upwelling systems (especially at large scales) could cause obvious disturbances to the ocean environment and potentially impact the ocean ecosystem [16,34]. Therefore, it is necessary to firstly deploy a test-and-trial artificial upwelling platform at sea to study the long-term impact of the artificial upwelling system on the marine environment. In particular, it is necessary to study the relationship between the physical operation of artificial upwelling and nutrients, primary productivity, dissolved oxygen content, pH, and $\mathrm{CO_2}$ air-sea interaction, as well as to evaluate the subsequent POC export and RDOC production in order to improve the capacity of ocean carbon storage [16,38]. As often encountered with other approaches, the exchange of other greenhouse gases (especially $\mathrm{N_2O}$) with the atmosphere must also be considered, which may have important roles in the carbon budget.

2.4. Increasing negative emissions through habitat restoration of coastal wetlands

Coastal wetlands (e.g., mangroves, salt marshes, seagrass beds) are considered as important habitats for negative emissions. Despite covering only 0.2% of the ocean surface, coastal wetlands contribute 50% of carbon burial due to the large carbon sequestration potential by soil accretion, and high carbon sequestration efficiency, holding the potential for reducing greenhouse gas emissions and increasing carbon sinks [39,40]. In particular, mangroves in coastal wetland have extremely high productivity and may play a vital role in climate change mitigation due to their efficiency as a carbon sink [41]. However, this high productivity also creates biogeochemical hotspots of enhanced microbial organic carbon oxidation mediated by different electron-accepting pathways [42,43]. In addition, RDOC and total alkalinity in mangrove wetlands may be horizontally transferred to the ocean [40,44]. As a result, the ultimate output of wetland negative emissions will depend on the effectiveness of organic carbon burial, transformation of labile DOC into RDOC, and conversion of respired CO_2 into solid forms of carbonate through MICP.

Recently, large areas of natural coastal wetlands have been damaged due to anthropogenic activities and sea leave rise [45], begging for urgent habitat restoration in order to make coastal wetlands function effectively for negative emissions. It needs to be noted that the land is used for many other purposes, which may limit the wetland's capacity for climate mitigation (see Section 3.4). Thus, comprehensive understanding and carbon budgeting are needed to verify any negative emissions from wetlands (e.g., [46,47]).

2.5. Increasing negative emissions through habitat restoration of coral reefs

Coral reefs constitute a significant component of carbon pool with potential for negative emissions in the marine ecosystem. The estimated global production of coral reefs is $\sim$0.9 Gt C/a [48], with the primary productivity (300–5000 g C/(m^2 a)) one order of magnitude higher than oceanic, non-reef ecosystems (50–600 g C/(m^2 a)) [49].

It is generally accepted that the relative contribution of the net organic and inorganic carbon production determines whether coral reefs are a source or sink of atmospheric CO_2. Because of the variations in local physicochemical parameters (e.g., reef morphology, water residence time, proximity to land, terrestrial nutrient input, etc.), the air-sea CO_2 fluxes, estimated using direct measurements of air–sea CO_2 partial pressure difference or community metabolism based on seawater carbonate chemistry, are not always consistent, leaving open a long-standing CO_2 "source or sink" debate [50–53]. Thus, the contribution of coral reef ecosystem to CO_2 sea-air exchange on a global scale needs to be better investigated.

What is promising is the rapid development of new tools and technologies powering research on the ecological health of coral reefs and their carbon "source-sink" effects. For example, compound-specific isotope analysis (such as δ^{13}C of amino acids or lipids) can be used to trace carbon flow and energy transfer in the food webs, effectively resolving the relationship between coral's trophic strategy and the carbon "sink-source" attribute of the reef ecosystem [54]. In addition, the carbon fingerprints of coral symbiosis can be traced and quantified *in situ* at the subcellular scale with the nano secondary ion mass spectrometry. In particular, the relative contributions of coral calcification, carbon fixation by zooxanthellae, and microbial metabolism to the carbon budget can be characterized more precisely [55].

