LETTER • OPEN ACCESS

Regional potential of coastal ocean alkalinization with olivine within 100 years

To cite this article: Murugan Ramasamy et al 2024 Environ. Res. Lett. 19 064030

View the article online for updates and enhancements.

You may also like

al

- Geoengineering impact of open ocean dissolution of olivine on atmospheric CO₂, surface ocean pH and marine biology Peter Köhler, Jesse F Abrams, Christoph Völker et al.
- <u>Electrochemical Capture and Conversion</u> of Carbon Dioxide into All-Carbon <u>Nanostructures</u> Anna Douglas, David Wood, Anna Klug et
- <u>Selective Conversion to Formic Acid from</u> <u>CO2 By Electrochemical Reduction over</u> <u>Cuxsny Intermetallic Compounds</u> <u>Hiroya Ochiai, Takao Gunji and Futoshi</u> Matsumoto

BREATH

BIOPS

Main talks

Early career sessions

Posters

Breath Biopsy Conference

Join the conference to explore the **latest challenges** and advances in **breath research**, you could even **present your latest work**!



Register now for free!

This content was downloaded from IP address 37.138.212.23 on 03/06/2024 at 09:44

ENVIRONMENTAL RESEARCH LETTERS

LETTER

OPEN ACCESS

CrossMark

RECEIVED 30 June 2023

REVISED 12 April 2024

ACCEPTED FOR PUBLICATION

2 May 2024 PUBLISHED

28 May 2024

Original content from this work may be used under the terms of the Creative Commons Attribution 4.0 licence.

Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.



Regional potential of coastal ocean alkalinization with olivine within 100 years

Murugan Ramasamy^{1,*}, Thorben Amann² and Nils Moosdorf^{1,3}

Department of Biogeochemistry and Geology, Leibniz Centre for Tropical Marine Research (ZMT), Bremen, Germany 2

Center for Earth System Sciences and Sustainability, Institute for Geology, Universität Hamburg, Hamburg, Germany 3

Institute of Geosciences, Kiel University, Kiel, Germany

Author to whom any correspondence should be addressed.

E-mail: murugan.ramasamy@leibniz-zmt.de

Keywords: CDR, alkalinity enhancement, CO2 sequestration, enhanced weathering, geochemical modeling Supplementary material for this article is available online

Abstract

The spreading of crushed olivine-rich rocks in coastal seas to accelerate weathering reactions sequesters atmospheric CO₂ and reduces atmospheric CO₂ concentrations. Their weathering rates depend on different factors, including temperature and the reaction surface area. Therefore, this study investigates the variations in olivine-based enhanced weathering rates across 13 regional coasts worldwide. In addition, it assesses the CO₂ sequestration within 100 years and evaluates the maximum net-sequestration potential based on varying environmental conditions. Simulations were conducted using the geochemical thermodynamic equilibrium modeling software PHREEQC. A sensitivity analysis was performed, exploring various combinations of influencing parameters, including grain size, seawater temperature, and chemistry. The findings reveal significant variation in CO₂ sequestration, ranging from 0.13 to 0.94 metric tons (t) of CO₂ per ton of distributed olivine-rich rocks over 100 years. Warmer coastal regions exhibit higher CO₂ sequestration capacities than temperate regions, with a difference of $0.4 \text{ t } \text{CO}_2/\text{t}$ olivine distributed. Sensitivity analysis shows that smaller grain sizes (10 μ m) exhibit higher net CO₂ sequestration rates (0.87 t/t) in olivine-based enhanced weathering across all conditions, attributed to their larger reactive surface area. However, in warmer seawater temperatures, olivine with slightly larger grain sizes (50 and 100 μ m) displays still larger net CO₂ sequestration rates (0.97 and 0.92 t/t), optimizing the efficiency of CO₂ sequestration while reducing grinding energy requirements. While relying on a simplified sensitivity analysis that does not capture the full complexity of real-world environmental dynamics, this study contributes to understanding the variability and optimization of enhanced weathering for CO₂ sequestration, supporting its potential as a sustainable CO₂ removal strategy.

1. Introduction

The continuous increase in atmospheric carbon dioxide (CO₂) concentrations over the past 120 years has raised concerns about its impact on climate change and its far-reaching consequences for human society (Keeling et al 2001, Gattuso et al 2015). In response to this global challenge, the COP21 climate change conference declared that global warming should be limited to well below 2 °C above preindustrial levels by 2100. Achieving this ambitious target requires not only reducing CO₂ emissions but also actively sequestering CO₂ from the atmosphere through negative emissions strategies (Friedlingstein et al 2020, IPCC 2021).

The Earth's carbon cycle is profoundly influenced by natural rock and mineral weathering, playing a crucial role in the long-term removal of CO₂ from the atmosphere (Berner 1991, Schlesinger 1991). Terrestrial and marine weathering processes significantly contribute to carbon sequestration over geological timescales (Lenton and Britton 2006), and rock weathering consumes approximately 0.3 gigatons of carbon (GtC) annually (Gaillardet *et al* 1999, IPCC 2021).

