A framework for understanding efficiency losses of Ocean Alkalinity Enhancement

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Abstract

Ocean alkalinity enhancement (OAE) is a proposed carbon dioxide removal (CDR) approach in which alkalinity is added to the surface ocean to facilitate atmospheric CO_2 uptake and long-term storage in the ocean's dissolved inorganic carbon pool. Carbonate system thermodynamics provide a stoichiometric upper bound on the amount of CO_2 that can be taken up per unit of alkalinity added, but realized uptake can be lower due to dynamic physical and (bio)geochemical processes that evolve over time. This Perspective presents a structured framework for understanding these potential efficiency losses. We identify loss pathways across four categories: incomplete mineral dissolution, loss of alkalinity to solid phases, incomplete air–sea exchange, and indirect impacts on ocean biogeochemical cycles. The potential significance of each loss varies across OAE approaches and across spatial and temporal scales. We highlight that substantial knowledge gaps persist across all of these processes, and we suggest targeted research priorities to improve fundamental understanding and reduce uncertainty. By providing a common structure and definitions, this framework aims to support coordination across oceanographic disciplines and to improve assessments of OAE efficiency at both the project and global scale.

Introduction

Ocean alkalinity enhancement (OAE) has been proposed as a method for carbon dioxide removal (CDR), given potential advantages in cost and scalability by using the global ocean as a CO_2 sorbent and storage reservoir. OAE works by increasing seawater alkalinity, defined as the excess of bases over acids, mimicking alkalinity addition from natural rock weathering. This increase in alkalinity shifts the speciation of dissolved inorganic carbon (DIC) away from CO_2, causing a deficit with respect to atmospheric CO_2. (See Zeebe and Wolf-Gladrow [2001], and Dickson [2010] for detailed explanations of seawater carbonate chemistry.[1,2]) For seawater in contact with the atmosphere, atmospheric CO_2 will consequently be absorbed to restore gas equilibrium. Or, for seawater with naturally high pCO_2, alkalinity addition will lead to reduced CO_2 outgassing. Considering carbonate system thermodynamics, between 0.77-0.96 mol of CO_2 may be taken up per mol of alkalinity added to seawater; this range is largely driven by temperature, and the global average is approximately 0.84.[3] This maximum, thermodynamically-defined uptake can only be achieved, however, if added alkalinity is not consumed by environmental processes and if it remains in surface waters long enough for complete air–sea equilibration. Because OAE operates in an open system—where both alkalinity and CO_2 concentrations evolve over time within a dynamic environment—the actual uptake can be lower than the thermodynamic limit.

How much atmospheric CO_2 is removed per unit of alkalinity added, and over what timescale, is a central question defining the *efficiency* of OAE interventions. The bulk of recent research on this topic focuses on how physical mixing and air-sea exchange rates may limit CO_2 uptake. While these physical processes are important, they represent only one class of potential efficiency loss; other chemical and biological processes can also reduce net CO_2 uptake.

In this Perspective, we present a framework for understanding the processes that impact the efficiency of OAE. We group losses into four categories: incomplete mineral dissolution, loss to solids, incomplete air-sea equilibration, and indirect impacts on ocean biogeochemical cycles that affect CO_2 uptake (Figure 1).

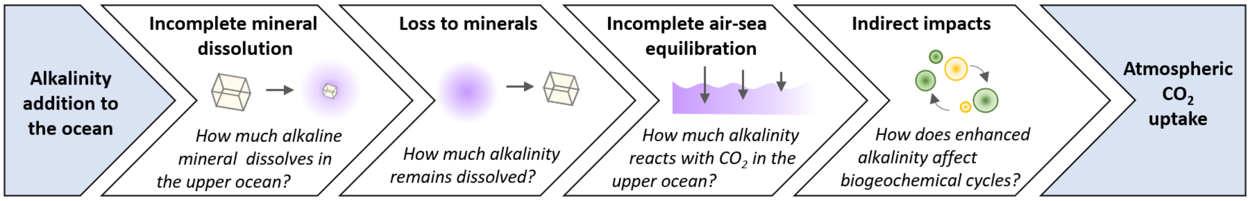


Figure 1. Four categories of processes that reduce OAE efficiency, i.e., the amount of atmospheric CO_2 uptake per unit of alkalinity added to the ocean. Losses can be considered in sequential order, however in practice they are often interactive.

We define an efficiency loss as any reduction in CO_2 uptake compared to the thermodynamically predicted value for a given alkalinity addition, or—for mineral applications—the *expected* alkalinity addition from mineral dissolution. (We note that alkalinity is, by definition, dissolved, i.e., a property of solutions; solid, alkaline feedstocks do not constitute alkalinity until they dissolve.) This definition refers specifically to losses occurring *after* addition of alkalinity or alkaline feedstock to the ocean. Ultimately, the net efficiency of OAE also depends on upstream emissions associated with alkalinity sourcing, energy use, and related processes; those factors must be considered via lifecycle assessment, and are beyond the scope of this work. From the perspective of lifecycle assessment, the loss pathways considered here inform the efficiency of *gross* CO_2 uptake, which is needed to calculate the net efficiency of the entire CDR intervention.