3. ONCE practical implementations

3.1. Coordinated land–ocean eco-engineering to achieve negative emissions in coastal waters by reducing terrestrial nutrient discharge

According to the Food and Agriculture Organization (FAO) of the United Nations, global fertilizer consumption has continuously increased in recent decades, e.g., from less than 1800 million tons in 2000 to 2600 million tons in 2015, a nearly 50% increase. The amount of fertilizers applied in China has increased nearly 30 times over the past 50 years, reaching a record high of 600 million metric tons in 2015 (National Bureau of Statistics of China). Excessive use of fertilizers not only alters the cohesive soil structure, but is also one of the major causes of environmental problems such as eutrophication in coastal waters [56,57].

Excessive input of terrestrial nutrients and organic matter can also affect the capacity of carbon storage in estuaries [58]. Investigations have shown that accumulated nitrogen derived from human activities is inversely correlated to organic carbon in a variety of natural environments [59], suggesting that excessive nutrients reduce the environmental capacity to retain organic carbon. This notion has been confirmed through experimental addition of excess nutrients in riverine, estuarine, shelf and offshore waters [60,61].

In shallow estuaries, tidal wave-induced resuspension processes intensify the interaction between the surface and the bottom layers of water, thus reducing the net downward carbon flux [62] and enhancing POC remineralization [63]. Under such situations, the efficiency of the BP is often low even for a highly productive coastal region. On top of that, the labile organic carbon from photosynthesis has priming effects on relatively recalcitrant terrestrial organic carbon [16,64,65], reducing the MCP efficiency. Therefore, reducing nutrients to a certain level is vital for efficient BP and MCP machineries to act in concert [16] in estuaries while achieving additional benefits such as slowdown or prevention of water deoxygenation.

The above goal can be accomplished by eco-engineering through coordinated land–ocean management strategies. In short, based on the results of large-scale spatio-temporal ecological surveys and simulation experiments, the main ecological processes driven by MCP and BP (e.g., primary productivity, heterotrophic respiration, grazing and growth, etc.) can be identified, and their relationship to nutrients and other environmental factors can be established.

Using statistical modeling tools, the nutrient concentration threshold can be inferred to implement public policies aimed at controlling the terrestrial nutrient discharge. Meanwhile, protocols for on-site tracking and monitoring of biotic and abiotic environmental parameters, such as sedimentation rate, RDOC production rate, RDOC to labile DOC ratio, can be used for the quantification and verification of marine carbon negative emissions. All these controls will be integrated to guide the practice of ecological engineering of the MCP- and BP-based marine negative emissions. Such analyses will also provide quantitative estimations for ecological compensation policy, which will in turn favor the practice of reducing terrestrial nutrient discharge to enhance negative carbon emission in the sea.

3.2. Eco-engineering of integrated BP, MCP and MICP in hypoxic and anoxic zones

Based on the above understanding (Section 2.2 in particular), a theoretical mechanism may be proposed to effectively draw down atmospheric CO_2 through integration of the BP, MCP and MICP (Fig. 2) in coastal waters including eutrophic areas or aquaculture regions. The proposed mechanism is based on the capability of charge-modified olivine and clay in mixture to absorb the microalgae grown in surface ocean, causing them to flocculate and settle rapidly. This process would reduce the residence time of algal biomass in the water column, which may be respired by heterotrophic organisms, leading to oxygen depletion and anoxic conditions in the bottom water.

