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## Marine carbon dioxide removal by alkalinization should no longer be overlooked

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## Marine carbon dioxide removal by alkalinization should no longer be overlooked

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## **Abstract**

To achieve the Paris climate target, deep emissions reductions have to be complemented with carbon dioxide removal (CDR). However, a portfolio of CDR options is necessary to reduce risks and potential negative side effects. Despite a large theoretical potential, ocean-based CDR such as ocean alkalinity enhancement (OAE) has been omitted in climate change mitigation scenarios so far. In this study, we provide a techno-economic assessment of large-scale OAE using hydrated lime ('ocean liming'). We address key uncertainties that determine the overall cost of ocean liming (OL) such as the CO2 uptake efficiency per unit of material, distribution strategies avoiding carbonate precipitation which would compromise efficiency, and technology availability (e.g., solar calciners). We find that at economic costs of 130–295 \$/tCO2 net-removed, ocean liming could be a competitive CDR option which could make a significant contribution towards the Paris climate target. As the techno-economic assessment identified no showstoppers, we argue for more research on ecosystem impacts, governance, monitoring, reporting, and verification, and technology development and assessment to determine whether ocean liming and other OAE should be considered as part of a broader CDR portfolio.

### **1. Introduction**

Net-zero emission targets, intended to stabilize the Earth's climate, require deep reductions in anthropogenic emissions [\[1](#page-9-0), [2\]](#page-9-1). In addition, carbon dioxide removal (CDR) is indispensable to compensate for emissions that are considered hard to abate, e.g. in the aviation or the agriculture sector, and also to achieve net-negative emissions that can offset historical emissions[[3\]](#page-9-2). Transformation pathways that reach net-zero targets so far depend on land-based CDR, mainly afforestation/reforestation and bioenergy with carbon capture and storage (BECCS) [\[2\]](#page-9-1), and to a lesser extent on direct air carbon capture and storage (DACCS) and enhanced rock weathering [[4\]](#page-9-3). However, all of these approaches have some drawbacks, such as requiring large areas of fertile land, and potentially also high water and fertilizer footprints [\[5](#page-9-4)], concerns about saturation and the durability of carbon removal, or high costs and energy requirements[[6](#page-9-5)]. While ocean-based CDR may address some of these concerns, it remains the least represented climate mitigation strategy in the literature, because it still lacks validation and parametrization to be included in the scenarios.

Ocean alkalinity enhancement (OAE) is considered to have one of the highest theoretical potentials among the marine CDR methods proposed in the literature so far[[7\]](#page-9-6). Several modelling experiments have investigated  $CO<sub>2</sub>$  removal potential of OAE and its effect on the pH and the carbonate saturation state  $[8-11]$  $[8-11]$  $[8-11]$ ,as well as its efficiency  $[12, 13]$  $[12, 13]$  $[12, 13]$  $[12, 13]$ , concluding

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that it could effectively remove gigatons of  $CO<sub>2</sub>$  from the atmosphere. The studies span a wide range of hypothetical deployment scenarios looking at various deployment patterns under different emission scenarios. However, a deep understanding of longterm as well as local impacts, potential ecological risks and feedback mechanisms associated with large-scale OAE deployment is still missing.

A variety of methods to enhance ocean alkalinity have been proposed in the literature including electrochemical processes (electrolysis or electrodialysis)[[14](#page-9-11), [15](#page-9-12)], adding natural silicate minerals to thecoastal or open ocean  $[16–18]$  $[16–18]$  $[16–18]$  $[16–18]$ , creating reactive materials through the calcination of limestone ('ocean liming')[[19](#page-9-15), [20](#page-9-16)], or the creation and addition of hydrated carbonate minerals [\[21](#page-9-17)]. The potential for large-scale deployment of ocean liming (OL) and the use of rather well-known technologies like calcination, suggested OL for a techno-economic assessment as conducted in this study. Recent studies have shown that, when enhancing alkalinity via liming, precipitation (crossing critical saturation thresholds of carbonate minerals, which reduces the atmospheric  $CO<sub>2</sub>$  uptake) occurs at much lower concentrations than previously thought [\[22](#page-9-18), [23\]](#page-9-19). Even though in reality the allowed concentration might be higher as the dissolution can be accelerated, e.g. by a propeller in the wake of a ship, these laboratory results must be taken into consideration as they lead to increased costs for distributing alkalinity. Moreover, the uptake efficiency, another important parameter, depends on the distribution region and from modeling experiments is lower than previously assumed [\[13](#page-9-10)]. While previous research has examined capital expenditures (CAPEX) and operating expenses (OPEX) costs of some approaches [\[20,](#page-9-16) [24](#page-10-0), [25](#page-10-1)], an analysis of distribution possibilities taking into account carbonate precipitation and the associated costs and the consideration of ocean uptake efficiency are still missing.

