An Analysis of Ocean Alkalinity Enhancement Models and Their Limitations.

by

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We acknowledge and respect the Lək̓ʷəŋən (Songhees and Xʷsepsəm/Esquimalt) Peoples on whose territory the university stands, and the Lək̓ʷəŋən and W̱SÁNEĆ Peoples whose historical relationships with the land continue to this day.

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Abstract

As atmospheric carbon dioxide (CO_2) levels continue to rise, increasing attention is being given to exploring mitigation techniques that could potentially enhance the natural drawdown of CO_2. One such mitigative intervention is ocean alkalinity enhancement (OAE), which has drawn the scientific community's attention over the past decade, as a natural form of carbon capture and a hopeful solution to the excess carbon dioxide in our atmosphere. OAE involves dissolving alkaline materials into ocean surface waters to increase its natural CO_2 buffering capacity. Limestone and lime have received plenty of attention given their widespread availability. Here, the following paper will address the order one policy-relevant question of whether OAE represents a viable CO_2 removal solution to global warming.

The UVic Earth System Climate Model (ESCM) was used to explore the potential of OAE interventions under representative concentration pathways (RCP) 2.6, 4.5, 6.0, and 8.5. For each RCP, three OAE interventions were implemented. First, it was assumed that the global annual production of limestone was crushed, uniformly distributed across the surface waters of the global ocean, where it immediately dissociates in the surface water. Second, it was assumed that the global production of limestone was converted to lime, with the CO_2 released in this process being added to the atmosphere. In the third intervention, the second intervention was repeated, though sequestering the CO_2 arising from lime production. Although lime and limestone OAE have been put forward as major solutions for carbon dioxide emissions, the results show that despite slightly raising pH, lowering CO_2 concentrations and lowering air surface temperatures, the change is small, suggesting that OAE interventions have little potential for mitigating global warming.

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List of Abbreviations

ABMT	area-based management tools
AOA	artificial ocean alkalinization
BBNJ	biological diversity beyond national jurisdiction
BECCS	biomass energy with carbon capture and sequestration
BFM	biogeochemical flux model
CB&TMT	capacity-building and the transfer of marine technology
CCS	carbon capture and storage
CCT	carbon capture technology
CCUS	carbon capture utilization and storage
CDR	carbon dioxide removal
COA	coastal ocean alkalinization
DAC	direct air capture
DACCS	direct air carbon capture and storage
DIC	dissolved inorganic carbon
DOR	direct ocean removal
DWT	deadweight tonnage
EIA	environmental impact assessments
EMT	emissions mitigation technique
ESCM	Earth System Climate Model
ESM	Earth System Model
GHG	greenhouse gas
HAMOCC	HAMburg Ocean Carbon Cycle Model

IPCC	Intergovernmental Panel on Climate Change
MPI	Max Planck Institute
NEMO	Nucleus for European Modelling of the Ocean
NET	negative emissions technologies
NPP	net primary productivity
NRCAN	Natural Resources Canada
NSERC	Natural Sciences and Engineering Research Council of Canada
NTA	normalized to a
OA	ocean alkalinization
OAE	ocean alkalinity enhancement
OIF	ocean iron fertilization
RCP	representative concentration pathway
SAT	surface air temperature
SRM	solar radiation modification
TA	total alkalinity
USA	United Stated of America
USD	United States dollar

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Dedication

To my parents, whose unwavering love, encouragement and support made this journey possible. Your belief in me never wavered, even in moments when I doubted myself. The values you instilled and the strength you gave me will guide me far beyond this achievement, and for that I am eternally grateful.

Chapter 1: Introduction

1.1 Climate Change

In 1990, the Intergovernmental Panel on Climate Change (IPCC) published its first Scientific Assessment of climate change (IPCC 1990). At the time, the atmospheric carbon dioxide concentration was 354 ppm, and human activity emitted 6.2 Gt/yr of fossil carbon to the atmosphere. Despite an increased understanding of the causes and profound consequences of unchecked global warming (IPCC 1990; IPCC 1992; IPCC 1995; IPCC 2001; IPCC 2007; IPCC 2013; IPCC 2021), global fossil carbon emissions in 2023 have increased 63% over 1990 levels to 10.1 Gt/yr (Friedlingstein et al. 2023). Compared to the 1750 pre-industrial level of 277 ppm, the atmospheric concentration of CO_2 has risen by 142 ppm as of 2023 to 419 ppm and is growing by about 2.4 ppm/yr (NOAA 2024 Apr 9). In total, human activities have emitted approximately 484 Gt of cumulative fossil carbon emissions over these 273 years (Friedlingstein et al. 2023). This observed increase in well-mixed greenhouse gas (GHG) emissions, specifically CO_2 emissions, is undeniably a result of human activities (IPCC 2023; NOAA 2025), see Figure 1. Between the mid-20th century and the end of it, emissions rose from about 5 to 35 Gt per year (NOAA 2024 Apr 9).

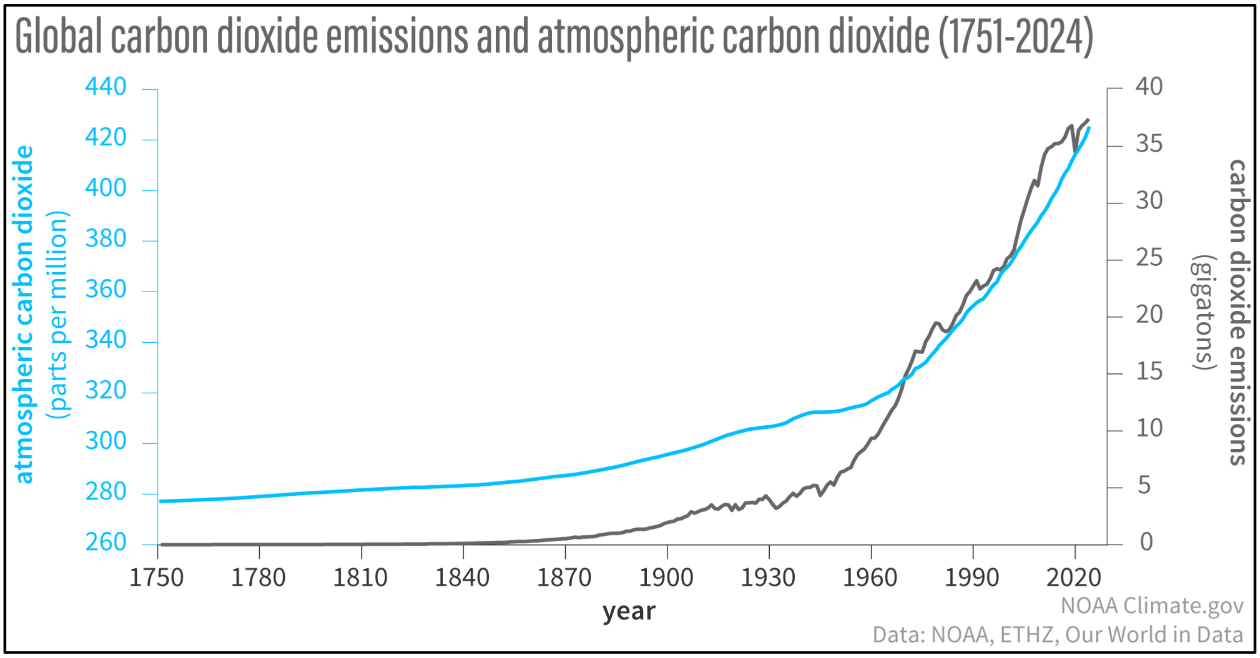

Figure 1: Global atmospheric carbon dioxide compared to annual emissions

Atmospheric carbon dioxide (blue line) increasing along with human emissions (grey line) since the start of the Industrial Revolution in 1750 (NOAA, 2024b)

This impact from human activity is also noticeable when looking at paleo ice core data over the past 800,000 years, in which the rapid increase from the start of the industrial era to 2024 of atmospheric CO_2 is evident (Figure 2) (Lüthi et al. 2008; NOAA 2024 Apr 9).

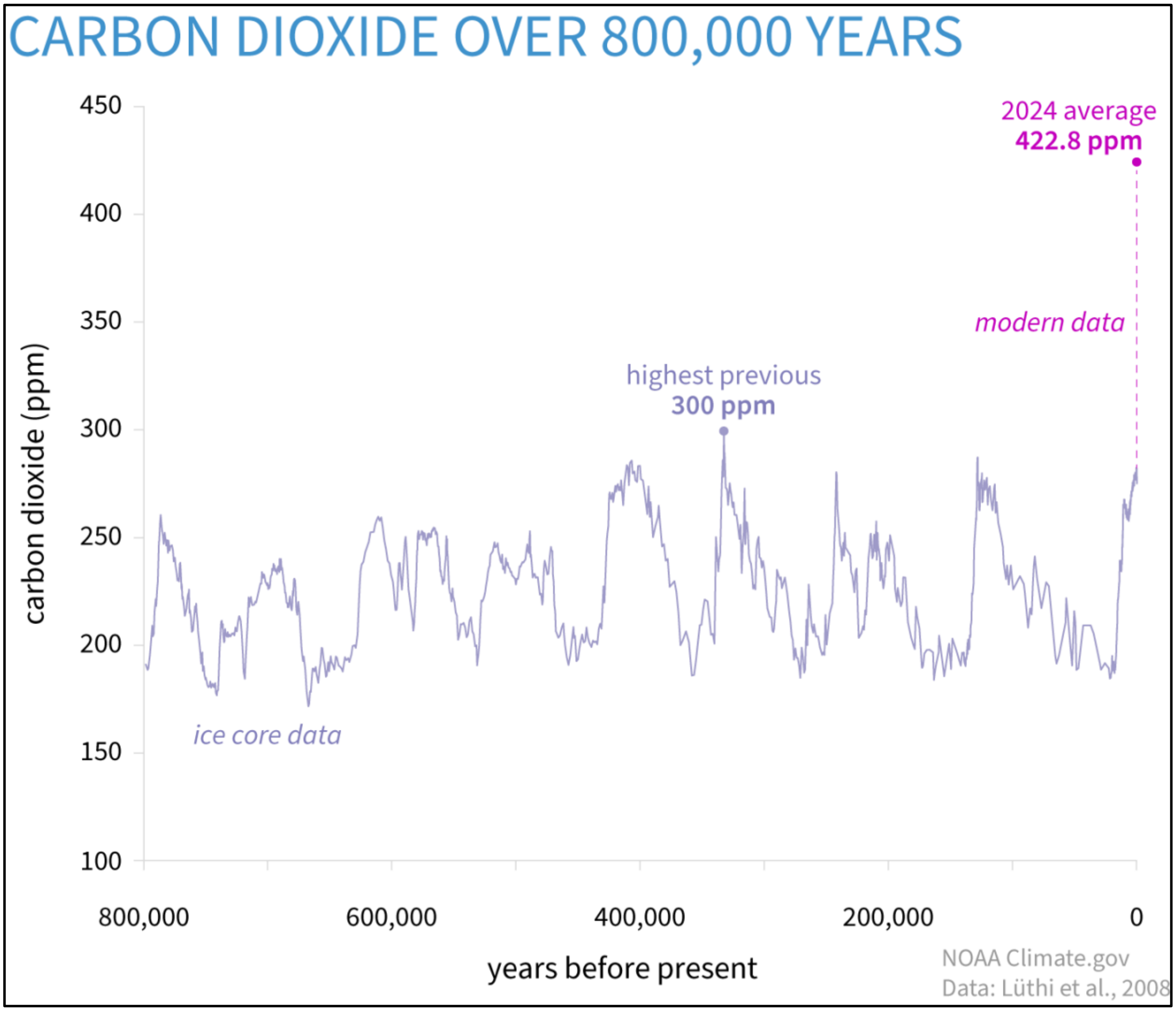

Figure 2: Carbon dioxide over 800,000 years

Atmospheric carbon dioxide (ppm) over the past 800,000 years based on ice-core (paleo) data (light purple line) compared to 2024 concentrations (bright magenta dot) (NOAA, 2024b; Lüthi et al., 2008)

As CO_2 and other GHG emissions increase in our atmosphere, so does our global surface temperature, where comparing the period of 1850-1900 to 2013-2022, we have seen a rise of 1.15 °C globally, 1.65 °C for land temperatures and 0.93°C for ocean temperatures (IPCC 2023). The increase in global surface temperature between 1970 and 2020 is the fastest increase of any other

50-year period over the last 2000 years (IPCC 2023). Suppose we want to keep global warming below a specific temperature. In that case, we can emit only so much carbon (i.e., carbon budget). For every 1000 Gt of CO_2 emitted through human activity, global mean temperature rises by roughly 0.27-0.63°C (IPCC 2023). Relative to 1850-1900 levels, a 1.5°C increase in global warming projects increased warm temperature extremes in all regions, an increase in precipitation, flooding events and frequency especially in the majority of Africa, Asia, North America and Europe and at 2°C, an increase in frequency of agricultural and ecological droughts are projected for Europe, Africa, Australasia, North America and South America (IPCC 2023). Further projected key risks include; 1) effects on food production, biodiversity, ecosystems, mortality and morbidity from heat and infectious diseases for Africa, 2) impacts from sea level rise, changes to water quality and availability and general health and wellbeing for the Mediterranean, 3) water scarcity, coastal flooding, heat stress, effects on mortality and morbidity in Europe, 4) loss and degradation of coral reefs for Australia, 5) impacts on cities and settlements for Australasia, 6) reduced viability of tourism-related activates, increased Lyme disease, reconstruction of infrastructure for North America, 7) sea ice change, effects on sea-ice ecosystems, changes in fisheries catch, losses of key infrastructure, and effects on sea-ice dependant ecosystems in the Arctic and 8) changes in fisheries in Antarctica (O'Neill et al. 2022). Regionally, the threat of more frequent heatwaves and droughts is likely, and other projected regional changes include intensification of cyclones, extratropical storms, aridity and fire weather (IPCC 2019; IPCC 2023). In regions with high temperatures, high vulnerability, or coastline exposure, climate change also threatens cities, settlements, and infrastructure in the long term (IPCC 2023).

As the complex systems of our climate change, so do the risks associated with exposure and vulnerability from climate-related hazards on humans, other species and ecosystems (IPCC 2023).

The global reasons for concern increase with each increment of warming. Likely impacts rise for wildfire damage, permafrost degradation, biodiversity loss, dryland water scarcity, tree mortality and carbon loss for land-based systems and warm-water corals, kelp forests seagrass meadows, epipelagic, rocky shores and salt marshes for ocean/coastal regions (IPCC 2023). Species loss is another projected impact. At the 1.5°C threshold, 3-14% of species will face a high risk of extinction and coral reefs are projected to decline by a further 70-90% (IPCC 2023). As warming increases, so does the irreversible loss of biodiversity in ecosystems such as forests, coral reefs and Arctic regions (IPCC 2023).

The ocean is the biggest carbon sink, absorbing 10 Gt of CO_2 emissions from the atmosphere yearly (Burns and Corbett 2020), and 20-30% of anthropogenic carbon emissions since the 1980s (IPCC 2019; Butenschön et al. 2021; Friedlingstein et al. 2023). The ocean acts as a sink of both CO_2 and heat due to its solubility, heat capacity and inertia (Schuckmann et al. 2020). Studies have estimated that it has taken up around 90% of the excess heat from the climate since 1970 (IPCC 2019; Schuckmann et al. 2020; Butenschön et al. 2021). As a result, of the absorbed anthropogenic emissions, ocean acidification has increased with the result that the ocean surface pH has lowered by 0.017-0.027 units per decade (Butenschön et al. 2021), threatening the balance of all ocean and coastal waters (NOAA Fisheries 2025). This rapid change, causing ocean acidification and changes to the carbonate chemistry system, is unlike anything the Earth has seen in 65 million years (IPCC 2022b). It is well known that human-caused CO_2 emissions are the primary driver of global surface ocean acidification, (IPCC 2023). As human-made CO_2 emissions increase, more CO_2 dissolves into the oceans (NOAA 2025). In the 200 years since the Industrial Revolution, the pH of surface ocean waters has decreased by 0.1 pH units, representing a 30% acidity increase (NOAA 2025). Based on future projections of carbon dioxide emissions, if emissions continue unabated, ocean surface waters could see a 150% increase in acidity by the end of this century, and a pH that the oceans haven't seen in 20 million years (NOAA Fisheries 2025). Where there once was a balance between CO_2 and pH in pre-industrial times, increase anthropogenic emissions means we now have more CO_2 dissolving than leaving the water (Weaver, 2008).

The increase in heat and acidification has detrimental effects on the oceans' ecosystems and their organisms, such as affecting metabolic regulation, the ability to form calcium carbonate shells, ecosystem destabilization and the threatening of important services (Butenschön et al. 2021). Many ocean species like oysters and corals are already feeling the effects of this, since the lack of available carbonate ions prevents these organisms from building and maintaining shells, skeletons and structures, and risks them dissolving if pH levels get too low (NOAA 2025). As for non-calcifying species, these changes in ocean chemistry can affect an organism's ability to detect predators or find suitable habitats thereby, affecting the entire food web (NOAA 2025). In addition, a risk to human health is posed due to increases in harmful toxin-producing algal species, contaminating and sickening marine organisms and the billion-plus people that rely on the ocean as their primary source of protein (NOAA Fisheries 2025).

International initiatives like the Paris Climate Agreement have set goals to limit warming to 2°C (ideally 1.5°C) (United Nations 2015). However, given our current trajectory, this would require a rapid and substantial (Jin and Cao 2023) reduction of emissions either through: 1) consuming less fossil fuels or; 2) capturing more human-produced CO_2 (Gasser et al. 2015). Sources suggest that to meet our targets we may need the removal of 10-20 Gt of CO_2 from the atmosphere per year by 2100 (Nemet et al. 2018; Caserini et al. 2021).

1.2 Carbon Dioxide Removal

If humanity chooses to stabilize the atmospheric level of CO_2 at any level, then net emissions of fossil carbon need to go to zero (Weaver et al. 2007; Weaver 2008; Allen et al. 2025). This could be accomplished by decarbonizing the world's energy systems or some combination of decarbonization alongside the widespread adoption of negative emission carbon dioxide removal (CDR) methods such as carbon capture and storage (CCS) and/or enhancing natural sinks. As mentioned, the ocean currently absorbs about 2.8 Gt/yr (26%) of anthropogenic carbon emissions (Friedlingstein et al. 2023) or about 10 Gt of carbon dioxide emissions from the atmosphere annually (Burns and Corbett 2020). Hence, it naturally makes sense to explore the potential for, and consequences of, artificially enhancing the natural process of oceanic CO_2 sequestration. CDR refers to "technology, practices and approaches that remove and durably store carbon dioxide from the atmosphere" (IPCC 2022a). It consists of human activities that intentionally remove and/or store carbon in reservoirs such as vegetation, soils, geological formations, the ocean or in manufactured products (IPCC 2022a) such as CO_2 injection in building materials like concrete (Bose et al. 2024). Occasionally, studies will use CDR and negative emissions technologies (NET) interchangeably (Feng et al. 2017; Butenschön et al. 2021). However, CDR is now considered the more appropriate term, as it's a broader umbrella term that can also include social science and humanities studies (Renforth et al. 2023).

Land-based biological CDR methods include: 1) forest management: afforestation, reforestation; 2) agricultural approaches: soil carbon sequestration, carbon farming, cover crops, grazing management; 3) biomass-based methods: biomass carbon removal, biochar production, direct biomass burial (IPCC 2022a; NOAA 2024; European Commission 2024; Vaughan 2024;

SARE). Ocean-based biological CDR methods include: 1) blue carbon approaches, such as wetland restoration, mangrove restoration, seagrass protection and the enhancement of salt marshes; 2) ocean fertilization; 3) artificial upwelling/downwelling (Burns and Corbett 2020; NOAA 2024; Vaughan 2024). A push for more natural solutions to carbon capture has resulted in high interest in using the oceans as a way of sequestering carbon. Until about 10 years ago, ocean iron fertilization (OIF) was the primary focus. However, this mitigative practice came into disfavour as many environmental issues arose within the research as well as its limited potential for permanent storage (Keller et al. 2014; Feng et al. 2017; Burns and Corbett 2020) though there are some newer efforts arising. Although both land-based carbon sinks and blue carbon are naturally occurring, the policies mentioned can be implemented to manage and maximize the natural uptake of carbon (EESI 2018). Nature-based solutions (afforestation, biochar, etc.) are considerably more affordable and ready for implementation compared to technological approaches (e.g. biomass with carbon removal and storage, upwelling, etc.). Technological approaches appear more scalable and permanent, though they need further development (C2ES).

Technological or engineered CDR approaches include: 1) Direct air capture/direct air carbon capture and storage (DAC/DACCS); 2) biomass energy with carbon capture and sequestration (BECCS); and 3) direct ocean removal (DOR) (Negative Emissions Technologies and Reliable Sequestration 2019; Burns and Corbett 2020; NOAA 2024; Carbon Gap; U.S. Department of Energy) but typically still have a long way to go in terms of large-scale storage, limitations and complications (National Research Council 2015; Feng et al. 2017; IPCC 2021). BECCS refers to carbon capture via biomass combustion electricity generation (EESI 2018), and DAC refers to the removal of CO_2 from ambient air via chemical bonding (Abouelnaga 2021).

Although not generally considered to be CDR but as an emissions mitigation technique (EMT) since they do not directly remove carbon from the atmosphere (EESI 2018), carbon capture technology (CCT) and carbon capture and storage (CCS) in conjunction with a shift to renewable energy and carbon capture is of great value to keep the Earth from continued warming. In the past decade, CCTs have drawn the scientific community's attention as a hopeful (and necessary) solution to reduce carbon dioxide emissions (Gasser et al. 2015). CCTs comprise four main technologies: pre-combustion, post-combustion, oxygen-rich fuel combustion and DAC, where carbon and other greenhouse gases are removed before or after entering the atmosphere (Zhang et al. 2024). CCTs traditionally work by undergoing a process where the carbon dioxide and hydrogen (or other gases) are separated from each other (Suleman et al. 2022; Zhang et al. 2024). More recently, we have also seen a shift in carbon capture and storage. CCS or carbon capture utilization and storage (CCUS) both function in the same way as CCTs, though storing or converting the captured carbon into valuable chemicals, commodities or products (Suleman et al. 2022). CCSs are considered to be methods for mitigation and storage of CO_2 emissions (Bandyopadhyay 2014) and are a more general type of carbon removal where carbon is trapped and stored underground (EESI 2018). CCUS follows a similar process to CCS but uses the trapped carbon dioxide for products and other purposes, making it profitable (EESI 2018).