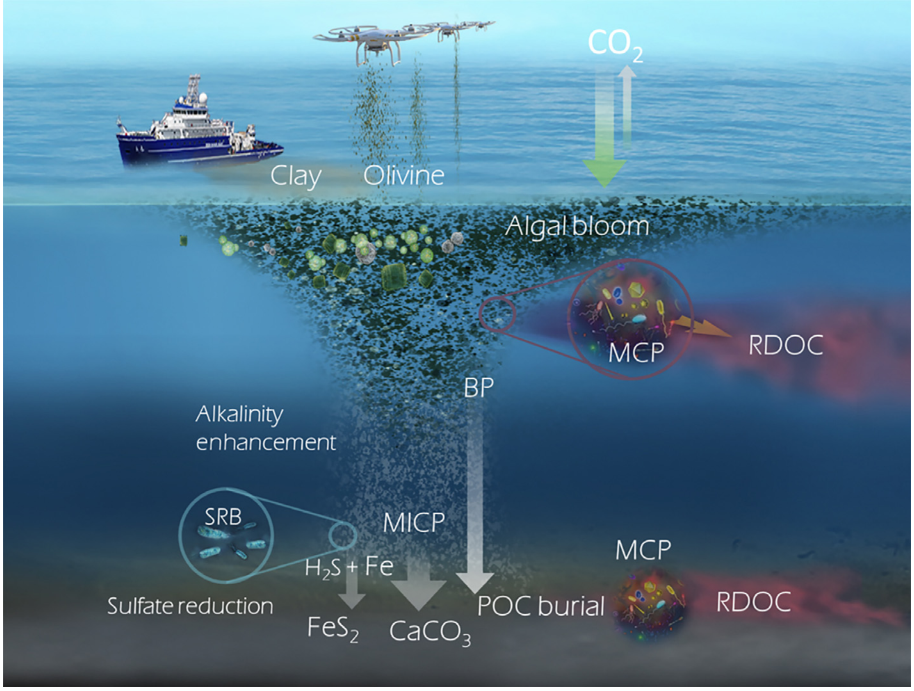


Fig. 2. A conceptual model of microbially induced carbonate precipitation (MICP) integrated with engineered CO_2 fixation by microalgae. Biological pump (BP) would be enhanced by the charge-modified olivine and clay in mixture to absorb the microalgae grown in the surface ocean, causing them to flocculate and settle rapidly for burial as particulate organic carbon (POC) in the sediment. Microbial carbon pump (MCP) in the water column or sediment porewater would function in converting labile organic carbon into recalcitrant dissolved organic carbon (RDOC). Alkalinity enhancement in the water column may be achieved through hydrolysis of olivine. MICP may be accomplished in anoxic sediment via sulfate reduction coupled to bicarbonate production.

Clay-bond olivine particles can also cover the algal biomass on the seafloor, allowing the anaerobic reduction of sulfate to occur and increase the alkalinity of the sediment porewater, which would favor the formation of calcium carbonate. H_2S gas generated by sulfate-reducing bacteria can react with the reduced iron in olivine to form pyrite (FeS_2), preventing H_2S from reacting with O_2 at the water–sediment interface to produce acidity. Furthermore, hydrolysis of olivine can also produce alkalinity, which may further strengthen the alkaline environment for carbonate precipitation (Fig. 2).

While this scheme is theoretically feasible, it has not been practically tested. In particular, stimulating sulfate reduction may have complicated impacts on ecological and biogeochemical processes of the benthic environment [66]. Thus, experiments and quantitative assays are needed to examine the chemical and mineralogical dynamics associated with the production of alkalinity through olivine hydrolysis and nucleation of carbonate crystals at supersaturation of ions in microbial cultures.

Models of MCP-MICP interactions may need to be established under controlled experimental conditions (CO_2 concentration, light intensity, sediment mineral composition, vertical redox gradient) for evaluating the carbon storage capacity and efficiency of MCP-MICP in a natural seabed environment. This may be complicated by oceanic processes such as water current variation, tidal cycles, as well as wind directions, all of which may impact the chemical conditions for MICP to occur at the bottom of the ocean. However, these uncertainties can be quantitatively evaluated by establishing a coastal water testbed supplemented with sophisticated monitoring in a well constrained environment.

