

# Supplementary Information for “Marine Carbon Dioxide Removal by alkanization should no longer be overlooked”

Katarzyna A. Kowalczyk<sup>1,\*</sup>, Thorben Amann<sup>2</sup>, Jessica Strefler<sup>1</sup>, Maria-Elena Vorrath<sup>2</sup>, Jens Hartmann<sup>2</sup>, Serena De Marco<sup>3</sup>, Phil Renforth<sup>4</sup>, Spyros Foteinis<sup>4</sup>, Elmar Kriegler<sup>1,5</sup>

<sup>1</sup> Potsdam Institute for Climate Impact Research (PIK), Member of the Leibniz Association, Potsdam, Germany

<sup>2</sup> Institute for Geology, Center for Earth System Research and Sustainability, Universität Hamburg, Hamburg, Germany

<sup>3</sup> Department of Civil and Environmental Engineering, Politecnico Di Milano, Milan, Italy

<sup>4</sup> Research Centre for Carbon Solutions, School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, United Kingdom

<sup>5</sup> Faculty of Economics and Social Sciences, University of Potsdam, Potsdam, Germany

\* Corresponding author

**Email:** kowalczyk@pik-potsdam.de

## Table of Tables

Table S1. Spreading alkalinity by different types of maritime fleets..... 7

## Table of Figures

Figure S1. Uptake efficiency vs temperature..... 8

Figure S2. Uptake efficiency vs atmospheric partial pressure..... 9

Figure S3. Energy demand vs discharge rate..... 9

# S1 Ocean alkalinity enhancement methods screening

A variety of methods to enhance ocean alkalinity have been proposed in the literature. However, so far, it was not clear which idea might be the most promising. Here, we discuss the methods in terms of feasibility, scalability, and the status of their technological development. We start with the simplest approach, which involves introducing alkalinity to the surface ocean in the form of powdered natural rocks, which are dissolved, a process similar to the chemical weathering of rocks on land. Then we look into generating an alkaline solution in a reactor and by electrochemical splitting. Lastly, we investigate the use of synthetic alkaline materials for ocean alkalinity enhancement (OAE), focusing on the spreading of calcium hydroxide in surface ocean waters, a process known as ocean liming (OL).

## S1.1 Rock powder distribution

Two typically evaluated materials for increasing the ocean's alkalinity are silicate and carbonate rocks, both highly abundant in the Earth's crust.<sup>1,2</sup> As they dissolve in the ocean, CO<sub>2</sub> is consumed and alkalinity is created, leading to a flux of CO<sub>2</sub> from the atmosphere to the ocean. The amount of CO<sub>2</sub> sequestered depends on the dissolution speed of alkaline materials, which, in turn, primarily depends on the type and grain size of the material, water temperature, salinity, and ocean chemistry. An alkaline material distributed in the ocean must dissolve within the mixed surface layer to maximize the atmospheric CO<sub>2</sub> uptake, thus has to be either ground to appropriate grain sizes to maximize the residence time in the surface waters of the open ocean,<sup>3</sup> or be distributed in coastal zones<sup>4,5</sup>.

**Silicate rocks**, e.g., dunite or peridotite, which are rich in olivine minerals, such as forsterite (Mg<sub>2</sub>SiO<sub>4</sub>), can be efficient in sequestering CO<sub>2</sub> but contain trace elements like nickel, chromium, and cobalt that are released during dissolution,<sup>6</sup> which may have harmful effects on the marine ecosystem.<sup>7,8</sup> In addition, due to the material's comparably low dissolution speed, it is estimated that it would have to be ground to a grain size of 1 μm<sup>3,9</sup> to fully dissolve within the mixed layer (that increases the electricity demand to 300–350 kWh t<sup>-1</sup><sup>10</sup>). Besides the significant energy requirements to achieve this, such a small grain size would be difficult to handle and comes with potential health hazards from respiration. When spreading olivine in coastal regions, grains could be larger than in the case of the open ocean distribution because coastal wave and tidal action facilitate grain collisions and, therefore, mineral dissolution.<sup>5,11</sup> However, materials spread in coastal areas might be buried in the sediment, which would slow down or cease dissolution and lead to an accumulation of trace metals.<sup>6,7</sup> Silicate rocks containing lower amounts of olivine such as basalt could reduce the risk of trace metals but have about 2/3 lower carbon capture potential compared to dunite.<sup>12</sup> Also, as basalt powder is used as a fertilizer in agriculture, the released potassium, phosphorus, and other micronutrients could lead to an elevated nutrient supply and therefore fuel algae blooms, thus directly impacting marine food webs.<sup>13</sup>

Nickel, in particular, can be harmful to many marine organisms, even in small quantities. The concentration of nickel in seawater is on average 5.4 μmol/kg and 4.5 μmol/kg seawater in coastal areas (< 200 m water depth) respectively<sup>14</sup> but olivine spreading could significantly increase the levels of nickel and other trace elements in the surrounding water. Meanwhile, the impacts of elevated marine nickel concentrations are yet not understood as many marine

organisms are sensitive to elevated trace element concentrations<sup>8</sup> but other studies do not show sensitivities of single phytoplankton species did not occur<sup>15</sup>. Further, the long-term effects as well as effects on benthic species in marine sediments have not been studied so far and need to be understood before olivine spreading can be considered a viable option for large-scale CDR.

**Carbonate rocks.** An alternative to silicate rocks containing olivine could be using carbonates, e.g., limestone,<sup>16–18</sup> which dissolves several orders of magnitude faster than forsterite and is not expected to contain potentially toxic trace metals.