Lastly, geochemical CDR approaches include: 1) enhanced weathering; 2) OAE (NOAA 2024; Vaughan 2024), also referred to as artificial ocean alkalinization (AOA) (González and Ilyina 2016; Feng et al. 2017; Jin and Cao 2023). The term coastal ocean alkalinization (COA) will occasionally be used in this thesis, although this generally refers to applying AOA/OAE to oceanic continental shelves shallower than 200 m (Feng et al. 2017). Enhanced weathering is one of the early foci on which current OAE practices are based. Enhanced weathering concentrates on the processing, grinding, and disposal of carbonate or silicate rocks (Rau and Caldeira 1999; Köhler et al. 2010; Rau 2010; Hartmann et al. 2013; Ilyina et al. 2013). Eventually, this would morph into OAE, in which olivine, lime or other processed minerals would be used. Both CCTs and CCSs would be of great use throughout the processes of OAE, whereby capturing the carbon produced during processing, transportation, grinding (i.e. reducing the size of particles for the purpose of quicker dissolution), etc., would keep these methods at net-zero emissions.

OAE has gathered much attention in recent years due to its 'vast' potential and myriad possible application methods (Fakhraee et al. 2022). Chapter 2 will focus on a review of what can be considered both an ocean-based and geochemical CDR solution, OAE. The general concept of OAE is the process of adding and dissolving basic minerals to the ocean with the hopes of increasing the seawater's buffering capacity (Palmiéri and Yool 2024). This is crucial as ocean acidification increases and lowers the ocean's pH, reducing the ocean's buffering capacity and therefore the ability to sequester, absorb and store carbon (Ilyina et al. 2013; NOAA 2019). OAE mimics the naturally occurring process of rock weathering, though speeding up the process, which would otherwise take tens to hundreds of thousands of years (Kheshgi 1995; Archer 2005; Ilyina et al. 2013; González and Ilyina 2016; Moras et al. 2022).

Although many CDR approaches could be considered, their consideration for future implementation often boils down to the potential scale, readiness and certainty for risks and benefits (Abouelnaga 2021; Fakhraee et al. 2022). Alkalinity additions, even in short-term additions, would have long-lasting effects on seawater carbonate chemistry due to slow equilibration (Ilyina et al. 2013). To stay below the thresholds and limit further warming, a combination of emission reduction, CDR and CCT/CCS is a potential way to shift our world into net negative emissions (Gasser et al. 2015; IPCC 2021; Fakhraee et al. 2022). However, it is important to note that although CDR is needed, it is not a replacement for cutting emissions (Abouelnaga 2021).

1.3 Carbonate Chemistry

To understand OAE, it's paramount to understand the chemistry first. The overarching idea is that as CO_2 is absorbed by the ocean, it reacts with water to form aqueous (aq), weak, carbonic acid (Harvey 2008) (Eq. 1), which subsequently dissociates, leading to an increase in hydrogen (H^+) and bicarbonate (HCO_3^-) ions (Eq. 2). If present in sufficient quantity, carbonate ions (CO_3^2) in surface waters neutralize the hydrogen ions, leading to an increase in bicarbonate ions at the expense of carbonate ions (reducing the concentration of carbonate ions) (Eq. 3; Caserini et al. 2022). The net reaction (Eq. 4) doesn't affect pH. However, the rate of carbonate flux to the ocean is limited, so in practice, the uptake of anthropogenic CO_2 by the ocean leads to increasing hydrogen and bicarbonate ion concentrations and decreasing carbonate concentrations. The resulting decrease in ocean pH and increased concentration of dissolved hydrogen ions is known as Ocean Acidification (Weaver 2008; NOAA 2025).

$$\mathrm{CO_2 + H_2O \leftrightarrow H_2CO_{3(\mathit{aq})}} \tag{1}$$

$$\mathrm{H_2CO_{3(\mathit{aq})} \leftrightarrow H^+ + HCO_3^-} \tag{2}$$

$$\mathrm{CO_3^{2-} + H^+ \leftrightarrow HCO_3^-} \tag{3}$$

$$\mathrm{CO_2 + H_2O + CO_3^{2-} \leftrightarrow 2HCO_3^-} \tag{4}$$

Inspired by the known buffering capacity that carbonate ions play in the natural carbon cycle, one mitigative intervention that has been proposed to reduce atmospheric CO_2 levels is to enhance carbonate availability through ocean alkalinity enhancement. As mentioned earlier, OAE is the process of adding and dissolving alkaline materials in the ocean to increase the seawater's natural CO_2 buffering capacity (Palmiéri and Yool 2024). Ocean alkalinity enhancement has the added benefit of allowing increased oceanic CO_2 uptake without changing the pH of surface waters (e.g. by making CO_3^{2-} more abundant in Eq. 3), where the carbonate ion acts as a buffer. This is not to say that there weren't enough carbonate ions prior to the preindustrial era, just that the oceans pH wasn't changing as there was no increase of CO_2. While Eq. 4 was the prevailing net reaction within the preindustrial natural ocean carbon cycle, anthropogenic emissions of CO_2 have upset this balance, with the net result that ocean surface pH has dropped by ~0.11 units between 1750 and 2000 (Caldeira and Wickett 2003; Canadell et al. 2021; Jiang et al. 2023).

Limestone is a sedimentary rock made of calcium carbonate ($CaCO_3$; Oates 1998) and largely comprises the minerals aragonite and calcite. Lime is a generic term for the dissociation product of calcium carbonate, used to denote either quicklime or slaked lime. Quicklime or calcium oxide (CaO) is obtained from limestone by calcination or "burning off" the CO_2 from $CaCO_3$ at high temperatures (thermal dissociation) (Oates 1998; Moras et al. 2022). Slaked or hydrated lime is produced when water is added to quicklime, thereby producing calcium hydroxide ($Ca(OH)_2$; Oates 1998). Quicklime and hydrated lime are both of high interest in the OAE field, given their relatively high solubility and quick dissolution (Moras et al. 2022). Limestone dissolves more slowly than lime in water, though lime requires more energy to produce and releases one molecule of CO_2 in the process of obtaining one molecule of CaO (Caserini et al. 2022). Given limestone's low solubility compared to lime (Moras et al. 2022), one method to possibly improve limestone application could be the dissolving of $CaCO_3$ prior to application. The ocean (for the most part) is supersaturated with respect to aragonite and calcite, where $\Omega > 1$ (i.e. omega; being the saturation state of $CaCO_3$)(Moras et al. 2022). Thus, dissolving the $CaCO_3$ in an undersaturated solution before ocean application results in the Ca^{2+} staying fully dissociated in the solution while OH^- and CO_3^{2-} react with H^+ to form water and HCO_3^-, increasing pH (lowering H+) and HCO_3^- and decreasing CO_2 (Ilyina et al. 2013). Limestone is extremely abundant across the Earth but is not readily soluble (Kheshgi 1995). If limestone is finely ground, it can then be sprinkled into the ocean's surface layer, sinking and eventually dissolving in unsaturated deep water (Harvey 2008). Limestone dissolves and precipitates at a rate of 1×10^{13} mol Ca/yr (Caserini et al. 2022). Quick lime and hydrated lime dissolve in seawater within a few hours (Moras et al. 2022).

Carbonates are incredibly effective in neutralizing acids, and this process in the past has taken place naturally through the dissolution of carbonate sediments at the bottom of the ocean and the weathering/erosion of carbonate rocks into the ocean (Weaver 2008). The natural process of rock weathering increases ocean alkalinity and sequesters 0.5 Gt of CO_2 each year (Renforth and Henderson 2017). Carbonate dissolution is the natural process that liming mimics. Carbon dioxide and calcium carbonate are mixed into water, producing a calcium ion and two bicarbonate ions (depending on the pH of the water) through the reaction:

$$\mathrm{CO_2 + CaCO_3 + H_2O \leftrightarrow Ca^{2+} + 2HCO_3^-} \tag{5}$$

The process of creating slaked lime from limestone involves the separation of calcium carbonate into lime and carbon dioxide, where lime and water can then create calcium hydroxide ($Ca(OH)_2$):

$$\mathrm{CaCO_3 \leftrightarrow CaO + CO_2} \tag{6}$$

$$\mathrm{CaO} + \mathrm{H_2O} \leftrightarrow \mathrm{Ca(OH)_2} \tag{7}$$

Ocean liming can occur when calcium hydroxide and carbon dioxide are dissociated into calcium ions and bicarbonate (depending on the pH of the water), shown by the following formula:

$$\mathrm{Ca(OH)_2} + 2\mathrm{CO_2} \leftrightarrow \mathrm{Ca^{2+}} + 2\mathrm{HCO_3^-} \tag{8}$$

Calcium carbonate already has CO_2 attached to it, so when one molecule of $CaCO_3$ is mixed into the water, it can only sequester one molecule of CO_2.

Lime, on the other hand, can theoretically absorb two CO_2 molecules; since the conversion of limestone to lime releases a molecule of CO_2 into the atmosphere. Lime/CaO dissolution of 1 mol increases total alkalinity (TA) by 2 moles and increases CO_2 uptake by 1.79 moles (Kheshgi 1995). Given that naturally occurring global silicate weathering contributes to about 0.012 Pmol yr^{-1}, an alkalinization scenario with a molar ratio of 2 TA to 1 CO_2 would mimic this natural process, increasing its scale by two orders of magnitude (Ilyina et al. 2013). For every mole of added slaked lime, the reaction will consume up to 2 moles of aqueous CO_2, the dissolution of the slaked lime then increases the alkalinity of the water body by 2 mole equivalents. However, it is essential to note that "the consumption of aqueous CO_2 does not correspond directly to a removal of atmospheric CO_2, which instead depends on the difference in partial pressure of CO_2 between sea and atmosphere at the water surface" (Butenschön et al. 2021). Increasing surface TA increases seawater pH, CO_2 uptake and CO_2 storage (González and Ilyina 2016). Surface ocean TA is influenced by the presence of freshwater through precipitation, sea ice melt and evaporation. Typically, the highest values are found in the Atlantic and Pacific Ocean gyres (Ilyina et al. 2013). These processes, along with temperature, salinity and pressure, determines the partial pressure of

CO_2 (pCO_2) and pH (Butenschön et al. 2021). Dissolved inorganic carbon (DIC) represents the sum of HCO_3 and CO_3^{2-}, whereas TA is the sum of HCO^{-}_3 and CO_3^{2-} (though ignoring smaller contributions from other components of TA).

As such, TA-DIC approximately indicates the amount of CO_3^{2}. Increasing TA at a constant DIC shifts carbonate chemistry to a higher pH, which then decreases CO_2 concentrations and pCO_2 (partial pressure of CO_2) and increases the atmospheric CO_2 uptake potential (Moras et al. 2022). However, the magnitude depends on the initial state of the marine carbonate system and its parameters (Köhler et al. 2010; Burt et al. 2021). Although OAE involving lime captures twice the CO_2 of OAE involving crushed limestone, unless the CO_2 released from limestone processing to lime is sequestered or captured, limestone and lime would be responsible for sequestering and producing equal amounts of CO_2.

1.4 Introduction to Thesis

Chapter 1, the introduction, has provided background on climate change, OAE as a CDR (carbon dioxide removal) method, and the general carbonate chemistry behind OAE. Chapter 2 serves as a literature review on OAE modelling and other OAE studies. Section 2.1 reviews the availability of alkaline materials (also considering the limitations of production), and Section 2.2 reviews the limitations of OAE as identified in previous works. This includes transportation, distribution, costs and environmental effects. Section 2.3 focuses on previous OAE modelling efforts and their differences, allowing for comparison between studies. This review will serve as the basis for understanding future model results. This is so that OAE can be assessed globally with general limitations in addition to the complications with mining, shipping, and disposal, as previous OAE simulations have been bound by. Chapter 3 is a rendition of the paper from Martin et al. (2025). It assesses the question: If the entire global resources of limestone mining and production were used for OAE, and if limestone and/or lime were to disassociate in surface waters entirely, would this be enough to reduce atmospheric carbon dioxide emissions significantly? That is, an estimate is provided of the maximum potential of OAE as a mitigative intervention to enhance CDR. Specifically, this paper will focus on using limestone and lime within OAE to assess it as a reliable green technology for sequestration.

Chapter 2: Literature Review of OAE Research & Modelling

2.1 Availability of Alkaline Resources

2.1.1 Limestone

The most crucial limiting factor to be considered when running OAE simulations is the global production and availability of the chosen mineral for alkalization. Several minerals are applicable to the general concept of OAE. However, few have the mining capabilities, global resource availability and current infrastructure readily available for increased production as that of limestone. Limestone, composed mainly of calcium carbonate, is found in large quantities in the Earth's crust (Moras et al. 2022). Limestone rocks are composed of more than 50% carbonate materials, made up primarily of calcite (pure $CaCO_3$) or dolomite ($CaMg[CO_3]_2$) or both (Chilingar 1967; Bliss et al. 2008). Limestone comes in many varieties such as chalk, travertine, marble, carbonatites, etc. (Bliss et al. 2008). While limestone covers about 10% of the Earth's surface and is found in most countries (Oates 1998), total global production and export inventories are not readily available. Limestone data is challenging to gather in terms of the total global production due to the lack of data sets in various countries with limestone exports and no published reserve and resource estimates on a worldwide scale.

Based on lime production, the largest limestone reserves are found in China, India, the United States and Russia (U.S. Geological Survey 2024). Limestone is an essential material for cement production (Caserini et al. 2022) and is often used in agriculture (soil management), metallurgy (capturing impurities) and sulphur removal (solidifying sulphur dioxide) (U.S. Geological Survey 2024). Materials processed from limestone are generally pulverized stone (used for mining or to neutralize acidified lakes), calcined stone (quicklime), dimension stone (for building purposes) and crushed stone aggregate (Keith and Webb 2015). A total of 6.6 Gt of limestone from global deposits is estimated to be produced annually as of 2017, similar to that of coal (7.3 Gt yr^{-1}), or about 9% out of the world's raw mineral production of 44 Gt yr^{-1} (Caserini et al., 2022). This estimation was based on a combination of data from limestone, lime, cement and crushed stone production (Caserini et al. 2022).

In 1998, it was estimated that 4,500 Mtpa were used globally (4.5 Gt yr^{-1}), with the US alone producing 800 Mt in limestone sales in 1994 (Oates 1998). Of the 4,500 Mtpa produced globally in 1998, the United States used 870 Mtpa, Japan used 208 Mtpa, and the United Kingdom used 208 Mtpa (Oates 1998).

Another problem in determining global lime and limestone production quantities is that data sets often classify it as a part of or as 'crushed stone'. In the United States alone, 1.5 Gt (1.5 Gt) of crushed stone were produced in 2023, 69% of which was limestone (U.S. Geological Survey 2024). Typical uses of crushed stone are: construction aggregate, cement manufacturing, lime manufacturing and agricultural uses (U.S. Geological Survey 2024). Approximately two-thirds of the crushed stone used in the United States yearly is limestone (Harvey 2008). Similarly, Natural Resources Canada (NRCAN) reported that Canadian limestone production in 2005 was approximately 117 Mt, of which 74% was composed of crushed stone and quicklime used for cement (Keith and Webb 2015). Excluding Prince Edward Island and Saskatchewan, limestone production occurs in each province, with Ontario and Quebec accounting for 84% of all active quarry operations, according to the 2006 Canadian statistics report (Keith and Webb 2015).

Limestone deposits can cover hundreds of square miles and be hundreds of feet thick with fairly uniform thickness and quality (Keith and Webb 2015). However, purities mostly range between 20% and 80% (Sverdrup et al. 2017) and up to 95% in some deposits (Bliss et al. 2008). Impurities in limestone include a spectrum of magnesium carbonates, dolomite, silica, glauconite, gypsum, fluorite, siderite, sulphides, iron and manganese oxides, phosphates, clays, and organic matter (Chilingar 1967). As for potential sources, one study estimated that in 2012, limestone theoretically available for extraction (based on areas underlain by limestone rock) could be as high as 150 million km^3 or 17,630 Mt/year, given an exploitation of 2-3% of known reserves (Sverdrup et al. 2017). This exploitation rate of only 2-3% is due to a lack of transportation, sources in developed areas, conflicts with land use like agriculture or protected areas and reserves and extraction challenges (Sverdrup et al. 2017).

2.1.2 Lime

Lime (Quicklime), obtained through calcination of the raw material of limestone, is used to treat soils, purify water and smelt copper, in addition to many uses in the chemical industry (Bliss et al. 2008; Moras et al. 2022). Quicklime applications include: cement production, flux for iron and steel, water purification, flue gas desulphurization, glass making, pulp and paper treatment and sugar refining (Keith and Webb 2015). The estimated world lime production is 0.43 $Gt\,yr^{-1}$ as of 2023 (U.S. Geological Survey 2024). The world's total lime production was 0.35 Gt in 2017 (U.S. Geological Survey 2018), which suggests a 22.8% increase in the last 6 years. In comparison, a 1998 study showed the global production of lime as 200 Mtpa (0.2 $Gt\,yr^{-1}$) (Oates 1998), suggesting a 46.5% increase in 25 years. About 420 Mt of lime were produced worldwide in 2024, a slight reduction from the previous year (U.S. Geological Survey 2024). The top producers of that

420 Mt were: 1) China, 310 Mt; 2) India, 17 Mt; 3) United States, 16 Mt; 4) Russia, 11 Mt (U.S. Geological Survey 2024). In Canada, lime is one of the top minerals produced, yielding 1.8 Mt of lime exports in 2018 (U.S. Geological Survey 2019; Natural Resources Canada 2023). Quicklime accounted for 19.4% of limestone production in Canada as of 2005 (Keith and Webb 2015). Though estimated in 2013, Ilyina describes how lime production would need to be increased by a minimum of "2 orders of magnitude" worldwide to implement OAE successfully (Ilyina et al. 2013). This assessment is based on the estimated average of 0.3 Pg yr^{-1} (0.3 Gt yr^{-1}) of lime produced between 2006 and 2010 (U.S. Geological Survey 2011), equivalent to 0.005 Pmol of $Ca(OH)_2$ yr^{-1} (Calcium hydroxide, i.e. slaked lime) (Ilyina et al. 2013). This was analyzed again in 2016, where to keep atmospheric CO_2 at RCP 4.5 levels under an RCP 8.5 scenario until year 2100, global surface TA concentrations would need to be increased by 30%, requiring 114 Pmol of added alkalinity (González & Ilyina, 2016). To achieve this, lime production would need to be increased annually by two orders of magnitude (increased to 4.22 Tt of total lime production from 2016-2100; González and Ilyina 2016). For a calcium oxide powder application, it has been suggested that 1.4-1.7 t of limestone would have to be processed for every t of CO_2 to be sequestered from the ocean liming (Renforth et al. 2013) and that for every g of lime, it could potentially sequester 1.57 g of CO_2 (Renforth and Henderson 2017). Given that CO_2 is released during processing, carbon capture and storage during lime production would be necessary for full OAE potential (Butenschön et al. 2021; Moras et al. 2022).

2.1.3 Carbonates

About 75% of the Earth is directly underlain by sedimentary rock, ranging in thickness up to 8-10 miles (Ham et al. 1962), and older estimates suggested that carbonate rocks make up 20-25%

of all sedimentary rocks (Ham et al. 1962; Flügel 2010). In recent years, studies have proposed that a third of the world's sediments are carbonate rock (Hartmann and Moosdorf 2012) and 15.2% of ice-free land surface is characterized by carbonate rocks (Goldscheider et al. 2020), suggesting that these resources are vast and have large availability (Harvey 2008; Caserini et al. 2022).

Carbonate rocks are those containing more than 50% carbonate materials (Ham et al. 1962). Carbonate rocks are classified into two divisions: limestones (composition of 50 %+ $CaCO_3$) and dolomites (composition of 50 %+ $CaMg(CO_3)_2$) (Flügel 2010). Associated with limestone and dolomite, common carbonate minerals include siderite, ankerite and magnesite (Caserini et al. 2022). Although other rocks contain carbonate, they are generally classified as non-carbonates, given a 50% or less make-up of carbonate, this includes calcareous (or dolomitic) sandstone, claystone or shale (Ham et al. 1962; Chilingar 1967). There are typically three classifications of rock from which calcium carbonate can be extracted: mixed sedimentary rocks, carbonate sedimentary rocks and pure carbonate rocks. Mixed sedimentary rocks represent sediments where carbonate is mentioned but not dominant, and are typically a mix of sandstone and limestone (Hartmann and Moosdorf 2012). Carbonate sedimentary rocks consist primarily of carbonate rocks, typically limestone, dolomite and marl (Hartmann and Moosdorf 2012). Pure carbonate rocks indicate rocks with low siliciclastic content (limestones and dolomites composed of calcite and dolomite) and represent about 9% of mapped sedimentary lithological classes (Hartmann and Moosdorf 2012).

About 15.2% or 20.3 million km^2 of the Earth's land surface is characterized by the presence of carbonate rocks (Goldscheider et al. 2020) and about 4.1 million km^2 of total pure carbonate outcrops are estimated to exist (Caserini et al. 2022). Mining pure carbonate outcrops for OAE, which are near the coastline and are below bare ground or shrub, is ideal for limiting costs and energy. Within 10 km of the coastline, deposits could provide approximately 5000 Gt of limestone, 15,000 Gt within 50 km and 40,000 Gt within 100 km (Caserini et al. 2022). Globally, carbonate rocks comprise 15.7% of marine coastlines (excluding Antarctica), with an estimated quarter of these coastal carbonate rocks here in Canada, spread through the Canadian Arctic and Hudson Bay (Goldscheider et al. 2020). The largest deposits of pure carbonate outcrops within 50 km of the coastline are found around the Mediterranean, Central and North Africa, the Middle East, South Asia and Central America, with Iran and Somalia being the countries with the highest amount of pure outcrops near the coast and below the bare ground or shrub (Caserini et al. 2022). Carbonate rocks exist on each continent, with Asia holding the largest surface area of continuous (65% or more carbonate) and discontinuous (15-65% carbonate) or mixed carbonate rock, and substantial amounts are also found in North America and Africa (Goldscheider et al. 2020). It's important to note that Caserini (2022) mentions that the presence of these carbonaceous units does not necessarily mean they can be exploited, given competition, transport availability, recreation areas and nature reserves.

2.1.4 Other Alkaline Resources

Many other minerals and materials have been considered for OAE. In addition to calcium-rich minerals/materials (quick lime, hydrated lime), magnesium-rich minerals/minerals (brucite, periclase, forsterite) are also considered to be the most suitable for OAE application (Renforth and Henderson 2017; Moras et al. 2022).

One mineral that has received much attention is olivine. Like limestone, olivine dissolution for OAE/AOA can be referred to as enhanced weathering, as it follows the natural process of olivine dissolution/weathering, which naturally occurs over thousands to millions of years (Kump et al.