In this study, we provide a techno-economic assessment of OL taking into consideration recent findings on precipitation and lower uptake efficiency, and identify remaining uncertainties, given the low technology readiness level (TRL). We discuss the impacts and limitations of the method and compare it to DACCS, which can be considered a fairly similar land-based CDR approach in terms of costs, energy requirements, and scalability. The study provides a basis for the integration of OAE into integrated assessment models and thus into transformation pathways for climate change mitigation.

## **2. Methods**

To determine which OAE options could make a significant impact in climate change mitigation scenarios, we performed a screening of OAE technologies in the literature. We examined the methods' feasibility, scalability, data availability, and the status of their technological development. The screening can be found in Note S1, supplementary material (SI). The steps required to produce hydrated lime for OL, including limestone extraction, comminution, calcination, and hydration, are already well-established and used at a large scale in the cement industry. This makes OL a promising option for gigaton-scale implementation, provided that it is scaled up sustainably and various challenges, such as precipitation, high energy demand, and CCS requirements (during quicklime production), are effectively addressed. Using the information gathered during the literature screening, we undertook a bottom-up technoeconomic assessment and provide the data, assumptions, and equations used throughout.

Quicklime (CaO) is produced via calcination, which involves heating limestone to typically around 900 *◦*C–1000*◦*C. The decomposition reaction (equation  $(1)$ ) of limestone releases  $CO<sub>2</sub>$ , which should be captured and stored to maximize the method's efficiency.

<span id="page-2-0"></span>
$$
CaCO3 + heat \rightarrow CaO + CO2.
$$
 (1)

CaO in contact with water forms hydrated lime  $(Ca(OH<sub>2</sub>), When CaO or Ca(OH<sub>2</sub>)$  is added to seawater (equation  $(2)$ ), it reacts with  $CO<sub>2</sub>$  dissolved in the water to produce  $Ca^{2+}$  and bicarbonate (HCO<sub>3</sub><sup>-</sup>), and to a smaller amount carbonate ions (CO<sub>3</sub><sup>−</sup>). This temporarily increases the pH and alkalinity and subsequently draws  $CO<sub>2</sub>$  from the atmosphere into the ocean. For effective atmospheric  $CO<sub>2</sub>$  uptake, alkaline materials distributed in the open ocean must dissolve within the mixed surface layer which requires grinding to appropriate grain sizes to maximize the residence time in the surface waters. The costs, energy, and material demands for quicklime/hydrated lime production are summarized in table [1](#page-3-0) and described in more detail in note S7 (SI)

<span id="page-2-1"></span>
$$
(CaO + H2O \rightleftharpoons Ca(OH)2) + 1.9CO2 \rightarrow Ca2+ + 1.8HCO3- + 0.1CO32-. (2)
$$

**Extraction and grinding.** Extraction and grinding account for a small amount of the energy needs and costs (note S7, SI). For comparison, over 40 Gt of raw materials are produced globally per year, of which 6.6 Gt is limestone [\[28\]](#page-10-2). It is estimated that 5 000 Gt of limestone are located within 10 km of the coasts and below bare ground or scrub [\[28\]](#page-10-2). It could thus be possible to minimize the cost of road transport (here estimated at 0.05 \$ tkm*−*<sup>1</sup> [[30](#page-10-3)]) for quicklime transportation, if the extraction and calcination facilities are close to ports. In this case, however, the  $CO<sub>2</sub>$  captured for geological storage might have to be transported, depending on where the CCS facilities are located. We assume that limestone deposits are no

<span id="page-3-0"></span>**Table 1.** Energy requirements, levelized capital cost per unit of CaO produced (CAPEX), operation and maintenance costs (OPEX), energy cost assumptions, and material demand of the production of hydrated lime, excluding the distribution of alkalinity. Step-specific data can be found in note S7 (SI).