One method for carbon dioxide removal (CDR) is to enhance naturally occurring rock weathering by dispersing finely ground silicate and/or carbonate minerals on land (enhanced weathering) or in the sea (ocean alkalinity enhancement, OAE) (Seifritz 1990, Oelkers 2001, Hartmann and Kempe 2008, Meysman and Montserrat 2017, Montserrat et al 2017, Renforth and Henderson 2017). Through chemical weathering, for example, olivine minerals $((Mg, Fe)_2SiO_4)$ initiate a slow dissolution process, producing alkalinity and binding CO₂ as bicarbonate in aqueous form (equation (1)), enabling the ocean to store more CO_2 (Schuiling and Krijgsman 2006, Hangx and Spiers 2009). Olivine-rich rocks have garnered significant attention due to their naturally occurring, fast dissolution rate and relative abundance (Griffioen 2017, Meysman and Montserrat 2017). Various rocks contain olivine in different concentrations, such as dunite (over 90% olivine) and peridotite (~40% olivine) (Philpotts and Ague 2022). Moosdorf et al (2014) outline the availability of global distribution of olivinerich ultramafic rocks. This OAE technique demonstrates significant potential in the open ocean (Ilyina et al 2013, Köhler et al 2013) and coastal areas (Hangx and Spiers 2009, Feng et al 2017). Coastal sea deployments of olivine-rich rocks offer cost-effective and energy-efficient logistics due to shorter transport distances than open ocean alternatives (Hangx and Spiers 2009, Köhler et al 2013, Feng et al 2017). Additionally, it could benefit from specific physicochemical conditions along coastlines,

$$(Mg,Fe)_2SiO_4 + 4H_2O + 4CO_2 \rightarrow 2(Mg,Fe)^{2+} + 4HCO_3^- + H_4SiO_4.$$
 (1)

Abundant mafic and ultramafic rocks on Earth's surface suggest that the material quantity may not be a limiting factor (Hartmann et al 2013); however, the weathering rate varies considerably depending on environmental conditions. While there is growing interest in OAE, studies on how regional environmental conditions affect olivine weathering remain scarce. For instance, chemical weathering rates of olivine-rich powder were addressed in lab settings over timescales of up to a year (e.g. Montserrat et al 2017, Fuhr et al 2022). The olivine chemical weathering rates are significantly influenced by temperature and grain size (Hangx and Spiers 2009, Köhler et al 2013, Feng et al 2017, Oelkers et al 2018), with larger particles dissolving more slowly than smaller ones (Rigopoulos et al 2018). While global models have explored olivine's overall weathering potential, they often overlook the time dimension (Hangx and Spiers 2009, Köhler et al 2010). The practical

applicability of enhanced weathering requires achieving specific CO_2 sequestration targets within policyrelevant timeframes, such as by 2100 (IPCC 2021). However, Feng *et al* (2017) emphasized the importance of conducting more refined regional studies to assess the technological and environmental feasibility of specific coastal deployment scenarios.

Therefore, this study aims to provide insights into the CO_2 sequestration efficiency of ocean alkalinization by olivine-rich rocks dissolution. The study covers different grain sizes and temperatures, representing 13 distinct coastal regions worldwide. By simulating weathering in a 100 year timeframe using geochemical thermodynamic equilibrium modeling (PHREEQC), we show the sequestration potential of olivine-rich rocks spreading under different conditions in policy-relevant timeframes. These findings support mitigation efforts by identifying regions with the highest physicochemical potential for coastal enhanced weathering, contributing to sustainable solutions to climate change challenges.

2. Methodology

2.1. Model description

Carbon sequestration was modeled utilizing the software PHREEQC version 2 (Parkhurst and Appelo 2013) following the approach outlined by Zhang et al (2019) utilizing its multicomponent geochemical model code and databases. For this study, the Lawrence Livermore National Laboratory thermodynamic database was used, which includes olivine (in the form of forsterite $(Mg_{1.682}Fe_{0.31}Ni_{0.008}SiO_4))$ equilibrium constants (Griffioen 2017). A box model was developed to simulate the dissolution of 1 g of forsterite with a grain size of 100 μ m in 1 kg of seawater. The 1:1000 water-rock ratio was chosen to balance realism, ensuring that the experimental conditions mirrored real-world scenarios as closely as possible, while also maintaining solubility control and practicality in laboratory settings. The model integrates forsterite mineralogy and incorporates thermodynamic data, including the dissociaton constant (log k =27.86) and enthalpy $(-205.61 \text{ kJ mol}^{-1})$ determined for forsterite (Zhang et al 2019).