2000; Köhler et al. 2010; Feng et al. 2017). Theoretically, under favourable conditions and complete dissolution, olivine could sequester 4 moles of atmospheric CO_2 for every 1 mole dissolved or about 0.34 g of carbon removal per g of olivine (Köhler et al. 2010; Montserrat et al. 2017; Wang et al. 2023). While limestone and lime produce calcium as a byproduct, olivine produces magnesium ions and silicic acid (Montserrat et al. 2017). When olivine is dissolved into seawater, it raises alkalinity and binds the CO_2 as bicarbonate (Wang et al. 2023), and compared to lime, it does not initially release CO_2 prior to increasing CO_2 uptake (Meysman and Montserrat 2017).

As will be explored further in Section 2.3, many models use total alkalinity (TA) rather than a simulated mineral input, as it directly indicates the ocean's buffering capacity. Keller explored how adding TA at a rate of 0.14 Pmol yr-1 would be equivalent to adding 5.19 Pg yr-1 of slaked lime or 4.92 Pg yr-1 of forsterite (a form of olivine) (Keller et al. 2018). For every gram of forsterite, it could potentially sequester 1.25 grams of CO_2 (Renforth and Henderson 2017). Although the theoretical potential for olivine is high, the final sequestration rate could be 20% or lower than suggested, given the required mining, transportation and grinding (Köhler et al. 2010; Wang et al. 2023).

The major limitation for olivine is mining, while 6.6 Gt of limestone is produced each year, olivine global production as of 2017 sat at only 8.4 Mt yr^{-1} (Caserini et al. 2022). As previously mentioned, to keep atmospheric CO_2 at RCP 4.5 levels under an RCP 8.5 scenario until year 2100, global surface TA concentrations would need to be increased, for olivine application. Olivine production would therefore need to increase by three orders of magnitude (increased to 4.01×10^{12} Tt) (González & Ilyina, 2016). In addition, olivine dissolution in surface waters, similar to limestone, could potentially mean a significant fraction of the input sinks undissolved to the ocean floor (Köhler et al. 2010). As such, olivine would need to be ground into small particles to avoid sinking before dissolution, thereby affecting net CO_2 efficiency due to the energy required to accomplish this crushing (Köhler et al. 2013; Feng et al. 2017).

Other materials analyzed for OAE include brucite, magnesite, soda ash, periclase, etc. Brucite ($Mg(OH)_2$) global production sits at 1.5 Mt yr^{-1} (Caserini et al. 2022); for every gram of brucite, 1.51 grams of CO_2 could potentially be sequestered (Renforth and Henderson 2017). Magnesium carbonate or magnesite ($MgCO_3$), a component of dolomite, also shows results similar to those of limestone (Harvey 2008). Annual magnesite production sits at 28 Mt yr^{-1} as of 2020, with Australia, Austria, Bosnia, Brazil and Canada as the top exporters (Reichl and Schatz 2022). For every gram of magnesite, 0.52 grams of CO_2 could potentially be sequestered (Renforth and Henderson 2017). Soda ash, or washing soda (NA_2CO_3), is another material that has been considered due to its readily soluble nature in seawater; however, supply is an issue. If the entire world's reserve were dissolved into the ocean, it would only offset 6 Gt of carbon (Kheshgi 1995; Harvey 2008). And for every gram of periclase, it could potentially sequester 2.18 grams of CO_2 (Renforth and Henderson 2017).

It’s important to note that as these other OAE minerals/materials do not make up large portions of the world’s deposits are mined on much smaller scales than limestone, and are produced on much smaller scales than lime, meaning both mining and production would need to be increased on a much larger scale with reserves that would quickly run out. And so, although synthetic alkalinity sources (like periclase) could suggest significant potential for carbon capture, it’s expected to be limited by feedstock supply (Fakhraee et al. 2022).

2.2 Processing and Application Limitations

In this section the limitations identified in the literature concerning the widespread adoption of OAE will be discussed. These limitations involve shipping and transportation capabilities, disposal options, emissions from processing and transportation, energy availability, costs, and market factors.

2.2.1 Transportation

Major limits facing the widespread adoption of limestone and lime for OAE are production and the ability to deliver the quantities required (Keller et al. 2014). The typical transportation methods for limestone are by tipper truck (dump truck), rail and ship. While trucks are cheap and flexible, they create high emissions (Oates 1998). Transport via rail is more environmentally acceptable, though less competitive (Oates 1998). Transport via shipping has a low cost per t (Oates 1998). Ideally, OAE would use limestone that doesn't require much transportation, resulting in lower emissions. This means limestone deposits located near coasts are ideal, as was previously explored. Harvey suggested using the largest oil supertankers to transport limestone powder, given their capacity to hold 650,000 t of oil (Harvey, 2008). As of 2000, globally, there was a world fleet of 6878 oil tankers with an average capacity of 39,300 t (IMO 2000; Harvey 2008). In reference to section 2.1.1, this would hold about 4.1% of the current global limestone mining output of 6.6 Gt/yr or 62.9% of the current global lime production of 0.43 Gt/yr.

2.2.2 Dissolution

The most common method suggested for the dispersal of alkaline materials in OAE simulations is using ships, which would slowly release the deposits in their travels. Harvey (2008) suggested that if limestone powder was applied at 200 g m^{-2} a^{-1} (if dissolved in the first 1000 m) in applications of 10-100 g m^{-2} at a time (requiring 2-20 applications in a particular region) in a 100 m wide path while the ship travels at 30 km h^{-1}, the powder could be applied at a rate of 30-300 t h^{-1} (Harvey 2008). This was considering a scenario where the limestone powder would be projected off the ship (Harvey 2008). On the other hand, dispersion in the ship's wake could prove an efficient technique given that rapid mixing is possible depending on the ship's waterline length, speed, and discharge (Caserini et al. 2021).

Limestone can only dissolve in $CaCO_3$ undersaturated oceanic water (Millero 2007; Harvey 2008; Feng et al. 2017). With limestone application, supersaturation and undersaturation with respect to calcite would need to be considered (Harvey 2008). Deep ocean waters are undersaturated with respect to calcium carbonate, due to the concentration of carbonate required for supersaturation increasing with depth/pressure (Harvey 2008). Organic matter dissolution in the ocean respires CO_2, reducing pH and lowering carbonate ion concentrations (Harvey 2008). In turn, the saturation horizon becomes shallower, allowing for more of the deep ocean to be capable of dissolving calcium carbonate (Harvey 2008). This is observable in our oceans, where the saturation depth in the North Atlantic is greater than 4000 m (younger water with less CO_2), whereas it's less than 500 m in the North Pacific (older water with more accumulated CO_2) and less than 250 m along the west coasts of North and South America (Harvey 2008).

However, limestone dissolution in these deep waters would not enhance oceanic-atmospheric CO_2 uptake unless processes like upwelling brought the water to the air-sea interface (Feng et al. 2017). Ensuring alkalinity dissolution in surface waters is of great importance, as the longer the alkalinized water is in contact with the air, the more effective it is in lowering atmospheric CO_2 (Ilyina et al. 2013). Harvey (2008) showed that adding limestone powder to areas with a shallow saturation depth and strong upwelling eventually spreads the absorbed CO_2 away from application regions, and nearly all of the added $CaCO_3$ participated in CO_2 absorption. This is observable where applications in ice-free regions lead to a modest increase in the Arctic and Southern Ocean due to horizontal transport via the vertical mixing processes of advection, convection and diffusion (Jin and Cao 2023).

Unless the water below the application site is driven to a saturation state, its ability to absorb CO_2 should not decrease (Harvey 2008). When the downwelled water ultimately reaches the application sites again, the carbonate ion concentrations will be exhausted through CO_2 absorption in the mixed layer, and so in theory, this process could continue indefinitely (Harvey 2008). Harvey also suggested that greater CO_2 absorption could occur if OAE were implemented in both regions with shallow saturation/high upwelling and with deep saturation/low upwelling (Harvey 2008). However, he mentioned that this is a somewhat idealistic scenario given the assumption of all upwelling water entering the mixed layer. Given the specific importance of the winter mixed layer depth, a limestone application could be most effective in high-latitude regions with shallow saturation and strong upwelling (Harvey 2008).

Another study showed that applying alkalinity gradually until year 2100 in accordance with the RCP 8.5 scenario of atmospheric CO_2 emissions, within the upper 12 m of the ocean, and allowing for natural mixing to occur through advection and diffusion suggests eventual TA increases by the end of the century (González and Ilyina 2016).

One of the major benefits of a lime OAE application is its rapid dissolution, especially compared to other OAE materials like olivine and basalt (Fakhraee et al. 2022). Quick lime and hydrated lime dissolve in seawater within a few hours and increase TA within minutes (Moras et al. 2022). Looking at water column dissolution and the marine biological pump, Fakhraee's (2022) study highlighted that MgO and CaO show rapid near-surface dissolution and that CaO alkalinity production is essentially completed in the upper ocean mixed layer regardless of application rate (Fakhraee et al. 2022). In comparison, despite high application rates and minimal grain sizes, olivine's alkalinity effectiveness near the surface is limited (Fakhraee et al. 2022). This suggests that basalt and olivine would be ineffective for much of the ocean, where dissolution would need to occur near the surface, as opposed to metal oxides, which could increase CO_2 uptake even in shallow depths (Fakhraee et al. 2022).

Surface water temperature is yet another possible barrier to the application of OAE. The calcium hydroxide in limewater (the aqueous solution of slaked lime) has poor solubility at 25 °C, and so using milk of lime, a slurry which could be prepared on ships by mixing crushed slaked lime with water, could be one way around this issue (Caserini et al. 2021). This slurry would speed up discharge, and if water from the vessel's engine cooling system was used, it could save energy from the pumping and the need for fresh seawater (Caserini et al. 2021). Temperature can also affect the CO_2 uptake potential. Moras et al (2022) demonstrated that atmospheric CO_2 removal could be enhanced by keeping seawater equilibrated with air or CO_2-enriched gases during the process of mineral dissolution. This is especially true for colder waters, where higher CO_2 solubility lowers Ω values (Moras et al. 2022). However, this process would consist of an additional step, leading to higher costs and a longer OAE distribution process (Moras et al. 2022). Another suggestion for application (hoping to avoid barriers to dissolution) is to react ground limestone with CO_2-rich gas streams in a water solution, before adding it to the ocean, ensuring dissolution prior to reaching open ocean (Rau et al. 2001).

2.2.3 Application

Many studies suggest using existing or new fleets of ships to spread ground limestone and lime through the ocean. Harvey (2008) indicated that 750-3000 dedicated smaller ships could deliver 4 Gt of $CaCO_3$ each year (750 central ships or 2,560 local delivery ships), given time and resupply. On the other hand, larger ships could also prove helpful. The majority of deadweight tonnage (DWT) is related to two categories of ships: 1) cargo, i.e. bulk carrier, container, general cargo and 2) tanker, i.e. chemical tanker, oil tanker (Caserini et al. 2021). Previous published research modelled OAE enhancement based on the total transport capacity of all large cargo ships and tankers, with an addition of 0.25 Pmol/y of alkalinity (Köhler et al. 2013). This was done in consideration of the total estimated deadweight tonnage of 0.33 Gt and an average of 32 port visits per ship per year (Caserini et al. 2021). The total transport capacity of all large cargo ships and tankers has been estimated at approximately 10 Pg y^{-1} (Keller et al. 2014). One study on the engineering challenges of ocean liming suggested loading bulk carrier ships with lime. Renforth assessed the need for 101 dedicated 300,000 deadweight tonnage bulk carriers discharging 1 Gt per year of slaked lime (Renforth et al. 2013). It showed that with approximately 11 days of discharge, open ocean discharge and return to port, 101 dedicated 300,000 DWT bulk carrier ships would be able to deliver 1 Gt of alkalinity (Renforth et al. 2013). Bulk carriers and container ships have been identified as ideal vessels due to their load capacity and global spatial coverage of shipping routes (Caserini et al. 2021). Together, they represent 17% of the international commercial fleet and 53% of the active tonnage fleet (IMO 2014; Caserini et al. 2021). Considering capacity, distance travelled, reloading, discharge rates and assuming a low discharge based on 15% of bulk carrier and container ship deadweight capacity, the maximum discharge potential for slaked lime using all active ships globally is 1.7-4 Gt per year (Caserini et al. 2021). Harvey (2008) estimated that 3000 ships would be needed to add 4 Gt of limestone, or 100 ships would be required to add 1 Gt of slaked lime (Renforth et al. 2013). Based on the 2012 global shipping capacity of 8 Gt a^{-1}, and the existing fleet of 50,000 ships, both scenarios would require a small expansion (Renforth and Henderson 2017).

Another possible application scenario is using the current network of cargo and tanker ships to address OAE (Butenschön et al. 2021). Although dedicated fleets for OAE would allow greater control of the application area and distribution rate, using existing fleets and routes could increase the area and potential scale for OAE disposal (Harvey 2008). Moras (2022) showed that using all available ship capacity and a slow discharge of 1.7- 4.0 of calcium hydroxide per year (Caserini et al. 2021) could increase atmospheric CO_2 absorption into the ocean by 1.2-2.8 Gt (Moras et al. 2022). If an agreement to share ship capacity took place and equipment designed for discharging the material was created, existing vessels could easily be used for OAE discharge (Caserini et al. 2021). As the Mediterranean already has large and established shipping routes, these commercial routes could provide a considerable opportunity for OAE to use existing traffic at a low cost (Butenschön et al. 2021). In the Mediterranean Sea alone, the potential for slaked lime could be 186 Mt discharge per year using existing vessels (Caserini et al. 2021). However, a new fleet of 1000 dedicated ships could discharge 1.3 Gt per year alone (although the construction or renewal of existing unused ships and emissions made during these processes would need to be considered)

(Caserini et al. 2021). Although adding weight on board would reduce the ship's performance, many container ships could store slaked lime in the hold, so 10-20% of the net cargo capacity could carry and discharge slaked lime (Caserini et al. 2021). Although this wouldn't require much modification to the ship itself, ports would have to install loading facilities (Caserini et al. 2021). Frequency of reload would be a major factor since the potential for OAE application would increase if lime could be reloaded during intermediate stops (Caserini et al. 2021).

Bulk carriers in ballast (ships empty of cargo and filled with ballast water for stability, typically awaiting docking, loading or on their return trip) could also be used to discharge lime (Caserini et al. 2021). Studies indicate that dry bulk carriers sail empty 42% of the time (Brancaccio et al. 2020). This is, of course, substantial unused capacity that could be utilized.

2.2.4 Location

Regional differences in carbon inventories and biogeochemical changes indicate regional sensitivity to OAE (González and Ilyina 2016; Burt et al. 2021). Considerations of TA and pH are also important. Some studies have focused on regional inputs in areas where less alkalinity would be required to raise pH. TA values are a result of freshwater input through precipitation, evaporation and sea ice melt and higher TA values are found in the gyres of both the Atlantic and Pacific (Ilyina et al. 2013). The North Pacific has higher values of normalized total alkalinity (NTA; normalized to a salinity of 35) than the North Atlantic, both increasing with depth, while surface water NTA is generally constant outside of polar deep water outcropping areas and coastal upwelling areas (Millero 2007). $CaCO_3$ solubility and dissolution are greater in the Pacific, given a much lower pH than the Atlantic (Millero 2007). When considering the location for applying OAE, the ocean temperature plays a role. Colder water can dissolve more CO_2 than warmer waters;

however, colder water has a lower buffering capacity (Ilyina et al. 2013), meaning it's more sensitive to change (an important consideration, especially for effects on ecosystems and organisms). Despite a scenario of uniform additions of alkalinity, surface alkalinity is not uniform between oceans due to the mixing and circulation characteristics of the ocean regimes, specifically when looking at the difference between latitudes given alkalinity addition in ice-free regions (Jin and Cao 2023).

Burt et al. (2021) found that OAE applications were more successful (sequestered more carbon) in the Southern Ocean and the Subpolar North Pacific, and less successful in the Subpolar North Atlantic compared to the Global simulation. Enhancing regionally could sequester 82-175 Pg of carbon into the ocean compared to 156 Pg of carbon when applied globally (Burt et al. 2021). Ilyina et al. (2013) also identified the North Atlantic as the least effective for increasing pH and lowering CO_2, given its circulation, where surface waters are brought to the deep. Ilyina further showed the greatest regional surface pH effect in the Southern Ocean (Ilyina et al. 2013).

Jin and Cao (2023) investigated the deep penetration of alkalinity as a result of North Atlantic deep water formation to depths as low as 1500 m. Targeting upwelling regions for limestone addition could prove beneficial, especially given the large occurrences of upwelling in the North Pacific (Harvey 2008).

Based on the global oceans' natural CO_2 sink and source hotspots, Ilyina et al. (2013) modelled alkalinity inputs in sink areas and coastal upwelling regions and although showing increased pH at the point of application, areas isolated from application saw continual decreases in pH, suggesting regional alkalinity enhancement could only mitigate ocean acidification at the point of application. In addition, the study found that alkalinity addition to source areas resulted in the largest global pH mitigation (Ilyina et al. 2013).

Application could also occur in rivers near where the mixing with ocean water occurs, especially given that most rivers are undersaturated with respect to calcite. COA is also of consideration, given that there are more minor requirements for transport and grinding (Feng et al. 2017).

2.2.5 Market

Although OAE is generally considered one of the most cost-efficient CDRs, further research, development, and implementation depend on investment opportunities and, therefore, restrictions on costs, market implementation, and energy consumption.

In 2022, the global limestone market was estimated to be valued at 72.32 billion United States dollar (USD) and expected to grow by 7.3 % by 2030 (Grand View Research 2021). The typical market segments of limestone production are: construction and building, cement, agriculture, metal refining, quicklime production and glass (Oates 1998). A large part of limestone production in the past has been used for cement, where of the total global extraction in 1998, 1420 tpa of the total 4500 tpa produced was used for cement production (Oates 1998). In the United Stated of America (USA) alone, 17 Mt of quicklime and hydrated lime valued at 2.6 billion USD were produced in 2023 (U.S. Geological Survey 2024).

The typical market segments for lime are chemical and industrial applications, construction, fertilizer, flue gas treatment, glass, nonferrous-metal mining, paper and pulp, precipitated calcium carbonate, steelmaking, sugar refining and water treatment (U.S. Geological Survey 2024). OAE implementation would require massive upscaling of both limestone mining and slaked lime production (Caserini et al. 2021).

2.2.6 Material and transportation costs

In the United States, the market costs for limestone were around \$5 t^{-1} as of 2007, and the grinding cost was about \$1–3 t^{-1} (Rau et al. 2007). Less expensive than lime, limestone is often used as a substitute in agriculture and for sulphur removal due to its less reactive material structure (U.S. Geological Survey 2024). In recent years, limestone has also increasingly been used in water treatment, as it can soften water by removing magnesium and calcium ions (Grand View Research 2021).

Limestone and limestone-product production are relatively inexpensive, meaning transport accounts for the vast majority of costs and can amount to over 50% of the final price (Oates 1998). Transportation is important as its cost equals the cost of material and production at about a distance of 50 km (Oates 1998), another reason why limestone extraction near coasts is of great value, in addition to keeping CO_2 emissions at a minimum. For road transport by dump truck/tipper wagon, the economic cost is between 40-60 km, rail transport can extend that to 200 km and transport by ship, extends this much further (Oates 1998).

Harvey (2008) considered using large ships for transportation prior to smaller vessels for disposal. Harvey suggested that costs for limestone transportation could be roughly \$0.034 t km^{-1} by train, \$0.010 t km^{-1} by barge, and \$0.003 t km^{-1} by freighter. Another consideration for the transportation of OAE materials is its place in the transportation of goods, especially when existing fleets are used. Unless carried on ballast ships or in the hull, alkaline materials would replace other goods, suggesting possible competition. Finally, a potential stream of revenue for spreading slaked lime (or other alkaline materials) could derive from carbon credits, increasing convenience (Caserini et al. 2021). For those ships whose trip or return trip did not carry full loads or carried lessened or empty loads, OAE could stand to benefit their bottom line.

2.2.7 Energy

Given lime's great potential for OAE, it's important to consider the costs and energy requirements around processing limestone to lime (and the capture of CO_2). The most significant limitation to OAE is the ability to ship the necessary material and the high energy cost associated with processing the materials.

For the thermal and electrical requirements, it is estimated that 0.6-5.6 GJ of thermal energy and 0.1-1.3 GJ of electrical energy would be required for every net t of CO_2 captured by lime, using flash calcination and carbon capture (Renforth et al. 2013). Renforth (2013) suggested that 4500 sites for limestone to lime processing and 100 dedicated ships for disposal could sequester 1 Gt of CO_2 annually, using approximately 1% of current global energy production. In his study, Renforth (2013) also suggested that thermal requirements could be up to 5.6 GJ t^{-1} CO_2 and electrical up to 1.2 GJ t^{-1} CO_2.

Grinding limestone would require about 15-45 kWh t^{-1} (Harvey 2008). In addition to mining limestone, for effective dissolution to occur, the limestone would have to be ground down to 40-80 µm particles (Harvey 2008). Harvey (2008) further suggested that different sizes would be required for different locations, although grinding in any form would create an energy penalty.

Assuming rates of application between 10 g m^{-2} and 100 g m^{-2}, and an application width of 100 m, ships with the capacity of 2500 t, 25,000 t and 250,000 t would use 0.23, 0.19 and 0.64 MJ t km^{-1} (energy use per t of applied limestone as initial load times the distance required to disperse the full load) respectively, given different energy intensities required, energy uses, power, etc. (Harvey 2008). Loads between 50-100% capacity indicated energy use per km is independent of the load (IMO 2000). For ships with a capacity of 2500 t and a 100 g m^{-2} application, the energy requirement could be as low as 0.0575 GJ $t^{-1,}$ and for ships with a 250,000 t capacity and a 10 g m^2 application, the energy requirement could be as high as 16 GJ t^{-1} per traverse (Harvey 2008). Due to the low average payload, Harvey suggested using a central larger ship with smaller ships that would service it, reducing energy requirements (Harvey 2008).

Energy requirements for disposal or the method of disposal would also need to be considered, where Harvey suggested projecting the powder, and Caserini suggested using the ship's wake to their advantage (Harvey 2008; Caserini et al. 2021). The energy requirement of Harvey's method, based on a 50 m projection of the powder, is approximately 0.36 MJ t^{-1} (Harvey 2008).

2.2.8 Cost analysis

Renforth estimated that it would roughly cost $72 USD for 159 t^{-1} CO_2 for the typical kiln processing of converting limestone to lime, $159 USD t^{-1} CO_2 using solar calcination or $261 USD per t of CO_2 captured if amine scrubbing (carbon capture) technologies are used (Renforth et al. 2013). This would rise considerably if direct carbon capture were employed. As the current uncertainty around DAC suggests, costs range from $100 to $ 1000 USD per t of CO_2 captured (Renforth et al. 2013). OAE using lime could potentially become a better option if technologies and expenses were to improve/lessen. Harvey (2008) also analyzed the cost of grinding limestone to 10 µm particles using coal-generated electricity. He noted that that grinding alone would cost \$0.90-2.70 per t in addition to the cost of the facilities. He further estimated that at the time (2008), the estimated cost for limestone OAE application could be \$40-45 billion per year at a 4 Gt per year application rate.