Clearly framing efficiency losses is important for several reasons. Foremost, OAE-driven CDR cannot be directly measured with high confidence, and ultimately requires some degree of modeling.[4] This reality results from both measurement challenges—such as low signal to noise ratio following rapid dilution—and the need to compare OAE interventions against a counterfactual scenario, which by definition must be modeled. The thermodynamic, physical, and biogeochemical processes governing CO_2 uptake must be represented in both the OAE and counterfactual scenarios. Even for OAE methods that deploy pre-equilibrated alkalinity, downstream losses within the ocean environment may occur, and these must also be considered. A clear framework for efficiency losses is therefore needed to specify which processes must be accounted for in a given deployment scenario, and to ensure that assumptions are comparable across models. Without such a framework, incomplete accounting or optimistic assumptions can lead to systematic overestimation of CO_2 uptake. At the global scale, this risks overestimating OAE's potential contribution to global CDR goals; and at the project level, such assumptions could lead to over-crediting. While current monitoring, reporting, and verification (MRV) protocols acknowledge some efficiency losses, many are not incorporated quantitatively due to knowledge gaps and limited data.[5] Finally, a consistent framework supports coordinated scientific research, and helps clarify which pathways require more targeted research and/or are insufficiently represented in relevant ocean models. Establishing common terminology and clear definitions also enables more effective coordination between experiments and model development, ensuring that both efforts address the same underlying questions.

Because efficiency losses will differ across deployment conditions, it is necessary to briefly outline the various approaches to OAE. OAE deployments can be organized along two primary dimensions: the source of alkalinity and the method of delivery to the ocean. From the perspective of mineral dissolution kinetics, alkalinity sources fall into three broad feedstock categories: slower-dissolving rocks and minerals that dissolve in typical seawater conditions on the scale of days to years (e.g., olivine, limestone, and some industrial waste products like steel slag); faster-dissolving rocks and minerals that dissolve on the scale of seconds to days (e.g., Ca-Mg- and potentially Na- oxides and hydroxides, and ikaite); and direct application of alkalinity, which can be produced electrochemically or via pre-dissolution of alkaline minerals (e.g., in reactors). For solid feedstocks, some alkalinity sources—steel slag for example—may be a mixture of both faster and slower-dissolving minerals.[6] For applications deploying (dissolved) alkalinity directly, the alkalinity may be either *unequilibrated* with respect to atmospheric CO_2 (i.e., ambient DIC and elevated pH), or *equilibrated* with respect to atmospheric CO_2 (i.e., enhanced DIC and circum-ambient pH) at the time of addition to the ocean. For approaches using equilibrated alkalinity, atmospheric CO_2 uptake occurs in engineered systems *prior* to addition to the ocean (Figure 2). Depending on alkalinity source, there are three possible methods for delivering alkalinity to the ocean: coastal seafloor placement (for slower-dissolving rocks and minerals); coastal surface discharge (for dissolved alkalinity or slurries of faster-dissolving minerals), and offshore surface discharge (e.g., dispersal from ships, likely only for faster-dissolving minerals). Each deployment scenario has different considerations for potential efficiency losses.

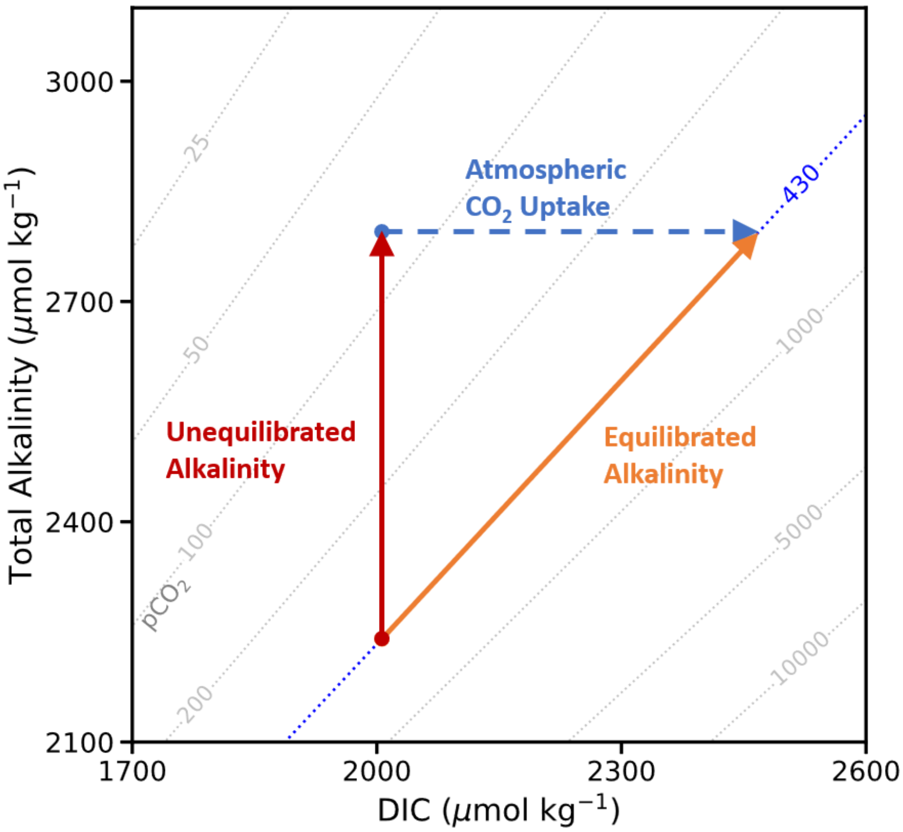


Figure 2. Relationship between total alkalinity, DIC, and pCO$_2$. When unequilibrated alkalinity (red line) is added to seawater, CO$_2$ uptake (blue dashed line) occurs until equilibration with the atmosphere is achieved (currently ~426 ppm). In engineered systems, alkalinity may be pre-equilibrated with the atmosphere prior to adding to the ocean (orange line). (Data modeled in PyCO2SYS at T=20, S=35).

In the following sections, we describe nine potential loss pathways, organized into four categories (Table 1). For each loss pathway, we describe the (bio)geochemical and physical processes that reduce expected CO$_2$ uptake, highlight the spatial and temporal scales at which they are most consequential, and suggest research priorities to constrain them. We do not attempt to quantify the magnitude of potential losses here, as these vary greatly across deployment scenarios and because research is ongoing.