3.3. Applications of artificial upwelling in seaweed cultivation and coral reef ecosystem

The global annual production of seaweeds was approximately 27.6 million tons in 2016 (FAO, 2016). Aquaculture is often blamed for its negative impacts on environments despite its critical contribution to food security [67]. Indeed, considering the seaweed cultivation alone, about one third of the photosynthetic products are released into the water as DOC or detritus [38] and decomposed rapidly, re-releasing as nutrients while consuming oxygen. Such successive processes ultimately lead to oxygen depletion and nutrients accumulated in bottom water as so called “nutrient bombs” [34]. Once the “nutrient bombs” are brought up to the surface water by perturbations such as storms, they trigger intense algal blooms [34,68].

Artificial upwelling may be a feasible approach to facilitate aquaculture operation in a context of sustainable ocean development. Artificial downwelling has additionally been advocated as a means to mitigate coastal hypoxia [69]. The combination of the two artificial processes in coastal waters or aquaculture areas has the potential to enhance production of organic matter in the euphotic zone and mitigate the formation of hypoxia and “nutrient bombs” in bottom water.

Artificial upwelling may also be applied to the coral reef ecosystem. On a natural coral reef island, interesting phenomena associated with upwelling of bottom waters were observed. On the reef side directly affected by upwelling, corals exhibited lower likelihood of bleaching and higher probability of recovery compared with corals on the side without the influence of upwelling [70], suggesting that applying artificial upwelling may help protect coral reef ecosystems whilst simultaneously promoting other carbon sinks.

3.4. Application of MCP and MICP in coastal “blue carbon” habitats

Blue carbon is defined as the carbon stored in coastal and marine ecosystems such as mangroves, tidal marshes and seagrass meadows, which sequester more carbon per unit area than terrestrial ecosystems (e.g., forests). Applications of MCP and MICP to such ecosystems are expected to mitigate climate change and enhance ecosystem functioning for negative emissions. Here, we propose a conceptual model (Fig. 3) based on previous studies and our current knowledge [21,71,72], which may guide us in using coastal wetlands as an effective negative emission system. The leaf and root litters of mangroves as well as root exudates are decomposed by microbial communities to form the microbial biomass [71]. Also, microbial necromass directly contributes to stable organic carbon, and can account for 30%–60% of soil organic carbon [73]. Furthermore, mangrove sediment microbial communities could convert labile organic carbon into RDOC [21], and anaerobic methane oxidation may accelerate carbonate precipitation [32,74,75]. Therefore, the mechanisms of microbial necromass production by BP, RDOC transformation by MCP, and carbonate precipitation by MICP are integral to carbon sinks in mangrove wetlands.

The full potential of blue carbon habitats for mitigation of climate change has not been well explored in vegetated coastal ecosystems including mangroves [76–80]. Also, it is noted that a competition between maintaining blue carbon ecosystems and food production may become severe in the future given human population increase and the projected increase in food requirements. Promotion of blue carbon practices may conflict with the sustainable development goal of "zero hunger" (https://www.un.org/sustainabledevelopment/hunger/). Therefore, blue carbon practices advocating for changes to land use should consider the net impacts on food security. For example, in many densely populated nations such as Germany, coastal waters are fully, if not over, exploited, such that no space for blue carbon measures can be generated without severe conflicts with other uses. However, conflict also creates opportunities. For example, wind farm platforms are regularly serviced areas that may provide a physical grid onto which marine geo-engineering approaches could be "mounted", highlighting the need for carefully designed co-utilization practices (marine protected areas or areas assigned to wind energy generation, Fig. 4). These potential conflicts or uncertainties demand creative approaches in utilizing blue carbon habitats for ocean negative carbon emissions to achieve carbon neutrality.