The production and decarbonization of calcium hydroxide from calcium carbonate has a relatively large CO_2 and energy footprint (Ilyina et al. 2013). Decarbonization, requiring heat to burn off the CO_2, would produce CO_2 unless said heat was produced from renewables (in addition to the CO_2 released during the burning) (Ilyina et al. 2013). In an ideal scenario for slaked lime application, high energy costs and CO_2 addition could be virtually eliminated using renewables and carbon capture. If a fuel source like coal is required for grinding and the limestone itself were not supplied from, and mined near a coast, this would significantly offset the CO_2 absorbed through OAE (Harvey 2008). Of course, renewable or non-emitting energy sources would change this.

2.3 Biological & Environmental Limitations

2.3.1 Current status and gaps

The biological and ecological effects of OAE are not well studied (Burt et al. 2021; Caserini et al. 2021). Despite being a limiting factor to future implementations of OAE, there is currently a significant lack of research on this subject, consequently restraining any future developments. Several environmental and biological issues could arise due to OAE. Alterations to the carbonate system and increases in pH could affect marine organisms and sensitive ecosystems, and some may be detrimental if alkalinity is enhanced further (Renforth et al. 2013). Localized impacts of

OAE need to be studied for safe application, and further experimental studies are necessary to better understand the ecological effects (Caserini et al. 2021).

2.3.2 Mining, scale and disposal considerations

The extraction of limestone is yet another barrier to OAE. If OAE is to be used on a large scale for climate change mitigation and carbon dioxide removal, mining efforts will have to increase dramatically. Terrestrial ecosystems could face risks due to mining and production processes (Harvey 2008; Köhler et al. 2013; Keller et al. 2014). In addition, about 6% of global limestone outcrops overlap with natural protected areas (Caserini et al. 2022), creating yet another barrier to the implementation of OAE. As for processing, CO_2 efficiency and effectiveness would significantly decrease depending on the grinding technique for limestone, especially if powered by coal, reaching emissions of 0.012-0.018 t of carbon produced per t of $CaCO_3$ for disposal (Harvey 2008).

The potential for CO_2 removal through OAE could also be limited by the availability of resources and ensuring that the application rate isn’t large enough to impact marine environments negatively (Caserini et al. 2021). Though some studies have suggested the dangers of global application, Ilyina et al. (2013) indicates that a worldwide application has higher mitigation potential as regional applications would only have local effects unless applied on very large scales.

Large-scale regional applications could potentially pose several environmental threats, which are also difficult to predict. Large-scale OAE applications, along with the warming of oceans, are likely to cause significant unintended side effects in our oceans (González and Ilyina 2016). The ship’s size and source of fuel are substantial, and CO_2 emissions produced by the vessel depending on these factors can increase or decrease the effectiveness of the OAE application. As previously mentioned, Harvey (2008) recommended using smaller ships to distribute limestone OAE. This was based on his analysis of using diesel-run ships and calculating the ratio of CO_2 emitted to CO_2 absorbed.

Smaller vessels with OAE application (2500 t capacity) enabled the removal of about 19 times more CO_2 from the atmosphere than they emitted, whereas ships with a 25,000 t capacity enabled the removal of approximately 2.3 times more CO_2 than they emitted, and large vessels (250,000 t capacity) emitted more CO_2 than they enabled to be removed (Harvey 2008).

2.3.3 Carbonate chemistry and precipitation risks

Kheshgi (1995) showed that the quantity of calcium added to increase TA would be nearly equivalent to the number of moles of CO_2 sequestered, so increased calcium concentration could be a possible concern. Given the rate of calcium added compared to our current emission rate, it would only account for a relatively small increase in the total concentration of calcium moles dissolved in the ocean, leading to the thought that OAE would benefit the oceans as opposed to harming them (Kheshgi 1995). Marine biota are sensitive to changes in temperature, mixing and pH; the change in carbonate ion concentrations and decrease in CO_2 may affect some species, especially those with calcium carbonate shells (Kheshgi 1995). Where one species if effected, others might be as well, given the interconnectedness of ecosystems. For this reason, we cannot assume that OAE would be harmless to organisms given the sensitivities of the carbonate system.

Runaway $CaCO_3$ precipitation is a possible condition that can occur where the addition of alkalinity triggers calcium carbonate formation, causing more TA to be removed than was added

(Moras et al. 2022). This process would require an induced change outside of the natural carbonate system and so isn't commonly found in our current oceans. $CaCO_3$ precipitation can occur through several pathways, relying on the presence of existing minerals, through the formation of $CaCO_3$ crystals or the formation of crystals on solid minerals (Moras et al. 2022). In turn, the precipitation of calcium carbonate influences DIC and TA (Burt et al. 2021). The inorganic precipitation of $CaCO_3$ as a result of the Ω threshold being disturbed is currently understudied (Moras et al. 2022).

The stoichiometry behind carbonate precipitation is particularly concerning for OAE applications, since when 1 mol of $CaCO_3$ is precipitated, the TA of the solution decreases by 2 mol due to the removal of 1 mol of carbonate ions (Moras et al. 2022). Under a hydrated lime application, if runaway $CaCO_3$ precipitation occurred, this could reduce the efficiency of OAE CO_2 uptake from 0.8 mol of CO_2 per mole of TA added to 0.1 mol of CO_2 per mole of TA added (Moras et al. 2022).

It is possible that undissolved $CaCO_3$ that reaches the sea floor causes the once undersaturated deep water to become saturated or to a nearly saturated state. Limestone particles in the upper and supersaturated part of the water column could lead to calcium carbonate precipitation, eventually having the opposite intended effect of OAE (Harvey 2008). Currently, there is no particular and widely accepted measurement for the upper limit of $CaCO_3$ saturation (Feng et al. 2017). This reiterates the further need for studies on this topic. Under typical ocean conditions, $CaCO_3$ precipitation does not occur, though depending on the Ω threshold, it is possible, and Moras et al. (2022) suggest that it might be dependent on the aragonite saturation state. Moreover, aragonite saturation Ω has been used in the past as a metric to understand the saturation state of seawater (Feng et al. 2017). Keller et al. (2014) not only reduced the rate of ocean acidification but were also able to keep saturation states of aragonite and calcite higher than the other climate engineering mitigations they considered.

Secondary runaway precipitation (reducing atmospheric CO_2 uptake) could result from the rapid increase of carbonate ions in the water caused by alkalinity enhancement (Wang et al. 2022). It is important to note that this does not currently occur naturally in our oceans. This secondary precipitation would decrease pH and Ω and increase the CO_2 concentration in the seawater, lowering the oceans' ability for CO_2 uptake (Moras et al. 2022). The elevated (and in some areas surpassed) pH compared to preindustrial levels could very well lead to carbonate precipitation, lowering the value of alkalinity enhancement (Renforth et al. 2013). This is reiterated by Wang et al. (2022), who emphasize that considerable TA additions could raise local pH and Ω past preindustrial conditions, possibly risking new stressors on ecosystems and inorganic mineral precipitation. Specifically, in application areas, it is possible that increased TA could incite TA loss through mineral precipitation (Wang et al. 2022).

Moras et al. (2022) suggested that precipitation needs to be avoided not only because it will drastically reduce CO_2 efficiency, but also because it will enhance ocean acidification. Carbonate precipitation could be avoided via the occurrence of dilution through the addition of alkalinity in the wake of ships or through natural mixing (Caserini et al. 2021; Moras et al. 2022). In addition, Moras et al. (2022) suggested that it could be avoided if a 1:1 dilution of TA-enriched seawater was performed, as the quicker the process, the less precipitation there would be. Although carbonate precipitation is of great concern, especially with limestone application, Moras et al. (2022) highlighted that not only do quick lime and hydrated lime both dissolve within a few hours, but that both dissolve without inducing $CaCO_3$ precipitation.

2.3.4 Marine ecosystem impacts

Depending on the material used for alkalinity addition, certain substances release toxic heavy metals, which could also impact biogeochemical cycles, marine ecosystems, and the food chain, possibly harming human populations (Hartmann et al. 2013; González and Ilyina 2016). Surprises between limestone application and marine organisms could arise, as the interactions have many uncertainties (Harvey 2008). The penetration of solar radiation into the mixed layer could be decreased through limestone OAE, increasing the albedo, and although this could have a cooling effect, it could also reduce the strength of the biological pump (Harvey 2008). This effect would lessen if the application were only done a few times a year (Harvey 2008). Minor additions of lime indicate smaller increases in pH, so there are minor environmental impacts (Renforth et al. 2013).

González and Ilyina (2016) submit that in their study, alkalinization can raise pH and Ω by 2100 to beyond modern levels. Although their simulation successfully mitigated ocean acidification, the pH and Ω levels were greatly exceeded, which could have adverse and detrimental effects on marine organisms (Renforth et al. 2013; González and Ilyina 2016). On the other hand, while pH and Ω were exceeded, specifically in shallow waters, this was not observed in deep water, where the reaction with carbonate sediments and undersaturated water would prevent AOA, preventing deep ocean acidification (González and Ilyina 2016).

The impact on biogeochemical cycles could especially arise in the Arctic and Tropical Oceans, possibly causing unknown ecological changes (González and Ilyina 2016). Potential effects could arise as OAE influences environmental conditions, such as increased calcite saturation states and pH (Feng et al. 2017; Renforth and Henderson 2017; Gore et al. 2019).

An increase in seawater pH could potentially have damaging effects on marine environments (Bach et al. 2019; Caserini et al. 2021). The current conditions of surface waters supersaturated with calcium carbonate are essential for many organisms (Harvey 2008). OAE could affect primary producers' nutrient limitation, which would in turn cause physiological damage and alter their metabolic balance (Cripps et al. 2013; Bach et al. 2019). The addition of alkalinity could threaten the acid-base balance of some marine organisms, specifically robust crustacean species (Cripps et al. 2013; González and Ilyina 2016). Harvey (2008) highlighted that with the use of limestone powder for OAE, the only likely impacts would be the neutralization of ocean acidity and the increase of $CaCO_3$ supersaturation; however, he mentioned the possibility of zooplankton ingesting the particles. Conservative discharge values should be applied to reduce the risk of the alkalinity discharge affecting marine life (Caserini et al. 2021).

Finally, Jin and Cao (2023) proposed that sudden termination of alkalinity addition would cause a rapid rebound of ocean acidification in the area of application, suggesting that OAE could be more of a band-aid or short-term solution.

2.3.5 Regulatory and legal considerations

Given the elevated pH within OAE simulations, further research into the environmental impacts is required to apply limestone and lime dumping safely. Originally put forth during the 1972 London Convention, the London Protocol Treaty on Prevention of Marine Pollution prohibits the dumping of wastes and other matter in our oceans (ECCC 2022). In 2006 and 2009, the protocol was amended to enable sub-seabed carbon storage (ECCC 2022). In 2019, a new resolution was adopted to allow CO_2 export for storage in other countries, in addition to expected further regulation on ocean fertilization (ECCC 2022). OAE does possibly pose a way around the permits required for dumping, where OAE is the treatment itself for wastewater and power plant cooling.

The United Nations Convention on the Law of the Sea was adopted in 1982 and put forth to govern the uses of our oceans and their resources (IMO 2019). The convention was signed to regulate the conservation and sustainable use of biological diversity beyond national jurisdiction (BBNJ), marine genetic resources, area-based management tools (ABMT), including marine protected areas, environmental impact assessments (EIA) and capacity-building and the transfer of marine technology (CB&TMT) (IMO 2019). Although it is imperative to regulate and be beneficial to the environment and aquatic creatures, as of right now, these treaties are one of the several barriers to advancing OAE implementation.

2.3.6 Biological & Environmental benefits & considerations

One of the major benefits of OAE is, of course, the potential to combat ocean acidification, which, if left unmitigated, would likely have many adverse effects on marine biodiversity and ecosystems. Several studies show these benefits. If OAE mitigated ocean acidification, it would protect marine ecosystems like tropical coral reefs (Feng et al. 2016; Feng et al. 2017). This in turn would prevent decreases in calcification rates and changes in reproduction and physiology in some aquatic organisms (Gattuso et al. 1998; Ilyina et al. 2013). Ilyina et al. (2013) suggested that, given the elevation in biogeochemical parameters as a result of ocean acidification, alkalinity addition would be able to combat this significantly (given a 2.4 Pmol yr $^{-1}$ alkalinity input from 2020-2030), lessening the environmental stress generated by said change.

For nearly a century, acetic acid and hydrated lime have been used as a way to control invasive species amongst shellfish aquacultures as a natural form of “pest management” (Locke 2009). Although considered as environmentally safe chemicals, they alter the pH and therefore affect biological and geochemical processes such as photosynthesis, metabolic processes, sediment diagenesis and calcium carbonate sedimentation and dissolution (Locke 2009). It has been suggested that the timing of lime treatments should consider the seasonal cycles of vulnerable organisms like larvae, especially those that are sensitive, like lobster larvae (Locke 2009). Alkalinity enhancement could also counter the reduction of net primary production caused by higher nutrient limitation (Bopp et al. 2013; González and Ilyina 2016).

2.4 OAE Modelling

The following section will review previous OAE modeling efforts and studies for atmospheric emission reduction. It is organized thematically with emphasis and detailed explanations on studies in relation to this thesis, beginning with limestone-based and lime-based global and regional studies analyzing future scenarios, see Table 1. It then highlights other works, still important to OAE literature, such as OAE models focused on TA additions based on other minerals, OAE models using other minerals, lab work, field work and pilot projects (Tables 2-4).

2.4.1 Limestone and Lime based

The work of Harvey (2008) was influenced by work from Caldeira and Rau (2000), who undertook quite extensive research and modelling into ocean alkalinity reactions when limestone was mixed with pumped seawater at a site on land and then piped and distributed over the ocean. Harvey (2008) proposed to sprinkle the crushed limestone directly over the ocean surface thereby avoiding outgassing and leveraging upwelling. He modelled the application of 4 Gt of $CaCO_3$ over several timescales (50 years, 100 years, 200 years). The intentions of the study were to 1) understand suitable conditions for limestone powder absorption; 2) find the maximum effectiveness of absorption of CO_2 for limestone addition; 3) understand the potential effects on pH and supersaturation in surface waters (Harvey 2008). The main areas of Harvey's focus in his analysis were the northern hemisphere polar downwelling regions and the southern hemisphere polar upwelling regions. Overall, Harvey (2008) found that this amount of $CaCO_3$ could absorb 1050 Mt of CO_2 after 200 years, although it would be quite expensive with high energy requirements. He also found that OAE works best in conjunction with other carbon-reducing technologies, and that although it is a promising approach, there would need to be significant investment, infrastructure development, further research on ecological impacts and the application of pilot studies before large-scale implementation.

The pioneering study of Ilyina et al. (2013) explored the consequences of the addition of alkalinity by a ratio of 2:1 (TA: CO_2). They used the Hamburg Ocean carbon cycle model under the IPCC A1B scenario (similar to the RCP 6.0 scenario) to model the consequences of adding 2.4 Pmol per year of alkalinity (approximately 89 Pg) from 2020 to 2030 using lime. Alkalinity was increased proportionally to anthropogenic CO_2 emissions, and in the first scenario, alkalinity was uniformly distributed over the surface of the entire ocean. The same was done in additional experiments, though TA was added to 47 million km^2 over the Atlantic and Pacific Oceans (Ilyina et al. 2013). The study found that adding alkalinity with a 2:1 ratio could keep atmospheric CO_2 below 520 ppm by 2100 compared to a minimum of 900 ppm in an unmitigated scenario. It was also found that pH levels would be maintained close to current levels, leading to the counteraction of ocean acidification. Although atmospheric CO_2 and pH did not return to unmitigated levels, they showed that the effects of alkalization persisted after the end of its application. Small-scale applications were shown to counteract ocean acidification on local scales and in upwelling systems, while global applications were the most effective and required massive amounts of alkalinity (Ilyina et al. 2013). Additionally, they found that large-scale applications altered ocean chemistry significantly and led to $CaCO_3$ precipitation. Ilyina et al. (2013) concluded that although alkalinity enhancement has excellent potential, due to the risks, the energy requirements and the massive upscaling of lime production required, it might be a better solution for local ocean acidification rather than global application.

Building on earlier work, Keller et al. (2014) used the UVic ESCM with an RCP 8.5 scenario to evaluate five different CO_2 removal methods, one being ocean alkalization based on maximum current transport capacity. The ocean alkalinization (OA) model used an alkalinity discharge of 10 Pg of $CaOH_2$ based on the maximum carrying capacity of large cargo and tanker ships; the total alkalinity was increased evenly in surface waters between 70° north and 60° south. By the year 2100, ocean alkalinity enhancement had lowered CO_2 atmospheric concentrations by 48 ppm, leading to slightly lower surface air temperatures (Keller et al. 2014). Carbon dioxide removals' effectiveness increased over time due to the ocean's buffering capacity remaining high. Despite this, the rate of ocean acidification was reduced. Keller et al. 2014 mentioned that side effects specifically on marine ecosystems, should be considered in addition to the impact of increased mining and production (Keller et al. 2014). Lastly, they noted that the main limitation was the production or capacity to deliver the limestone from which the $CaOH_2$ originated, in addition to the large energy requirements throughout production, transportation and distribution.

Gonzalez and Ilyina (2016) took a geoengineering perspective where they modelled ocean alkalinity enhancement under an 8.5 RCP scenario, aiming to stabilize atmospheric carbon levels to RCP 4.5 levels. Here, the Max Planck Institute Earth System Model (MPI-ESM) and the HAMburg Ocean Carbon Cycle Model (HAMOCC) were used with alkalinity added to the surface (first 12 m of the ocean) and gradually increasing over time from 2018 to 2100, eventually totalling an addition of 114 Pmol of alkalinity. They found that 940 Gt of carbon could be removed from the atmosphere; that marine DIC content would double; that RCP 8.5 levels would stabilize to RCP 4.5 levels; and that there would be a 1.5 K reduction in global mean air surface temperature. In addition, they found that TA concentrations were increased by around 30%, and that carbon uptake showed more promise within latitudes from 30° to 80° and that Arctic and tropical oceans are ‘hot spots’ for biogeochemical change. They concluded that olivine or lime production would require a massive upscaling of 2 to 3 orders of magnitude, that many unknown ecological and biological challenges could come with this, that it only partially addresses deep ocean acidification and that although AOA shows potential, regional impacts need to be carefully considered.

Feng et al. (2016) further explored the AOA potential on ocean acidification and the protection of tropical coral ecosystems. Their regional study looked at the Great Barrier Reef, the Caribbean and South China Sea regions, accounting for oceanic surface, aragonite saturation, and surface pCO_2 (Feng et al. 2016). Once more, they used the UVic model under an RCP 8.5 scenario with alkalinity added in the upper 50 m of the specified regions based on lime addition. Their study found that AOA could prevent local acidification in these regions, although the areas require different amounts of lime, with a total of 356 Gt of lime implemented over the 80-year scenario (Feng et al. 2016). Importantly, it was mentioned that AOA needs to continue, otherwise the ocean chemistry would rapidly return to high acidification. Although the primary focus of their study was ocean acidification, their simulations showed atmospheric CO_2 drawdowns by 2099, where the Great Barrier Reef saw a 7 ppm reduction, the Caribbean Sea saw a 15 ppm reduction, and the South China Sea saw a 16 ppm reduction (Feng et al. 2016). They suggested that although AOA is not a replacement for CO_2 atmospheric reductions, it could act as a short-term Band-Aid solution with respect to ocean acidification.

The study of Sonntag et al. (2018) compared climate engineering methods: 1) solar radiation modification (SRM) through stratospheric sulphur injection; 2) CDR through ocean alkalization; 3) CDR through afforestation (Sonntag et al. 2018). The MPI–ESM model was also used under an RCP 8.5 scenario for all three studies, with the aim to eventually reach RCP 4.5 levels (Sonntag et al. 2018). For their ocean alkalinity study, 114 Pmol of alkalinity was added uniformly to the surface layer (upper 12 m) of the ocean by 2100, which was based on earlier studies (González and Ilyina 2016; Sonntag et al. 2018). They showed that surface air temperature could be reduced by 1.55 K (average between 2081-2100 and atmospheric CO_2 by 430 ppm (by 2100). They further found that 905 Gt of carbon was sequestered from the atmosphere and 36 Gt from land, resulting in 941 Gt of carbon added to the ocean. Lastly, they showed increased ocean buffering capacity, enhanced CO_2 solubility, reduced marine & terrestrial net primary productivity (NPP), and a better nutrient supply in surface waters due to lower stratification.

On the other hand, Lenton et al. (2018) used the CSIRO-Mk3L-COAL Earth System Model (ESM) to compare an RCP 8.5 scenario and an RCP 2.6 scenario. The model was run from 2020 to 2100 with an annual global discharge of 0.25 Pmol per year (9.3 Pg $CaOH_2$) over the latitude bands: Subpolar, Subtropical, and Tropical. They found that under the high emission scenario, atmospheric CO_2 could be reduced by 16%, although it was less effective in reducing ocean acidification. The low emission scenario showed more promise in reducing CO_2 and ocean acidification, eventually maintaining temperatures near 2020 levels by 2100 (Lenton et al. 2018). Further, their study found similar responses to alkalinity between global and regional applications, with tropical applications showing the best results for preserving alkalinity in surface waters and subpolar regions with the least retention. Again, Lenton et al. (2018) found that OA alone would not be enough under a high-emission scenario to address atmospheric CO_2 emissions.

Butenschon et al. (2021) built on earlier research using the Nucleus for European Modelling of the Ocean (NEMO) and biogeochemical flux model (BFM) models under a 4.5 RCP scenario. They modelled two OAE scenarios in the Mediterranean basin between 2021 and 2050 using lime as the alkalinity source. Compared to previous studies, this model did not use an idealized, spatially homogeneous scenario, as it was practically hard to implement. The study wanted to use a heterogenous approach, making it more realistic by not assuming uniform addition to the entire ocean. Instead, their OAE interventions were based on current shipping routes throughout the Mediterranean to analyze the total possible efficiency of alkalization using the existing network of cargo ships and tankers. For the first scenario (CTS200), a total of 6.0 Gt of $Ca(OH)_2$ was released using a constant discharge flux of 200 Mt/yr (Butenschön et al. 2021). This corresponds to 7.58 kg of $Ca(OH)_2$ added to the surface total alkalinity for each ship (Butenschön et al. 2021). They found that surface alkalinity could increase by 80 μmol kg^{-1} during the first few years, gradually increasing to 2760 μmol kg^{-1} by 2050. For the second scenario, 3.2 Gt of $CaOH_2$ was added, with the amount annually increasing gradually, proportional to surface pH in the Mediterranean. This gradual increase eventually reached 2740 μmol kg^{-1} by 2050 (Butenschön et al. 2021). This was done to allow the mean surface pH to stabilize. This resulted in an increase of the carbon uptake potential, efficiency and air sea flux where at the end of the simulation air sea flux on an annual increase of 80 Gt of carbon per year at the end of the simulation. Scenario 2 showed stronger results for ocean acidification than scenario 1. Although both scenarios had increased CO_2 uptake and pH, the first scenario showed a more substantial increase in CO_2 uptake and pH (Butenschön et al. 2021). However, the second scenario was deemed as more effective as it used less lime, avoided sudden pH changes that could possibly harm wildlife, better stabilized the pH levels and counteracted acidification per mole of lime more efficiently. Due to their high load capacity and coverage of the Mediterranean, they identified container ships as the most ideal disposal method for OAE. Lastly, the study showed that dense water around gyres in the Mediterranean could only increase carbon absorption efficiency, and these locations might be more beneficial than other parts of the Mediterranean (Butenschön et al. 2021).