The model considered various factors for forsterite dissolution in seawater across 13 regions (table 1). This included the composition of seawater, including concentrations of Na, K, Mg, Ca, Cl, SO₄, HCO₃, and Si, in addition to pH and temperature. Seawater pH and their chemistry data were sourced from literature and governmental datasets (table S1 in supplementary material). Sea surface temperature (SST) data were obtained from NOAA's daily satellite observations (Huang *et al* 2021). Table 1 summarizes input seawater temperatures and pH used in the models for the thirteen regional seas, representing a range of

Climate	Location/Sea	SST (°C)			рН		
		Mean SST of the coldest month	Annual mean SST	Mean SST of the warmest month	Cold	Average	Warm
Temperate Region	Black Sea	3.1	7.2	26.7	8.35	8.38	8.41
	Mediterranean	7.6	14.7	29.2	8.03	8.34	8.53
	Sea						
	North Sea	3.0	11.1	18.7	7.87	8.15	8.49
	Baltic Sea	1.6	10.9	20.2	7.90	8.20	8.50
	Marmara Sea	8.8	17.0	25.1	7.90	8.20	8.50
Tropical Region	Mexico Gulf	13.2	22.1	30.9	8.20	8.22	8.22
	Caribbean Sea	24.8	27.7	30.5	7.93	8.00	8.16
	Red Sea	19.5	26.0	32.5	8.20	8.45	8.70
	Arabian Gulf	22.1	27.6	33.0	7.80	8.20	8.10
	Bay of Bengal	25.3	28.4	30.9	7.47	7.75	7.93
	Arabian Sea	25.3	28.4	31.2	7.87	8.03	8.10
	South China Sea	21.5	26.6	30.0	7.66	8.04	8.40
	Banda Sea	27.7	29.3	31.2	8.20	8.20	8.20

Table 1. Modeled locations and input temperature for different scenarios.

coastal environments, including annual averages and extremes.

In PHREEQC, kinetic modeling involves two primary data blocks: KINETICS and RATES (Parkhurst and Appelo 2013). The KINETICS data block defines the initial moles of the reacting phase, in this case, forsterite (0.0066 mol), and initial mineral surface area (0.2291 m² g⁻¹) calculated based on Strefler *et al* (2018) with a scaling factor of 0.131. The RATES data block specifies kinetic parameters, including rate constants, activation energies, and reaction orders (Parkhurst and Appelo 2013). This model utilizes BASIC language scripts developed by Zhang *et al* (2019) for forsterite.

The model simulates the gradual dissolution of forsterite while maintaining equilibrium with atmospheric O_2 and CO_2 concentrations in the form of gases and minerals. It also allowed for the precipitation of goethite, calcite, aragonite, magnesite, talc, sepiolite, and amorphous silica if they become saturated. The model prescribes that water and atmosphere are in equilibrium (p $CO_2 = 400$ ppm and $pO_2 = 20\,000$ ppm), as described by Griffioen (2017).

2.2. Calculation of CO₂ sequestration

PHREEQC tracked the moles of reactants, representing the amount of olivine that had reacted over time, following the dissolution kinetics defined for the system (Parkhurst and Appelo 2013, Zhang *et al* 2019). The stoichiometry of the reactions governing olivine dissolution and the subsequent conversion of dissolved species to solid carbonate minerals was considered, allowing for the calculation of CO_2 sequestration. The corresponding moles of CO_2 involved in the reactions were determined for each mole of forsterite consumed. These mole-based calculations were then converted to mass units using the molar mass of CO_2 .

2.3. Sensitivity analysis

A sensitivity analysis was conducted using 42 simulations, varying temperature from 5 °C to 30 °C in 5 °C increments and grain size from 1 to 1000 μ m (1, 10, 50, 100, 300, 500, 1000), to determine weathering efficiency within a 100 year timeframe in annual time steps. The specific surface area for the material of the mentioned grain sizes was calculated based on Strefler *et al* (2018). For example, the calculated specific surface area ranges from 69.18 m² g⁻¹ for 1 μ m grain size down to 0.013 m² g⁻¹ for 1000 μ m grain size.

2.4. Mapping of CO₂ sequestration

To calculate the annual CO_2 sequestration potentials of olivine-based enhanced weathering in coastal areas, those were identified by masking the GEBCO bathymetry dataset (GEBCO 2021) with depths between 0 and -200 m, representing shelf areas. Monthly mean SST data from NOAA NCDC OISST 1991–2020 (Huang *et al* 2021) were combined to create an annual mean SST map. All rasters were resampled to a 10 km resolution to harmonize the datasets and improve computing efficiency. SST data were related to the bathymetry raster using zonal statistics. Spatial data were processed with ArcGIS Pro 3.

3. Results

3.1. Regional variability of CO₂ sequestration over 100 years

Sequestration of CO_2 exhibits significant regional variability during the simulated period (figure 1). The annual average conditions sequestration capacity ranges from 0.13 to 0.94 t CO_2/t olivine. CO_2 sequestration through olivine weathering depends on the forsterite content. The results presented here assume an olivine composition of 80% forsterite



and 20% fayalite, as calculated using Equation in the methodology. If the forsterite content were 90%, CO₂ sequestration would range from 0.14 to 1.04 tons per ton of olivine. In practice, olivine-rich rock, rather than pure olivine, would be applied to the waters. The amount of olivine-rich rock needed to achieve the same sequestration rates is inversely proportional to the rock's olivine content. For example, for a peridotite containing 40% olivine, 2.5 times more rock would be needed compared to pure olivine to achieve the same CO2 sequestration. The Banda Sea exhibits the highest sequestration rates, while the lowest are calculated for the Black Sea (figure 1). Sequestration rates in temperate seas range 0.13-0.32 t CO₂/t olivine, in the Black and Mediterranean Sea, respectively. Warmer seas show higher sequestration rates, with the Red Sea and South China Sea averaging 0.84 t CO₂/t olivine.