However, dissolving carbonates increases the carbonate concentration in seawater, which is already supersaturated with respect to carbonate minerals.<sup>19</sup> When limestone dissolves in seawater and is not counterbalanced by CO<sub>2</sub> uptake (CO<sub>2</sub>-equilibration) fast enough, the crossing of critical thresholds of the saturation indices of calcite and aragonite lead to rapid precipitation of carbonates, the so-called runaway precipitation.<sup>20</sup> The precipitation of carbonates, in turn, leads to the release of CO<sub>2</sub> and significantly reduces the efficiency of CO<sub>2</sub> removal or even results in a net release of CO<sub>2</sub>.<sup>19,20</sup>

## S1.2. Reactor-produced alkalinity

Using an alkaline solution that is equilibrated with CO<sub>2</sub> instead of powdered minerals could minimize the risk of post-deployment precipitation.<sup>19</sup> The alkaline solution could be generated by mixing minerals and CO<sub>2</sub> with seawater in a reactor, where temperature, pressure, and CO<sub>2</sub> concentration can be controlled. An alkaline solution could be diluted and distributed in rivers and oceans to store the CO<sub>2</sub> on a geological timescale.<sup>18</sup> Although extensive literature in this field is still lacking, few pilot studies have attempted to investigate this approach.<sup>21–23</sup>

Olivine and limestone dissolve faster in water that is exposed to a CO<sub>2</sub>-rich gas stream leading to a pH decrease that accelerates the chemical reaction.<sup>18,24,25</sup> With limestone as feedstock, the method is called accelerated weathering of limestone (AWL). AWL could utilize a gas stream from a direct air capture (DAC) or a biomass plant (to store the CO<sub>2</sub>, an alternative to geological storage), as well as an industrial process, such as cement production, for emissions avoidance. If combined with DAC or bioenergy with carbon capture, the system would be a long-term CDR, however, OAE is rather a storage part of the operation. For this method to be considered CDR alone, ambient air would have to be used in the reactor. However, under ambient conditions, the time required for the reaction to occur would be significantly longer, leading to very low efficiency.

Increasing alkalinity via ground minerals highly depends on the dissolution as shown in S1.1. The dissolution could be accelerated by mixing minerals with seawater in a reactor, where the temperature, pressure, and CO<sub>2</sub> concentration can be regulated. That being the case, the resulting alkaline solution could be diluted and disposed of into the ocean to store the CO<sub>2</sub> permanently. Few pilot studies have attempted to investigate this approach. However, the literature regarding the process is still lacking.

It has been shown that olivine and limestone dissolve faster in water that is exposed to a CO<sub>2</sub>-rich gas stream, which has a much higher concentration of CO<sub>2</sub>, leading to a pH increase that drives the chemical reaction.<sup>18,24</sup> If limestone is used, the method is referred to as accelerated

weathering of limestone (AWL). AWL could utilize a gas stream from a direct air capture plant (to store the CO<sub>2</sub>, alternatively to geological storage), or an industrial process, such as cement production, for emissions avoidance. However, in both cases, AWL is a carbon storage method, not carbon dioxide removal, thus we will not consider it further in this study. For this method to be considered CDR, the gas stream would have to be the ambient air.

Raising the partial pressure of CO<sub>2</sub> to >5000 µatm (at a total pressure of 1 atm) in contact with seawater creates the conditions in which carbonate minerals can spontaneously dissolve.

### S1.3 Electrochemical splitting

Adding limestone to an acidic anolyte, part of a water electrolysis cell, increases the mineral's dissolution as well as the water's alkalinity.<sup>18</sup> This leads to a drawdown of CO<sub>2</sub> and durable storage if the solution is released into the ocean. The technology requires a significant amount of electricity (8 GJ per tCO<sub>2</sub> consumed). However, 45 kg of hydrogen is produced per 1 tCO<sub>2</sub> captured, which corresponds to 5.4 GJ of energy.<sup>26</sup> Additionally, electrochemical splitting does not require carbon storage (CCS), unlike several other CDR technologies such as DACCS, BECCS, or further discussed ocean liming. The advantages of no carbon storage requirements and low additional energy requirements on top of the hydrogen production could make electrochemical splitting an attractive OAE option. Despite the advantages, we do not consider the method in this study as more research is needed to reduce uncertainties around costs and potential limitations as well as the need for technological development.

### S1.4 Ocean Liming

The reaction of CaO in contact with water is quick and exothermic, thus the transportation of quicklime must be handled carefully. Dissolving one mol of CaO in 1kg of water at 20°C increases the water's temperature by 15.5°C. However, such a temperature increase would not occur, as the reaction is limited by the saturation of Ca(OH)<sub>2</sub>, which limits the temperature increase to 0.36°C. The resulting rise in water temperature is thus negligible.

Lime could be distributed as a powder if a safe concentration that avoids precipitation is not exceeded. Another option is to distribute the alkalinity as a slurry of Ca(OH)<sub>2</sub> since rock powder (order of 10 µm) might be hazardous to human health and the environment.<sup>27</sup> The rapid, exothermic, and volume increasing hydration reaction results in physical decomposition of the material such that commercially produced hydrated lime has a small particle size (typically 2-5 µm, but <1 µm possible) and a large surface area (7 – 15 m<sup>2</sup> g<sup>-1</sup>).<sup>28,29</sup> For comparison, particles of sizes 40-60 µm are used in the literature.<sup>19,20,30</sup> To minimize the weight loaded on the ship, the slurry could be created shortly before the discharge. Ships already pump significant amounts of water, which could be used to create the slurry, for their cooling system to remove the unused heat from the ships' engines and other machinery. The slurry would have to stay away from contact with the atmosphere to avoid precipitation on deck. The discharge rate should not exceed the safe limit above which precipitation might occur. The dilution would happen in the ocean as the ship moves.