Burt et al. (2021) examined the impact of ocean patterns on alkalinity distribution and carbon uptake efficiency (Burt et al. 2021). Their study explored regional versus global OAE applications, specifically looking at the non-linear marine carbonate system and its sensitivity to OAE (Burt et al. 2021). They used the MPI Ocean Model for 75-year simulations with total alkalinity increasing by 0.25 Pmol annually both globally and in 8 regions. Their study found that 82–175 Pg of carbon was sequestered regionally compared to the 156 Pg of carbon sequestered globally. They further found that the sequestration efficiency was highest in the Southern Ocean, even compared to the global experiment, despite a surface area 40 times smaller, and therefore, the surface pattern of total alkalinity played a massive role in carbon uptake. Overall, the study showed that surface currents, vertical mixing, and circulation patterns were critical, and that local conditions and transport mechanisms must be carefully considered in OAE regional applications (Burt et al. 2021).

Caserini et al. (2021) analyzed the potential of existing and new cargo ships for deploying calcium hydroxide (slaked lime) OAE on both regional and global scales. Their seminal study explored the restraints around potential discharge, such as the number of existing vessels, load capacity, routes, reloading and discharge rates. They recognized the potential of using “milk of lime”, a slurry of slaked lime mixed with water, which could be prepared on board ships. As this study focuses on the shipping and disposal potential of lime, much of the methods section reviews the differences between ships, their emissions and the dilution rates in the wake of ships (Caserini et al. 2021). Their study also used a three-dimensional non-reactive model to assess discharge and flow in the wake of a vessel, in addition to a one-dimensional reactive model to determine the dispersion of lime particles. The study showed that given the current traffic for bulk carriers and container ships, the maximum potential on a global scale for OAE was between 1.7 and 4 Gt of lime discharge per year, potentially removing 1.5-3.3 Gt of atmospheric CO_2 per year. For the Mediterranean, they calculated that potential discharge could be 186 Mt per year (Caserini et al. 2021). Lastly, by adding 1000 new ships dedicated to OAE, the potential Mediterranean discharge rose to 1.3 Gt per year. Lastly, Caserini et al. (2021) recognized the need for further understanding of OAE's ecological and biological implications and the potential constraints of limestone mining, slaked lime production and further technological challenges like port and ship modifications.

Fakhraee et al. (2022) also compared the environmental impacts and effectiveness of different OAE methods. This synopsis will focus mainly on the CaO aspects of the paper. The IOTA biogeochemical model was designed and used to assess the impacts of carbon capture through mineral feedstock alkalinity addition (Fakhraee et al. 2022). Their study shows that with small grain sizes and high dispersion rates, olivine's near-surface release of alkalinity is limited however, MgO and CaO show prompt near-surface dissolution with ideal depth-integrated alkalinity. Their CaO results showed an ideal feedstock application where alkalinity production was nearly complete in the upper mixed layer, independent of application rate. This showed that alkaline metal oxides like CaO could promote CO_2 uptake in regions with shallow mixed layer depths. In addition, CaO shows little impact on particle cycling in the shallow ocean, whereas MgO shows more complex potential adverse reactions and implications on surface marine ecosystems, and natural silicates like olivine and basalt showed significant negative potential for surface ecosystems (Fakhraee et al. 2022). Next, the cGENIE model was used with the IOTA to analyze the carbon chemistry and global potential for deployment scenarios where a benchmark application rate of 300 g m^{-2} y^{-1} corresponds to an overall max flux between ~6-30 Gt of feedstock. These simulations suggested that the CaO and MgO deployments would promote biotic calcification while avoiding calcium carbonate abiotic nucleation and precipitation. By 2100, the CaO simulation resulted in a CDR rate amounting to 5.7-20.4 Gt of CO_2 annually and the authors suggest that although the potential for carbon capture with synthetic materials is significant, it will most likely be limited by constraints on feedstock supply (Fakhraee et al. 2022).

Finally, Jin and Cao (2023) assessed the response of the carbon cycle to both CDR and SRM. They also used the UVic ESCM to conduct model integration from 2020-2100 using forced historical emissions. They examined results from an AOA scenario driven by RCP 8.5 with an alkalinity addition flux based on Keller et al. (2014). They applied the alkalinity flux to ice-free oceans in order to lower and maintain atmospheric CO_2 at RCP 4.5 levels. In additional scenarios, they also followed Lenton (2018) by applying AOA regionally at different latitudinal bands and in another, terminating the simulations by 2075 to analyze the effects of ceasing AOA. The AOA scenario, where 127 Pmol of alkalinity was added, increased CO_2 uptake from 397 PgC to 1550 PgC, raising global mean surface pH by 0.42 (Jin and Cao 2023). Although the sudden termination of AOA in 2075 led to a gradual increase in surface air temperature (SAT), a greater effect on acidification was noted, where pH and aragonite saturations decrease rapidly. The regional scenarios found the lowest increase in global surface alkalinity in subpolar regions (likely due to strong surface-to-seep mixing) and the lowest CO_2 uptake in subtropical areas (Jin and Cao 2023).

Author	Type of Model	Focus	Mineral
(Harvey 2008)	Regional (North Atlantic)	Absorption, effectiveness & pH	Limestone ($CaCO_3$ slurry)
(Ilyina et al. 2013)	Global	General, acidification	TA based on lime
(Keller et al. 2014)	Global	CDR's, max carrying capacity	TA based on lime
(González and Ilyina 2016)	Global & Regional	Carbon cycle and climate	TA
(Feng et al. 2016)	Regional	Coastal protection and acidification	Hydrated lime
(Sonntag et al. 2018)	Global	Global carbon cycle impacts	TA
(Lenton et al. 2018)	Global	Acidification mitigation & efficiency	TA based on lime
(Butenschön et al. 2021)	Regional	Shipping routes, disposal	Lime TA
(Burt et al. 2021)	Global	Efficiency, distribution and timing	TA (assumed from lime)
(Caserini et al. 2021)	Regional	Shipping routes, disposal, ability to dissolve	Lime-based TA
(Fakhraee et al. 2022)	Global & Regional	Effectiveness, dissolution, ecological risks	Lime & other
(Jin and Cao 2023)	Global & Regional	Termination effects	Lime-based TA

Table 1: Limestone-based and lime-based modelling studies

2.4.2 Other minerals

Table 2 presents an overview of OAE models outside of limestone-based and lime-based scenarios. A large focus is placed on olivine simulations and TA applications based on olivine.

Author	Type of Model	Focus	Mineral
(Hangx and Spiers 2009)	Regional	Coastal viability	Olivine
(Köhler et al. 2010)	Box model	Enhanced silicate weathering	Olivine
(Köhler et al. 2013)	Global	Dissolution kinetics and long-term impacts	Olivine
(Hauck et al. 2016)	Regional	Biogeochemical cycles	TA, Olivine
(Feng et al. 2017)	Regional/global coastal	Coastal ocean alkalinization	Olivine TA
(González et al. 2018)	Global	Differences in modelling	Several
(Köhler 2020)	Global	Updated dissolution kinetics	Olivine
(Wang et al. 2023)	Regional	Ecosystem responses	TA, Sodium hydroxide
(Kwiatkowski et al. 2023)	Regional	Ecosystem impacts	Olivine
(Yang et al. 2023)	Regional	Dissolution effectiveness	Magnesium hydroxide
(Nagwekar et al. 2024)	Global & regional	Deep water formation	Olivine
(Palmiéri and Yool 2024)	Global/Coastal regions	Coastal efficacy	Olivine TA

Table 2: Total alkalinity & other mineral modelling studies

2.4.3 Lab & field work

Table 3 documents experimental OAE studies with both lab and field work included.

Author	Type	Focus	Mineral
(Cripps et al. 2013)	Lab control tests	Testing impact on crabs and Ca concentrations	Calcium hydroxide
(Gore et al. 2019)	Lab work	Effect on algae	TA
(Bach et al. 2019)	Lab experiment	Shellfish response	Sodium hydroxide
(Moras et al. 2022)	Lab work	Calcium carbonate precipitation	Quick lime and hydrated lime
(Ferderer et al. 2022)	Lab mesocosm	Phytoplankton responses	Sodium hydroxide
(Hartmann et al. 2023)	Lab experiment	Carbonate precipitation	Several
(González-Santana et al. 2023)	Lab/field hybrid	Follow up study	Several
(Fuhr et al. 2023)	Field trial	Sediment application	Calcite
(Ringham et al. 2024)	Lab work	Efficiency, precipitation and potential	Sodium hydroxide
(Kitidis et al. 2024)	Field test	Wastewater application	Magnesium hydroxide
(Suitner et al. 2024)	Lab work	Precipitation, application, safety	TA
(Paul et al. 2025)	Lab work, mesocosm	Ecosystem impacts, C:N changes, durability	Carbonate based alkalinity

Table 3: Lab & field work

2.4.4 Pilot projects

Table 4 presents real-world pilot projects testing OAE, highlighting the shift in recent years from lab work as seen in Table 4 to operational deployment. The purpose of this table was not for completion but to demonstrate recent efforts.

Project	Year	Where	Focus	Mineral
Project Vesta	2019-2022	Beach trials, Dominican Republic	Olivine weathering, dissolution, ecosystem	Olivine sand
Planetary technologies	2022-current	Halifax Harbour, Canada	Harbour deployment, carbon uptake, biological effects	Mine tailings alkaline solution Brucite (Magnesium hydroxide)
Coastal Minerals	2022	UK coastal waters	Weathering, dissolution and chemistry changes	Crushed limestone
Port of Rotterdam olivine project	2022-current	Netherlands, coastal	Runoff and weathering	Olivine sand
COTEC mesocosms	2022-2023	Offshore Portsmouth, UK	Ecosystem responses	Alkaline materials, several
OceanNETs KOSMOS	2023	Gran Canaria, Spain	Offshore mesocosm platforms, biological monitoring	Calcium hydroxide
Vesta Portland	2023-current	Oregon coastal	Chemistry changes	Olivine sand
Seafield Carbon	2023	Scotland	High energy coastal environments	Agricultural lime
Blue Sybiosis	2024	Great Barrier reef, Australia	Coral calcification responses	Calcium hydroxide

Table 4: Pilot projects

Chapter 3: Assessing the Effectiveness of OAE

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3.1 Methods

3.1.1 UVic Model

In this study, we use Version 2.9 of the UVic ESCM comprising a two-dimensional atmospheric energy-moisture balance model (Fanning and Weaver 1996; Weaver et al. 2001), a three-dimensional ocean general circulation model (Pacanowski 1995), a dynamic-thermodynamic sea ice model (Bitz et al. 2001) with elastic visco-plastic rheology (Hunke and Dukowicz 1997), a comprehensive carbon cycle model, and a land surface and dynamic vegetation model based on the Hadley Centre Met Office Surface Exchange Scheme (MOSES) coupled to the Top-down Representation of Interactive Foliage and Flora Including Dynamics (TRIFFID) model with five plant functional types (Cox 2001; Meissner et al. 2003). The model resolution is 3.6° (in the zonal direction) and 1.8° (in the meridional direction) and the ocean model has 19 vertical levels, with thickness varying from 50m near the surface to 500m in the deep ocean. Surface wind stress and winds are specified from 40-year climatological reanalysis data from the National Centers for Environmental Prediction (NCEP) project (Kalnay et al. 1996). An oxic-only ocean sediment model has been added to the UVic ESCM, whereby simulations of $CaCO_3$ preservation in deep-sea sediments are represented (Archer 1996), in addition to a nutrient-phytoplankton-zooplankton-detritus marine ecosystem model component (Schmittner et al. 2008). Eby et al. (2009) provide a more comprehensive discussion of the carbon cycle and how it is incorporated into the UVic ESCM. In addition, the UVic ESCM model, along with its simulations of past, present and future climates, has been extensively evaluated against observations, other intermediate complexity and more comprehensive models, and used as an assessment tool in the IPCC 3rd (IPCC 2001), 4th (IPCC 2007), 5th (IPCC 2013), and 6th (IPCC 2021), scientific assessments.

3.1.2 Experimental Design

In total, we used the UVic ESCM for 16 separate experiments. First, the model was initialized with a 10,000-year spin-up at the year 850. From 850 to the year 2000 historical forcing (land cover change, solar, volcanic, aerosol, other GHGs) was used to force the model. The atmospheric CO_2, globally-averaged surface air temperature and globally-averaged ocean surface pH at year 2000, the end of this spin-up period, are 367.7 ppm, 14.1 °C, and 8.07, respectively.

The model was then integrated from 2000 to 2300 with changing atmospheric CO_2, land surface, aerosol and other greenhouse gas forcing following each of RCP 2.6, 4.5, 6.0 and 8.5 extended scenarios (Moss et al. 2008). From 2000-2300, the equivalent CO_2 emissions were diagnosed (Zickfeld et al. 2013) and then used in subsequent sensitivity experiments. From 2300 to 2500, atmospheric CO_2 and other RCP forcings were held fixed (at their year 2300 values) so that the diagnosed emissions of CO_2 gradually reach equilibrium and approach zero after year 2300. This process was followed to ensure that our OAE intervention experiments allowed atmospheric CO_2 to evolve freely away from prescribed RCP values to see the true effectiveness of OAE without the constraints of the RCP values.

The diagnosed emissions from the four RCP simulations described in the previous paragraph were then used in three OAE intervention experiments conducted for each RCP scenario. In the first set of experiments (Figure 3), termed *$CaCO_3$* below, the global mean production of limestone (6.6 Gt/yr) was distributed uniformly over the entire ocean annually for 500 years. In doing so, it was assumed that all the calcium carbonate dissolved immediately once applied to the ocean surface, thereby offering an upper-bound estimate of OAE potential.

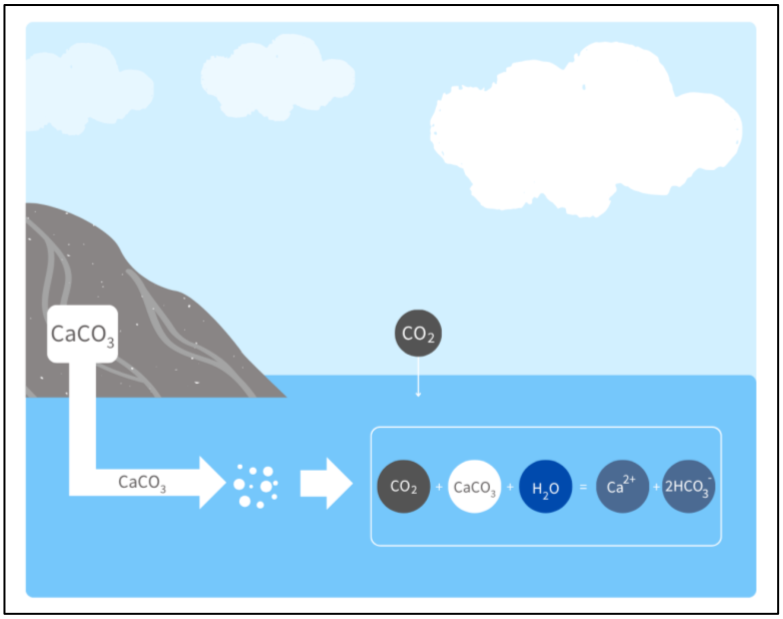

Figure 3: *$CaCO_3$* run diagram

In the second set of experiments (Figure 4), termed *$CaO+CO_2$*, we assume the global annual mean limestone production was entirely converted to lime on land and this lime was then distributed uniformly over the entire ocean annually for 500 years. The CO_2 arising from the production of lime was added to the atmosphere.

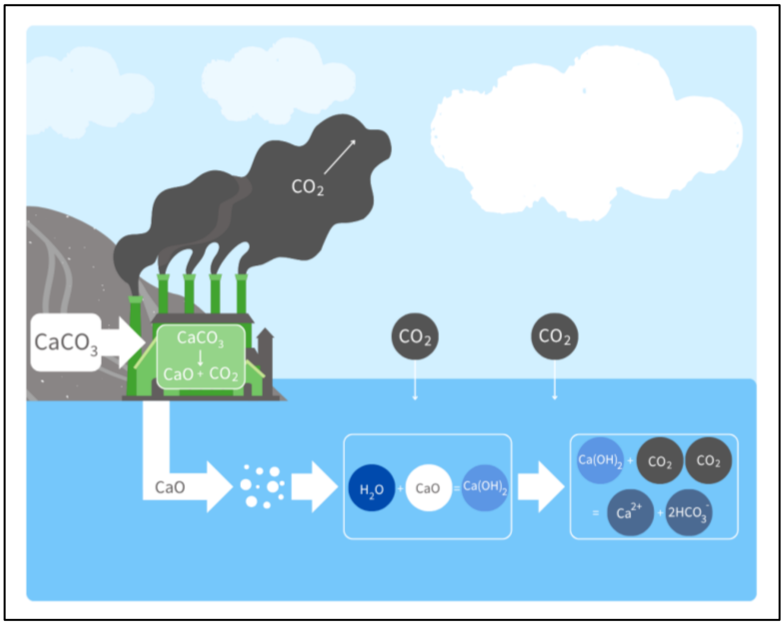

Figure 4: *CaO* + *CO₂* run diagram

In the third set of experiments (Figure 5), termed *CaO*, we repeated the *CaO+CO₂* experiment but now assumed that all CO$_2$ produced in making lime was sequestered.

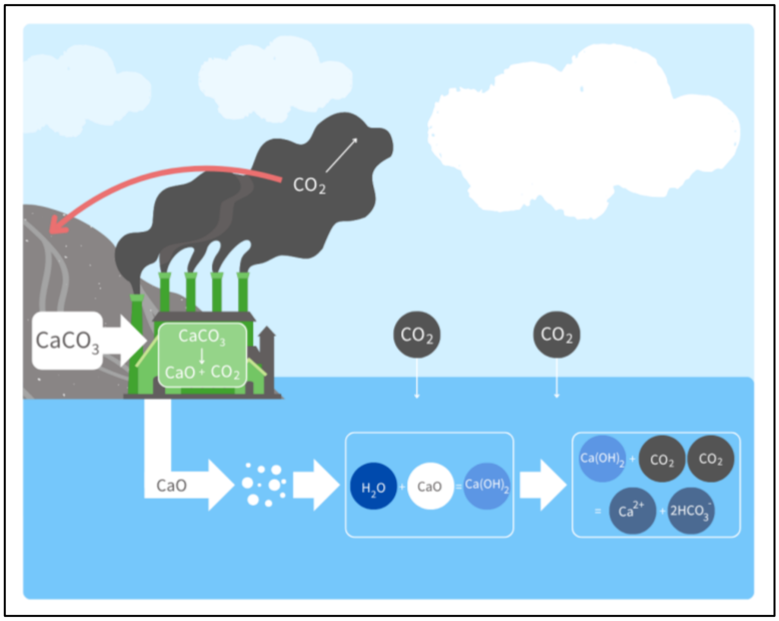

Figure 5: *CaO* run diagram

Finally, we also conducted four control integrations (one for each RCP) where the UVic ESCM was forced with the diagnosed emissions and no OAE interventions.

3.2 Results

The atmospheric concentration of CO_2, globally averaged surface air temperature, and globally averaged ocean surface pH over the 500-year integrations for each RCP control experiment, and each of the three OAE intervention experiments associated with each RCP are illustrated in Figure 1. Table 5 summarizes the year 2500 atmospheric CO_2 concentration, globally averaged surface air temperature, and globally averaged ocean surface pH for each RCP as an anomaly relative to the respective control integration at year 2500. Atmospheric CO_2 and surface air temperature anomalies are both negative as each OAE intervention reduces these metrics from their respective control integrations. On the other hand, the OAE interventions increase pH from the respective control integrations.

In the first OAE intervention, the present-day global and annual production of limestone ($CaCO_3$) was distributed uniformly across the surface of the ocean and assumed to dissolve immediately. The net effect after 500 years was to reduce the atmospheric CO_2 concentration by 38-145ppm (depending on the RCP) from the control simulation. Surface temperatures responded by being 0.27-0.49°C cooler, and the ocean surface pH became 0.06-0.075 higher than the control simulation, depending on the RCP. While the RCP 8.5 simulation had the largest drop in atmospheric CO_2 associated with the $CaCO_3$ OAE intervention, it also had the smallest response in temperature. This is attributed to the logarithmic response of radiative forcing to atmospheric CO_2 concentration, such that at high CO_2 levels, the addition of one CO_2 molecule has less effect on radiative forcing than at lower CO_2 levels.

RCP scenario	Atmospheric CO_2 (ppm)			Surface air temperature (°C)			Sea surface pH		
	$CaCO_3$	$CaO + CO_2$	CaO	$CaCO_3$	$CaO + CO_2$	CaO	$CaCO_3$	$CaO + CO_2$	CaO
RCP 2.6	-38	-31	-89	-0.46	-0.36	-1.2	0.066	0.062	0.13
RCP 4.5	-63	-62	-141	-0.50	-0.49	-1.3	0.073	0.075	0.14
RCP 6.0	-86	-84	-191	-0.49	-0.49	-1.2	0.075	0.076	0.15
RCP 8.5	-145	-144	-309	-0.27	-0.26	-0.58	0.060	0.061	0.10

Table 5: Run results

Annually averaged global mean atmospheric CO_2, surface air temperature and sea surface pH changes from the RCP 2.6, 4.5, 6.0 and 8.5 control integrations at year 2500, 500 years after the mitigative interventions were initiated. *$CaCO_3$* corresponds to the limestone OAE experiment, *$CaO+CO_2$* corresponds to the lime OAE experiment with the CO_2 produced in creating lime from limestone added to the atmosphere, and *CaO* corresponds to the lime OAE experiment with the CO_2 produced in creating lime from limestone captured and sequestered permanently.