<b>Energy demand</b>				
Heat (high-temperature) [MJ tCaO $^{-1}$ ]	3100 (2910-3492)	$\left[20\right]$		
Electricity [MJ tCaO <sup><math>-1</math></sup> ]	995 (992-1188)	$[20, 25 - 27]$		
Diesel [MJ tCa $O^{-1}$ ]	$103(73 - 943)$	$\left[20\right]$		
Costs w/o energy				
CAPEX [ $$tCaO^{-1}]$ ]	15.8 (31 if electric calciner)	[20, 25, 28]		
OPEX [ $$tCaO^{-1}]$ ]	58.1	[20, 26]		
<b>Energy costs</b>				
Natural gas $[$GJ^{-1}]$	8	$\left[29\right]$		
Electricity [ $\$$ GJ <sup>-1</sup> ]	31	$\left[29\right]$		
Diesel [ $I^{-1}$ ]	0.7	$\left[29\right]$		
<b>Materials</b>				
Limestone demand [tCaCO <sub>3</sub> tCaO <sup>-1</sup> ]	1.82			
Hydrated lime output $[tCa(OH)_2 tCaO^{-1}]$	1.32			

**Table 2.** Carbon intensity factors and efficiency-related assumptions.

<span id="page-3-2"></span>

further than 20 km from the ports, and the  $CO<sub>2</sub>$  storage sites within 250 km.

**Calcination.** Producing one ton of quicklime requires 1.82 tons of limestone and 2.9–3.5 GJ of heat [[20](#page-9-16)]. For the calcination process, high-temperature (HT) heat is necessary, which as of now has been supplied by coal or natural gas, e.g. in the cement industry. Fossil heat could be replaced by biogenic fuels or hydrogen[[31](#page-10-9), [32\]](#page-10-10). However, concerns have been raised that the caloric density of biogenic fuels alone might be too low to provide a high enough temperature in the kiln[[31](#page-10-9), [33\]](#page-10-11). Similarly, hydrogen is not expected to fully replace fossil fuel requirements. For the moment, there are no large-scale examples of using electricity, hydrogen, or waste heat in the calcination or the cement production process. The  $TRL<sup>6</sup>$  $TRL<sup>6</sup>$  $TRL<sup>6</sup>$ of electric calciners is higher (TRL 5) than of hydrogen calciners (TRL 4) [\[34\]](#page-10-12). Oxyfuel fired kilns are assumed as they produce high-purity  $CO<sub>2</sub>$  flue gases, which facilitates geological storage. We consider two calciner scenarios depending on the energy used natural gas calciner and electrical calciner. Their main differences in parameters are the capex (table [1](#page-3-0)) and energy-related emissions (table [2](#page-3-2)).

**Emissions and CCS.** The unavoidable emissions from the decomposition of limestone during calcination (process emissions) amount to  $0.78$  tCO<sub>2</sub> per ton of quicklime. For OL to be effective, these emissions must be sequestered. If the heat demand is to be supplied by natural gas alone, there will be 0.21 t of additional  $CO<sub>2</sub>$  emissions per ton of quicklime [\[35\]](#page-10-7), which are assumed to be stored geologically.

**Measures to avoid precipitation.** Originally, it has been suggested that lime is pumped to the ocean or spread in the open ocean by ships [\[19\]](#page-9-15). Recent findings indicate that this method might lead to solid carbonate precipitation, significantly decreasing or even canceling out the efficiency of OL[[22](#page-9-18), [23](#page-9-19)]. First laboratory experiments suggest that secondary precipitation is not induced if the powder minerals are dissolved and diluted fast enough such that a critical aragonite saturation threshold is not exceeded[[22](#page-9-18)]. The suggested safe concentration is 250  $\mu$ mol Ca(OH)<sub>2</sub> discharged per 1 kg seawater,

<span id="page-3-1"></span><sup>6</sup> TRL measured from 1 ('Initial Idea') to 11 ('Proof of stability reached'). TRL 4 corresponds to 'Early prototype' and TRL 5 to 'Large prototype'[[34](#page-10-12)].

which corresponds to 20 g Ca $(OH)_2$  per m<sup>3</sup> (note S4, SI), which is further used in this study.

**Discharge rate.** Bulk carriers and container ships have been proposed as the main means for lime spreading since they allow for reaching vast areas of the ocean and could be easily fitted for distributing powdered minerals or slurry [\[20](#page-9-16), [24\]](#page-10-0). The discharge rate and the distribution system determine the alkalinity concentration after adding lime to the ocean. The concentration must not exceed the limit above which spontaneous precipitation occurs [\[36\]](#page-10-13). For ships, a system of nozzles covering a certain area could be used, that distributes the material as the ship moves. The amount of alkalinity that can be discharged depends on the allowed concentration limit (gCa(OH)<sup>2</sup> m*−*<sup>3</sup> ) and the volume of water reached in a unit of time (m<sup>3</sup> h *−*1 ), which in our approach depends on the speed of the ship and the discharge area (equation([3\)](#page-4-0)). Here, we consider distribution via dedicated ships of type 'Panamax' (note S3, SI)

$$
\begin{aligned} \text{Discharge rate} &= \text{(concentration limit)}\\ &\times \text{(discharge area)} \text{ (ship's speed)}\\ &\cdot \text{ (3)} \end{aligned}
$$