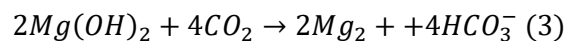
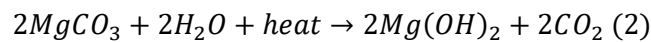
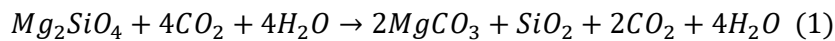
There exist ways to avoid geological storage if OL is combined with other processes. For instance, an alternative approach to ocean liming couples ocean liming with mineral carbonation

(MC-OL),<sup>31</sup> which allows for CO<sub>2</sub> recycling (described below). Alternatively, CO<sub>2</sub> emissions from calcination in OL could be utilized in a reactor (Note S1.2) also avoiding geological storage.

An alternative is to inject alkalinity within coastal areas using networks of pipes. This method has been proposed to lower the cost of distribution, however, at the same time, it limits the destined area significantly. For large-scale deployment, the utilization of ships might be necessary to avoid excessive local pH and aragonite saturation increases in the coastal areas.<sup>32</sup>

### Combined Mineral Carbonation and Ocean Liming

An alternative approach to ocean liming couples ocean liming with mineral carbonation (MC-OL).<sup>31</sup> Its advantage over OL is that it doesn't require CCS for the emissions from the chemical process (0.44 tCO<sub>2</sub> per 1t of limestone). In the first step, called mineral carbonation, silicate rocks are reacted with CO<sub>2</sub> to form stable carbonate minerals, i.e., magnesite (MgCO<sub>3</sub>) (Reaction 1). The second step is the calcination of MgCO<sub>3</sub>, which decomposes to CO<sub>2</sub> and brucite (Mg(OH)<sub>2</sub>) (Reaction 2), the latter used as the feedstock to ocean liming instead of quicklime (Reaction 3). The MC process could use the CO<sub>2</sub> produced during calcination, thus recycling emissions from the chemical decomposition and avoiding CCS. MC-OL would therefore be preferred over ocean liming in scenarios with limited CCS availability.



The MC-OL steps consist of olivine extraction, grinding, mineral carbonation, and calcination (no CCS required), comminution, and production and distribution of the alkaline solution. Olivine deposits are estimated to be in the order of hundreds of gigatons, while 8.4Mt of olivine and 1.5Mt of brucite are produced globally every year.<sup>2</sup>

## S2 Distribution in coastal areas

Alkaline material distributed as powder in coastal areas might lead to excessive local pH and aragonite saturation increases, which limits the potential significantly.<sup>32</sup> To minimize the local mineral concentration when alkalinity is added to coastal areas, which are often characterized by slow mixing could be using reactors. An example of a reactor exists in Lausitz, Germany, and has been used to neutralize acid-mining lakes. The reactor is included in a closed system made of four major components: a standard slake lime production unit, a carbon dioxide dissolution reactor, a gas recovery reactor, and an injection system. The alkalinity is produced by mixing a stream of CO<sub>2</sub>-enriched water coming from the carbon dioxide dissolution reactor with pit lake water and a lime suspension.

Previously, the treatment of coal mine drainage had been addressed in<sup>33</sup>, using almost similar instrumentation but intermittently fluidized beds of limestone as alkalinity reactors. The main components of the system were a packed column to mix CO<sub>2</sub> with acidic lake water, a fluidized bed reactor to allow the reaction between calcite and CO<sub>2</sub> exiting the carbonator, and a system of air stripping column and tanks to remove the excess CO<sub>2</sub> from the treated water. Overall, at

its simplest, an alkalinity reactor can be thought of as a tubular flow reactor equipped with an agitator or a circulation device to maintain turbulence. Achieving an adequate degree of agitation is crucial as different reaction phases (i.e., atmospheric CO<sub>2</sub> usually mixed with water before entering the reactor, a slurry of alkaline material, and water) must be properly blended to generate a suitable CO<sub>2</sub>-equilibrated alkaline solution.

The maximum concentration of CO<sub>2</sub>, alkaline material, and seawater flow rate introduced in the reactor and the production time (i.e., residence time to allow the mixing plus required time to achieve the equilibration of the solution) affect the efficiency with which all the processes are carried out. To make this method a CDR alone, the stream would have to come from ambient air, so the CO<sub>2</sub> concentration would be much lower than used thus far. Using limestone in this case would result in a low efficiency due to slow dissolution. Quicker dissolving materials, e.g., CaO might be used instead. However, this method has not been tested yet. To create water equilibrated with the atmosphere, the alkalinity added should be the result of precalculated ratios of HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup> instead of the pure form of OH<sup>-</sup>.<sup>19</sup> However, the way and the magnitude with which factors such as the quality of particle surface and the amount of surface area per volume of water to be chosen, and the dilution rate of treated and untreated water representative of the application site, contribute to triggering the alkalinity loss remain unclear and under investigation.<sup>19</sup>

### S3 Distribution on ships

The use of a fleet completely devoted to CDR purposes instead of an existing and partially dedicated fleet allows a list of advantages. Among them, flexibility in the management and higher discharge rates are the most relevant. In addition, if the fleet was composed of existing and unutilized ships, the purchase capital expenditures could be curtailed by the total cost of the spreading. Such a decision is also supported by other legal and practical reasons.