For the second intervention, we assumed that the annual mean production of limestone was converted to lime on land and then sprinkled uniformly across the global ocean for 500 years. The CO_2 produced in the conversion of $CaCO_3$ to CaO was released into the atmosphere. After 500 years and relative to the respective RCP control runs, the *$CaO+CO_2$* experiments led to a reduction in atmospheric CO_2 of 31-144 ppm, a reduction in surface air temperature between 0.26 and 0.49°C, and an increase in surface pH of 0.061-0.076. The results are almost identical to those obtained in the *$CaCO_3$* OAE intervention experiments for RCP 4.5, RCP 6.0 and RCP 8.5 (Table 5 and Figures 6-11), with only minor differences occurring in the RCP 2.6 case. This result was to be expected as the CO_2 arising from the production of CaO was released into the atmosphere, and the equilibration time of CO_2 across the air-sea interface is rapid. While each CaO molecule can draw down two molecules of CO_2 (Eqs 7 & 8) and each $CaCO_3$ can only draw down one molecule of CO_2 (Eq. 5), there is an additional molecule of CO_2 in the atmosphere. That is, adding CaO instead of $CaCO_3$ to the surface waters of the ocean will make little difference unless the CO_2

released in the production of CaO is also sequestered (not accounting for differences in dissolution).

In the case of RCP 2.6, there are slight differences between the results of the *$CaCO_3$* and *$CaO+CO_2$* experiments both during (Figures 6-11) and at the end (Table 1) of the 500-year integration. Once more, this should be expected as RCP 2.6 implies the introduction of negative emission technology to draw down the atmospheric levels of CO_2 seen in the RCP 2.6 control run shown in Figure 6. The positive radiative forcing in RCP 4.5, RCP 6.0 and RCP 8.5 tends to increase the stratification of the ocean surface waters whereas when negative emissions are introduced, this would tend to increase the likelihood of convective overturn (a nonlinear process). Convective overturn is more likely in a negative emissions scenario (RCP 2.6) due to CO_2 undersaturated surface waters changing the air-sea gradient and so vertical mixing, possibly enhancing CO_2 uptake. Zickfeld et al. (2021) have noted this asymmetry in the climate-carbon cycle response to positive and negative CO_2 emissions.

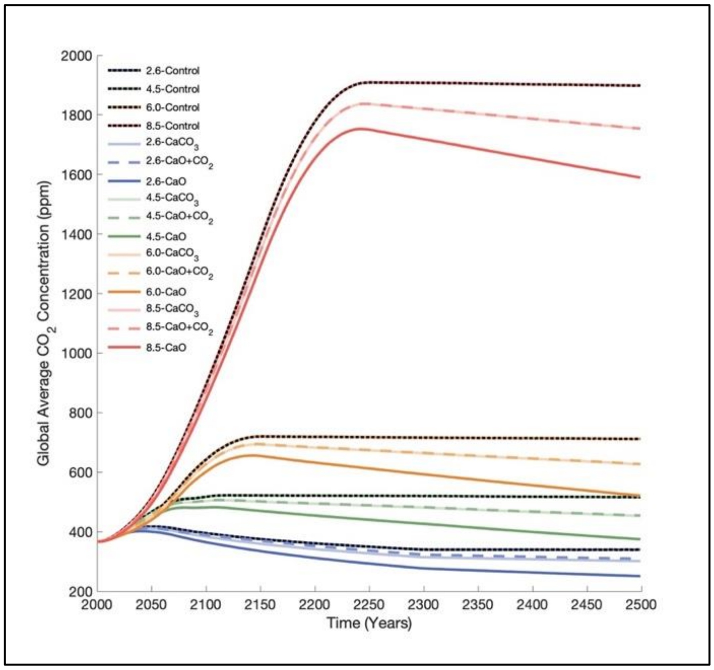

Figure 6: Globally averaged atmospheric CO2 concentration (ppm) from year 2000 to year 2500 as simulated in the 16 simulations.

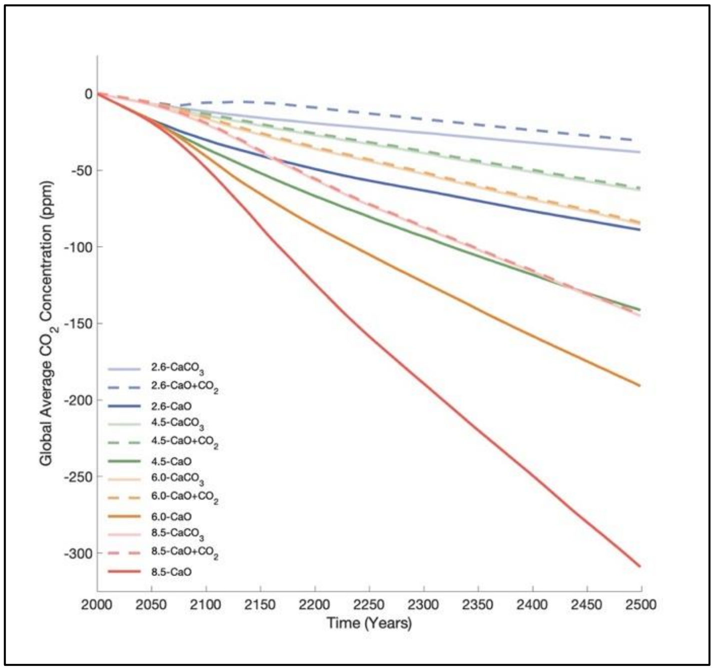

Figure 7: Globally averaged atmospheric CO2 concentration (ppm) control from year 2000 to year 2500 as simulated in the 12 OAE experiments and shown as an anomaly from its respective control simulation.

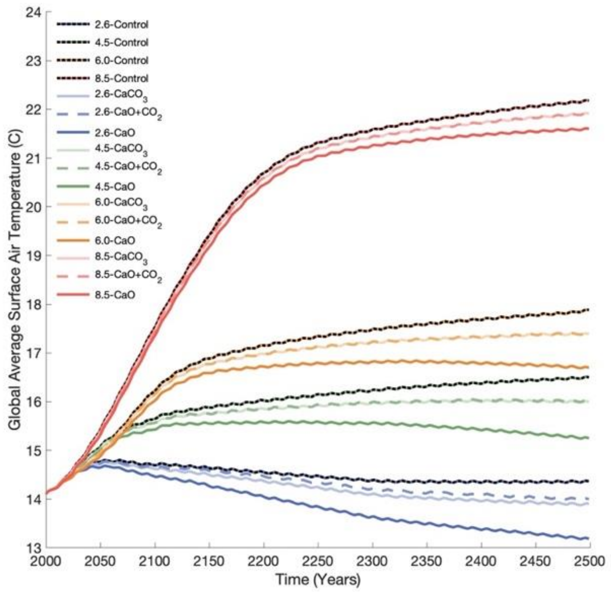

Figure 8: Surface air temperature (°C)
from year 2000 to year 2500 as simulated in the 16 simulations.

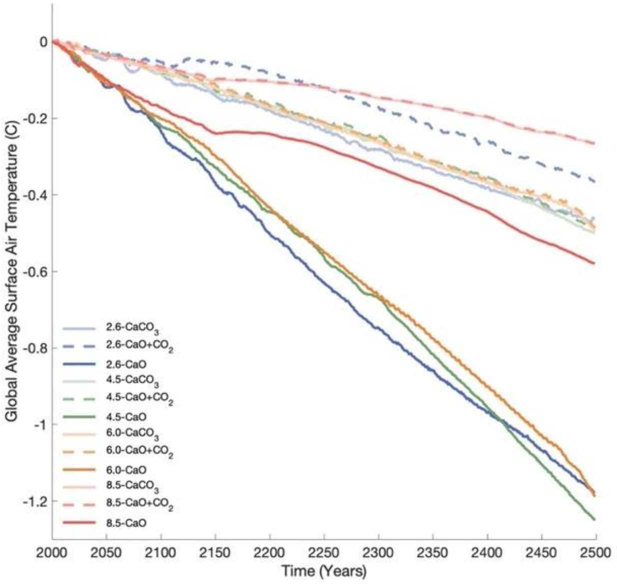

Figure 9: Surface air temperature (°C) control
from year 2000 to year 2500 as simulated in the 12 OAE experiments and shown as an anomaly from its respective control simulation.

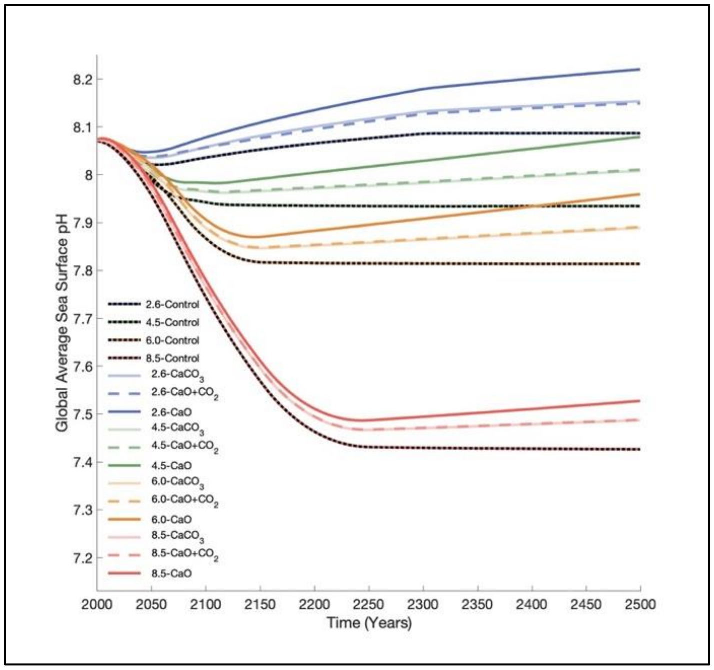

Figure 10: Surface pH
from year 2000 to year 2500 as simulated in the 16 simulations.

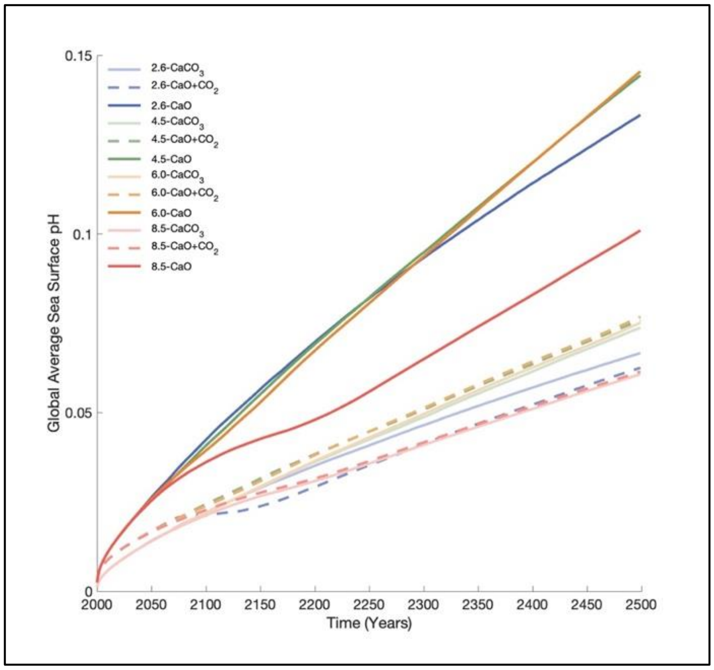

Figure 11: Surface pH control
from year 2000 to year 2500 as simulated in the 12 OAE experiments and shown as an anomaly from its respective control simulation.

The third intervention is the same as our second intervention, except now we also assume that the CO_2 produced in the conversion of $CaCO_3$ to CaO is sequestered. In addition, and as was the case in the *CaO+*CO_2 and *Ca*CO_3 experiments, we assume that all transportation of CaO to the ocean, all the energy required to mine the limestone and all energy used to heat $CaCO_3$ to produce CaO come from non-GHG-emitting sources. In this case, both the range of responses across the RCPs, as well as the mitigative potential, is larger than in the *Ca*CO_3 and *CaO+*CO_2 cases. After 500 years and relative to the respective RCP control runs, the *CaO* OAE experiments led to a reduction in atmospheric CO_2 of 89-309 ppm (depending on the RCP), a reduction in surface air temperature between 0.58 and 1.3°C, and an increase in surface pH of 0.10-0.15. Evidently, this third intervention produced the most advantageous results. Figures 6-11 show a noticeable difference between the *CaO+*CO_2 and *Ca*CO_3 runs and the *CaO* run, with CaO having a much larger impact on surface pH, surface temperature and atmospheric CO_2 concentrations. This was to be expected, given the sequestration of the CO_2 associated with the production of lime meant that each CaO molecule dissolved in the surface waters could draw down up to two CO_2 molecules without adding an additional CO_2 molecule to the atmosphere. Once more, the OAE intervention is more effective in reducing the warming in RCP 2.6, RCP 4.5 and RCP 6.0 than in RCP 8.5 due to the logarithmic response of radiative forcing to atmospheric CO_2.

3.3 Discussion

Previous studies into OAE have focused on exploring factors such as accessibility, transportation, shipping, disposal, discharge, costs, market and energy (e.g., Harvey 2008; Renforth and Henderson 2017; Harvey and Zhang 2001; Renforth et al. 2013; Butenschön et al. 2021; Caserini et al. 2021) or the potential effect of the restoration of blue carbon ecosystems (e.g., Fakhraee et al. 2023). These studies have suggested that OAE represents a promising form of anthropogenic carbon removal. Yet, the order one policy-relevant question has been left unaddressed: Does OAE represent a viable CDR solution to global warming given the current restraints imposed upon it? This analysis suggests that it does not. The overstated promise that this intervention poses a viable solution to global warming is a distraction from the reality that to stabilize the atmospheric level of CO_2 at any level, humanity needs to transition rapidly to zero-emitting energy systems.

To illustrate the previous conclusion, consider the cases of RCP 4.5 and RCP 6.0 (Table 1; Figure 6), the trajectories of human emissions are most closely tracking. If the entire world production of limestone were somehow mined, transported, shipped and distributed throughout the ocean's surface waters without emitting any greenhouse gases for 500 years, and if all the $CaCO_3$ dissolved in the surface waters with none of it sinking to greater depths, then only about a 0.5°C reduction in an otherwise 3-5°C warming above preindustrial levels would be realized (Figs 1c, d). Of course, that would also mean that limestone was no longer available for cement production. Even in the more promising case of converting all the world's annual limestone production to lime prior to its application to surface waters, all the energy used in this process (i.e., heating the $CaCO_3$ to >900°) would have to come from non-emitting sources, and the CO_2 released in this process would have to be sequestered. And even in this case, the 1.2-1.3°C reduction of warming while significant, would still be small relative to the overall 3-5°C warming above preindustrial levels that would be otherwise realized.

Chapter 4: Conclusion

With 2024 being the warmest year on record and each consecutive month from June 2023-August 2024 setting a new monthly global mean temperature record (NOAA 2024), clearly mitigation efforts to reduce global warming are failing. Although it can seem hopeless, in truth, achieving net-zero emissions is achievable so long as there is continued support for CDRs, government leadership, encouragement for green technologies and a push for global policy reducing carbon-intensive energy sources.

Within OAE simulations, there is a need for research into lime and limestone scenarios using only the upper bound of global production and further research into the environmental and biological effects of ocean alkalization. Though showing slight potential, a key aspect of our results to consider is its basis on the current total global limestone mining and production. Despite a 1.25°C drop in surface temperature for the RCP 4.5 scenario and a reduction of 309 ppm in the RCP 8.5 scenario, which seems impactful, these are only achievable if all limestone processing and mining are directed entirely to OAE use only for the next 500 years. Although there are limitations with accessibility, transportation, shipping, disposal, discharge, costs, market and energy, limitations of limestone global production are the utmost barrier in future OAE limestone applications.

Albeit showing slight potential, the constraint of total global limestone mining suggests OAE cannot make an impactful difference on CO_2 emissions, as it would require a massive upscaling in mining and the transference of all current limestone mining to OAE solely. OAE can be one of many solutions used to reduce atmospheric carbon emissions, but unless the extraction of limestone is increased dramatically, OAE cannot be a stand-alone solution to climate change.

Though research into these solutions should be continued, it's important to understand that OAE does not show large-scale climate-saving potential and should only be considered as one of many solutions needed for climate mitigation.

The magnitude of the task and the rates required to achieve net carbon removal suggest that many mitigative options should be implemented, including other CDRs and forms of carbon capture. In the race to reduce CO_2 emissions, research should be devoted not only to current frontrunners, i.e., bioenergy and carbon capture and storage, but also to approaches less formally evaluated in terms of cost, effectiveness, resource availability, and acceptability.

Rather than rapidly embracing the immediate transition to emission-free energy systems, far too much attention has been focused on perturbing the natural carbon cycle to maintain the status quo. As Allen et al. (2024) convincingly show, nature-based solutions cannot solve the challenge of global warming. Though OAE interventions could play a minor role in a large basket of climate solutions, tinkering with the natural carbon cycle alone will not keep warming to below 2°C, or even 3°C, above preindustrial levels. In fact, investments in these areas would be better utilized towards decarbonizing energy systems, which are the cause of global warming.

References

Abouelnaga M. 2021. Carbon Dioxide Removal Pathways and Policy Needs. Centre for Climate Energy Solutions. https://www.c2es.org/wp-content/uploads/2021/06/carbon-dioxide-removal-pathways-and-policy-needs.pdf.

Allen MR, Frame DJ, Friedlingstein P, Gillett NP, Grassi G, Gregory JM, Hare W, House J, Huntingford C, Jenkins S, Jones CD, Knutti R, Lowe JA, Matthews HD, Meinshausen M, Meinshausen N, Peters GP, Plattner G-K, Raper SCB, Rogelj J, Stott PA, Solomon S, Stocker TF, Weaver AJ, Zickfeld K. 2025. Geological Net Zero and the need for disaggregated accounting for carbon sinks. Nature. 638(8050):343–350. doi:10.1038/s41586-024-08326-8.

Archer D. 2005. Fate of fossil fuel CO2 in geologic time - Journal of Geophysical Research: Oceans - Wiley Online Library. [accessed 2025 May 20]. https://doi.org/10.1029/2004JC002625.

Archer DE, 1996. An atlas of the distribution of calcium carbonate in sediments of the deep sea. Global Biogeochemical Cycles, 10(1): 159-174.

Archer D, Khesgi H, Maier-Reimer E. 1998. Dynamics of fossil fuel CO2 neutralization by marine CaCO3. Global Biogeochemical Cycles - Wiley Online Library. [accessed 2024 Apr 15]. https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/98GB00744.

Bach LT, Gill SJ, Rickaby REM, Gore S, Renforth P. 2019. CO2 Removal With Enhanced Weathering and Ocean Alkalinity Enhancement: Potential Risks and Co-benefits for Marine

Pelagic Ecosystems. Frontiers in Climate. 1. doi:10.3389/fclim.2019.00007. [accessed 2025 Jun 18]. https://www.frontiersin.org/journals/climate/articles/10.3389/fclim.2019.00007/full

Bandyopadhyay A, editor. 2014. Carbon Capture and Storage, CO2 Management Technologies. New York: Apple Academic Press.

Bitz CM, Holland MM, Weaver AJ, Eby M. 2001. Simulating the ice-thickness distribution in a coupled climate model. Journal of Geophysical Research: Oceans, 106(C2): 2441-2463.

Bitz CM, Lipscomb WH. 1999. An energy-conserving thermodynamic model of sea ice. Journal of Geophysical Research: Oceans. 104(C7):15669–15677. doi:10.1029/1999JC900100. [accessed 2024 Jun 11]. https://onlinelibrary.wiley.com/doi/abs/10.1029/1999JC900100.

Bliss JD, Hayes TS, Orris GJ. 2008. Limestone; A Crucial and Versatile Industrial Mineral Commodity. [accessed 2025 Jun 3]. https://pubs.usgs.gov/fs/2008/3089/.

Bopp L, Resplandy L, Orr JC, Doney SC, Dunne JP, Gehlen M, Halloran P, Heinze C, Ilyina T, Séférian R, et al. 2013. Multiple stressors of ocean ecosystems in the 21st century: projections with CMIP5 models. Biogeosciences. 10(10):6225–6245. doi:10.5194/bg-10-6225-2013. [accessed 2025 Jun 19]. https://bg.copernicus.org/articles/10/6225/2013/bg-10-6225-2013-discussion.html.

Bose D, Bhattacharya R, Kaur T, Pandya R, Sarkar A, Ray A, Mondal S, Mondal A, Ghosh P, Chemudupati RI. 2024. Innovative approaches for carbon capture and storage as crucial measures for emission reduction within industrial sectors. Carbon Capture Science & Technology. 12:100238. doi:10.1016/j.ccst.2024.100238. [accessed 2024 Sep 12]. https://www.sciencedirect.com/science/article/pii/S2772656824000502.

Brancaccio G, Kalouptsidi M, Papageorgiou T. 2020. Geography, Transportation, and Endogenous Trade Costs. Econometrica. 88(2):657–691. doi:10.3982/ECTA15455. [accessed 2025 Jun 20]. https://onlinelibrary.wiley.com/doi/abs/10.3982/ECTA15455.

Burns W, Corbett CR. 2020. Antacids for the Sea? Artificial Ocean Alkalinization and Climate Change. One Earth. 3(2):154–156. doi:10.1016/j.oneear.2020.07.016. [accessed 2024 Apr 30]. https://www.sciencedirect.com/science/article/pii/S2590332220303602.

Burt DJ, Fröb F, Ilyina T. 2021. The Sensitivity of the Marine Carbonate System to Regional Ocean Alkalinity Enhancement. Frontiers in Climate. 3. doi:10.3389/fclim.2021.624075. [accessed 2024 May 6]. https://www.frontiersin.org/articles/10.3389/fclim.2021.624075.

Butenschön M, Lovato T, Masina S, Caserini S, Grosso M. 2021. Alkalinization Scenarios in the Mediterranean Sea for Efficient Removal of Atmospheric CO2 and the Mitigation of Ocean Acidification. Frontiers in Climate. 3. doi:10.3389/fclim.2021.614537. [accessed 2024 Apr 15]. https://www.frontiersin.org/articles/10.3389/fclim.2021.614537.

C2ES. Carbon Dioxide Removal. Centre for Climate Energy Solutions. [accessed 2025 May 21]. https://www.c2es.org/content/carbon-dioxide-removal/.

Caldeira K, Rau GH. 2000. Accelerating carbonate dissolution to sequester carbon dioxide in the ocean: Geochemical implications. Geophysical Research Letters. 27(2):225–228. doi:10.1029/1999GL002364. [accessed 2025 May 6]. https://onlinelibrary.wiley.com/doi/abs/10.1029/1999GL002364.

