



SEQUESTERING ATMOSPHERIC CARBON DIOXIDE BY INCREASING OCEAN ALKALINITY

HAROON S. KHESHGI

Corporate Research Laboratories, Exxon Research and Engineering Company, Annandale, NJ 08801, U.S.A.

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Abstract—We present a preliminary analysis of a geoengineering option based on the intentional increase of ocean alkalinity to enhance marine storage of atmospheric CO₂. Like all geoengineering techniques to limit climate change, with today's limited understanding of the climate system, this approach must be regarded as a potential strategic option that requires ongoing assessment to establish its potential benefits and side effects. CO₂ would be absorbed from the atmosphere by the oceans at an increased rate if ocean alkalinity were raised. Ocean alkalinity might be raised by introducing the dissolution products of alkaline minerals into the oceans. The limited deposits of naturally occurring soda ash (Na₂CO₃) are readily soluble and easily mined. Limestone (CaCO₃) is abundant in the Earth's crust but is not readily soluble. This analysis explores the potential feasibility and limits of such approaches.

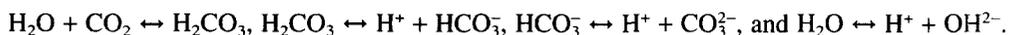
1. INTRODUCTION TO GEOENGINEERING RESPONSES TO CLIMATE CHANGE

A number of response options have been proposed to limit concerns that increasing concentrations of greenhouse gases may lead to future climate change. Among the options that might become necessary to deploy at some time in the future, should climate change prove to be serious, are those that involve geoengineering techniques to control greenhouse gas concentrations or to limit potential impacts.¹ While scientific understanding of climate change is far too limited today to contemplate deploying response options based on geoengineering, they should be investigated to improve our understanding of their potential to contribute to development of a portfolio of strategic options to address climate change. In this paper we consider geoengineering approaches to increase ocean alkalinity in order to draw CO₂ from the atmosphere to the oceans.

2. THE RELATIONSHIP BETWEEN OCEAN ALKALINITY, THE OCEAN CARBON SINK AND ENVIRONMENTAL IMPACTS

The oceans contain 3.2×10^{18} moles of dissolved inorganic carbon, predominantly in the form of bicarbonate ion.² Exchange of CO₂ between the atmosphere and the mixed layer (roughly the top 100 m) of the ocean is rapid compared with exchange between the mixed layer and deeper layers of the ocean. The characteristic time scale for the atmosphere to exchange carbon with the mixed layer is on the order of one year, whereas the characteristic time scale for the abyssal oceans to mix with the surface layers is greater than a century. Furthermore, atmosphere/mixed-layer exchange is rapid compared to the trend of atmospheric CO₂ caused by anthropogenic emissions which is increasing at roughly $1 \mu\text{atm/yr}$ with the current $p\text{CO}_2 \approx 350 \mu\text{atm}$.³ Therefore, the partial pressure of CO₂ in the mixed layer is *near equilibrium* with that in the atmosphere on average, although there are regional deviations.^{2,4}

The pH of the oceans has a strong effect on the partial pressure of CO₂ for a given inorganic carbon content TC, which is the sum of the concentrations of carbonic acid [H₂CO₃], bicarbonate ion [HCO₃⁻] and carbonate ion [CO₃²⁻]. Carbon is partitioned between H₂CO₃, HCO₃⁻ and CO₃²⁻ through the reactions.



Additional reactions for borate chemistry as well as the effects of temperature and salinity also influence the partitioning of carbon in sea water. The pH of the oceans is affected by what is known as titration alkalinity TA, which is the molar charge equivalent of anions such as Na⁺ or Ca²⁺ in excess of those

balanced by strong cations such as Cl^- . The effect of introducing the dissolution products of alkaline minerals would be to increase TA, buffer the oceans to decreasing $p\text{H}$ and, thereby, increase the solubility of CO_2 in sea water. Baes,⁵ and Broeker and Peng² give a good overview of ocean carbonate chemistry. The sensitivity of TC, $p\text{H}$ and $[\text{CO}_3^{2-}]$ to TA calculated with a model^{6,7} for equilibrium chemistry is

$$\begin{aligned} (\partial\text{TC}/\partial\text{TA})_{p\text{CO}_2=350\mu\text{atm}} &= 0.89, (\text{TA}) \times (\partial p\text{H}/\partial\text{TA})_{p\text{CO}_2=350\mu\text{atm}} = 0.39, \\ (\partial[\text{CO}_3^{2-}]/\partial\text{TA})_{p\text{CO}_2=350\mu\text{atm}} &= 0.087. \end{aligned} \quad (1)$$

These sensitivities were calculated for current mixed layer conditions, $\text{TC} = 2255 \mu\text{mol}/\text{kg}$, $\text{TA} = 2313 \mu\text{mol}/\text{kg}$, $p\text{CO}_2 = 350 \mu\text{atm}$, salinity = 34.87‰, and temperature = 18°C.^{4,8} Therefore, adding one mole equivalent of alkalinity increases the $p\text{H}$ and increases the carbon content of the seawater by 0.89 mole carbon when at equilibrium with an atmosphere at fixed $p\text{CO}_2 = 350 \mu\text{atm}$. Of the 0.89 mole of carbon absorbed or added to seawater, 0.087 mole is in the form of carbonate ion with the bulk of the remaining 0.80 mole in the form of bicarbonate ion.