Using existing ships nowadays implies compliance with the new environmental regulations imposed by IMO in terms of Energy Efficiency Existing Ship Index (EEXI) and Carbon Intensity Indicator (CII). Such obligations came into force on 1<sup>st</sup> January 2023 and set limitations on the sailing speed (reduced) to save fuel. In the future, this may entail some ships being retrofitted, some others (the oldest ones) being put out of the market, and overall, a reduction of the fleet capacity.<sup>34</sup> The average age of bulk carriers is 11-14 years.<sup>34</sup>

Betting on newly built ships instead of recycling or re-organizing the existing ones, seems to be an easy way to cope with both environmental restrictions and implementation of OAE. However, uncertainties on future fuel and carbon prices, as well as on which technology is the most dominant could add complexity to the decision process. Moreover, even when the decision has been made, the required time for a shipping line to get new ships is not short; it could take from two to four years.<sup>34</sup> Adding new dedicated fleets may also have an impact on the ports' capacity. Expanding the current capacity may require building new terminals, providing additional highway lanes, rail service, new and larger cranes, dredging equipment, reinforced quay walls, and increasing the number of berths at terminals.<sup>35</sup>

Building new ports, on the other hand, takes years or decades to carry through due to the establishment of inland connections and compliance with local and/or global environmental and trade regulations.<sup>34</sup> Examples of local limiting factors may be the tariff increases imposed by some

countries, labor shortages, and shortage of infrastructures and facilities such as ship loading and unloading platforms, onshore storage, and warehouse spaces.

Table S1. Spreading alkalinity by different types of maritime fleets.

Type of fleet	Advantages	Disadvantages
Existing and partially designed	No purchase cost of the ship	Limited space onboard Competition in the transport of goods onboard Fixed trade routes to follow Fixed discharge rate Need of intermediate reloads to increase the amount of alkaline material onboard Need of better logistics Cost of utilization of the ship
Existing and specifically designed	No space-saving problem No competition in the transport of goods onboard Flexible management No fixed trade routes to follow Flexible discharge rate Higher discharge rate No purchase cost of the ship	Possible ship retrofitting cost Competition with the trade shipping Cost of utilization of the ship
New dedicated	No space-saving problem No competition in the transport of goods onboard Flexible management No fixed trade routes to follow Flexible discharge rate Higher discharge rate No ship retrofitting cost	Purchase cost of the ship Increase of the ship traffic Possible port congestion Need to increase the current ports' capacity Need of better management to mitigate port congestion No cost of utilization of the ship

A strategy to avoid the installation of pumping and piping systems on purpose could be the functional use of ballast water tanks to store and discharge the alkaline solution.<sup>9,30</sup> Ballast water inside a ship is commonly used to provide stability to the ship and accounts for around 30%-40% of the dwt of a bulk carrier or a container ship.<sup>30</sup> Its utilization produces harmful organisms (e.g., bacteria, microbes, small invertebrates) and substances (e.g., mud, silk), requiring specific treatments across the route and a controlled discharge at arrival. Alkaline solutions could be adopted in the treatment, thereby avoiding the installation of additional equipment for the discharge. The main problem is that the use of alkaline solutions for ballast water treatment is not regulated yet and the real benefit and practical limitations induced (e.g., the effective amount of water required to avoid precipitation of the alkaline solution inside the ballast water tank) need to be evaluated.

According to the work done by <sup>30</sup>, the most suitable ships to spread alkalinity into the ocean are bulk carriers and container ships because of technical and logistic reasons. Between them, the former, in particular, has a lower average emission factor and is almost twice the latter in terms of the number of operative ships on the sea.

## S4 Precipitation

According to <sup>20</sup>, the total alkalinity (TA) generation of about 500  $\mu\text{M}$  by  $\text{Ca}(\text{OH})_2$  is possible w/o following precipitation if diluted 1:1 after  $\text{Ca}(\text{OH})_2$  addition. We derive the safe concentration to avoid precipitation as follows: 500  $\mu\text{M}$  TA increase requires 250  $\mu\text{M}$   $\text{Ca}(\text{OH})_2$ , which corresponds to 18.5 mg  $\text{Ca}(\text{OH})_2 \text{ L}^{-1}$  ( $\sim 20 \text{ g Ca}(\text{OH})_2 \text{ m}^{-3}$ ). The concentration limit of 20 g  $\text{Ca}(\text{OH})_2 \text{ m}^{-3}$  is used throughout the paper.

## S5 Discharge Rate

The discharge rate is the multiplication of the ship's speed, the discharge area, and the concentration limit to avoid precipitation. Detailed calculations can be found the excel sheet attached to the manuscript.

## S6 Efficiency

The efficiency (uptake of  $\text{CO}_2$  per unit of material or alkalinity added) depends on temperature and atmospheric  $\text{pCO}_2$  (Figures S1, S2). From these, we derive that the efficiency varies between 1.6-1.8  $\text{molCO}_2 \text{ molCaO}^{-1}$  (Figures S1, S2). Since model exercises report that the actual efficiency is lower (1.3-1.6  $\text{molCO}_2 \text{ molCO}_2^{-1}$ ),<sup>32</sup> in our study, we consider values between 1.2-1.8  $\text{molCO}_2 \text{ molCaO}^{-1}$ , which correspond to 0.9-1.4  $\text{tCO}_2 \text{ tCaO}^{-1}$ .