In the annual average cold conditions across different seas, the CO₂ sequestration within 100 years ranges from 0.07 to 0.88 t CO₂/ t olivine (figure 1). In temperate seas, the CO₂ sequestration rates range from 0.07 t CO₂/t olivine removed in the Baltic Sea to 0.17 t CO₂/t olivine removed in the Marmara Sea. For tropical seas, the range extends from 0.53 t CO₂/t olivine removed in the Red Sea to 0.88 t CO₂/t olivine removed in the Banda Sea.

In the annual warm conditions, CO_2 sequestration rates range from 0.46 to 0.98 t $CO_2/$ t olivine dissolution (figure 1). The Baltic Sea exhibits a sequestration rate of 0.46 t CO_2/t olivine, while the Red Sea and the Persian Gulf demonstrate the highest rates at 0.98 t CO_2/t olivine. In temperate seas, rates range from 0.46 (Baltic Sea) to 0.94 (Mediterranean Sea) t CO_2/t olivine. For tropical seas, rates are 0.94 (Caribbean Sea) and 0.98 (Red Sea and Persian Gulf) t CO_2/t olivine.

The percentage of olivine weathered in the warmer regions was between 13% and 18% after 10 years, 54%–69% after 50 years, and 84%–95%

after 100 years (figure 1). In temperate regions, the cumulative weathering of olivine would be 2%-5% after 10 years, 7%-22% after 50 years, and 13%-40% after 100 years.

3.2. Sensitivity analysis

The sensitivity analysis shows that smaller grain sizes and higher temperatures result in significantly higher CO₂ sequestration, while larger grain sizes and lower temperatures lead to lower CO₂ sequestration (figure 2). Among the tested scenarios, grain sizes $\leq 10 \,\mu m$ sequestered CO₂ the fastest (figures 2(a) and (b)). For instance, with a grain size of 1 μ m and various temperature combinations, complete dissolution occurs within 1-10 years, resulting in a CO₂ sequestration of 0.98 t CO_2 /t olivine (figure 2(a)). Similarly, for a grain size of 10 μ m and temperature combinations ranging from 10 °C to 30 °C, complete dissolution occurs within 9-84 years, while a grain size of 10 μ m and temperature 5 °C result in 90% dissolution at 100 years, with a CO_2 sequestration of 0.9 t CO_2/t olivine (figure 2(b)).

For larger grain sizes, the dissolution rates decrease. For a grain size of 100 μ m, dissolution ranges from 10%–90% at the end of the 100 year simulation period (figure 2(c)). In the case of 1000 μ m grain size, the dissolution is only 8%, even at temperatures of 30°C, leading to a relatively lower CO₂ consumption ranging from 0.01 to 0.08 t CO₂/t olivine throughout the simulation (figure 2(d)).

4. Discussion

4.1. Sequestration variability

The regional differences in CO_2 sequestration rates across the 13 studied seas indicate that the geographical location strongly controls the effectiveness of olivine-based enhanced weathering. Each region has its unique combination of climate and environmental factors, contributing to the observed variability



in CO₂ sequestration. Seawater temperature was the variable that controlled most of the process. Warmer waters provide more favorable conditions for enhanced weathering (Köhler *et al* 2013). The increased temperature accelerates chemical reactions, leading to faster dissolution of olivine particles and more efficient conversion of CO₂ into carbonate minerals (Oelkers *et al* 2018). Our results underline that these differences amount to an additional 0.4 t CO₂/t olivine dissolved in seawater in warm seas in 100 years (figure 1).

Furthermore, the simulations estimated the total mass of weathered olivine (figure 1), which aligns with the analytical solutions given by Hangx and Spiers (2009) and the Earth system model assessed by Feng et al (2017). Grain size was the second variable strongly influencing olivine dissolution. Hangx and Spiers (2009) found that 100 μ m olivine grains would require more than 100 years to dissolve at a tropical SST of 25 °C, consistent with the results obtained for the Red Sea in the current simulation (SST: 26 °C). Similarly, the complete dissolution of olivine in temperate regions at the same grain size may take more than 1000 years (similar to the Black Sea). This highlights the importance of reducing the grain size to accelerate the dissolution and achieve significant CO₂ sequestration, even in colder regions.

4.2. Impact of seawater temperature

CO₂ sequestration rates of enhanced olivine-rich rock weathering are susceptible to seawater temperature

(figure 3(a)). Lower temperatures lead to slower reactions, and higher temperatures accelerate the process. The CO₂ sequestration rates follow an exponential function, which can be approximated by a linear relation of a seawater temperature range of 15 °C–25 °C (figure 3(b)). At these temperatures, an additional 1.5 kg CO₂/t olivine is sequestered per year in the sea per degree of temperature increase (figure 3(b)). The linear correlation can be used to compare or correct field results at different temperature levels.