Table 1. Specific loss mechanisms vary by feedstock and deployment type, as well as environmental parameters.

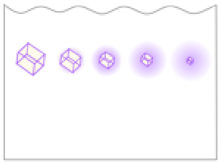
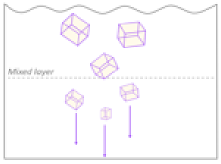
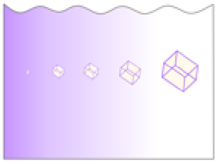
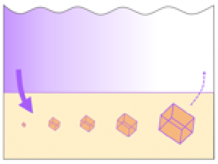
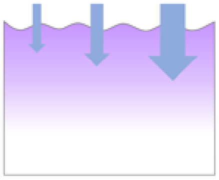
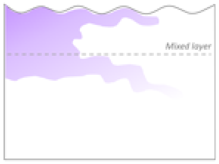
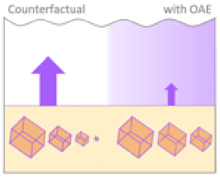
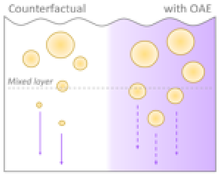
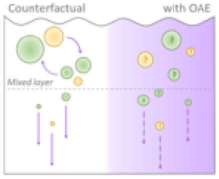
Category	Sub-category	Description	Most relevant feedstock and deployment types	Relevant parameters
Incomplete mineral dissolution	**Mineral dissolution kinetics** 	Feedstock dissolves more slowly than anticipated, and/or incompletely.	Rock and mineral feedstocks in all deployment methods	Feedstock mineralogy; particle size distribution and reactive surface area; seawater Ω near the reactive surface area, and other chemical factors; seawater temperature; sinking or advection rate
	Particle sinking 	Feedstock and/or secondary CaCO3 or other autochthonous precipitates sink out of mixed layer before fully dissolving.	Faster-dissolving feedstocks added to rivers, coastal water, or the open ocean via ships, or floor environments with (re)suspended sediments	Particle density, size distribution, and reactive surface area; mineral dissolution rate as affected by water advection close to the reactive surface area; turbulence; aggregation
Alkalinity loss to minerals	**Water column** 	Secondary minerals or solid phases form out of solution in the water column and may not redissolve.	Dissolved alkalinity and faster-dissolving feedstocks in all deployment methods	Feedstock mineralogy, particle size distribution and reactive surface area; seawater Ω (bulk and near-surface) and other chemical factors; temperature; suspended solid quality and density (nucleation sites)
	Sediments 	Alkalinity is lost within sediments via mineral precipitation, clay formation, and/or ion exchange.	All alkalinity types in coastal deployments	Porewater Ω; sediment characteristics (mineralogy, permeability, grain size distribution, etc.); porewater exchange rates; biological activity (e.g., bioturbation)

Table 1. (Continued)

Category	Sub-category	Description	Most relevant feedstock and deployment types	Relevant parameters
Incomplete air-sea equilibration	**Kinetics of air-sea exchange** 	Air-sea CO2 equilibration takes time. Early timepoints have lower apparent efficiency than later timepoints.	All (unequilibrated) alkalinity types, all deployment methods	Physical factors controlling air-sea gas exchange, including factors affecting ΔpCO2 (e.g., mixing)
	Subduction and diapycnal mixing 	Complete air-sea equilibration may take years; during this time, unequilibrated alkalinity may subduct or mix into the ocean interior.	All alkalinity types, all deployment methods	Downwelling; diapycnal mixing; air-sea exchange rate
Indirect impacts	**Decreased sediment alkalinity flux** 	Dissolution of carbonate minerals in sediments is suppressed by elevated Ω, decreasing benthic flux of alkalinity to the water column.	All alkalinity types in coastal deployments (near-term); potentially relevant for all deployment methods (long-term)	Porewater Ω; sediment characteristics (mineralogy, permeability, grain size distribution, etc.); porewater exchange rates
	Increased carbonate counterpump 	Export of carbonate minerals from the surface ocean increases due to enhanced production and/or reduced dissolution.	All alkalinity types, all deployment methods	Seawater Ω; plankton community
	Altered organic carbon cycling 	Export of organic carbon from the surface ocean is altered due to changes in productivity, respiration, and/or ballasting.	All alkalinity types, all deployment methods	Seawater Ω; plankton community

1. Incomplete mineral dissolution

For OAE applications using solid alkaline feedstocks, it cannot be assumed that alkalinity embodied within minerals fully dissolves in the upper ocean. Slower than expected or incomplete dissolution results in discrepancies between expected and actual alkalinity addition, lowering efficiency. For open-ocean applications, particle sinking speed, in combination with dissolution rate, also dictates how much alkalinity is added to the surface ocean versus how much sinks below the mixed layer. Because these two interrelated mechanisms—dissolution rate and sinking speed—may require different analytical approaches, we discuss them here separately.

Mineral dissolution kinetics

Dissolution kinetics, or the rate at which particles dissolve, determines how quickly, and how much, alkalinity is added to seawater over a certain timescale. Dissolution kinetics are well-parameterized for pure minerals in ideal solutions, and—in theory—rates can be calculated given particle surface area (or grain size), temperature, salinity, and mineral saturation state (Ω), etc.[7–9] However, environmental conditions at the mineral surface may differ from bulk seawater, precluding direct calculation of dissolution rates. For example, in OAE applications, hydrodynamic conditions can be an important control on seawater chemistry. Optimal dissolution kinetic parameters are generally derived in experiments with high advection energy, reducing Ω at the mineral surface (c.f. Pokrovsky & Schott, 2004).[10] In the ocean however, advection energy is typically lower than in a stirred reactor, resulting in higher Ω at the mineral surface and slower dissolution rates.[11] For faster-dissolving feedstocks, high Ω around particles can also result in a dynamic interplay between dissolution and secondary mineral precipitation (described in the next section). If dissolution is sufficiently rapid but transport of resulting alkalinity away from the grain surface is limited, supersaturation with respect to carbonate minerals may occur, leading to mineral precipitation (aragonite in seawater and calcite in freshwater).[12–15] Further, precipitation on the feedstock surface potentially slows dissolution rates. Therefore, the role of dissolution kinetics on OAE efficiency must be considered in tandem with loss of alkalinity to carbonate formation, especially for applications with fast-dissolving feedstock.