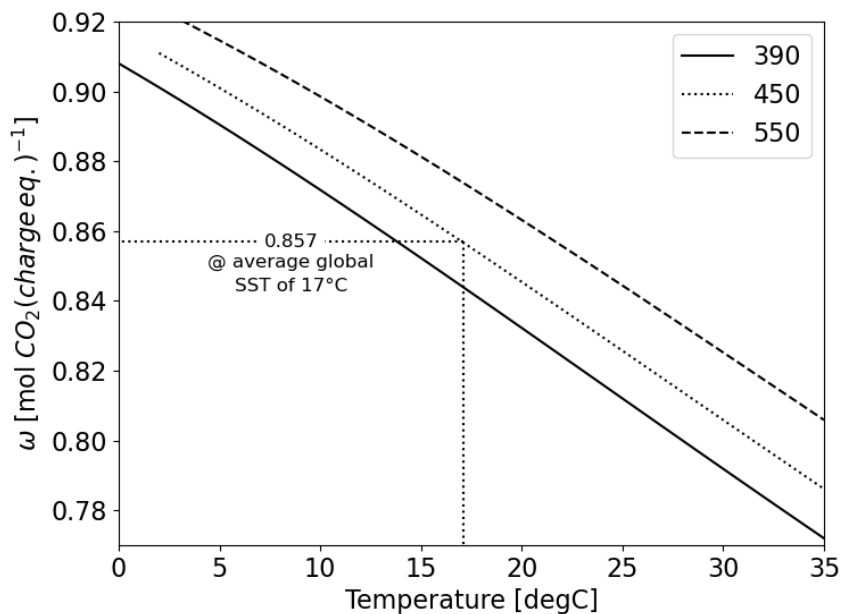


Figure S1. Uptake efficiency vs temperature.



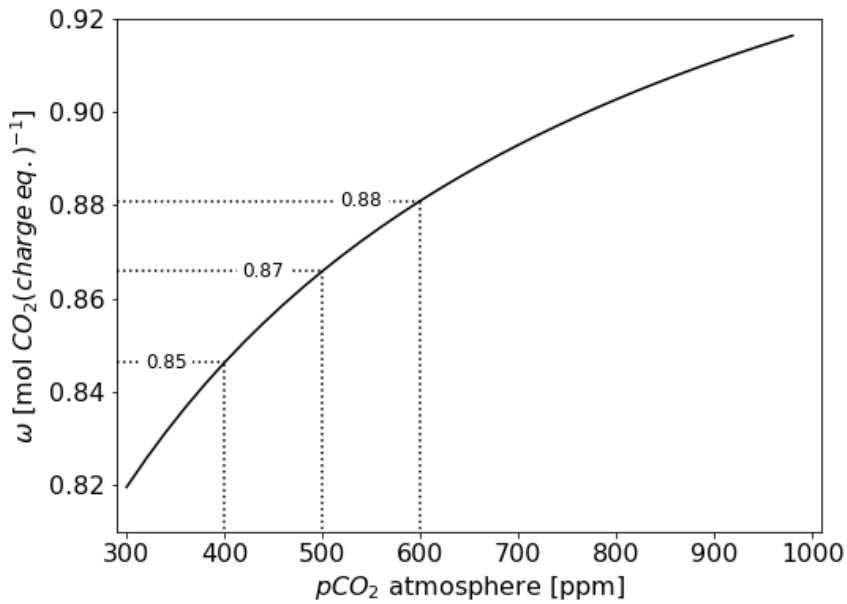


Figure S2. Uptake efficiency vs atmospheric partial pressure.

## S7 Energy

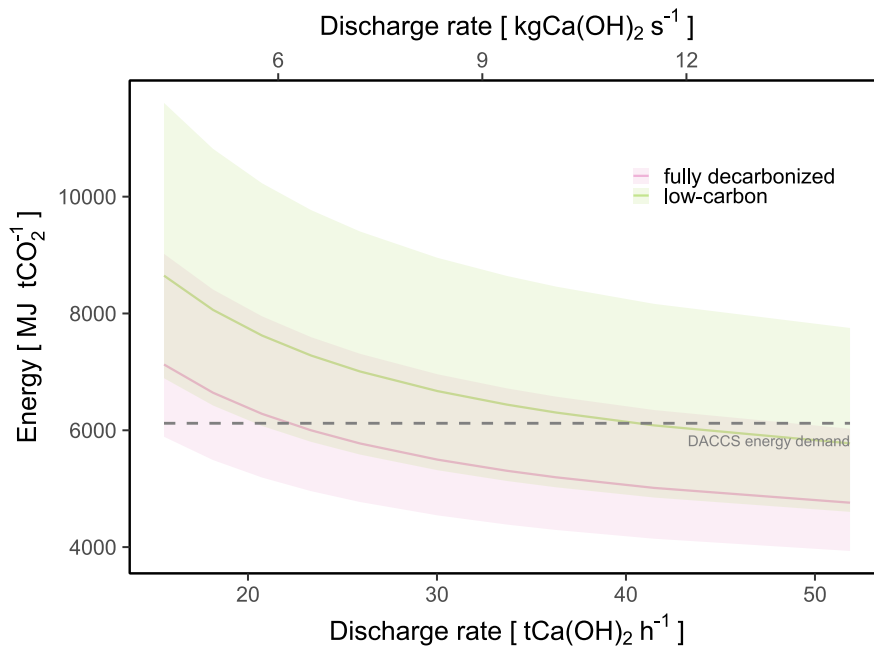


Figure S3. Energy demand vs discharge rate.

Detailed and step-specific data on energy and costs can be found in sheets “Production of Ca(OH)<sub>2</sub>” and “Distribution of 1Gt Ca(OH)<sub>2</sub>” in the excel sheet attached to the manuscript.