3.5. Turning coral reefs from "sources" to "sinks" of atmospheric CO_2

The global coral reef crisis reflects climate change and anthropogenic activities, in parallel with the poor outcomes of traditional management methodologies [81]. This has led to the development of active restoration approaches. One of the most widely used tactics is based on the "coral gardening" tenet, where coral colonies are farmed in designated underwater coral nurseries, preferably floating mid-water nurseries (Fig. 5), and then transplanted onto degraded reef areas [82–84]. Together with the application of the gardening tenet, farmed corals within the nurseries could also be used to optimize carbon sequestration [85] by employing silviculture concepts in coral transplantation [86].

Nursing coral colonies in floating reef nurseries has become a common practice [82,87]. Farming of stocks of corals can be easily adopted as a novel approach under the ONCE tenet. Newly established coral nurseries possess almost zero carbon as the starting coral nubbins are only a few mm each in dimension, but could develop within a year to about 10 cm in size (e.g., the branching coral colony *Stylophora pistillata*). The average dry weight of such a colony is 250 g, equivalent to 110 g CO_2. A 10 m × 10 m floating nursery may contain 10,000 colonies, or 10^6 coral colonies per 1 km^2 of coral nursery, equivalent to 110 t of sequestered CO_2 per year. Eventually, the mid-water nurseries develop into floating reef ecosystems (Fig. 5), with a continuous flux of propagules, algae, fish and invertebrates that arrive, settle and become part of the floating reefs [88]. The photosynthetically fixed carbon translocated to the coral skeletons is about 10% of the carbon stored in animal tissues and algal cells [89]. A systematic analysis of coral reef biogeochemical processes, including community