Slower-dissolving feedstocks used in coastal seafloor applications may also experience slower than expected dissolution rates. At the most fundamental level, silicate dissolution is well known to slow over time as mineral surfaces age; thus, ideal rates quickly become inapplicable.[16–18] Olivine, for example, may develop passivation layers that slow dissolution on relatively short timescales, and/or experience incomplete dissolution (e.g., serpentinization) on longer timescales.[19] In addition, porewater chemistry typically differs from the overlying water in Ω, pH, and redox state, which can increase or decrease dissolution rates relative to the water column, depending on conditions. In cohesive sediment with little porewater advection, porewater is likely to become saturated with respect to the feedstock, slowing dissolution of buried particles.[20,21] High energy environments like sandy beaches may be less prone to these issues, via increased porewater flushing that decreases Ω, and/or prevention of passivation layers by physical abrasion. However, as with fast-dissolving minerals, a dynamic interplay between dissolution and secondary precipitation can occur, resulting in less alkalinity addition than expected.[22] Because dissolution kinetics change over time, potential efficiency losses must be integrated over (potentially long) timescales for slow-dissolving minerals. Measuring small dissolution signals, that become increasingly small over time, against a dynamic background in field deployments is challenging.[21] Thus, modeling will realistically be necessary to extrapolate local measurements across larger areas and over long time periods. Developing confidence in such extrapolations will require improved understanding of sediment diagenesis across a range of sediment types – e.g., with varying levels of organic matter, mineralogy, porosity, kinetic energy, etc.

Particle sinking

In open ocean applications, faster-dissolving minerals can be used for OAE if they dissolve sufficiently rapidly before sinking below the mixed layer.[23] Sinking speed can be modelled simplistically with Stokes Law; however, in real-world conditions, turbulence can significantly affect settling, potentially increasing sinking speed by orders of magnitude.[24] Rapid sinking can result in negligible dissolution in the mixed layer, even for small particles of fast-dissolving minerals (e.g., MgO with $< 10\ \mu m$ diameter).[25] Settling rates may also increase as a result of particle aggregation.[26,27] On the other hand, dissolving alkaline particles may form carbonate precipitates at the µm scale, which may remain suspended for longer periods of time within the mixed layer.[28] Studies of particle sinking have thus far focused on observation of natural particles and/or numerical simulations. Experimental lab and field observations are needed to confirm these results for OAE applications, as well as improve models with relevant processes. Proper modelling must consider interactions between particle dissolution, alkalinity generation, potential carbonate formation (discussed in the next section), and aggregation, coupled to the movement of particles through the mixed layer. Such models do not yet exist.

Settling rate is also an important consideration for near-shore applications, as particles may sink to the seabed.[29] Fast-dissolving feedstocks may dissolve over time on the sediment surface, with resulting alkalinity entrained into nearshore water. However, complex sediment interactions may result in alkalinity loss not anticipated in the water column (described in next section). For these reasons, mineral sinking rates should also be considered in the design of near-shore OAE deployments.

2. Alkalinity loss to minerals

Following mineral dissolution, alkalinity can potentially be lost via subsequent precipitation to, or reaction with, solid minerals. This concept is sometimes referred to as alkalinity *stability*.[15] Loss to solids can occur with both equilibrated and unequilibrated alkalinity, although thresholds are crossed more readily with unequilibrated alkalinity.[30] Loss can occur over dynamic timescales – ranging from spontaneous, runaway precipitation for high Ω conditions, to delayed loss for more moderate conditions, and slower interactions with sediments over timescales of porewater cycling. The mineralogy of the precipitated solid phase will also affect loss timescales, and determine whether the loss is transient (e.g., potentially for $Mg(OH)_2$) or effectively permanent (e.g., potentially for carbonate minerals).[28] The potential, rate, and durability of alkalinity loss to minerals depend on environmental conditions and feedstock properties, so understanding these pathways mechanistically is essential for evaluating the efficiency of specific OAE deployments.