From a global perspective, fast weathering is evident in East Asian Seas (e.g. Banda Sea, South China Sea), while the coastal seas of the Northwest Pacific (e.g. Yellow Sea, East China Sea) and North-East Atlantic (e.g. Baltic, and North Sea) exhibit relatively slower weathering but encompass extensive areas (figure 4). The total considered coastal area is 26.6 Mkm² (figure 4), of which 6.53 Mkm² are kinetically favorable for EW by allowing more than 80% of olivine (grain size 100 μ m) to dissolve within 100 years. Despite reduced weathering speeds, other regions still hold significant potential for olivine-based enhanced weathering due to their extensive coastal coverage (see figure S2.1 in supplementary material for regional CO₂ sequestration). The observed trends in various regions on the global map contribute to the overarching theme emphasizing the importance of a tailored, regionspecific approach in olivine-based enhanced weathering strategies (figure 4).







4.3. Other controlling factors of CO₂ sequestration efficiency

The second key influencing factor is the grain size of the olivine-rich rocks. A smaller grain size increases the CO₂ sequestration rate of olivine weathering (figure 2) due to the exponential increase of mineral surface area with a decrease in grain size (Strefler et al 2018). Grains with a size of $\leq 10 \ \mu m$ show a very high CO_2 sequestration efficiency (figures 2(a) and (b)). Larger grain sizes, especially in temperate conditions, experience a decline in CO₂ sequestration due to slower reaction rates in colder waters (figures 2(c)) and (d)). The reduced efficiency of olivine weathering in these conditions leads to a decreased rate of CO₂ sequestration. Thus, in cold regions, more energy needs to be invested in milling to allow weathering rates high enough to be relevant at timescales of decades.

Another influence factor is seawater chemistry, which may affect CO_2 sequestration. The model incorporated a comprehensive range of seawater chemistry data collected from 13 distinct regional



seas. The simulations maintained a constant average seawater temperature of 17 °C and a pH of 8.1. The range spanned minimum, maximum, and average seawater conditions (based on major ion content, see table S1), with corresponding sequestration rates of 0.34, 0.40, and 0.42 t CO_2/t olivine (figure 5). This



variation is attributed to the availability of reactants in the surrounding water, which influences olivine dissolution rates. In regions where seawater chemistry deviates from the average, whether due to natural variations (geological and climatic differences) or anthropogenic influences (like industrial discharges), the efficiency of olivine-based enhanced weathering can be notably impacted.

4.4. Optimizing CO₂ sequestration: grain size and temperature interplay

Selecting the appropriate grain size based on specific environmental conditions becomes crucial for maximizing CO₂ sequestration efficiency (Strefler *et al* 2018). Figure 6 shows a 2D mapping of CO₂ sequestration rates as a function of grain size and seawater temperature over 100 years. For instance, enhanced weathering with olivine in the Black Sea would require a grain size of 15 μ m to reach a CO₂ consumption ratio of 0.8 t CO₂/t olivine after 100 years, while the Banda Sea achieves the same ratio with a grain size of 150 μ m (figure 6). This aligns with previous findings emphasizing the significance of small-grained olivine (2–6 μ m) to achieve substantial CO₂ sequestration rates over several decades (Hangx and Spiers 2009).

However, reducing the material grain size requires energy input from crushing, grinding, or milling. This energy requirement will to some extent incur CO_2 emissions, a penalty that can diminish the efficiency of olivine-rich rocks spreading for CO_2 sequestration (Hangx and Spiers 2009, Moosdorf *et al* 2014). Renforth (2012) highlights the impracticality of grinding olivine to a size below 10 μ m in most locations due to cost and time constraints. Despite this, the CO₂ emissions resulting from the grinding and crushing of olivine are generally low compared to the achieved sequestration (Moosdorf *et al* 2014). The emissions associated with primary crushing and secondary grinding of olivine range from 3.1 to 110.5 kg of CO₂ per t of CO₂ sequestered, depending on olivine particle size (Hangx and Spiers 2009). In an energy mix with more renewables, the associated CO₂ penalty would be lower.

Net CO₂ sequestration (i.e. sequestration after 100 years (figure 7(a)) minus the above-mentioned CO_2 emissions) of grain sizes of 10 μ m was consistently 0.87 t CO2 /t olivine for all scenarios (figure 7(b)). Warmer regions, such as the Banda Sea and Red Sea, show higher maximum net CO2 sequestration rates (0.97 t CO₂ /t olivine) at a grain size of 50 μ m. In the Banda Sea, even net CO₂ sequestration rates of olivine with a grain size of 100 μ m (0.93 t CO₂ /t olivine) exceed that of 10 μ m particles (figure 7(b)). These findings highlight that in warmer regions, higher net-CO₂ sequestration ratios per time can be reached using coarser grains, thereby optimizing the efficiency of CO₂ sequestration. Extrapolating these ratios and material application rates across diverse coastal surface areas in various regions allows for a robust estimate of the broader-scale CO₂ sequestration potential.