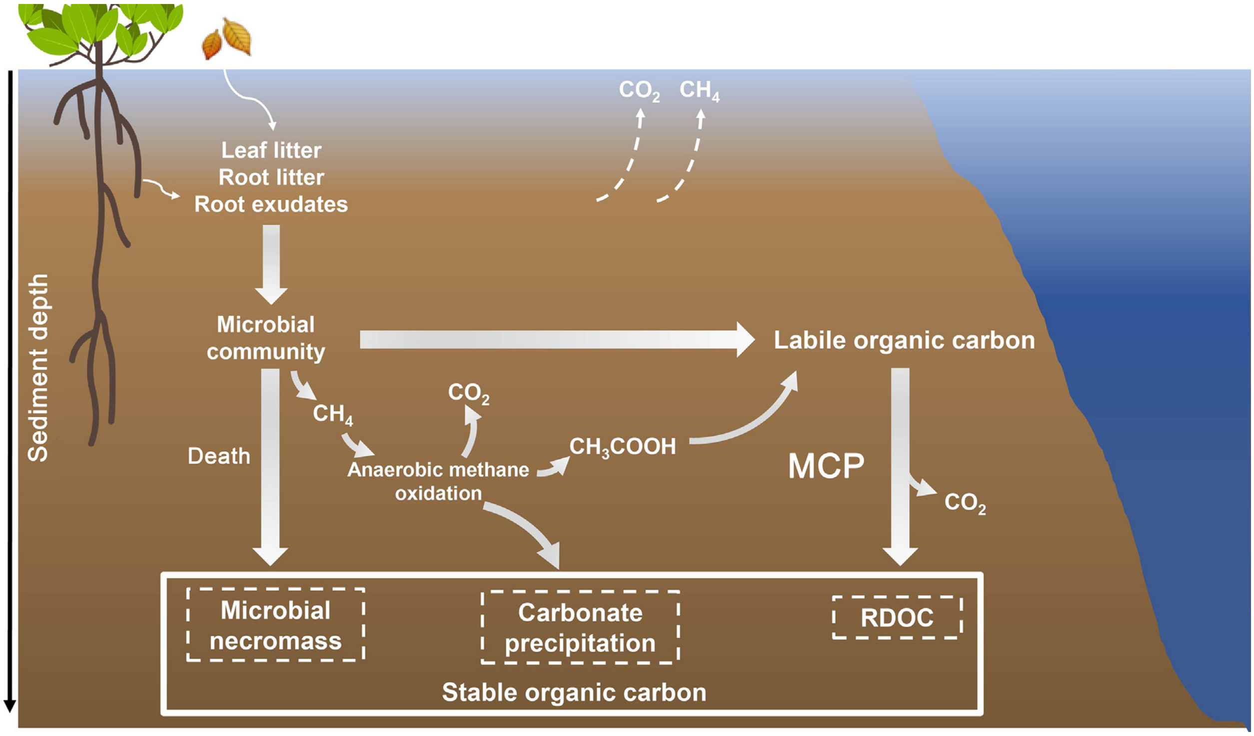
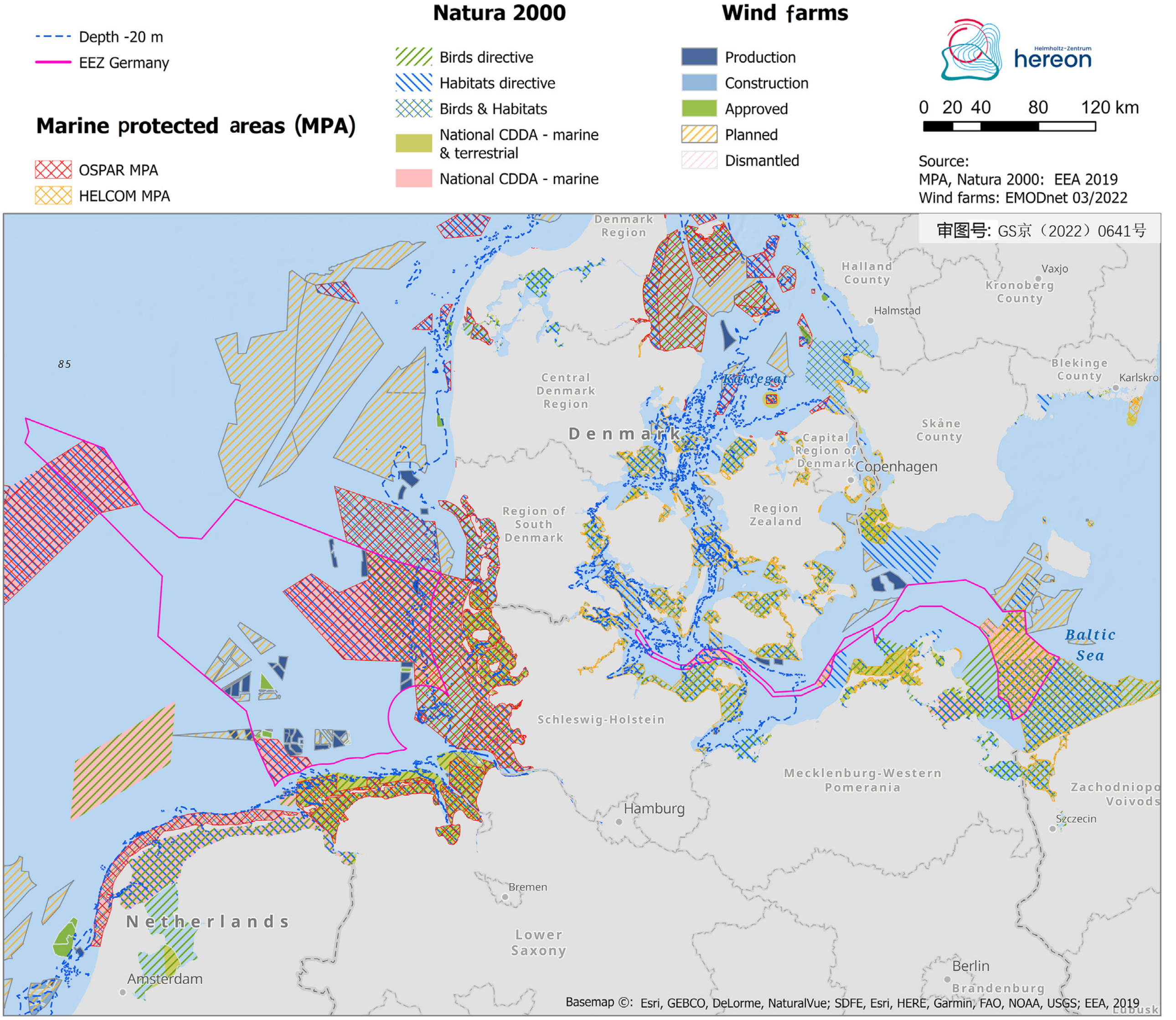