Water column: mineral precipitation

In the water column, mineral precipitation may occur when seawater becomes sufficiently oversaturated following alkalinity addition. In an OAE context, the most likely minerals to precipitate include calcium carbonate ($CaCO_3$) polymorphs and magnesium hydroxide ($Mg(OH)_2$).[14,15,28,30–34] Although seawater is naturally oversaturated with respect to $CaCO_3$ in most locations, precipitation is kinetically inhibited, and significant oversaturation is required to facilitate spontaneous precipitation.[35–37] If sufficiently oversaturated, $CaCO_3$ precipitation may then occur either homogeneously, i.e., from the association of dissolved calcium (Ca^{2+}) and carbonate ions (CO_3^{2-}) without the need for a pre-existing mineral surface area, or (pseudo-)heterogeneously, i.e., by precipitation on suspended particles and colloids.[38,39] The latter is more likely to occur at lower Ω, and will occur more quickly on surfaces with higher lattice compatibility, e.g., $CaCO_3$ or calcium-rich particles. Therefore, available surface area plays a key role in the precipitation process.[14,28] Similarly, temperature and salinity also have a role in triggering precipitation.[40] Because seawater is typically oversaturated with respect to $CaCO_3$, precipitated $CaCO_3$ may constitute a long-term loss of alkalinity from the water column.[41,42] The second mineral likely to precipitate is $Mg(OH)_2$. Seawater is typically undersaturated with respect to $Mg(OH)_2$, however alkalinity addition can result in a rapid rise in pH, and therefore OH^- concentration. Once reaching a pH of ~10.5, $Mg(OH)_2$ precipitates concomitantly with alkalinity increase.[30–34,43] The stability of precipitated $Mg(OH)_2$ is not well understood, though some research suggests it may dissolve over longer timescales, i.e., months to years.[11] For both $CaCO_3$ and $Mg(OH)_2$, two moles of alkalinity are lost per mole of mineral formed. For $CaCO_3$, one mole of DIC is also lost; this sequestration of carbon (C) in mineral form may still constitute CDR, albeit at a reduced efficiency than if the DIC had remained dissolved. (In seawater with a thermodynamic efficiency of 0.8 mols atmospheric CO_2 sequestered per mol alkalinity addition, precipitation of alkalinity as $CaCO_3$ results in an efficiency of 0.3 mols CO_2 per mol alkalinity.) In contrast, no C is sequestered when alkalinity is lost permanently to $Mg(OH)_2$. Thus, the mineral fate of lost alkalinity must be considered in assessments of OAE efficiency.

Potential loss to mineral precipitation will vary by feedstock and deployment scenario. Applications deploying (dissolved) unequilibrated alkalinity may trigger immediate $Mg(OH)_2$ precipitation if rapid pH spikes exceed the critical pH >10. In these conditions, $CaCO_3$ precipitation may be limited, as the absence of mineral surface area allows for higher Ω thresholds.[15,30] In coastal applications, however, suspended sediments may facilitate (pseudo-)heterogeneous $CaCO_3$ precipitation.[14,44–47] Mineral precipitation is less likely when using equilibrated alkalinity, as reaching critical thresholds requires much higher alkalinity addition. In contrast to direct release of alkalinity, using solid, faster-dissolving feedstocks should result in $CaCO_3$ precipitation before $Mg(OH)_2$ buffering. $CaCO_3$ may precipitate on the surface of dissolving feedstock particles (pseudo-)heterogeneously or in the diffusive boundary layers of these particles where microenvironments may promote homogeneous precipitation.[14] Such precipitation may enter a runaway process, where more alkalinity is lost than the amount added.[11,14,15,30,48,49] The exact pH and Ω of precipitation thresholds will vary with environmental conditions. Tailored research is required to understand these thresholds across salinities and temperatures gradients, and the effect of potential inhibitors like Mg or dissolved organic matter, all of which may vary significantly across the land-ocean transition zone (Saez preprint).[13,50,51] Further, the effect of dilution, i.e., lowering precipitation thresholds, following runaway carbonate formation should also be better understood.

Sediments: mineral precipitation and ion exchange

Spontaneous mineral precipitation can also occur when seawater interacts with sediments. Precipitation thresholds may be lower in sediments compared to the water column, due to buffering by in situ minerals, alkalinity production by anaerobic respiration, and the potential for heterogenous precipitation onto mineral surfaces. This has only been studied with respect to OAE in a few publications to date, with observed alkalinity loss presumed to be from mineral precipitation.[22,52] Alkalinity may be lost within sediments to $CaCO_3$ precipitation on relatively short timescales, and/or to silicate minerals (clays, i.e., reverse weathering) on longer timescales.[19,53] For iron-containing feedstocks like olivine, iron hydroxide precipitation may also result in indirect alkalinity loss.[54] Formation of more soluble minerals like $Mg(OH)_2$ is also possible if porewater Ω is sufficiently elevated, although this may be transient for pulsed deployments, redissolving upon the return of less saturated seawater. As in the water column, precipitation of $CaCO_3$ sequesters CO_2, while silicate and hydroxide minerals are alkalinity sinks with no associated C sequestration.

A related loss pathway is ion exchange, in which dissolved ions exchange with ions adsorbed to mineral surfaces. For example, cation exchange can occur if elevated concentrations of base cations (Na^+, Mg^{2+}, Ca^{2+}, etc.) displace H^+ from sediment grain surfaces, thus reducing alkalinity. This phenomenon is increasingly identified as a major alkalinity sink for terrestrial enhanced rock weathering.[55] Although there have not yet been targeted studies in OAE applications, similar processes have been shown experimentally in sediments, where proton buffering and exchange of bicarbonate complexes can occur following changes in pH or CO_2 concentration.[56] The effect of ion exchange on alkalinity may be ephemeral, as OAE-impacted seawater dilutes or is flushed out of sediments, and natural seawater returns. Alternatively, it could lead to more permanent alkalinity loss if elevated porewater alkalinity results in mineral precipitation. Clearly identifying the fate of alkalinity loss, between ion exchange and precipitation of different minerals, is important since each possibility has a different impact on net CO_2 uptake over different timescales.

Alkalinity losses to sediments would logically be most significant for coastal deployments, where alkalinity is elevated in water masses with prolonged sediment contact, and/or for mineral deployments on the seafloor. The magnitude of loss is likely a function of the level of alkalinity addition (as above, with water column precipitation) and sediment properties like permeability and mineralogy. Experimental research is needed across these variables. Furthermore, observational hydrodynamic data are needed to define the extent to which seawater interacts with sediments. Ultimately, these data could be integrated into coupled sediment-water column models to estimate alkalinity loss to sediments near the deployment site and at regional scales.