Caldeira K, Wickett M. 2003. Anthropogenic carbon and ocean pH. Nature, 425(6956): 365. https://doi.org/10.1038/425365a

Canadell JG, Monteiro PMS, Costa MH, Cotrim da Cunha L, Cox PM, Eliseev AV, Henson S, Ishii M, Jaccard S, Koven C, Lohila A, Patra PK, Piao S, Rogelj J, Syampungani S, Zaehle S, Zickfeld K. 2021. Global carbon and other biogeochemical cycles and feedbacks. *In* Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change. *Edited by* V Masson-Delmotte, P Zhai, A. Pirani, SL Connors, C Péan, S Berger, N Caud, Y Chen, L Goldfarb, MI Gomis, M Huang, K Leitzell, E Lonnoy, JBR Matthews, TK Maycock, T Waterfield, O Yelekçi, R Yu and B Zhou. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp. 673–816, doi:10.1017/9781009157896.007.

Carbon Gap. Carbon Dioxide Removal 101. Carbon Gap. [accessed 2025 May 20]. https://carbongap.org/carbon-dioxide-removal-101/.

Caserini S, Pagano D, Campo F, Abbà A, De Marco S, Righi D, Renforth P, Grosso M. 2021. Potential of Maritime Transport for Ocean Liming and Atmospheric CO2 Removal. Frontiers in Climate. doi:10.3389/fclim.2021.575900. https://www.frontiersin.org/journals/climate/articles/10.3389/fclim.2021.575900/full.

Caserini S, Storni N, Grosso M. 2022. The Availability of Limestone and Other Raw Materials for Ocean Alkalinity Enhancement. Global Biogeochemical Cycles. 36(5):e2021GB007246. doi:10.1029/2021GB007246.

Chilingar GV. 1967. Carbonate rocks. Amsterdam ; Elsevier Pub. Co. (Developments in sedimentology).

Climate change: the 1990 and 1992 IPCC assessments, IPCC first assessment report overview and policymaker summaries and 1992 IPPC supplement. 1992. Geneve: IPCC.

Cox PM. 2001. Description of TRIFFID dynamic global vegetation model. Hadley Cell Technical Note 24: 1-16.

Cripps G, Widdicombe S, Spicer JI, Findlay HS. 2013. Biological impacts of enhanced alkalinity in *Carcinus maenas*. Mar Pollut Bull. 71(1):190–198. doi:10.1016/j.marpolbul.2013.03.015. [accessed 2025 Jun 18]. https://www.sciencedirect.com/science/article/pii/S0025326X13001550.

CSIRO. 2023. Oceans absorb 30% of our emissions, driven by a huge carbon pump. Tiny marine animals are key to working out its climate impacts. 2023. Commonwealth Scientific and Industrial Research Organization. [accessed 2024 Apr 30]. https://www.csiro.au/en/news/All/Articles/2023/June/oceans-absorb-emissions.

ECCC. 2022. London protocol on prevention of marine pollution. Environment and Climate Change Canada, Government of Canada. [accessed 2024 May 9]. https://www.canada.ca/en/environment-climate-change/corporate/international-affairs/partnerships-organizations/london-protocol-prevention-marine-pollution.html.

EESI. 2018. Carbon Removal Strategies: A Broad Overview | Article | Environmental and Energy Study Institute. [accessed 2025 May 21]. https://www.eesi.org/articles/view/carbon-removal-strategies-a-broad-overview.

European Commission. 2024. Carbon Removals and Carbon Farming - European Commission. Energy, Climate Change, Environment, EU Climate Action. [accessed 2025 May 20]. https://climate.ec.europa.eu/eu-action/carbon-removals-and-carbon-farming_en.

Fakhraee M, Li Z, Planavsky N, Reinhard C. 2022. Environmental impacts and carbon capture potential of ocean alkalinity enhancement. doi:10.21203/rs.3.rs-1475007/v1. [accessed 2025 May 6]. https://www.researchgate.net/publication/359888483_Environmental_impacts_and_carbon_capture_potential_of_ocean_alkalinity_enhancement.

Fanning AF, Weaver AJ. 1996. An atmospheric energy-moisture balance model: Climatology, interpentadal climate change, and coupling to an ocean general circulation model. Journal of Geophysical Research: Atmospheres. 101(D10):15111–15128. doi:10.1029/96JD01017. [accessed 2024 Jun 11]. https://onlinelibrary.wiley.com/doi/abs/10.1029/96JD01017.

Feick K. Aggregate | Earth Sciences Museum. Univ Waterloo. [accessed 2024 May 1]. https://uwaterloo.ca/earth-sciences-museum/resources/detailed-rocks-and-minerals-articles/aggregate.

Feng EY, Keller DP, Koeve W, Oschlies A. 2016. Could artificial ocean alkalinization protect tropical coral ecosystems from ocean acidification? Environ Res Lett. 11(7):074008. doi:10.1088/1748-9326/11/7/074008. [accessed 2025 May 6]. https://dx.doi.org/10.1088/1748-9326/11/7/074008.

Feng EY, Koeve W, Keller DP, Oschlies A. 2017. Model-Based Assessment of the CO2 Sequestration Potential of Coastal Ocean Alkalinization - Feng - 2017 - Earth’s Future -

Wiley Online Library. doi:10.1002/2017EF000659. [accessed 2024 May 13]. https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2017EF000659.

Ferderer A, Chase Z, Kennedy F, Schulz KG, Bach LT. 2022. Assessing the influence of ocean alkalinity enhancement on a coastal phytoplankton community. Biogeosciences. 19(23):5375–5399. doi:10.5194/bg-19-5375-2022. [accessed 2025 Jun 20]. https://bg.copernicus.org/articles/19/5375/2022/.

Flügel E. 2010. Microfacies of Carbonate Rocks: Analysis, Interpretation and Application. Berlin, Heidelberg: Springer. [accessed 2025 Jun 4]. https://link.springer.com/10.1007/978-3-642-03796-2.

Friedlingstein P, O'Sullivan M, Jones MW, Andrew RM, Bakker DCE, Hauck J, Landschützer P, Le Quéré C, Luijkx IT, Peters GP, Peters W, Pongratz J, Schwingshackl C, Sitch S, Canadell JG, Ciais P, Jackson RB, Alin SR, Anthoni P, Barbero L, Bates NR, Becker M, Bellouin N, Decharme B, Bopp L, Brasika IBM, Cadule P, Chamberlain MA, Chandra N, Chau T-T-T, Chevallier F, Chini LP, Cronin M, Dou X, Enyo K, Evans W, Falk S, Feely RA, Feng L, Ford DJ, Gasser T, Ghattas J, Gkritzalis T, Grassi G, Gregor L, Gruber N, Gürses Ö, Harris I, Hefner M, Heinke J, Houghton RA, Hurtt GC, Iida Y, Ilyina T, Jacobson AR, Jain A, Jarníková T, Jersild A, Jiang F, Jin Z, Joos F, Kato E, Keeling RF, Kennedy D, Klein Goldewijk K, Knauer J, Korsbakken JI, Körtzinger A, Lan X, Lefèvre N, Li H, Liu J, Liu Z, Ma L, Marland G, Mayot N, McGuire PC, McKinley GA, Meyer G, Morgan EJ, Munro DR, Nakaoka S-I, Niwa Y, O'Brien KM, Olsen, A, Omar, A M, Ono, T, Paulsen, M, Pierrot, D, Pocock, K, Poulter, B, Powis CM, Rehder G, Resplandy L, Robertson E, Rödenbeck C, Rosan TM, Schwinger J, Séférian R, Smallman TL, Smith SM, Sospedra-Alfonso R, Sun Q, Sutton AJ, Sweeney C, Takao S, Tans PP, Tian H, Tilbrook B, Tsujino H, Tubiello F, van der Werf GR, van Ooijen E, Wanninkhof R, Watanabe M, Wimart-Rousseau C, Yang D, Yang X, Yuan W, Yue X, Zaehle S, Zeng J, Zheng B. 2023. Global Carbon Budget 2023. Earth Syst Sci Data. 15(12):5301–5369. doi:10.5194/essd-15-5301-2023. [accessed 2025 Mar 6]. https://www.research.ed.ac.uk/en/publications/global-carbon-budget-2023.

Fuhr M, Wallmann K, Dale AW, Diercks I, Kalapurakkal HT, Schmidt M, Sommer S, Böhnke S, Perner M, Geilert S. 2023. Disentangling artificial and natural benthic weathering in organic rich Baltic Sea sediments. Front Clim. 5. doi:10.3389/fclim.2023.1245580. [accessed 2025 Jun 20]. https://www.frontiersin.org/journals/climate/articles/10.3389/fclim.2023.1245580/full.

Gasser T, Guivarch C, Tachiiri K, Jones CD, Ciais P. 2015. Negative emissions physically needed to keep global warming below 2 °C. Nat Commun. 6(1):7958. doi:10.1038/ncomms8958. [accessed 2025 May 16]. https://www.nature.com/articles/ncomms8958.

Gattuso J-P, Frankignoulle M, Bourge I, Romaine S, Buddemeier RW. 1998. Effect of calcium carbonate saturation of seawater on coral calcification. Glob Planet Change. 18(1):37–46. doi:10.1016/S0921-8181(98)00035-6. [accessed 2025 Jun 19]. https://www.sciencedirect.com/science/article/pii/S0921818198000356.

Goldscheider N, Chen Z, Auler AS, Bakalowicz M, Broda S, Drew D, Hartmann J, Jiang G, Moosdorf N, Stevanovic Z, et al. 2020. Global distribution of carbonate rocks and karst water resources. Hydrogeol J. 28(5):1661–1677. doi:10.1007/s10040-020-02139-5. [accessed 2025 Jun 5]. https://doi.org/10.1007/s10040-020-02139-5.

González MF, Ilyina T. 2016. Impacts of artificial ocean alkalinization on the carbon cycle and climate in Earth system simulations. Geophys Res Lett. 43(12):6493–6502. doi:mil.

González MF, Ilyina T, Sonntag S, Schmidt H. 2018. Enhanced Rates of Regional Warming and Ocean Acidification After Termination of Large-Scale Ocean Alkalinization. Geophys Res Lett. 45(14):7120–7129. doi:10.1029/2018GL077847. [accessed 2025 May 9]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2018GL077847.

González-Santana D, Segovia M, González-Dávila M, Ramírez L, González AG, Pozzo-Pirotta LJ, Arnone V, Vázquez V, Riebesell U, Santana-Casiano JM. 2023. Ocean alkalinity enhancement using sodium carbonate salts does not lead to measurable changes in Fe dynamics in a mesocosm experiment. Biogeosciences. 21(11):2705–2715. doi:10.5194/bg-21-2705-2024. [accessed 2025 Jun 20]. https://bg.copernicus.org/articles/21/2705/2024/.

Gore S, Renforth P, Perkins R. 2019. The potential environmental response to increasing ocean alkalinity for negative emissions | Mitigation and Adaptation Strategies for Global Change. [accessed 2025 Jun 18]. https://link.springer.com/article/10.1007/s11027-018-9830-z.

Grand View Research. 2021. Limestone Market Size, Share & Trends Analysis Report By Product (High Calcium, Magnesian) By End-use (Building & Construction, Chemical, Agriculture, Chemical), By Region (North America, Europe), And Segment Forecasts, 2025 - 2030. [accessed 2024 May 1]. https://www.grandviewresearch.com/industry-analysis/limestone-market.

Ham WE, American Association of Petroleum Geologists Research Committee, Society of Economic Paleontologists and Mineralogists. 1962. Classification of carbonate rocks: a symposium. Tulsa, Okla: American Association of Petroleum Geologists. (Memoir (American Association of Petroleum Geologists)).

Hangx SJT, Spiers CJ. 2009. Coastal spreading of olivine to control atmospheric CO2 concentrations: A critical analysis of viability. Int J Greenh Gas Control. 3(6):757–767. doi:10.1016/j.ijggc.2009.07.001. [accessed 2025 May 9]. https://www.sciencedirect.com/science/article/pii/S1750583609000656.

Hartmann J, Moosdorf N. 2012. The new global lithological map database GLiM: A representation of rock properties at the Earth surface. Geochem Geophys Geosystems. 13(12):n/a. doi:10.1029/2012GC004370. [accessed 2024 May 7]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2012GC004370.

Hartmann J, Suitner N, Lim C, Schneider J, Marín-Samper L, Arístegui J, Renforth P, Taucher J, Riebesell U. 2023. Stability of alkalinity in ocean alkalinity enhancement (OAE) approaches – consequences for durability of CO_2 storage. Biogeosciences. 20(4):781–802. doi:10.5194/bg-20-781-2023. [accessed 2025 Jun 20]. https://bg.copernicus.org/articles/20/781/2023/.

Hartmann J, West AJ, Renforth P, Köhler P, De La Rocha CL, Wolf-Gladrow DA, Dürr HH, Scheffran J. 2013. Enhanced chemical weathering as a geoengineering strategy to reduce atmospheric carbon dioxide, supply nutrients, and mitigate ocean acidification. Rev Geophys. 51(2):113–149. doi:10.1002/rog.20004. [accessed 2025 Jan 21]. https://onlinelibrary.wiley.com/doi/abs/10.1002/rog.20004.

Harvey LDD. 2008. Mitigating the atmospheric CO2 increase and ocean acidification by adding limestone powder to upwelling regions. Journal of Geophysical Research: Oceans. 113(C4). doi:10.1029/2007JC004373. [accessed 2024 Apr 18]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2007JC004373.

Harvey LDD, Huang Z. 2001. A quasi-one-dimensional coupled climate-change cycle model: 1. Description and behavior of the climate component. Journal of Geophysical Research: Oceans. 106(C10):22339–22353. doi:10.1029/2000JC000364. [accessed 2024 May 13]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2000JC000364.

Hauck J, Köhler P, Wolf-Gladrow D, Völker C. 2016. Iron fertilisation and century-scale effects of open ocean dissolution of olivine in a simulated CO2 removal experiment. Environ Res Lett. 11(2):024007. doi:10.1088/1748-9326/11/2/024007. [accessed 2025 Jun 20]. https://dx.doi.org/10.1088/1748-9326/11/2/024007.

He J, Tyka MD. 2023. Limits and CO_2 equilibration of near-coast alkalinity enhancement. Biogeosciences. 20(1):27–43. doi:10.5194/bg-20-27-2023. [accessed 2025 May 9]. https://bg.copernicus.org/articles/20/27/2023/.

Hibler WD. 1979. A Dynamic Thermodynamic Sea Ice Model. J Phys Oceanogr. 9(4):815–846. doi:10.1175/1520-0485(1979)009<0815:ADTSIM>2.0.CO;2. [accessed 2024 Jun 11]. https://journals.ametsoc.org/view/journals/phoc/9/4/1520-0485_1979_009_0815_adtsim_2_0_co_2.xml.

Hoffmann J, Jaimurzina A, Premti A, Valentine V, Youssef F. 2012. Review of Maritime Transport 2012. Rev Marit Transp.

Hunke EC, Dukowicz JK. 1997. An Elastic–Viscous–Plastic Model for Sea Ice Dynamics. J Phys Oceanogr. 27(9):1849–1867. doi.org/101029/2000JC000364 [accessed 2024 Jun 11]. https://journals.ametsoc.org/view/journals/phoc/27/9/1520-0485_1997_027_1849_aevpmf_2.0.co_2.xml.

Ilyina T, Wolf-Gladrow D, Munhoven G, Heinze C. 2013. Assessing the potential of calcium-based artificial ocean alkalinization to mitigate rising atmospheric CO2 and ocean acidification. Geophys Res Lett. 40(22):5909–5914. doi:10.1002/2013GL057981. [accessed 2024 Apr 15]. https://onlinelibrary.wiley.com/doi/abs/10.1002/2013GL057981.

IMO. 2000. First IMO GHG Study on GHG emissions from ships. [accessed 2025 Jun 12]. http://unfccc.int/files/methods_and_science/emissions_from_intl_transport/application/pdf/imoghgmain.pdf.

IMO. 2014. Third IMO GHG Study 2014, Executive Summary and Final Report. International Maritime Organization. [accessed 2024 Apr 15]. https://www.imo.org/en/ourwork/environment/pages/greenhouse-gas-studies-2014.aspx.

IMO. 2019. United Nations Convention on the Law of the Sea. [accessed 2024 May 9]. https://www.imo.org/en/ourwork/legal/pages/unitednationsconventiononthelawofthesea.aspx.

IPCC. 1990. FAR Climate Change: Scientific Assessment of Climate Change — IPCC. [accessed 2025 May 15]. https://www.ipcc.ch/report/ar1/wg1/.

IPCC. 1992. Climate Change 1992: The Supplementary Report to the IPCC Scientific Assessment — IPCC. *Edited by* JT Houghton, BA Callander and SK Varney. Cambridge

University Press, Cambridge, United Kingdom and New York, NY, USA, 200 p. [accessed 2025 May 15]. https://www.ipcc.ch/report/climate-change-1992-the-supplementary-report-to-the-ipcc-scientific-assessment/.

IPCC. 1995. SAR Climate Change 1995: Synthesis Report — IPCC. *Edited by* JT Houghton, LG Meira Filho, BA Callander, N Harris, A Kattenberg and K. Maskell. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 572 p. [accessed 2025 May 15]. https://www.ipcc.ch/report/ar2/syr/.

IPCC. 2001. TAR Climate Change 2001: Synthesis Report — IPCC. *Edited by* JT Houghton, Y Ding, DJ Griggs, M Noguer, PJ van der Linden, X Dai, K Maskell and CA Johnson. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881pp. [accessed 2025 May 15]. https://www.ipcc.ch/report/ar3/syr/.

IPCC. 2007. AR4 Climate Change 2007: Synthesis Report — IPCC. Edited by S Solomon, D Qin, M Manning, Z Chen, M Marquis, KB Averyt, M Tignor, and HL Miller. Cambridge University Press, Cambridge, United Kingdom, and New York, NY, USA, 996 pages. [accessed 2025 May 15]. https://www.ipcc.ch/report/ar4/syr/.

IPCC. 2013. AR5 Climate Change 2013: The Physical Science Basis — IPCC. *Edited by* TF Stocker, D Qin, G-K Plattner, M Tignor, SK Allen, . Boschung, A Nauels, Y Xia, V Bex and PM Midgley. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 p. [accessed 2025 May 15]. https://www.ipcc.ch/report/ar5/wg1/.

IPCC. 2019. Summary for Policymakers — Special Report on the Ocean and Cryosphere in a Changing Climate. [accessed 2025 May 15]. https://www.ipcc.ch/srocc/chapter/summary-for-policymakers/.

IPCC. 2021. AR6 Climate Change 2021: The Physical Science Basis — IPCC. *Edited by* V Masson-Delmotte, P Zhai, A Pirani, SL Connors, C Péan, S Berger, N Caud, Y Chen, L Goldfarb, MI Gomis, M Huang, K Leitzell, E Lonnoy, JBR Matthews, TK Maycock, T Waterfield, O Yelekçi, R Yu and B Zhou. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2391 p. doi: 10.1017/9781009157896. [accessed 2025 May 15]. https://www.ipcc.ch/report/sixth-assessment-report-working-group-i/.

IPCC. 2022a. Carbon Dioxide Removal. Climate Change 2022: Impacts, Adaptation and Vulnerability. Contribution of Working Group III to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change. *Edited by* Shukla PR, Skea J, Slade R, Al Khourdajie A, van Diemen R, McCollum D, Pathak M, Some S, Vyas P, Fradera R, et al..

IPCC. 2022b. Global Warming of 1.5°C: IPCC Special Report on Impacts of Global Warming of 1.5°C above Pre-industrial Levels in Context of Strengthening Response to Climate Change, Sustainable Development, and Efforts to Eradicate Poverty. 1st ed. Cambridge University Press. [accessed 2025 May 15]. https://www.cambridge.org/core/product/identifier/9781009157940/type/book.

IPCC. 2023. AR6 Synthesis Report: Climate Change 2023 — IPCC. [accessed 2025 May 15]. https://www.ipcc.ch/report/sixth-assessment-report-cycle/.

IPCC, Berger S, Connors SL, Allan RP, Berntsen T, Bock L, Cerezo-Mota R, Cobb K. 2021. IPCC AR6 Chapter 5 FAQ. https://www.ipcc.ch/report/ar6/wg1/resources/frequently-asked-questions.

IPCC_AR6_WGI_SPM_final.pdf. [accessed 2024 Apr 25]. https://www.ipcc.ch/report/ar6/wg1/downloads/report/IPCC_AR6_WGI_SPM_final.pdf.

Jiang L-Q, Dunne J, Carter BR, Tjiputra JF, Terhaar J, Sharp JD, Olsen A, Alin S, Bakker DCE, Richard A. Feely RA, Gattuso J-P, Hogan P, Ilyina T, Lange N, Lauvset SK, Lewis ER, Lovato T, Palmieri J, Santana-Falcón Y, Schwinger J, Séférian R, Strand G, Swart N, Tanhua T, Tsujino H, Wanninkhof R, Watanabe M, Yamamoto A, Ziehn T. 2023. Global surface ocean acidification indicators from 1750 to 2100. Journal of Advances in Modeling Earth Systems, 15(3): e2022MS003563. https://doi.org/10.1029/2022MS003563

Jin X-Y, Cao L. 2023. Comparison of the carbon cycle and climate response to artificial ocean alkalinization and solar radiation modification. Adv Clim Change Res. 14(2):322–334. doi:10.1016/j.accre.2023.03.002. [accessed 2025 May 6]. https://www.sciencedirect.com/science/article/pii/S1674927823000357.

Kalnay E, Kanamitsu M, Kistler R, Collins W, Deaven D, Gandin L, Iredell M, Saha S, White G, Woollen J, Y. Zhu Y, Leetmaa A, Reynolds B, Chelliah M, Ebisuzaki W, Higgins W, Janowiak J, Mo KC, Ropelewski C, Wang J, Jenne R, Joseph D. 1996. The NCEP/NCAR 40-Year Reanalysis Project. Bulletin of the American Meteorological Society, 77(3): 437-472.

Keith E, Webb T. 2015. Limestone; Mineral Commodity Profile No. 11. [accessed 2025 Jun 3]. https://www.google.com/search?client=safari&rls=en&q=Keith%2C+E.A.+and+Webb%2C+T.C.+(compilers)+2015.+Limestone.+New+Brunswick+Department+of+Energy+and+Mines%3B+Resources%2FExploration%2FDevelopment%2FManagement+Division%2C+Mineral+Commodity+Profile+No.+11%2C+7+p.&ie=UTF-8&oe=UTF-8.

Keller DP, Feng EY, Oschlies A. 2014. Potential climate engineering effectiveness and side effects during a high carbon dioxide-emission scenario. Nat Commun. 5(1):3304. doi:10.1038/ncomms4304. [accessed 2025 Jan 19]. https://www.nature.com/articles/ncomms4304.