Fig. 3. A conceptual mode for microbially driven carbon sink formation and transformation in mangrove wetlands. The possible carbon sinks in mangrove wetlands include microbial necromass production by biological pump (BP), RDOC transformation by microbial carbon pump (MCP), and carbonate precipitation by microbially induced carbonate precipitation (MICP).



Source:
MPA, Natura 2000: EEA 2019
Wind farms: EMODnet 03/2022

Basemap ©: Esri, GEBCO, DeLorme, NaturalVue; SDFE, Esri, HERE, Garmin, FAO, NOAA, USGS; EEA, 2019

Fig. 4. Selected uses of the German Exclusive Economic Zone (EEZ), enclosed by the purple line, denoting Germany's territorial waters. Please note that shipping lanes are not shown. The dashed blue line denotes the 20 m isobaths. Windfarms are areas assigned to power generation using wind energy, with different status of the individual "farms" indicated. Natura 2000 indicates different nature protection areas. Marine protected areas (MPA) are officially protected space by the respective regulatory bodies for the North Sea (OSPAR), and the Baltic Sea (HELCOM). Graphics: Hereon/Ulrike Kleeberg.

metabolism, organic production, calcium carbonate precipitation and RDOC export in the long run could showcase that carefully tended coral reefs can be a sink of CO_2.

Such practice carries additional benefits in tradable goods and services that enhance its applicability. The benefits include the maintenance and even enhancement of reef biodiversity, the possible extraction of pharmaceuticals and other commercially valuable compounds and materials from reef organisms, enhanced fisheries, sustainable green economy-tourism, new jobs, and not least an extensive source material for coral reef restoration [90–92].

3.6. Establishing a framework of marine carbon fingerprints and footprints at both the micro- and macro-levels

To date, the marine carbon sink has not been included in the carbon trade system due to difficulties in tracking and accounting for the budgets, fluxes, and incremental additions of carbon. A clear example of a critical but poorly defined budget component is the missing RDOC in the coral reef, aquaculture and coastal blue carbon systems. Additionally, quantifying the origin and the fate of organic carbon compounds in the variable marine environment is challenging, which necessitates the establishment of a framework of marine carbon fingerprints and footprints at both the micro- and macro-levels.

Carbon fingerprints provide mechanical insights into the biogeochemical processes and metabolic products of carbon cycling driven by microbial communities at the genetic and molecular (chemical) levels, whereas ecosystem-level footprints calculate total carbon emissions throughout specific processes. Two types of fingerprints can be implemented: one is the genetic fingerprint that characterizes the taxonomic composition and functional processes of microbial communities driving carbon cycling [93]; the other is a molecular chemical fingerprint that characterizes the form of organic production in the ocean [94]. In particular, RDOC can be measured or evaluated by using Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) that is capable of assessing the ageing of DOMs such as the carboxyl-rich alicyclic molecules (CRAMs) widely existed in the deep ocean ([95]; see review [23] for other methods of quantifying RDOC). By linking genetic fingerprints with chemical fingerprints, models can be built and trained to predict the microbially mediated carbon storage potential in ocean ecosystems, supporting ocean carbon negative emissions at genetic and molecular chemical levels.