3. Incomplete air-sea CO_2 equilibration

Air-sea CO_2 exchange is a primary control on OAE efficiency following the addition of unequilibrated alkalinity to seawater; incomplete equilibration can depress CO_2 uptake over CDR-relevant timescales (years to decades). Approaches deploying equilibrated alkalinity avoid this inefficiency (although equilibration rates in reactors are still controlled by gas exchange rates, even under optimal reactor conditions). In this loss category, we distinguish between two key controls on air–sea CO_2 equilibration: 1) surface air–sea gas exchange processes, and 2) the physical redistribution of alkalized waters through mixing and subduction that limits their atmospheric exposure. These two processes are intricately related and are estimated with the same global circulation modeling tools. We distinguish them here to highlight their different consequences and timescales: air-sea gas exchange is associated with increasing efficiency over time as full equilibration is reached over years to decades, whereas loss of alkalinity to mixing and subduction may result in effectively permanent efficiency loss.

Kinetics of air-sea CO_2 exchange

With unequilibrated alkalinity, equilibration between atmospheric CO_2 and surface seawater is not instantaneous following alkalinity addition. The flux, or exchange of CO_2 across the air-sea interface, occurs gradually and is controlled by many environmental factors. This flux is often described as F (mol m^{-2} s^{-1}):

$$F = k\, K_o\, (pCO_w - pCO_a) \qquad \text{Equation 1}$$

where k (m s^{-1}) is the gas transfer velocity, K_o (mol m^{-3} Pa^{-1}) is the aqueous-phase solubility of CO_2, pCO_a (Pa) is the partial pressure of CO_2 in air, and pCO_w (Pa) is the partial pressure of CO_2 in water.[57]

Key kinetic drivers of the gas transfer velocity (k) include: 1) wind dynamics (i.e., velocity, direction, and fetch), 2) wave dynamics (i.e., breaking and bubble entrainment), and 3) surface microlayer properties (i.e., surfactant concentrations, micro-organism presence and activity, and gel particles). In practice, k is typically parameterized by wind speed, although recent research suggests this approach may significantly underestimate air-sea exchange rate in some coastal waters.[58] The key factors that affect the air-sea pCO_2 difference (ΔpCO_2) are temperature, biological C cycling, and alkalized water transport (including mixed layer depth). Both k and ΔpCO_2 can vary strongly across location and season.

Following alkalinity addition, CO_2 uptake is initially rapid but slows as the air-sea pCO_2 difference decreases. Complete equilibration can take anywhere from months to decades, depending on local and regional conditions.[59] This equilibration timescale is longer than that for the atmospheric pCO_2 perturbation from anthropogenic emissions, where the global average equilibration time is approximately 1 year.[60] In ideal locations where lateral flow retains alkalized water at the ocean surface (e.g., subpolar latitudes), near-complete equilibration may be reached relatively rapidly, within a few years. However, in less ideal locations, equilibration may take significantly longer (>15 years) as shallow overturning and a deep mixed layer decrease ΔpCO_2.[59] This delay in complete equilibration has two important implications for the efficiency of OAE deployments. First, on shorter timescales prior to complete equilibration, estimates of CDR will inherently have lower effective efficiency than those after complete equilibration. After complete equilibration, maximum efficiency is reached, and the "loss" for this category approaches zero. Second, alkalized waters may subduct or mix into the ocean interior before they can fully equilibrate with the overlying atmosphere (described in the next section).

CO_2 uptake curves can be modeled for specific deployment scenarios to estimate efficiency at any given timepoint. Accurate parameterization of such uptake will require site- and condition-specific hydrodynamic modeling (e.g., nearfield turbulent models and regional ocean models), coupled to global ocean models. Experimental work is also needed to better constrain k values, especially in coastal waters.

Subduction and diapycnal mixing

For air-sea CO_2 exchange to occur, alkalized waters must remain at the ocean surface. Alkalinity that moves into the ocean interior prior to full equilibration cannot interact with atmospheric CO_2, and may be effectively lost on timescales beyond the intent of CDR intervention. This loss may result from either subduction of the alkalized water mass, or from diapycnal mixing with deeper water masses. Both processes depend on oceanographic conditions that vary by location, as well as seasonally, interannually, and with weather.[59,61] Losses can be minimized by deploying in locations without significant downwelling, and at times of year with less turbulent mixing between water masses.

The timescale of alkalinity subduction (or mixing) and potential return to the ocean surface also varies by location. If OAE is deployed in regions where deep water is formed, it may be hundreds of years to millennia before the water resurfaces and the equilibration process can resume.[62,63] During that time, biological processes within the subducted water mass may consume some of the unequilibrated alkalinity, however this will have no impact on atmospheric CO_2 until the water mass resurfaces.[64] At that time, air-sea equilibration and CO_2 uptake may proceed, and/or CO_2 outgassing may be decreased relative to the counterfactual scenario without the OAE intervention. Estimating the impact on atmospheric CO_2 upon resurfacing requires coupled physical-biogeochemical ocean models.

Advancements in such coupled models are needed to better constrain both the controls on air-sea CO_2 equilibration related to CO_2 exchange kinetics, and the surface residence time of alkalized water set by subduction and mixing. This will require coordinated efforts across a model hierarchy, from nearfield turbulence and plume models, to regional circulation and biogeochemistry models, to global Earth system models. Most relevant models have not yet been configured for OAE applications, and parameters important for long-term OAE performance, like interannual variability, must be integrated.[65] In addition, more complex deployment scenarios – e.g., coastal sites, seafloor applications, timed pulses – must also be represented. Finally, standardized, model intercomparisons are needed to help identify biases and reduce uncertainties.

4. Indirect Impacts

Potential efficiency losses from indirect impacts include decreased alkalinity flux from sediments, an increase in the $CaCO_3$ counterpump, and perturbation to organic carbon cycling, e.g., the soft tissue pump. In contrast to the pathways described above that mediate the availability of added alkalinity, these indirect processes alter ocean biogeochemical processes that influence atmospheric CO_2 uptake. For example, OAE may drive changes to Ω_{CaCO3}, particle dynamics, and/or ecosystem processes that modulate inorganic and organic C cycling. These impacts are likely to operate over large spatial and temporal scales.