Previous studies assign a CO<sub>2</sub> penalty to the electricity demand associated with grinding.<sup>9,10</sup> However, if OAE is to be deployed on a large scale later in the century, as expected in the transformation pathways, electricity generation is expected to be decarbonized.<sup>36</sup>

## S8 Sensitivity analysis

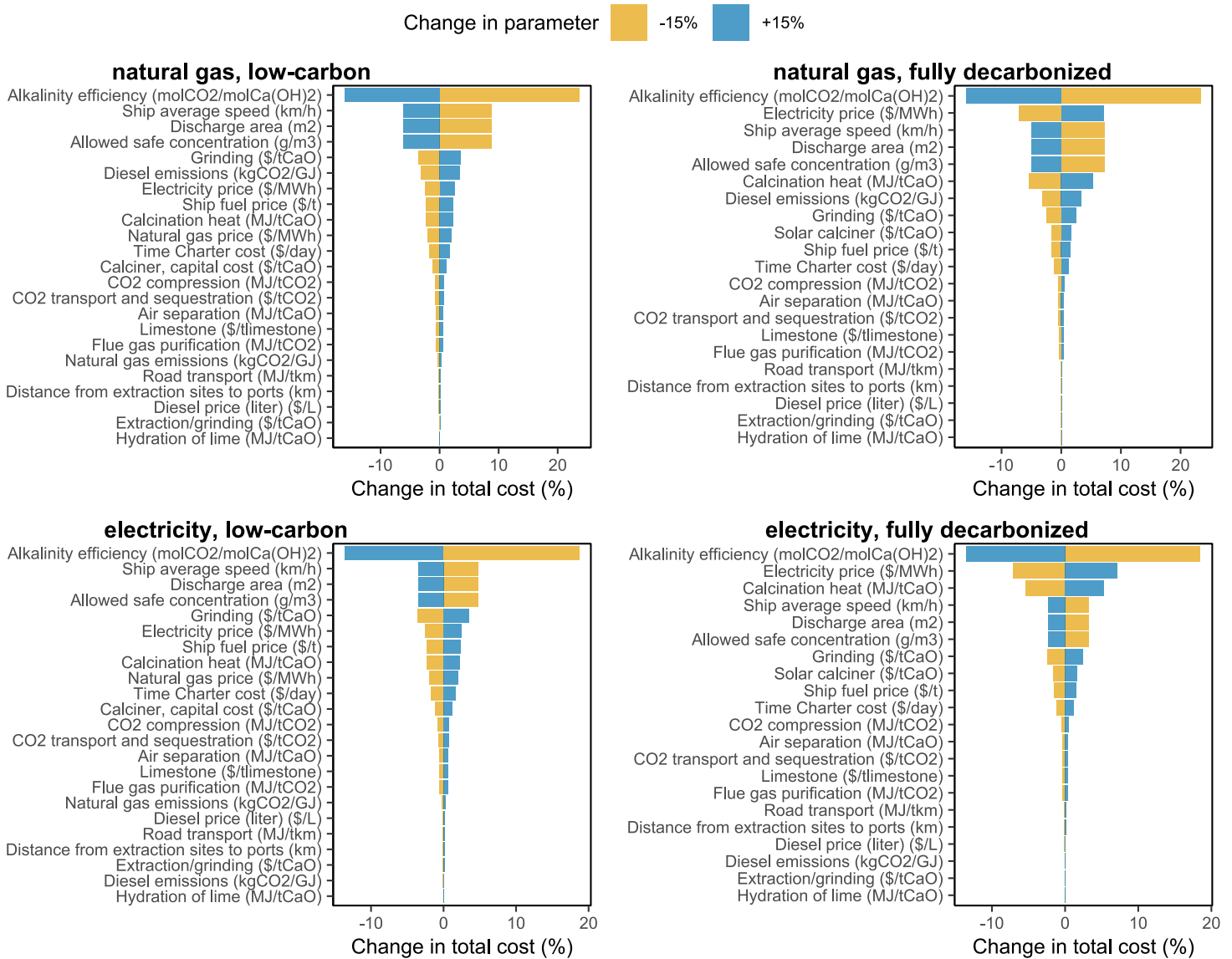


Figure S4. Sensitivity analysis of the total cost. Parameters with a change smaller than 0.04% were filtered out for figure's clarity.

The potential CAPEX of the distribution system has not been included in the estimates, as it is not yet known. However, the cost of DACCS might serve as an upper limit of the total cost for OAE to be cost-competitive. For instance, for a discharge rate of 30 tCa(OH)<sub>2</sub> h<sup>-1</sup> and the efficiency of 1.5 molCO<sub>2</sub> molCa(OH)<sub>2</sub><sup>-1</sup>, the additional capital cost could be 73-144 \$ tCO<sub>2</sub><sup>-1</sup> (natural gas case) and no larger than 80 \$ tCO<sub>2</sub><sup>-1</sup> (electricity case) for the method to be cost-competitive with DACCS under an optimistic learning rate and fully decarbonized energy system.