Fig. 5. A submerged floating coral nursery in Eilat, Israel (6 m below the water surface, 20 m above the substrate), in blue, oligotrophic waters, several kilometers away from the closest natural coral reef. Farmed *Acropora* spp. colonies attract a wide range of marine life from the plankton, fish and invertebrate eggs and larvae that settle and grow, transforming the floating coral nursery into an oasis in blue waters. Part of the nursery bed is exposed (the right side of the figure) to reveal the initial status. All farmed corals started from coral nubbins (fragments of about 0.5 cm in size). Photographed by S. Shafir.

Advanced FT-ICR-MS technologies have enabled us to identify millions of DOM molecules from the marine environment, which have molecular weights ranging from a few hundred to 100,000 Da [68]. These DOM compounds can be clustered into thousands of molecule groups in FT-ICR-MS profiles, leading to the establishment of a molecular database based on reference RDOC samples [24]. Second, characterizing the complex microbial processes driving carbon cycling and RDOC production has enabled us to decipher complex links between microbial communities and DOM compositions using methods like co-occurrence networks [94,96]. In addition, other approaches such as high throughput functional gene arrays [97,98] can be developed and considered for standard assessment of microbially mediated carbon cycling in the ocean. Characterizing the micro-fingerprints and macro-footprints is essential for carbon storage estimation and calculation, with quantitative molecular indices being used to underpin potential carbon trade.

4. Conclusion and outlook

Reaching carbon neutrality is a goal that we cannot afford to fail. Yet, carbon neutrality via negative emissions is plagued by uncertainties in carbon capture and carbon storage [99] as well as blue carbon science [77]. These issues must be considered in any action planned or to be taken. The ocean has a huge potential for CO_2 removal from the atmosphere and the underlying science of carbon fluxes through the ocean system must be fully explored to help achieve that goal. Ocean-based carbon neutrality is possible only when carbon storage is achieved and verified through sustainable ecological engineering processes.

The mechanisms for CO_2 removal reviewed here are mechanistically viable and accomplishable, based on field observations and sound scientific research. However, there will be uncertainties and potential risks when applying natural principles to engineering practices. Thus, careful testing and validation of any approach must be carried out before measures are executed at scale. The comprehensive approaches proposed in this review aim to help reach carbon neutrality, focusing not only on increased carbon sequestration itself but also to the alleviation of other environmental stresses such as eutrophication, red tides, green tides, anoxia, and acidification. We believe that approaches with multifaceted advantages will help cope with climate change and support the vision of the United Nations Decade of Ocean Science for Sustainable Development (2021–2030) that emphasizes "the science we need for the ocean we want" (https://en.unesco.org/ocean-decade).

As an international program, the purpose of the ONCE project is to promote coordinated efforts in developing sustainable methodologies focused on ecologically sound engineering approaches. All cutting-edge ecological/biogeochemical manipulations to support ONCE must be carefully tuned to fit into the framework of BP, MCP and MICP for maximum sequestration of carbon in inorganic/organic, biotic/abiotic, and particulate/dissolved forms. Combined with experimental simulations and field studies, the ONCE program is expected to lay the foundation for a variety of negative emission models and measures with quantifiable, reproducible, and verifiable features that can be used for carbon trade in the near future. Global efforts in a cooperative and collegial spirit are required to make the ONCE program effective in serving the grand goal of carbon neutrality.

Conflict of interest

The authors declare that they have no conflict of interest.

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Author contributions

Nianzhi Jiao conceptualized the study and led the discussion of the review. Chuanlun Zhang, Nianzhi Jiao, and Tuo Shi drafted the manuscript and the response letter to reviewers' comments, with contribution from all the other co-authors. All authors read and approved the final version of the manuscript.

Appendix A. Supplementary materials

Supplementary materials to this review can be found online at https://doi.org/10.1016/j.scib.2022.11.016.

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