<span id="page-4-0"></span>*.*

**OL efficiency.** According to reaction [\(2\)](#page-2-1), one mol of CaO or  $Ca(OH)_2$  captures about 1.9 molCO<sub>2</sub>. In reality, the efficiency is lower due to the carbonate system re-equilibration in the ocean[[20](#page-9-16), [37](#page-10-14)], and depends on temperature and the partial pressure of  $CO<sub>2</sub>$  in the atmosphere. Efficiency varies across ocean regions [\[12\]](#page-9-9). Additionally, model experiments show that the efficiency might be even lower than previously thought[[13](#page-9-10)]. We assume that the uptake efficiency of OL (tons of  $CO<sub>2</sub>$  removed per ton of alkaline material) varies between  $1.2-1.8$  mol $CO<sub>2</sub>$ molCa(OH)<sub>2</sub><sup>-1</sup> (0.7–1.1 tCO<sub>2</sub> tCa(OH)<sub>2</sub><sup>-1</sup>) discharged into the ocean (note S6, SI). The maximum efficiency will be hard to achieve as ships need to cross less efficient areas.

The method's final efficiency (equation [\(4](#page-4-1))) also depends on the uncaptured emissions produced during deployment.  $CO<sub>2</sub>$  is created as an unavoidable product from lime production (equation([1\)](#page-2-0)). The rest of the emissions, coming from the energy supply, depend on the mitigation efforts in the energy system (table [3](#page-4-2))

efficiency<sub>OL</sub>

= efficiencyuptake *−* energy emissionsdistribution *−* (1 *−* CCS capture rate) (process emissions + energy emissionscalcination) (4)

**Scenarios.** To capture how the assumptions of the future energy system affect the total cost of OL, we consider two emission scenarios: (a) fully decarbonized and (b) low-carbon energy system. In scenario

**Table 3.** Distribution of alkalinity via ships.

<span id="page-4-2"></span>

Parameter/Assumption	Comment		
Hiring a ship $\lceil \frac{1}{3} \text{ day}^{-1} \rceil$ Fuel consumption $[t h^{-1}]$ Fuel cost $[$t^{-1}]$ Average speed [ $km h^{-1}$ ] Ship's tonnage [t] DWCC usable for alkaline material [%]	11 250 1.41 450 25 75 000 85	Market report [39] [40] Market report $[41]$ $\left[24\right]$ $\lceil 24 \rceil$ $\lceil 24 \rceil$	
Concentration limit $\left[\text{gCa(OH)}_{2} \text{ m}^{-3}\right]$	20	$[22]$ ; calculations in Note S4, SI	

(b), it is assumed that decarbonized electricity is available and that the emissions from burning natural gas can be stored geologically. On top of that, scenario (a) assumes that the ships run on emission-free fuel, so the unabated emissions only come from a 95% capture rate on the calcination emissions (table [2\)](#page-3-2). For each emissions scenario, we consider the two calciner technologies mentioned above.

**Costs**. Distribution costs are determined by the amount of alkalinity that can be distributed into the ocean per ship and year and the costs of these ships, which consist of OPEX (costs of hiring a ship) and fuel costs (table [3](#page-4-2)); both depend on the discharge rate. The final cost of producing and distributing alkalinity is calculated according to equation [\(5\)](#page-4-3)

<span id="page-4-3"></span>
$$
CostCaO (discharge rate)
$$
  
= CAPEX<sub>prod</sub> + OPEX<sub>prod</sub> + energy cost<sub>prod</sub>  
+ OPEX<sub>distr</sub> (discharge rate)  
+ energy cost<sub>distr</sub> (discharge rate). (5)

We calculate the cost of removing a net ton of  $CO<sub>2</sub>$ via OL using equation ([6\)](#page-4-4), where the efficiency is the OL efficiency calculated in equation [\(4](#page-4-1)) after discounting the unabated emissions. The cost of removal of a gross  $tCO<sub>2</sub>$  can be calculated if the uptake efficiency is used instead.