In the context of policy making, with specific CO_2 sequestration targets to be met in a defined timeframe, this study shows that by prioritizing regions with higher net CO_2 sequestration rates





(nearly 1 t CO₂ /t olivine), legislation can strategically focus efforts on maximum CO₂ sequestration. Additionally, adopting smaller grain sizes ($\leq 10 \ \mu$ m) in colder regions (like the European Union) presents an adaptive approach to optimizing weathering rates under less favorable temperature conditions at the cost of efficiency. By incorporating these scientific insights into policy frameworks, policymakers can make informed decisions and allocate resources effectively to regions and strategies with the highest potential for achieving CO₂ sequestration goals.

4.5. Model assumptions and limitations

The applied model substantially simplifies natural systems, designed only to highlight concepts and trigger discussions. It does not represent exact and detailed conditions at individual locations or cover the complexities of the natural world.

Like others studying marine enhanced weathering, one limitation of this approach is that it assumes the weathering takes place in the water column and the system is far from saturation. As shown, olivine weathering in seawater typically occurs relatively slowly, spanning decades for individual grains (figure 1). Applying the simplified Stokes' Law equation (Stokes 1851), allows to calculate whether the olivine stays in the water column for that time. Round olivine grains with a diameter of 10 μ m take a month (26.8 d for 1000 m) to settle, while grains with a diameter of 100 μ m take just days (0.26 d for 1000 m). In real-world complexities, ocean currents and turbulences substantially influence settling behavior. The comparison of days to settle and decades to weather indicates that most olivine would weather on the seafloor, especially considering that the general coastal oceans have a water depth of no more than 200 m (Tyler 2003). Therefore, the specific conditions for weathering on the seafloor deserve closer consideration in future analyses.

Additionally, this study neglects dynamic coastal currents, which could impact the olivine weathering rates. The study does not account for the evolving chemistry of seawater over time due to alkalization through advection, especially in open coastal systems. Furthermore, variations in seawater parameters such as $_{P}CO_{2,P}O_{2}$, seawater temperatures, and pH with depth are not fully addressed.

Another important aspect is that understanding environmental and ecological factors is vital for successfully and sustainably implementing coastal enhanced weathering. As olivine-rich rocks dissolves, it can increase seawater alkalinity locally, mitigating ocean acidification caused by excess CO2 absorption (Feng et al 2017). Nevertheless, the introduction of olivine-rich rocks, rich in silicon, iron, and trace heavy metals, may lead to eutrophication, harmful algal blooms, and oxygen depletion, necessitating careful evaluation and effective management strategies (Blain et al 2007, Hauck et al 2016). Thus, the environmental and ecological implications of coastal enhanced weathering, including benefits and potential risks, must be carefully considered to ensure responsible and sustainable implementation (Bach et al 2019). On the other hand, the relevance of microbiology on weathering rates is known from studies of natural chemical weathering on land (Rogers and Bennett 2004). However, this aspect has been neglected in analyses of enhanced weathering in the marine realm. Integrating microbiological aspects into this model would require additional data and a more intricate framework, exceeding the scope of the current investigation.

An increase in alkalinity may lead to calcium carbonate precipitation in the water column. This could counteract the intended benefits of ocean alkalinization by removing alkalinity, potentially reducing olivine dissolution efficiency (Moras *et al* 2022, Hartmann *et al* 2023). However, as per the geochemical character as silicate, the re-precipitation only releases 50% of the captured CO_2 (e.g. Hartmann *et al* 2013). Precipitation can be triggered during spreading and would impact olivine dissolution rates (Fuhr *et al* 2022), which is not considered here. Numerous intricate environmental factors (i.e. nutrient availability and microbial activity) affect the mineral reactivity and weathering rates, requiring consideration for specific enhanced weathering evaluations (Fuhr *et al* 2022, Wallmann *et al* 2022).

Lastly, the OAE linked CO_2 costs of mining and transport operations are not considered, given their demonstrated comparably small impact on coastal enhanced weathering (Moosdorf *et al* 2014). Also, internal transport processes like diffusion within the rock matrix are beyond the study's scope.

5. Conclusion

This study highlights the potential of olivine-rich rocks enhanced weathering to reduce atmospheric CO_2 levels along the coast within a defined time frame (100 years). The model simulated the weathering effect of 13 coastal seas worldwide, providing valuable insights into the variability of CO_2 sequestration rates. Each location has a maximum CO_2 sequestration capacity determined by its unique environmental conditions. In warmer regions, the maximum CO_2 sequestration capacity is higher than in cooler regions. For example, in the Banda Sea, the CO_2 sequestration capacity is estimated to be 0.94 t CO_2/t olivine in 100 years, while in the Baltic Sea, it is estimated to be 0.13 t CO_2/t olivine, using 100 μ m grain size.

The faster reaction rates observed in warmer areas highlight the importance of strategically focusing sequestration efforts on these regions to maximize CO₂ sequestration. Globally, 6.53 Mkm² of coastal seas allow for dissolution of 80% of olivine of a grain size of 100 μ m within 100 years. Additionally, the selection of the grain size plays a critical role in optimizing weathering rates. For instance, achieving a sequestration efficiency of 80% (0.8 t CO2 /t olivine) within 100 years requires a grain size of 15 μ m in the temperate Baltic Sea, while a larger grain size of 150 μ m is needed in the warmer Banda Sea. Achieving fast weathering rates through grain size reduction means putting more energy into crushing, which may introduce a CO₂ penalty if no renewable energy sources are available.