The potential environmental impact of altering ocean alkalinity is of primary concern. The impact of increased calcium concentration could be one concern. However, the quantity of calcium introduced to increase ocean TA in order to sequester atmospheric CO_2 will be comparable to the number of moles of CO_2 sequestered. Even if calcium were added to the ocean at a rate comparable to the current emission rate of CO_2 of $5 \times 10^{14} \text{ mol CO}_2/\text{yr}$, this is quite small relative the total quantity 1.5×10^{19} moles Ca dissolved in the oceans.⁹ While there may be an impact of such a small relative increase in the Ca content of the oceans, this is not expected. There could, of course, be important impacts of the specific operational method for altering ocean alkalinity.

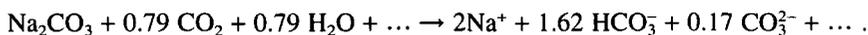
There is reason to believe that there could be a net environmental benefit from increasing ocean alkalinity, in addition to removing CO_2 from the atmosphere. Marine biota are sensitive to temperature, ocean mixing and $p\text{H}$. Some may be sensitive to carbonate ion concentration especially when $[\text{CO}_3^{2-}]$ is decreased to the point at which sea water becomes undersaturated with respect to CaCO_3 , since many species of biota require shells of CaCO_3 . Increases in atmospheric CO_2 decrease both $[\text{CO}_3^{2-}]$ and the $p\text{H}$ of ocean water. Using equilibrium chemistry again,

$$\begin{aligned} (\text{TC}/p\text{CO}_2) \times (\partial p\text{CO}_2/\partial\text{TC})_{\text{TA}=2313\mu\text{mol}/\text{kg}} &= 14., (\text{TC}) \times (\partial p\text{H}/\partial\text{TC})_{\text{TA}=2313\mu\text{mol}/\text{kg}} = -5.4, \\ (\partial[\text{CO}_3^{2-}]/\partial\text{TC})_{\text{TA}=2313\mu\text{mol}/\text{kg}} &= -0.56 \end{aligned} \quad (2)$$

for mixed layer conditions. Calculated at ocean mixed-layer conditions, the current $p\text{H}$ is 8.2 and $[\text{CO}_3^{2-}]$ is $114 \mu\text{mol}/\text{kg}$. For sea water at equilibrium with the atmosphere, increases in atmospheric CO_2 from a pre-industrial value of 285 to 350 μatm would have already decreased the $p\text{H}$ by 0.079 and $[\text{CO}_3^{2-}]$ by $20 \mu\text{mol}/\text{kg}$ (15% of the pre-industrial value). Addition of alkalinity to the mixed layer of the ocean could be used to keep ocean $p\text{H}$ and $[\text{CO}_3^{2-}]$ closer to their pre-anthropogenic values, since the effect is counter that of increasing CO_2 (and TC) as is evident by comparing Eqs. (1) and (2).

3. THE EFFECTIVENESS OF METHODS TO INCREASE OCEAN ALKALINITY

Few alkaline minerals present in the Earth's crust are readily soluble, and none of these are recoverable in large quantities. Soda ash (Na_2CO_3) is soluble in sea water and its dissolution results in two mole equivalents of TA increase per mole of Na_2CO_3 dissolved. There would be 0.79 mole of CO_2 uptake per mole Na_2CO_3 dissolved through the net reaction

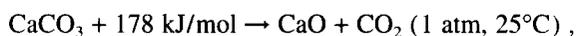


The fractionation of carbon between HCO_3^- and CO_3^{2-} follows from Eq. (1). Two moles of Na^+ lead to an increase in alkalinity by two moles which leads to an increase in TC of 1.79 moles, given by Eq. (1), of which one mole is supplied by the Na_2CO_3 and 0.79 mole is supplied by dissolution of atmospheric CO_2 . Changes in OH^- and H^+ as well as minor constituents such as H_2CO_3 , H_2BO_3^- and H_3BO_3

not listed in the net reaction account for the small discrepancy in the stoichiometry of hydrogen. The largest reserves of soda ash are found in the form of trona in Wyoming, U.S.A. The estimated world recoverable reserves of soda ash are 4.9×10^{14} moles Na₂CO₃.¹⁰ If the entire world's recoverable reserves of Na₂CO₃ were dissolved in sea water, this would offset only one year's emissions from the combustion of fossil fuels at the current rate of 6 Gton C/yr = 5×10^{14} mol CO₂/yr.