Indirect impacts are sometimes referred to as "additionality" considerations.[52] In general, additionality refers to the extent to which an intervention results in CDR above and beyond what would have happened in a counterfactual (no intervention) scenario. At the broadest level, additionality considerations range from regulatory and financial incentives to impacts on ecological processes (as discussed here). Some approaches to calculating the efficacy of CDR interventions decompose net CDR into a) observed CO_2 uptake resulting directly from the activity itself, and b) indirect impacts on baseline ecological processes (i.e., additionality considerations), which are typically assumed to decrease C sequestration.[66] For open system CDR approaches like OAE, the climate benefit of an intervention always requires comparing the CO_2 uptake in the intervention with that of a counterfactual (non-intervention) scenario. This comparison must be done with models that integrate all relevant physical and biogeochemical processes. So-called direct CO_2 uptake and indirect impacts may physically and temporally overlap, and the resulting net CO_2 uptake is an emergent result of interactive physical and bio(geochemical) processes spread over large spatial areas and temporal scales. Thus, it is functionally difficult to separate CO_2 uptake resulting *directly* from the alkalinity enhancement itself from *indirect* processes that mediate this uptake. We therefore consider so-called direct losses and indirect losses equally within our framework, and do not ascribe the latter alone to additionality considerations. Ultimately, defining discounts to gross CO_2 uptake as integrated efficiency losses (as done here), or changes to a baseline (additionality considerations), is simply an accounting choice; either approach is appropriate as long as all potential losses are accounted for.

Decreased sediment alkalinity flux

Sediments can be a significant source of dissolved alkalinity to the water column, due in part to dissolution of carbonate minerals.[67–69] OAE-induced elevation of Ω may suppress this dissolution, effectively reducing the natural flux of benthic alkalinity. Thus, the unperturbed, counterfactual scenario will have a higher input of benthic alkalinity to the water column, relative to the OAE scenario. This difference effectively reduces the net alkalinity addition of the OAE scenario. There has been little research on this subject to date, so the magnitude of potential efficiency loss remains unconstrained. In extreme, experimental scenarios, alkalinity addition can suppress sediment $CaCO_3$ dissolution such that the OAE scenario results in lower net alkalinity addition than natural sediments alone.[52]

Functionally, suppression of benthic $CaCO_3$ dissolution and $CaCO_3$ precipitation within sediments may be difficult to disentangle: precipitation and dissolution are in dynamic equilibrium with one another, representing two ends of a spectrum. Here, we include them as separate loss pathways since end-member conditions may be relatively more important in different situations. For example, $CaCO_3$ precipitation may be significant near deployments, whereas suppressed dissolution is more likely as a diffuse impact over the continental shelf. The two mechanisms may be integrated in different ways into nearfield and/or global ocean models. For deployments using solid minerals, another consideration is the interaction between benthic alkalinity flux and feedstock dissolution rates: mutual buffering between OAE feedstock and $CaCO_3$ in sediments likely reduces the dissolution kinetics of both alkalinity sources.

Constraining losses associated with these processes requires measurements and experiments that resolve how benthic alkalinity flux responds to changes in bottom water chemistry. Experiments with both high and low alkalinity additions are needed across sediment types and hydrodynamic conditions to assess local impacts near deployments, as well as diffuse impacts at regional to global scales. These data can then be used to inform nearfield and global models. Projections of OAE scalability must also consider that benthic alkalinity flux from $CaCO_3$ dissolution is expected to increase in the future in response to ocean acidification, in both shallow coastal and deep ocean sediments.[70–72] Future OAE deployments may increasingly suppress this alkalinity flux, resulting in greater effective efficiency loss.

Increased carbonate counterpump

In the natural C cycle, the sinking of biogenic carbonate minerals exports an estimated 1 Gt of C from the surface ocean per year (equivalent to approximately 10 Mmol of alkalinity).[68,73] This export of alkalinity is often referred to as the "carbonate counterpump", as it *increases* pCO_2 at the ocean surface (in contrast to the biological or soft tissue pump–described in the next section—that *decreases* pCO_2 at the ocean surface). OAE may enhance the carbonate counterpump by increasing calcification and/or decreasing dissolution of particles before sinking, in both cases resulting in alkalinity loss from the surface ocean. While there is currently no evidence of either process in OAE-specific research, global ocean observations show a clear, positive correlation between seawater alkalinity and calcification, suggesting that OAE may indeed lead to increased formation of $CaCO_3$.[74] Dissolution of biogenic $CaCO_3$ has also been shown to slow at higher Ω.[75] Palaeoceanographic data indicate a shift toward more heavily calcified planktonic species and increased carbonate export in response to warming and subsequent weathering over geologic (10^5 y) timescales; whether such ecological changes could occur on CDR-relevant timescales remains unknown.[76] Despite these observations, the exact relationship between saturation state and carbonate export remains difficult to define, and likely depends on other processes (e.g., temperature, nutrients, respiratory-driven dissolution, etc.).[68] Given the early stages of research on this topic, much of what is known is extrapolated from ocean acidification studies, which do not show clear directional impacts between carbonate export and seawater pH.[77]

Species-level assays can help inform the mechanistic impacts of OAE on calcification. However, micro- and mesocosm experiments at the community-level are needed to understand the net impact on both calcification and dissolution, and the emergent impact on export rates. Identifying alkalinity thresholds, below which there is no measurable impact on export, would be useful for constraining deployment design. Recent experiments are starting to be published on this topic, but more work is needed across different marine biogeographies.[78] Systematic efforts like the Ocean Alkalinity Enhancement Pelagic Impact Intercomparison Project (OAEPIIP) will help fill this gap.[79] If significant results are observed, incorporating experimental data into global biogeochemical components of ocean models is critical.[65]