<span id="page-4-4"></span>
$$
CostCO2 (discharge rate, efficiencyOL)
$$
  
= 
$$
\frac{CostCaO (discharge rate)}{efficiencyOL}
$$
 (6)

The break-even carbon price determines the carbon price at which the technology becomes economically viable (equation([7\)](#page-4-5))

<span id="page-4-5"></span>Cost<sub>CO<sub>2</sub></sub> (discharge rate, uptake efficiency)  
+ 
$$
(C_{\text{price}})
$$
 (unabated emissions)  
 $\leq C_{\text{price}}$  (7)

<span id="page-4-1"></span>**Sensitivity analysis**. In section [3.2](#page-5-0), we investigate the local sensitivity of the results by varying a set of 28 input variables around the base values by  $\pm$  15% as described in Strunge *et al* [\[42\]](#page-10-18).

<span id="page-5-1"></span>

energy costs consider two scenarios—if the calcination is fueled by natural gas or electricity, with the natural gas price of 28 \$ MW<sub>heat</sub><sup>-1</sup> and electricity of 110 \$ MWh<sub>el</sub><sup>-1</sup> [\[29](#page-10-6)]. Emissions from limestone decomposition and burning natural gas are captured and stored.

### **3. Results**

#### **3.1. Techno-economic assessment**

In this analysis, we first discuss the cost of the production and distribution of quicklime, going beyond previous assessments by Renforth *et al*[\[20\]](#page-9-16) by considering the latest experiment results regarding runaway precipitation [\[22,](#page-9-18) [23,](#page-9-19) [36](#page-10-13)]. Second, we discuss the total cost of CDR via OL depending on the future emissions scenarios and uptake efficiency, which from the modeling experiments is lower than previously thought [\[13\]](#page-9-10).

#### *3.1.1. Production and distribution of quicklime*

The cost of distribution increases with decreasing discharge rate, however, compared to the costs of quicklime production, it is not a large contribution for discharge rates higher than 30 t Ca(OH)<sup>2</sup> h *−*1 (figure [1\)](#page-5-1). A discharge rate of 30 t Ca(OH)<sup>2</sup> h *<sup>−</sup>*<sup>1</sup> would require a discharge area of 60  $m^2$ , assuming an average speed of 25 km h*−*<sup>1</sup> . Such a discharge area should pose a simple engineering challenge given that current container ships can be up to 30 m wide [\[43\]](#page-10-19). It is unclear whether a distribution system attached to a ship induces additional costs. We discuss the capital cost for distribution note S8 (SI) in the context of cost competitiveness.

#### *3.1.2. Total cost estimate*

The uncertainties in discharge rate and uptake efficiency, as well as the choice of calcination technology, result in wide OL cost spreads from 120 \$ tCO<sup>2</sup> *−*1 to almost 350 \$ tCO<sup>2</sup> *−*1 (fully decarbonized scenario; figure [2](#page-6-0)(a)). The total cost rises to 145–445 \$ tCO<sub>2</sub> $^{\rm -1}$ if emissions from freight transport, which is a sector challenging to decarbonize [\[44\]](#page-10-20), are accounted for (low carbon scenario; figure  $2(b)$  $2(b)$ ). However, the upper end of costs seems rather unlikely as discharge rates of at least 30 t Ca(OH)<sub>2</sub> h<sup>-1</sup> are likely feasible from a technical perspective. Additionally, larger discharge rates may be possible if dilution in the wake of a ship allows for a larger concentration limit. Our best estimate of total costs using current calciner technologies, medium uptake efficiency of  $1.5$  mol  $CO<sub>2</sub>$ mol Ca(OH)<sub>2</sub><sup>-1</sup>, and a discharge rate of 30 t Ca(OH)<sub>2</sub> h *−*1 is 161 \$ tCO<sup>2</sup> *−*1 (fully decarbonized scenario) and 195 \$  $tCO_2^{-1}$  if the efficiency is reduced by the unabated emissions from freight transport (lowcarbon scenario). Unabated emissions account for 0.22 tCO<sub>2</sub> per gross ton of CO<sub>2</sub> uptaken by the ocean (0.18–0.27 depending on the uptake efficiency). We calculate the break-even carbon price as the carbon price equal to the total cost of removing a gross ton of  $CO<sub>2</sub>$  via OL plus the cost of unabated emissions. For the low-carbon scenario, it is 205 \$ tCO<sub>2</sub><sup> $-1$ </sup> (171–257) for a natural gas calciner and 298 \$ $tCO_2^{-1}$  (248–372) for an electric calciner.