Adding the CO_2 costs of crushing procedures into the equation leads to a theoretical maximum amount of net CO_2 sequestration in a region in 100 years, which is 0.87 t CO_2 /t olivine in colder areas such as the Baltic Sea and close to its stoichiometric maximum with 0.97 t CO_2 /t olivine in warmer regions like the Banda Sea. These findings show that it strongly matters where olivine-rich rocks spread for enhanced weathering to meet specific political CO_2 sequestration targets within designated timeframes.

Data availability statement

The manuscript contains original contributions that are presented in detail within the article/Supplementary Materials. For additional information or inquiries, we encourage contacting the corresponding author.

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

This study was supported by the German Ministry for Education and Research through the Project RETAKE-L, which was granted to NM (BMBF Grant # 03F0895L). The funding was provided within framework of the Deutsche Allianz für Meeresforschung (DAM) mission CDRmare. Additional funding provided by Germany's Excellence Strategy-EXC 2037 Climate, Climatic Change, and Society-Project Number 390683824, contribution to the Center for Earth System Research and Sustainability (CEN) of the Universität Hamburg. The reseach leading to these results has received funding from the European Union's Horizon Europe research and innovation programme under the UPTAKE project (g.a. no. 101081521). The views and opinions expressed are, however, those of the author(s) only and do not necessarily reflect those of the European Union or CINEA.

Conflict of interest

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

ORCID iDs

Murugan Ramasamy b https://orcid.org/0000-0002-3023-9165

Thorben Amann (b) https://orcid.org/0000-0001-9347-0615

Nils Moosdorf
https://orcid.org/0000-0003-2822-8261

References

- Bach L T, Gill S J, Rickaby R E, Gore S and Renforth P 2019 CO₂ removal with enhanced weathering and ocean alkalinity enhancement: potential risks and co-benefits for marine pelagic ecosystems *Front. Clim.* 1
- Berner R 1991 A model for CO₂ over Phanerozoic time *Am. J. Sci.* **291** 339–76
- Blain S *et al* 2007 Effect of natural iron fertilization on carbon sequestration in the Southern Ocean *Nature* **446** 1070–4
- Feng E Y, Koeve W, Keller D P and Oschlies A 2017 Model-based assessment of the CO₂ sequestration potential of Coastal Ocean Alkalinization *Earth's Future* 5 1252–66
- Friedlingstein P *et al* 2020 Global carbon budget 2020 *Earth Syst. Sci. Data* **12** 3269–340
- Fuhr M, Geilert S, Schmidt M, Liebetrau V, Vogt C, Ledwig B and Wallmann K 2022 Kinetics of olivine weathering in seawater: an experimental study *Front. Clim.* **4** 39

Gaillardet J, Dupré B, Louvat P and Allègre C J 1999 Global silicate weathering and CO₂ consumption rates deduced from the chemistry of large rivers *Chem. Geol.* **159** 3–30

Gattuso J-P *et al* 2015 Contrasting futures for ocean and society from different anthropogenic CO₂ emissions scenarios *Science* **349** aac4722

GEBCO Bathymetric Compilation Group 2021 The GEBCO_2021 Grid - a continuous terrain model of the global oceans and land NERC EDS British Oceanographic Data Centre NOC (https://doi.org/10.5285/C6612CBE-50B3-0CFF-E053-6C86ABC09F8F)

Griffioen J 2017 Enhanced weathering of olivine in seawater: the efficiency as revealed by thermodynamic scenario analysis *Sci. Total Environ.* **575** 536–44

Hangx S J T and Spiers C J 2009 Coastal spreading of olivine to control atmospheric CO₂ concentrations: a critical analysis of viability *Int. J. Greenhouse Gas Control* **3** 757–67

Hartmann J and Kempe S 2008 What is the maximum potential for CO₂ sequestration by "stimulated" weathering on the global scale? *Naturwissenschaften* **95** 1159–64

 Hartmann J, Suitner N, Lim C, Schneider J, Marín-Samper L, Arístegui J, Renforth P, Taucher J and Riebesell U 2023
 Stability of alkalinity in ocean alkalinity enhancement (OAE) approaches—consequences for durability of CO₂ storage *Biogeosciences* 20 781–802

Hartmann J, West A J, Renforth P, Köhler P, De La Rocha C L, Wolf-Gladrow D A, Dürr H H and Scheffran J 2013
Enhanced chemical weathering as a geoengineering strategy to reduce atmospheric carbon dioxide, supply nutrients, and mitigate ocean acidification *Rev. Geophys.* 51 113–49

Hauck J, Köhler P, Wolf-Gladrow D and 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** 024007

Huang B, Liu C, Banzon V, Freeman E, Graham G, Hankins B, Smith T and Zhang H-M 2021 Improvements of the daily optimum interpolation sea surface temperature (DOISST) version 2.1 J. Clim. 34 2923–39