Keller DP, Lenton A, Scott V, Vaughan NE, Bauer N, Ji D, Jones CD, Kravitz B, Muri H, Zickfeld K. 2018. The Carbon Dioxide Removal Model Intercomparison Project (CDRMIP): rationale and experimental protocol for CMIP6. Geosci Model Dev. 11(3):1133–1160. doi:10.5194/gmd-11-1133-2018. [accessed 2025 Jun 11]. https://gmd.copernicus.org/articles/11/1133/2018/gmd-11-1133-2018.html.

Keller DP, Oschlies A, Eby M. 2012. A new marine ecosystem model for the University of Victoria Earth System Climate Model. Geosci Model Dev. 5(5):1195–1220. [accessed 2024 Jun 16]. https://gmd.copernicus.org/articles/5/1195/2012/.

Kheshgi HS. 1995. Sequestering atmospheric carbon dioxide by increasing ocean alkalinity. Energy. 20(9):915–922. doi:10.1016/0360-5442(95)00035-F. [accessed 2024 Apr 16]. https://www.sciencedirect.com/science/article/pii/036054429500035F.

Kitidis V, Rackley S, Burt W, Rau G, Fawcett S, Taylor M, Tarran G, Woodward E, Harris C, Fileman T. 2024. Magnesium hydroxide addition reduces aqueous carbon dioxide in wastewater discharged to the ocean. Commun Earth Environ. 5. doi:10.1038/s43247-024-01506-4.

Köhler P. 2020. Anthropogenic CO2 of High Emission Scenario Compensated After 3500 Years of Ocean Alkalinization With an Annually Constant Dissolution of 5 Pg of Olivine. Front Clim. 2. doi:10.3389/fclim.2020.575744. [accessed 2025 May 9]. https://www.frontiersin.orghttps://www.frontiersin.org/journals/climate/articles/10.3389/fclim.2020.575744/full.

Köhler P, Abrams JF, Völker C, Hauck J, Wolf-Gladrow DA. 2013. Geoengineering impact of open ocean dissolution of olivine on atmospheric CO2, surface ocean pH and marine biology. Environ Res Lett. 8(1):014009. doi:10.1088/1748-9326/8/1/014009. [accessed 2025 May 9]. https://dx.doi.org/10.1088/1748-9326/8/1/014009.

Köhler P, Hartmann J, Wolf-Gladrow DA. 2010. Geoengineering potential of artificially enhanced silicate weathering of olivine. Proc Natl Acad Sci. 107(47):20228–20233. doi:10.1073/pnas.1000545107. [accessed 2025 May 20]. https://www.pnas.org/doi/full/10.1073/pnas.1000545107.

Krausmann F, Gingrich S, Eisenmenger N, Erb K-H, Haberl H, Fischer-Kowalski M. 2009. Growth in global materials use, GDP and population during the 20th century. Ecol Econ. 68(10):2696–2705. doi:10.1016/j.ecolecon.2009.05.007.

Kump LR, Brantley SL, Arthur MA. 2000. Chemical Weathering, Atmospheric CO2, and Climate. Annu Rev Earth Planet Sci. 28(Volume 28, 2000):611–667. doi:10.1146/annurev.earth.28.1.611. [accessed 2025 Jun 11]. https://www.annualreviews.org/content/journals/10.1146/annurev.earth.28.1.611.

Kwiatkowski L, Berger M, Bopp L, Doléac S, Ho DT. 2023. Contrasting carbon dioxide removal potential and nutrient feedbacks of simulated ocean alkalinity enhancement and macroalgae afforestation. Environ Res Lett. 18(12):124036. doi:10.1088/1748-9326/ad08f9. [accessed 2025 May 9]. https://dx.doi.org/10.1088/1748-9326/ad08f9.

Lenton A, Matear RJ, Keller DP, Scott V, Vaughan NE. 2018. Assessing carbon dioxide removal through global and regional ocean alkalinization under high and low emission pathways. Earth Syst Dyn. 9(2):339–357. doi:10.5194/esd-9-339-2018. [accessed 2025 Jan 19]. albr.

Locke A. 2009. Preliminary evaluation of effects of invasive tunicate management with acetic acid and calcium hydroxide on non-target marine organisms in Prince Edward Island, Canada. Aquat Invasions. 4(1):221–236. doi:10.3391/ai.2009.4.1.23.

Lüthi D, Le Floch M, Bereiter B, Blunier T, Barnola J-M, Siegenthaler U, Raynaud D, Jouzel J, Fischer H, Kawamura K, et al. 2008. High-resolution carbon dioxide concentration record 650,000–800,000 years before present. Nature. 453(7193):379–382. doi:10.1038/nature06949. [accessed 2025 May 16]. https://www.nature.com/articles/nature06949.

MacIsaac AJ, Mengis N, Zickfeld K, Nzotungicimpaye C-M. 2021. New applications for an EMIC coupled to an atmospheric chemistry model - The University of Victoria Earth system climate model version 2.10 + FAIR chemistry module. Copernicus Meetings Report No.: EGU21-13072. [accessed 2025 Apr 23]. https://meetingorganizer.copernicus.org/EGU21/EGU21-13072.html.

Martin KR, Nickoloff A, Moffat L, Weaver AJ, Eby M. 2025. Assessing the effectiveness of ocean alkalinity enhancement on carbon sequestration and ocean acidification. FACETS. 10:1–8. doi:10.1139/facets-2024-0171. [accessed 2025 Sep 12]. https://www.researchgate.net/publication/395195043_Assessing_the_effectiveness_of_ocean_alkalinity_enhancement_on_carbon_sequestration_and_ocean_acidification.

Meissner KJ, Weaver AJ, Matthews HD, Cox PM. 2003. The role of land surface dynamics in glacial inception: a study with the UVic Earth system climate model. Climate Dynamics, 21: 519-537.

Meysman FJR, Montserrat F. 2017. Negative CO2 emissions via enhanced silicate weathering in coastal environments. Biol Lett. 13(4):20160905. doi:10.1098/rsbl.2016.0905. [accessed 2025 Jun 5]. https://royalsocietypublishing.org/doi/10.1098/rsbl.2016.0905.

Millero FJ. 2007. The Marine Inorganic Carbon Cycle. Chem Rev. 107(2):308–341. doi:10.1021/cr0503557. [accessed 2025 Jun 4]. https://doi.org/10.1021/cr0503557.

Montserrat F, Renforth P, Hartmann J, Leermakers M, Knops P, Meysman FJR. 2017. Olivine Dissolution in Seawater: Implications for CO2 Sequestration through Enhanced Weathering in Coastal Environments. Environ Sci Technol. 51(7):3960–3972. doi:10.1021/acs.est.6b05942. [accessed 2025 Jun 5]. https://doi.org/10.1021/acs.est.6b05942.

Moras CA, Bach LT, Cyronak T, Joannes-Boyau R, Schulz KG. 2022. Ocean alkalinity enhancement – avoiding runaway $CaCO_3$ precipitation during quick and hydrated lime dissolution. Biogeosciences. 19(15):3537–3557. doi:10.5194/bg-19-3537-2022. [accessed 2024 Apr 15]. https://bg.copernicus.org/articles/19/3537/2022/.

Moss R, Babiker M, Brinkman S, Calvo E, Carter T, Edmonds J, Elgizouli I, Emori S, Erda L, Hibbard K, Roger Jones J, Kainuma M, Kelleher J, Lamarque J-F, Manning M, Matthews B, Meehl J, Meyer L, Mitchell J, Nakicenovic N, O'Neill B, Pichs R, Riahi K, Rose S, Runci P, Stouffer R, van Vuuren D, Weyant J, Wilbanks T, van Ypersele J-P, Zurek M. 2008. Towards New Scenarios for Analysis of Emissions, Climate Change, Impacts, and Response Strategies. Intergovernmental Panel on Climate Change, Geneva, 132 pp.

Nagwekar T, Nissen C, Hauck J. 2024. Ocean Alkalinity Enhancement in Deep Water Formation Regions Under Low and High Emission Pathways- Earth's Future - Wiley Online Library. doi:10.1029/2023EF004213. https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2023EF004213.

National Academies of Sciences, Engineering, and Medicine. 2019. Negative Emissions Technologies and Reliable Sequestration: A Research Agenda; Chapter 4: Bioenergy with Carbon Capture and Sequestration. doi:10.17226/25259. [accessed 2024 Apr 30]. https://nap.nationalacademies.org/read/25259/chapter/6.

National Research Council. 2015. Climate Intervention: Carbon Dioxide Removal and Reliable Sequestration. doi:10.17226/18805. [accessed 2025 May 21]. https://nap.nationalacademies.org/read/18805/chapter/1.

Natural Resources Canada. 2023. Production of Canada's Leading Minerals. Gov Can. [accessed 2025 Jun 3]. https://mmsd.nrcan-rncan.gc.ca/prod-prod/mon-men-eng.aspx.

Negative Emissions Technologies and Reliable Sequestration: A Research Agenda. 2019. Washington, D.C.: National Academies Press. [accessed 2024 Apr 30]. https://www.nap.edu/catalog/25259.

Nemet GF, Callaghan MW, Creutzig F, Fuss S, Hartmann J, Hilaire J, Lamb WF, Minx JC, Rogers S, Smith P. 2018. Negative emissions—Part 3: Innovation and upscaling. Environ Res Lett. 13(6):063003. doi:10.1088/1748-9326/aabff4. [accessed 2025 May 16]. https://dx.doi.org/10.1088/1748-9326/aabff4.

Vaughan N, Fuss S, Buck H, Schenuit F, Pongratz J, Schulte I, Lamb WF, Probst B, Edwards M, Nemet GF, et al. 2024. The State of Carbon Dioxide Removal - 2nd Edition. doi:10.17605/OSF.IO/F85QJ. [accessed 2025 Jul 22]. https://osf.io/f85qj/.

NOAA. 2019. Surface ocean pH and buffer capacity. [accessed 2024 Apr 30]. https://www.pmel.noaa.gov/co2/story/Surface+ocean+pH+and+buffer+capacity.

NOAA. National Centers for Environmental Information, Monthly Global Climate Report for June 2024. 2024. Climate Change: Atmospheric Carbon Dioxide | NOAA Climate.gov. [accessed 2025 Mar 6]. https://www.climate.gov/news-features/understanding-climate/climate-change-atmospheric-carbon-dioxide.

NOAA. 2024. Carbon dioxide removal: NOAA State of the Science factsheet | NOAA Climate.gov. [accessed 2025 May 20]. https://www.climate.gov/news-features/understanding-climate/carbon-dioxide-removal-noaa-state-science-factsheet.

NOAA. 2025. Ocean acidification | National Oceanic and Atmospheric Administration. [accessed 2025 May 16]. https://www.noaa.gov/education/resource-collections/ocean-coasts/ocean-acidification.

NOAA Fisheries. 2025. Understanding Ocean Acidification | NOAA Fisheries. [accessed 2025 May 16]. https://www.fisheries.noaa.gov/insight/understanding-ocean-acidification.

Oates JAH. 1998. Lime and Limestone. 1st ed. John Wiley & Sons, Ltd. [accessed 2024 Apr 15]. https://onlinelibrary.wiley.com/doi/10.1002/9783527612024.

O'Neill B, van Aalst M, Ibrahim ZZ, Berrang Ford L, Bhadwal S, Buhaug H, Diaz D, Frieler K, Garschagen M, Magnan A, et al. 2022. Key risks across sectors and regions. In: Pörtner H-O, Roberts DC, Tignor MMB, Poloczanska ES, Mintenbeck K, Alegría A, Craig M, Langsdorf S, Löschke S, Möller V, et al., editors. Climate Change 2022: Impacts, Adaptation and Vulnerability. Contribution of Working Group II to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press.

Pacanowski RC. 1995. Pacanowski: MOM 2 documentation, user's guide and... - Google Scholar. [accessed 2024 Jun 11]. https://scholar.google.com/scholar_lookup?hl=en&publication_year=1995&pages=232&author=R.+Pacanowski&title=MOM+2+Documentation+User%27s+Guide+and+Reference+Manual%2C+GFDL+Ocean+Group+Technical+Report.

Palmiéri J, Yool A. 2024. Global-Scale Evaluation of Coastal Ocean Alkalinity Enhancement in a Fully Coupled Earth System Model. Earths Future. 12(3):e2023EF004018.

doi:10.1029/2023EF004018. [accessed 2024 Apr 30]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2023EF004018.

Paul AJ, Haunost M, Goldenberg SU, Hartmann J, Sánchez N, Schneider J, Suitner N, Riebesell U. 2025. Ocean alkalinity enhancement in an open-ocean ecosystem: biogeochemical responses and carbon storage durability. Biogeosciences. 22(12):2749–2766. doi:10.5194/bg-22-2749-2025. [accessed 2025 Jun 20]. https://bg.copernicus.org/articles/22/2749/2025/.

Rau G, Caldeira K, Knauss K, Downs B. 2001. (PDF) Enhanced carbonate dissolution as a means of capturing and sequestering carbon dioxide. ResearchGate. [accessed 2025 Jun 12]. https://www.researchgate.net/publication/228416335_Enhanced_carbonate_dissolution_as_a_means_of_capturing_and_sequestering_carbon_dioxide.

Rau GH. 2010. CO2 Mitigation via Capture and Chemical Conversion in Seawater. Environ Sci Technol. 45(3):1088–1092. doi:10.1021/es102671x. [accessed 2025 May 20]. https://doi.org/10.1021/es102671x.

Rau GH, Caldeira K. 1999. Enhanced carbonate dissolution:: a means of sequestering waste CO2 as ocean bicarbonate. Energy Convers Manag. 40(17):1803–1813. doi:10.1016/S0196-8904(99)00071-0. [accessed 2025 May 20]. https://www.sciencedirect.com/science/article/pii/S0196890499000710.

Rau GH, Knauss KG, Langer WH, Caldeira K. 2007. Reducing energy-related CO2 emissions using accelerated weathering of limestone. Energy. 32(8):1471–1477.

doi:10.1016/j.energy.2006.10.011. [accessed 2025 Jun 20]. https://www.sciencedirect.com/science/article/pii/S0360544206002982.

Reichl C, Schatz M. 2022. World Mining Data 2022. International Organizing Committee for the World Mining Congresses, Federal Ministry Republic of Austria. https://www.world-mining-data.info/wmd/downloads/PDF/WMD2022.pdf.

Reichl C, Schatz M, Zsak G. 2017. World Mining Data 2017 - OD Mekong Datahub. [accessed 2024 May 1]. https://data.laos.opendevelopmentmekong.net/library_record/world-mining-data-2017.

Renforth P, Bellamy R, Beerling D, Boettcher M, Bonalumi D, Brandão M, Fridahl M, Fuss S, Hansson A, Heyward C, et al. 2023. Specialty grand challenge: renaming our section to "Carbon Dioxide Removal." Front Clim. 5. doi:10.3389/fclim.2023.1279109. [accessed 2025 May 21]. https://www.frontiersin.org/journals/climate/articles/10.3389/fclim.2023.1279109/full.

Renforth P, Henderson G. 2017. Assessing ocean alkalinity for carbon sequestration - Renforth - 2017 - Reviews of Geophysics - Wiley Online Library. [accessed 2024 Apr 15]. https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2016RG000533.

Renforth P, Jenkins BG, Kruger T. 2013. Engineering challenges of ocean liming - ScienceDirect. [accessed 2024 Apr 15]. https://www-sciencedirect-com.ezproxy.library.uvic.ca/science/article/pii/S0360544213006816?via%3Dihub.

Ringham MC, Hirtle N, Shaw C, Lu X, Herndon J, Carter BR, Eisaman MD. 2024. An assessment of ocean alkalinity enhancement using aqueous hydroxides: kinetics, efficiency, and precipitation thresholds. Biogeosciences. 21(15):3551–3570. doi:10.5194/bg-21-3551-2024. [accessed 2025 May 9]. https://bg.copernicus.org/articles/21/3551/2024/.

SARE. Cover Crops and Carbon Sequestration. SARE. [accessed 2025 May 20]. https://www.sare.org/publications/cover-crops-ecosystem-services/cover-crops-and-carbon-sequestration/.

Schmittner A, Oschlies A, Matthews HD, Galbraith ED. 2008. Future changes in climate, ocean circulation, ecosystems, and biogeochemical cycling simulated for a business-as-usual CO_2 emission scenario until year 4000 AD, Global Biogeochemical Cycles, 22: GB1013, doi:10.1029/2007GB002953.

Schuckmann K, Cheng L, Palmer MD, Hansen J, Tassone C, Aich V, Adusumilli S, Beltrami H, Boyer T, Cuesta-Valero FJ, et al. 2020. Heat stored in the Earth system: where does the energy go? Earth Syst Sci Data. 12(3):2013–2041. doi:10.5194/essd-12-2013-2020. [accessed 2025 May 15]. https://essd.copernicus.org/articles/12/2013/2020/.

Sonntag S, Ferrer González M, Ilyina T, Kracher D, Nabel JEMS, Niemeier U, Pongratz J, Reick CH, Schmidt H. 2018. Quantifying and Comparing Effects of Climate Engineering Methods on the Earth System. Earths Future. 6(2):149–168. doi:10.1002/2017EF000620. [accessed 2025 Jan 21]. https://onlinelibrary.wiley.com/doi/abs/10.1002/2017EF000620.

SREX_fact_sheet.pdf. [accessed 2024 Apr 25]. https://www.ipcc.ch/site/assets/uploads/2018/04/SREX_fact_sheet.pdf.

Suitner N, Faucher G, Lim C, Schneider J, Moras CA, Riebesell U, Hartmann J. 2024. Ocean alkalinity enhancement approaches and the predictability of runaway precipitation processes: results of an experimental study to determine critical alkalinity ranges for safe and sustainable application scenarios. Biogeosciences. 21(20):4587–4604. doi:10.5194/bg-21-4587-2024. [accessed 2025 Jun 20]. https://bg.copernicus.org/articles/21/4587/2024/.

Suleman H, Fosbøl PL, Nasir R, Ameen M, editors. 2022. Sustainable Carbon Capture: Technologies and Applications. Boca Raton: CRC Press.

Sverdrup HU, Koca D, Schlyter P. 2017. A Simple System Dynamics Model for the Global Production Rate of Sand, Gravel, Crushed Rock and Stone, Market Prices and Long-Term Supply Embedded into the WORLD6 Model. Biophys Econ Resour Qual. 2(2):8. doi:10.1007/s41247-017-0023-2. [accessed 2025 Jun 3]. https://doi.org/10.1007/s41247-017-0023-2.

United Nations. 2015. The Paris Agreement. U N. [accessed 2025 May 16]. https://unfccc.int/files/essential_background/convention/application/pdf/english_paris_agreement.pdf.

U.S. Department of Energy. Carbon Dioxide Removal. Energy.gov. [accessed 2025 May 20]. https://www.energy.gov/fecm/carbon-dioxide-removal.

U.S. Geological Survey. 2011. Mineral Commodity Summaries 2011. https://d9-wret.s3.us-west-2.amazonaws.com/assets/palladium/production/mineral-pubs/mcs/mcs2011.pdf.

U.S. Geological Survey. 2018. Mineral commodity summaries 2018: U.S. Geological Survey. https://doi.org/10.3133/70194932.

U.S. Geological Survey. 2019. Mineral commodity summaries 2019: U.S. Geological Survey. https://doi.org/10.3133/70202434.

U.S. Geological Survey. 2024. Mineral commodity summaries 2024: U.S. Geological Survey. U.S. Geological Survey Report No.: 2024. [accessed 2024 May 1]. https://pubs.usgs.gov/publication/mcs2024.

Wang B, Gao X, Song J, Li X, Yuan H, Xie L, Zhao J, Xing Q, Qin S. 2023. Feasibility of increasing marine carbon storage through olivine addition. J Environ Chem Eng. 11(6):111221. doi:10.1016/j.jece.2023.111221. [accessed 2025 Jun 5]. https://www.sciencedirect.com/science/article/pii/S2213343723019607.

Wang H, Pilcher DJ, Kearney KA, Cross JN, Shugart OM, Eisaman MD, Carter BR. 2022. Simulated Impact of Ocean Alkalinity Enhancement on Atmospheric CO2 Removal in the Bering Sea. Earths Future. 11(1):e2022EF002816. doi:10.1029/2022EF002816. [accessed 2025 Jun 19]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2022EF002816.

Weaver AJ. 2008. *Keeping our Cool: Canada in a Warming World* (Hardcover), Viking Canada, Toronto, Ontario, ISBN:978-0-670-06800-5, 323 p.

Weaver AJ, Eby M, Wiebe EC, Bitz CM, Duffy PB, Ewen TL, Fanning AF, Holland MM, MacFadyen A, Matthews HD, Meissner KJ, Saenko O, Schmittner A, Wang H and Yoshimori M. (2001. The UVic earth system climate model: Model description, climatology, and applications to past, present and future climates. Atmosphere-Ocean, 39(4): 361–428. https://doi.org/10.1080/07055900.2001.9649686

Weaver AJ, Zickfeld K, Montenegro A, Eby M. 2007. Long term climate implications of 2050 emission reduction targets. Geophysical Research Letters, 34: L19703, doi:10.1029/2007GL031018. [accessed 2025 May 15]. https://onlinelibrary.wiley.com/doi/abs/10.1029/2007GL031018.

Yang B, Leonard J, Langdon C. 2023. Seawater alkalinity enhancement with magnesium hydroxide and its implication for carbon dioxide removal. Mar Chem. 253:104251. doi:10.1016/j.marchem.2023.104251. [accessed 2025 May 9]. https://www.sciencedirect.com/science/article/pii/S0304420323000476.

Zhang G, Li Y, Jin Z, Dykas S, Cai X. 2024. A novel carbon dioxide capture technology (CCT) based on non-equilibrium condensation characteristics: Numerical modelling, nozzle design and structure optimization. Energy. 286:129603. doi:10.1016/j.energy.2023.129603. [accessed 2024 Apr 30]. https://www.sciencedirect.com/science/article/pii/S0360544223029973.

Zickfeld K, Eby M, Weaver AJ, Alexander K, Crespin E, Edwards NR, Eliseev AV, Feulner G, Fichefet T, Forest CE, Friedlingstein P, Goosse H, Holden PB, Joos F, Kawamiya M, Kicklighter D, Kienert H, Matsumoto K, Mokhov II, Monier E, Olsen SM, Pedersen JOP, Perrette M, Philippon-Berthier G, Ridgwell A, Schlosser A, Schneider von Deimling T, Shaffer G, Sokolov AP, Spahni R, Steinacher M, Tachiiri K, Tokos K, Yoshimori M, Zeng N, Zhao F. 2012. Long-term climate change commitment and reversibility: An EMIC intercomparison. Journal of Climate, 16: 5782-5809.

Zickfeld K, Azevedo D, Mathesius S, Matthews HD. 2021. Asymmetry in the climate–carbon cycle response to positive and negative CO_2 emissions. Nature Climate Change, 11: 613–617. https://doi.org/10.1038/s41558-021-01061-2