#### <span id="page-5-0"></span>**3.2. Sensitivity analysis**

The sensitivity analysis of the scenario with the discharge rate of 30 tCa(OH)<sub>2</sub>  $h^{-1}$  and the efficiency of 1.5 molCO<sub>2</sub> molCa(OH)<sub>2</sub><sup> $-1$ </sup> (figure [3](#page-7-0)) shows the largest relative change to the total cost is caused by the uptake efficiency and the discharge rate (determined by discharge area, speed of a ship, and safe alkalinity concentration). In addition, all factors determining the discharge rate are still uncertain and may vary much more than the 15% shown in figure [3](#page-7-0). This highlights that OL deployment should take place in the areas where the uptake efficiency is high and that a distribution system should allow for a possibly larger discharge rate. The costs for  $CO<sub>2</sub>$  transport

<span id="page-6-0"></span>

and sequestration are still very uncertain as well, but given the lower relative importance even a doubling of these costs would increase total costs by less than 5%.

#### **4. Discussion**

#### **4.1. Impacts and limitations**

This study focuses on techno-economic considerations and the question if OL is in principle a CDR option that has the potential to scale and to be economically competitive. However, several other aspects could limit deployment. These aspects are beyond the scope of this study but should be considered in further research.

**Physical limitations.** To avoid the loss of alkalinity due to precipitation, the dissolution of minerals should be kept below critical saturation thresholds. This requires determining the safe concentrations of alkaline materials. Laboratory experiments by Moras *et al* [[22\]](#page-9-18) suggest that the increase in total alkalinity by 500 *µ*mol kg*−*<sup>1</sup> induces precipitation (corresponding to a safe aragonite saturation threshold of 5). The safe concentration will differ regionally [\[46\]](#page-10-22) and conditions as dilution in the wake of a ship could allow for a larger initial concentration. Therefore, further research is needed to determine regional safety guardrails. Assuming no technical hurdles, the discharge rate is then limited by the regional safe concentration that prevents precipitation.

**Marine ecosystems.** It has been shown that some organisms seem sensitive to the addition of  $Ca(OH)_2$ , e.g. crustacean and shrimp species [\[47,](#page-10-23) [48\]](#page-10-24). More research and field experiments are required to understand the impacts of alkalinity addition on marine organisms and the durability of potential ocean acidification mitigation. Negative environmental consequences could be a potentially limiting factor for OAE, e.g. if it significantly reduces the deployment area or the allowed alkalinity concentration.

**Industry scale-up.** When it comes to technoeconomic constraints, a potential limitation to achieving gigatons of CDR via OL could be scaling up the industry. In 2019, 439 Mt of quicklime was produced worldwide (of which over 70% is in China) and 4.1 Gt of cement (half of which is in China) [\[49\]](#page-10-25). As reported by Caserini *et al* [[28](#page-10-2)] limestone deposits are abundant, thus, the resource supply would not be a limitation. However, the expansion of the mining industry could have potential negative impacts on the local environment (e.g. changes in land use patterns, dust pollution) [\[50](#page-10-26)] and thus has to be conducted sustainably. Another limitation comes from the scaleup of maritime transport. The global fleet's carrying capacity in 2022 was 2.2 billion deadweight tons, 64 Mt more than the previous year, while container port traffic reached 11 Gt[[43](#page-10-19)]. For comparison, to remove a gigaton of  $CO<sub>2</sub>$  via OL, one would need to extract 1.3–1.9 Gt of limestone and transport 0.9– 1.4 Gt of material  $(Ca(OH<sub>2</sub>)$  via ships depending on the efficiency of  $CO<sub>2</sub>$  uptake per unit of material.

**Governance.** As of now, there are no legal frameworks that specifically address the use of the oceans for OAE[[51](#page-10-27), [52\]](#page-10-28). However, activities applied in the ocean are subject to international and/or domestic laws, which might be a potential barrier to OAE deployment and field research. One such law is the London Protocol [\[53\]](#page-10-29), an international agreement that regulates the deliberate disposal of waste or other matter into the ocean. However, when it was created, it did not intend to inhibit ocean-based CDR.

**Monitoring, reporting, and verification** (MRV) of CDR are crucial measures to constitute a reliable carbon market. Determining how much  $CO<sub>2</sub>$  will be sequestered via OL presents a challenge, as  $CO<sub>2</sub>$  equilibration in the ocean cannot be measured directly. It

<span id="page-7-0"></span>

is possible to measure the  $CO<sub>2</sub>$  uptake in a reactor, however, as the solution is mixed in the ocean, the water re-equilibrates and might lose or gain some CO<sup>2</sup> depending on the water chemistry. Models will play a crucial role in determining how much  $CO<sub>2</sub>$  is captured for a given OL operation [\[54,](#page-10-30) [55](#page-10-31)].