Ilyina T, Six K D, Segschneider J, Maier-Reimer E, Li H and Núñez-Riboni I 2013 Global ocean biogeochemistry model HAMOCC: model architecture and performance as component of the MPI-Earth system model in different CMIP5 experimental realizations J. Adv. Modeling Earth Syst. 5 287–315

IPCC 2021 IPCC, 2021: Climate Change 2021: The Physical Science Basis. Contribution of Working Group I to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change ed V Masson-Delmotte et al (Cambridge University Press) (https://doi.org/10.1017/9781009157896)

Keeling C D, Piper S C, Bacastow R B, Wahlen M, Whorf T P, Heimann M and Meijer H A 2001 Exchanges of atmospheric CO₂ and ¹³CO₂ with the terrestrial biosphere and oceans from 1978 to 2000. I Global Aspects (SIO Reference, Scripps Institution of Oceanography, 2001) (available at: https:// escholarship.org/uc/item/09v319r9)

Köhler P, Abrams J F, Völker C, Hauck J and Wolf-Gladrow D A 2013 Geoengineering impact of open ocean dissolution of olivine on atmospheric CO₂, surface ocean pH and marine biology *Environ. Res. Lett.* 8 014009

Köhler P, Hartmann J and Wolf-Gladrow D A 2010 Geoengineering potential of artificially enhanced silicate weathering of olivine Proc. Natl Acad. Sci. 107 20228–33

Lenton T M and Britton C 2006 Enhanced carbonate and silicate weathering accelerates recovery from fossil fuel CO₂ perturbations *Glob. Biogeochem. Cycles* **20** Meysman F J and Montserrat F 2017 Negative CO₂ emissions via enhanced silicate weathering in coastal environments *Biol. Lett.* **13** 20160905

Montserrat F, Renforth P, Hartmann J, Leermakers M, Knops P and Meysman F J R 2017 Olivine dissolution in seawater: implications for CO₂ sequestration through enhanced weathering in coastal environments *Environ. Sci. Technol.* 51 3960–72

Moosdorf N, Renforth P and Hartmann J 2014 Carbon dioxide efficiency of terrestrial enhanced weathering *Environ. Sci. Technol.* **48** 4809–16

Moras C A, Bach L T, Cyronak T, Joannes-Boyau R and Schulz K G 2022 Ocean alkalinity enhancement—avoiding runaway CaCO₃ precipitation during quick and hydrated lime dissolution *Biogeosciences* **19** 3537–57

Oelkers E H 2001 General kinetic description of multioxide silicate mineral and glass dissolution *Geochim. Cosmochim. Acta* 65 3703–19

Oelkers E H, Declercq J, Saldi G D, Gislason S R and Schott J 2018 Olivine dissolution rates: a critical review *Chem. Geol.* **500** 1–19

Parkhurst D and Appelo T 2013 Description of input and examples for PHREEQC version 3—a computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations US Geological Survey Techniques and Methods vol 6 (U.S. Geological Survey) ch A43, p 497

Philpotts A R and Ague J J 2022 *Principles of Igneous and Metamorphic Petrology* (Cambridge University Press)

Renforth P 2012 The potential of enhanced weathering in the UK Int. J. Greenhouse Gas Control 10 229–43

Renforth P and Henderson G 2017 Assessing ocean alkalinity for carbon sequestration *Rev. Geophys.* **55** 636–74

Rigopoulos I, Harrison A L, Delimitis A, Ioannou I, Efstathiou A M, Kyratsi T and Oelkers E H 2018 Carbon sequestration via enhanced weathering of peridotites and basalts in seawater *Appl. Geochem.* **91** 197–207

Rogers J Roberts and Bennett P C 2004 Mineral stimulation of subsurface microorganisms: release of limiting nutrients from silicates *Chem. Geol.* **203** 91–108

Schlesinger W 1991 Biogeochemistry: An Analysis of Global Change (Academic)

Schuiling R D and Krijgsman P 2006 Enhanced weathering: an effective and cheap tool to sequester CO_2 Clim. Change 74 349–54

Seifritz W 1990 CO₂ disposal by means of silicates *Nature* **345** 486

Stokes G G 1851 Transactions of the Cambridge Philosophical Society vol IX p 8 (Reprinted in Mathematical and Physical Papers, Sir George Gabriel Stokes and Sir J Larmor vol 3 pp 1880–905)

Strefler J, Amann T, Bauer N, Kriegler E and Hartmann J 2018 Potential and costs of carbon dioxide removal by enhanced weathering of rocks *Environ. Res. Lett.* 13 034010

Tyler P A 2003 *Ecosystems of the Deep Oceans* (Elsevier)

Wallmann K, Diesing M, Scholz F, Rehder G, Dale A W, Fuhr M and Suess E 2022 Erosion of carbonate-bearing sedimentary rocks may close the alkalinity budget of the Baltic Sea and support atmospheric CO₂ uptake in coastal seas *Front. Mar. Sci.* 9 968069

Zhang Y, Hu B, Teng Y, Tu K and Zhu C 2019 A library of BASIC scripts of reaction rates for geochemical modeling using PHREEQC Comput. Geosci. 133 104316