Altered organic carbon cycling

The biological pump exports approximately 10 Gt of organic C per year from the ocean's surface to the deep sea – approximately ten times greater than inorganic C export via the carbonate counterpump.[80] This export is considered a long-term sink of atmospheric CO_2. OAE could potentially alter the magnitude of the biological pump directly via changes to primary productivity and respiration, and/or by changes to mineral ballasting. The net impact of these changes on C export is unclear, and early research has not shown a significant impact in either direction.[81,82] The impact is likely to be different across locations and OAE deployments. For example, the use of silicate feedstocks containing micro- and macro-nutrients, such as olivine, may stimulate primary productivity more than electrochemically-produced alkalinity.[83] For all OAE scenarios, changes to ballasting must be considered because organic C and $CaCO_3$ export are intricately linked.[84,85] While an enhanced carbonate counterpump would *decrease* atmospheric CO_2 uptake, enhanced organic C export would *increase* atmospheric CO_2 uptake. Thus, the balance of the two processes, if affected by OAE, is difficult to predict. Changes to organic C cycling in rivers, estuaries, and coastal ecosystems (e.g., burial in sediments) must also be considered for river and coastal applications.

As with considerations for the carbonate counterpump, species-level assays can help inform mechanistic impacts on primary productivity and respiration, although these are expected to be insignificant at dilute levels of enhanced alkalinity. Micro- and mesocosm experiments are needed to understand the net impact of OAE on organic and inorganic C cycling, and the resulting impact on export rates. Data from systematic efforts like OAEPPIIP across different biogeographies will help improve the biogeochemical modules used to account for these processes in ocean models.

Discussion

The CDR efficiency of OAE, at a given point in time following alkalinity addition, is often described as:

$$\eta(t) = \Delta DIC(t)/\Delta Alk \qquad \text{Equation 2}$$

where ΔAlk is the increase in seawater alkalinity, and ΔDIC is the corresponding increase in DIC resulting from uptake of atmospheric CO_2.[61] In most applications, ΔAlk is typically considered equal to the amount of alkalinity added to the ocean, omitting consideration of loss pathways that consume alkalinity. We propose to use instead a slightly broader definition of alkalinity, Alk_{Added}, that considers the amount of alkalinity (or assumed alkalinity from mineral dissolution) added to the ocean, rather than the subsequent increase in alkalinity. Using ΔAlk or Alk_{Added} may be functionally equivalent for modeling studies focused solely on air-sea exchange, but differences may result when including alkalinity-related loss pathways (dissolution, loss to solids, and indirect impacts) described here.

Incorporating all potential losses into a single metric is not straightforward because each process impacts efficiency in different ways and over different timescales. While the kinetics of air-sea exchange impact ΔDIC(t) directly, other losses affect ΔDIC(t) *indirectly* by changing ΔAlk and/or altering the sequestration of organic C. Conceptually, all losses can be integrated into an efficiency formulation as:

$$\eta(t) = (\Delta DIC_{max} - L_{Dissolution}(t) - L_{Minerals}(t) - L_{Air\text{-}sea\ flux}(t) - L_{Indirect}(t)) / Alk_{Added} \qquad \text{Equation 3}$$

Where ΔDIC_{max} is the maximum potential CO_2 uptake for Alk_{Added} considering carbonate thermodynamics, and L(t) factors represent losses for the four loss categories described here, estimated at a given point in time.

Efficiency must be assessed for a specific point in time because losses act at different temporal scales. For example, efficiency losses associated with mineral sinking and precipitation to solids may occur essentially immediately, over the course of seconds to days, whereas losses to incomplete mineral dissolution, and incomplete air-sea flux will gradually decrease over time, on the scale of years.[15,59] In addition to the loss pathways discussed here, long-term changes to Earth system C fluxes, for example by shifting the burden of atmospheric CO_2 uptake between oceanic vs terrestrial reservoirs, will also impact OAE efficiency at scale.[86] However, these unavoidable feedbacks between global C reservoirs affect all CDR pathways, and are typically not considered in CDR efficiency assessments. Of course, what ultimately matters to climate mitigation is the total impact on radiative forcing, so changes to non-CO_2 greenhouse gas fluxes and albedo should also be considered in OAE efficiency.[87,88] However, there is no research to date on these potential impacts.

Losses also have different spatial footprints. Those associated with short timescales – mineral sinking and precipitation to solids – are primarily a concern in proximity to OAE deployments, and can likely be avoided with careful deployment design, for example by keeping alkalinity below critical thresholds. Other losses occur at larger spatial scales, and must be assessed in regional and global models. Incomplete air-sea flux is currently the focus of most modeling efforts, which remain under active development.[89] Additional losses can be integrated into these modeling efforts, but this is currently challenging given incomplete scientific understanding and lack of generalizable loss factors. Ultimately, all relevant losses should be incorporated into both local and global models to better constrain OAE efficiency across scales.

Reducing uncertainty in estimates of OAE efficiency will require coordination across experimental, observational, and modeling efforts. The framework presented here is intended to help align these communities by providing a common set of loss definitions that can be used across disciplines. Given that gaps in understanding persist across all the loss pathways described here, a consistent framework also helps standardize assumptions in current approaches to estimating efficiency, and also helps identify priority research areas. Looking forward, progress will depend not only on improved measurements and process understanding, but also on consistent reporting and targeted model intercomparisons that test the significance of specific losses across deployment conditions. Ultimately, informed accounting of all potential losses is required, both for technically defensible C accounting at the project level, and for avoiding systematic overestimation of OAE's contribution to global climate mitigation.

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