#### **4.2. Comparison with DACCS**

DACCS and OL are comparable in a number of aspects. They both grant long-term storage and have a large estimated  $CO<sub>2</sub>$  removal potential with the constraining resources being costs, energy, and CCS availability (table [4\)](#page-8-0), instead of land as e.g. for BECCS [[2,](#page-9-1) [6\]](#page-9-5). Despite many similarities, they differ in some aspects, which might be crucial in deciding which of these two options is more desirable, given the limited time left to reach carbon neutrality to stabilize global mean temperature.

In a DACCS system, the  $CO<sub>2</sub>$  under increased air flow, reacts with an aqueous solvent or a solid sorbent and is later regenerated in the presence of heat and stored geologically. Solution-based approaches require 900 *◦*C heat, hence are also called HT DACCS. Using solid sorbents, e.g., amines, allows to regenerate the CO<sup>2</sup> under lower temperatures, below 100 *◦*C, thus are called low-temperature (LT) DACCS. Input requirements for chemical absorbents do not limit DAC scale-up [\[56\]](#page-10-32).

The LT heat required for LT DAC could easily be supplied by electricity, waste heat, or district heat, and thus has the advantage over OL, as the latter requires a HT calcination process. Electric or hydrogen calciners havea low TRL  $(4–5$  out of 11  $[34]$  $[34]$  $[34]$ ;), which increases the probability of continued use of fossil fuels in the cement industry and also for OL to provide HT heat [[31](#page-10-9)].When it comes to geological storage, OL requires up to 45% less CCS, depending on the efficiency. However, OL requires marine transport, which is a more difficult sector to decarbonize [\[44,](#page-10-20) [57](#page-10-33)]. If we assume that the emission factor of shipping does not change (low-carbon scenario), then there are 0.17– 0.27 tons of unabated  $CO<sub>2</sub>$  emissions per ton of  $CO<sub>2</sub>$ taken up by the ocean (table [4](#page-8-0)). This underlines the importance of decarbonizing all of the energy system, including transportation, even when CDR methods like OL become available.

Besides the high energy demand, the cost of DACCS is seen as a primary barrier to implementation, at least on a larger scale[[6\]](#page-9-5). Current costs are estimated to be within the range of 600–1000 \$ per 1 tCO<sub>2</sub> [\[6,](#page-9-5) [58](#page-10-34)]. The CAPEX of DACCS is expected to decrease due to learning to  $80-200$  \$/tCO<sub>2</sub> by 2050 [[45](#page-10-21)], while the total cost of removing a ton of  $CO<sub>2</sub>$ might decrease to 100–600 \$ tCO<sub>2</sub><sup>-1</sup> [[59](#page-10-35)] or 100– 300 \$  $tCO_2^{-1}$  according to Fuss *et al* [\[6](#page-9-5)]. For OL, there is less potential to reduce costs as the largest cost component is the calcination process, a well-known and mature technology. The overall cost depends on the efficiency of the method. As of now, the range of estimated CAPEX of OL is significantly lower

<span id="page-8-0"></span>**Table 4.** Techno-economic comparison of a net ton CO<sub>2</sub> (unabated emissions were discounted from the uptake efficiency) taken up by DACCS and OL with the discharge rate of 30 tCa(OH)<sub>2</sub> h<sup>-1</sup>. The ranges correspond to varying uptake efficiency (1.2–1.8 molCO<sub>2</sub> molCa(OH)<sup>2</sup> *−*1 ). For OL, the two emissions scenarios are presented—a fully decarbonized energy system and, in parenthesis, low-carbon.

Technology	Energy (GJ per $1tCO2$ sequestered)			Fixed costs $(CAPEX + OPEX)$	Total cost <sup>a</sup>	<b>CCS</b> requirement	Unabated emissions
	Heat	Electricity	Shipping fuel	\$ tCO $_2^{-1}$	\$ tCO <sub>2</sub> <sup>-1</sup>	tCCS tCO <sub>7</sub> <sup>1</sup>	tCO <sub>2</sub> tCO <sub>2</sub> <sup>-1</sup>
OL, natural gas calciner	$2.2 - 3.3$	$0.7 - 1.1$	$1.9 - 2.8$	$71 - 109(83 - 140)$	$133 - 204$ $(156 - 263)$	$0.68 - 1.04$ $(0.79 - 1.34)$	$0.04 - 0.06$ $(0.22 - 0.27)$
OL, electric calciner	$\mathbf{0}$	$2.9 - 4.4$		$80 - 123(94 - 157)$	$211 - 322$ $(227 - 380)$	$0.55 - 0.83$ $(0.64 - 1.07)$	$0.03 - 0.05$ $(0.17 - 0.26)$
LT DACCS	$\mathbf{0}$	$6.1 - 8.3$	$\Omega$	Now: $\sim$ 700 Floor cost: $90-200b$	$287 - 353$		$\Omega$
<b>HT DACCS</b>	$5.1 - 8.1$	$1.3 - 2.7$	$\Omega$	Now: $\sim 800$ Floor cost: $100-220b$	$170 - 206$		$\mathbf{0}$