## References

- (1) USGS. *Mineral Commodity Summaries 2019*; U.S. Geological Survey, 2019. <https://doi.org/10.3133/70202434>.
- (2) Caserini, S.; Storni, N.; Grosso, M. The Availability of Limestone and Other Raw Materials for Ocean Alkalinity Enhancement. *Glob. Biogeochem. Cycles* **2022**, *36* (5), e2021GB007246. <https://doi.org/10.1029/2021GB007246>.
- (3) Renforth, P.; Henderson, G. Assessing Ocean Alkalinity for Carbon Sequestration. *Rev. Geophys.* **2017**, *55* (3), 636–674. <https://doi.org/10.1002/2016RG000533>.
- (4) Hangx, S. J. T.; Spiers, C. J. Coastal Spreading of Olivine to Control Atmospheric CO<sub>2</sub> Concentrations: A Critical Analysis of Viability. *Int. J. Greenh. Gas Control* **2009**, *3* (6), 757–767. <https://doi.org/10.1016/j.ijggc.2009.07.001>.
- (5) Meysman, F. J. R.; Montserrat, F. Negative CO<sub>2</sub> Emissions via Enhanced Silicate Weathering in Coastal Environments. *Biol. Lett.* **2017**, *13* (4), 20160905. <https://doi.org/10.1098/rsbl.2016.0905>.
- (6) Flipkens, G.; Blust, R.; Town, R. M. Deriving Nickel (Ni(II)) and Chromium (Cr(III)) Based Environmentally Safe Olivine Guidelines for Coastal Enhanced Silicate Weathering. *Environ. Sci. Technol.* **2021**, *55* (18), 12362–12371. <https://doi.org/10.1021/acs.est.1c02974>.
- (7) Flipkens, G.; Horoba, K.; Bostyn, K.; Geerts, L. J. J.; Town, R. M.; Blust, R. Acute Bioaccumulation and Chronic Toxicity of Olivine in the Marine Amphipod *Gammarus Locusta*. *Aquat. Toxicol.* **2023**, *262*, 106662. <https://doi.org/10.1016/j.aquatox.2023.106662>.
- (8) Wang, Z.; Yeung, K. W. Y.; Zhou, G.-J.; Yung, M. M. N.; Schlekot, C. E.; Garman, E. R.; Gissi, F.; Stauber, J. L.; Middleton, E. T.; Lin Wang, Y. Y.; Leung, K. M. Y. Acute and Chronic Toxicity of Nickel on Freshwater and Marine Tropical Aquatic Organisms. *Ecotoxicol. Environ. Saf.* **2020**, *206*, 111373. <https://doi.org/10.1016/j.ecoenv.2020.111373>.
- (9) Köhler, P.; Abrams, J. F.; Völker, C.; Hauck, J.; Wolf-Gladrow, D. A. Geoengineering Impact of Open Ocean Dissolution of Olivine on Atmospheric CO<sub>2</sub>, Surface Ocean pH and Marine Biology. *Environ. Res. Lett.* **2013**, *8* (1), 014009. <https://doi.org/10.1088/1748-9326/8/1/014009>.
- (10) Renforth, P. The Potential of Enhanced Weathering in the UK. *Int. J. Greenh. Gas Control* **2012**, *10*, 229–243. <https://doi.org/10.1016/j.ijggc.2012.06.011>.
- (11) Montserrat, F.; Renforth, P.; Hartmann, J.; Leermakers, M.; Knops, P.; Meysman, F. J. R. Olivine Dissolution in Seawater: Implications for CO<sub>2</sub> Sequestration through Enhanced Weathering in Coastal Environments. *Environ. Sci. Technol.* **2017**, *51* (7), 3960–3972. <https://doi.org/10.1021/acs.est.6b05942>.
- (12) Strefler, J.; Amann, T.; Bauer, N.; Kriegler, E.; Hartmann, J. Potential and Costs of Carbon Dioxide Removal by Enhanced Weathering of Rocks. *Environ. Res. Lett.* **2018**, *13* (3), 034010. <https://doi.org/10.1088/1748-9326/aaa9c4>.