<sup>a</sup> Cost assuming energy prices: 28 \$ MWh<sub>heat</sub><sup>-1</sup>, 110 \$ MWh<sub>el</sub><sup>-1</sup> [\[29](#page-10-6)], and 450 \$ t<sub>fuel</sub><sup>-1</sup> [[41\]](#page-10-17).

<sup>b</sup> DACCS floor costs are the estimates for 2050 according to Fasihi *et al* [[45](#page-10-21)].

compared to the current costs of DACCS (table [4\)](#page-8-0). However, DACCS could become competitive for the most optimistic cost reduction estimates.

OL has been criticized for a potentially harmful effect on the marine ecosystem, and negative impacts related to the mining and pre-processing of large amounts of rock. When applied locally (for example in protected bays, lagoons), OL could restore the preindustrial pH levels in the water  $[60-62]$  $[60-62]$  $[60-62]$ , as emissions mitigation efforts alone do not suffice. DACCS, on the other hand, has little impact on the environment besides the impacts associated with the energy demand and CCS [\[6](#page-9-5), [63\]](#page-11-2).

Further potential deployment barriers specific to OL relate to MRV, the governance of international waters, and the current legal framework (e.g., the London Protocol[[53](#page-10-29)]), which prohibits the release of all substances not explicitly allowed into the ocean.

## **5. Conclusions**

OAE has the potential to be an economically competitive gigaton-scale CDR option. Given the current energy prices and a fully decarbonized energy system including freight transport, we estimate the total cost of removing a ton of  $CO<sub>2</sub>$  via OL to be 133– 204 \$ tCO<sub>2</sub><sup>-1</sup> and 194–296 \$ tCO<sub>2</sub><sup>-1</sup>, if the heat is supplied by natural gas or by electricity, respectively. The uncertainty comes from the method's efficiency, here assumed to vary between  $1.2-1.8$  molCO<sub>2</sub> molCa(OH)<sub>2</sub><sup>-1</sup>. However, if freight transport continues to rely on fossil fuels, then costs increase to 156–263 \$ tCO<sub>2</sub><sup>-1</sup> and 227–380 \$ tCO<sub>2</sub><sup>-1</sup>, respectively, thus again underlining the importance of full decarbonization of the energy system, even when CDR options like OL become available. Compared to DACCS, OAE is economically competitive if discharge rates and efficiencies are high. This result highlights the importance of localized application where the efficiency is favorable and of addressing the engineering challenges to maximize distribution efficiency.

However, we identify three types of critical issues that must be addressed before OL can be deployed at a large scale:

- (1) The most critical knowledge gap that could stop deployment is the impact of OAE on the marine ecosystem. So far, research has not indicated negative impacts, but the size and importance of the marine ecosystem warrant a thorough investigation before actual deployment.
- (2) Uncertainties that could delay deployment relate to the availability of CCS, speed and depth of energy system decarbonization, social acceptance, and MRV. Delayed availability of CCS and energy system decarbonization make OL less efficient and therefore increase the costs, which would most likely delay deployment. Difficulties in verification of carbon removal would most likely also delay a subsidy for carbon removal, which would impede economic efficiency and therefore also lead to delays. However, these uncertainties apply to almost all CDR technologies [\[6,](#page-9-5) [64](#page-11-3)].
- (3) Further challenges that need to be addressed are legal and governance frameworks as well as the distribution system. Currently the London protocol prevents adding alkalinity to the ocean, and a governance framework that incentivizes CDR including OAE and regulates MRV is lacking as well. The distribution system presents an engineering challenge that needs to be solved, and which currently adds to the uncertainty of total costs for OAE.

We find that the limitations of this method are not in the technical potential of the removals or its costs, but rather in its unresolved sustainability concerns, technological uncertainties, and governance and MRV challenges. The large potential and competitive costs make OL an interesting CDR option for many countries, which calls for further research

and international cooperation to fill the gaps outlined above.

## **Data availability statement**

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

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