- (13) Bach, L. T.; Gill, S. J.; Rickaby, R. E. M.; Gore, S.; Renforth, P. CO<sub>2</sub> Removal With Enhanced Weathering and Ocean Alkalinity Enhancement: Potential Risks and Co-Benefits for Marine Pelagic Ecosystems. *Front. Clim.* **2019**, *1*.
- (14) GEOTRACES Intermediate Data Product Group. The GEOTRACES Intermediate Data Product 2021 (IDP2021)., 2021. <https://doi.org/10.5285/CF2D9BA9-D51D-3B7C-E053-8486ABC0F5FD>.
- (15) Guo, J. A.; Strzepek, R.; Willis, A.; Ferderer, A.; Bach, L. T. Investigating the Effect of Nickel Concentration on Phytoplankton Growth to Assess Potential Side-Effects of Ocean Alkalinity Enhancement. *Biogeosciences* **2022**, *19* (15), 3683–3697. <https://doi.org/10.5194/bg-19-3683-2022>.
- (16) Kheshgi, H. S. Sequestering Atmospheric Carbon Dioxide by Increasing Ocean Alkalinity. *Energy* **1995**, *20* (9), 915–922. [https://doi.org/10.1016/0360-5442\(95\)00035-F](https://doi.org/10.1016/0360-5442(95)00035-F).
- (17) Harvey, L. D. D. Mitigating the Atmospheric CO<sub>2</sub> Increase and Ocean Acidification by Adding Limestone Powder to Upwelling Regions. *J. Geophys. Res. Oceans* **2008**, *113* (C4). <https://doi.org/10.1029/2007JC004373>.
- (18) Caldeira, K.; Rau, G. H. Accelerating Carbonate Dissolution to Sequester Carbon Dioxide in the Ocean: Geochemical Implications. *Geophys. Res. Lett.* **2000**, *27* (2), 225–228. <https://doi.org/10.1029/1999GL002364>.
- (19) Hartmann, J.; Suitner, N.; Lim, C.; Schneider, J.; Marín-Samper, L.; Aristegui, J.; Renforth, P.; Taucher, J.; Riebesell, U. Stability of Alkalinity in Ocean Alkalinity Enhancement (OAE) Approaches – Consequences for Durability of CO<sub>2</sub> Storage. *Biogeosciences* **2023**, *20* (4), 781–802. <https://doi.org/10.5194/bg-20-781-2023>.
- (20) Moras, C. A.; Bach, L. T.; Cyronak, T.; Joannes-Boyau, R.; Schulz, K. G. Ocean Alkalinity Enhancement – Avoiding Runaway CaCO<sub>3</sub> Precipitation during Quick and Hydrated Lime Dissolution. *Biogeosciences* **2022**, *19* (15), 3537–3557. <https://doi.org/10.5194/bg-19-3537-2022>.
- (21) Haas, S.; Weber, N.; Berry, A.; Erich, E. Limestone Powder Carbon Dioxide Scrubber as the Technology for Carbon Capture and Usage. *Cem. Int.* **2014**, *12*, 34–45.
- (22) Chou, W.-C.; Gong, G.-C.; Hsieh, P.-S.; Chang, M.-H.; Chen, H.-Y.; Yang, C.-Y.; Syu, R.-W. Potential Impacts of Effluent from Accelerated Weathering of Limestone on Seawater Carbon Chemistry: A Case Study for the Hoping Power Plant in Northeastern Taiwan. *Mar. Chem.* **2015**, *168*, 27–36. <https://doi.org/10.1016/j.marchem.2014.10.008>.
- (23) Kirchner, J. S.; Berry, A.; Ohnemüller, F.; Schnetger, B.; Erich, E.; Brumsack, H.-J.; Lettmann, K. A. Reducing CO<sub>2</sub> Emissions of a Coal-Fired Power Plant via Accelerated Weathering of Limestone: Carbon Capture Efficiency and Environmental Safety. *Environ. Sci. Technol.* **2020**, *54* (7), 4528–4535. <https://doi.org/10.1021/acs.est.9b07009>.
- (24) Caserini, S.; Cappello, G.; Righi, D.; Raos, G.; Campo, F.; De Marco, S.; Renforth, P.; Varliero, S.; Grosso, M. Buffered Accelerated Weathering of Limestone for Storing CO<sub>2</sub>: Chemical Background. *Int. J. Greenh. Gas Control* **2021**, *112*, 103517. <https://doi.org/10.1016/j.ijggc.2021.103517>.
- (25) Oelkers, E. H.; Declercq, J.; Saldi, G. D.; Gislason, S. R.; Schott, J. Olivine Dissolution Rates: A Critical Review. *Chem. Geol.* **2018**, *500*, 1–19. <https://doi.org/10.1016/j.chemgeo.2018.10.008>.

- (26) Møller, K. T.; Jensen, T. R.; Akiba, E.; Li, H. Hydrogen - A Sustainable Energy Carrier. *Prog. Nat. Sci. Mater. Int.* **2017**, *27* (1), 34–40. <https://doi.org/10.1016/j.pnsc.2016.12.014>.
- (27) Patra, A. K.; Gautam, S.; Kumar, P. Emissions and Human Health Impact of Particulate Matter from Surface Mining Operation—A Review. *Environ. Technol. Innov.* **2016**, *5*, 233–249. <https://doi.org/10.1016/j.eti.2016.04.002>.
- (28) Yakymechko, Y.; Lutsyuk, I.; Jaskulski, R.; Dulnik, J.; Kropyvnytska, T. The Effect of Vibro-Activation Time on the Properties of Highly Active Calcium Hydroxide. *Buildings* **2020**, *10* (6), 111. <https://doi.org/10.3390/buildings10060111>.
- (29) Romagnoli, M.; Gualtieri, M. L.; Hanuskova, M.; Rattazzi, A.; Polidoro, C. Effect of Drying Method on the Specific Surface Area of Hydrated Lime: A Statistical Approach. *Powder Technol.* **2013**, *246*, 504–510. <https://doi.org/10.1016/j.powtec.2013.06.009>.
- (30) Caserini, S.; Pagano, D.; Campo, F.; Abbà, A.; De Marco, S.; Righi, D.; Renforth, P.; Grosso, M. Potential of Maritime Transport for Ocean Liming and Atmospheric CO<sub>2</sub> Removal. *Front. Clim.* **2021**, *3*.
- (31) Renforth, P.; Kruger, T. Coupling Mineral Carbonation and Ocean Liming. *Energy Fuels* **2013**, *27* (8), 4199–4207. <https://doi.org/10.1021/ef302030w>.
- (32) He, J.; Tyka, M. D. Limits and CO<sub>2</sub> Equilibration of near-Coast Alkalinity Enhancement. *Biogeosciences* **2023**, *20* (1), 27–43. <https://doi.org/10.5194/bg-20-27-2023>.
- (33) Watten, B. J.; Sibrell, P. L.; Schwartz, M. F. Acid Neutralization within Limestone Sand Reactors Receiving Coal Mine Drainage. *Environ. Pollut.* **2005**, *137* (2), 295–304. <https://doi.org/10.1016/j.envpol.2005.01.026>.
- (34) UNCTAD. *Review of Maritime Transport*; Review of maritime transport / United Nations Conference on Trade and Development, Geneva; United Nations: Geneva, 2022.
- (35) Corbett, J. J.; Winebrake, J. J. International Trade and Global Shipping. *Chapters* **2008**.
- (36) *Climate Change 2022 - Mitigation of Climate Change: Working Group III Contribution to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change*; Intergovernmental Panel on Climate Change (IPCC), Ed.; Cambridge University Press: Cambridge, 2023. <https://doi.org/10.1017/9781009157926>.