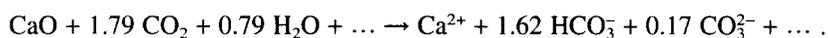
Limestone (CaCO₃), on the other hand, is one of the most prevalent minerals in the Earth's crust. It is mined at a rate of 3 Gton CaCO₃/yr (3×10^{13} mol/yr), a mass rate comparable to coal, primarily for use as a building material.¹¹ In nature, limestone dissolves and precipitates at a slow rate (about 1×10^{13} mol Ca/yr) and this would be slower if it were not for biological activity.^{2,8,9} Technology has long existed to extract soluble calcium from limestone. While more effective methods may be discovered for this purpose, the known technology for thermal decomposition of limestone to lime (CaO) will now be considered.

Limestone decomposes to CO₂ and lime, which is readily soluble, at a temperature of roughly 1200°C. The decomposition temperature depends on *p*CO₂ within the limestone which is often high because of mass-transfer limitation. Limestone calcination has long been carried out in kilns fueled by fossil or biomass fuels. Production of lime has three steps: mining of limestone, milling of the limestone to stones of the optimal size, and calcination in a kiln. The cost of lime production is dominated by kiln operation because it is energy intensive. Energy requirements stem from the latent heat of the decomposition reaction



and the net sensible heat losses which have been reduced to roughly 10% of the heat requirements in modern kilns.¹¹

Lime, once produced, could then be either shipped to the ocean for dissolution, or dissolved and pumped to the ocean if there were a local water supply. The details and local environmental impacts of the lime distribution system have yet to be addressed. The dissolution of one mole of CaO results in the increase of TA by two mole equivalents. There would be 1.79 moles of CO₂ uptake per mole CaO dissolved through the net reaction



The fractionation of carbon between HCO₃⁻ and CO₃²⁻ follows again from Eq. (1). The dissolution of 1.79 moles of CO₂ to form 1.79 moles of HCO₃⁻ plus CO₃²⁻ requires 1.79 moles of O. The oxygen is provided by 0.79 mole H₂O and 1 mole CaO. Changes in OH⁻, H⁺, H₂CO₃, H₂BO₃, and H₃BO₃ not listed in the net reaction account for the small discrepancy in the stoichiometry of hydrogen. Examination of CO₂ exchange with the atmosphere and mixing between the mixed layer and the deep ocean shows that if CaO were dissolved in the mixed layer, then the CO₂ would be absorbed from the atmosphere before the Ca²⁺ had time to mix to the deep ocean.⁴ As the high-alkalinity water eventually mixed with the deep ocean, dissolved carbon would be carried along to the deep ocean as well. The system to produce soluble lime will require considerable energy, and the beneficial absorption of CO₂ by the oceans can be (partially) offset by CO₂ emissions during limestone processing. The effectiveness of conceptual systems will be considered next.

In existing kilns, the limestone is mixed with the fuel and air is passed through the mixture to allow combustion. The CO₂ produced by limestone decomposition is emitted along with that produced by combustion. If existing kiln designs were used to produce lime destined to increase ocean alkalinity (System 1), then the emissions of CO₂ offset much of the additional CO₂ sequestered by the oceans. Another option is to capture the CO₂ produced by decomposition and/or combustion and store this CO₂ in, e.g., underground or undersea reservoirs as has been considered¹²⁻¹⁵ for power plant emissions. Two approaches to capture of CO₂ are proposed here. In the first (System 2), the combustion process is kept separate from decomposition, which will require some effective (new) technology for transferring combustion heat to calcine limestone. The decomposition gases will be primarily CO₂ (and water) which can be dried and compressed for transport to a storage site, thus avoiding the energy-intensive separation of a large gas stream of CO₂ mixed with nitrogen. The combustion gases in System 2 are released to the atmosphere, although technology similar to that used for the capture of power plant

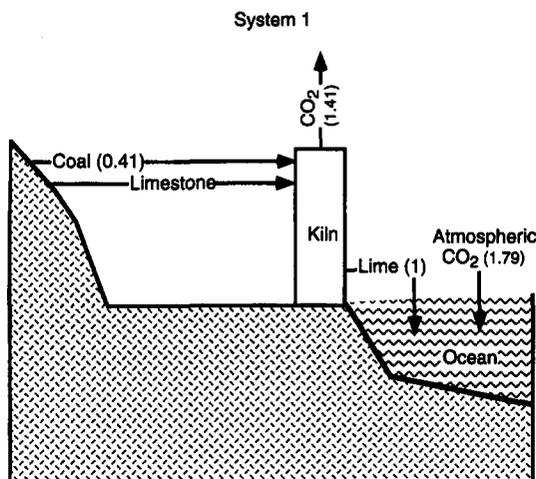


Fig. 1. Schematic of a conceptual process with limestone converted to lime by a high-efficiency kiln using existing technology. Combustion and decomposition gases are emitted to the atmosphere. Dissolution products of lime are put into the ocean, resulting in enhancement of the oceanic uptake of atmospheric CO₂. The estimated mole flow rates relative to that of produced lime are based on Table 1 and are shown in parentheses. The production of 1 mole of CaO requires 0.41 mole of carbon in coal, and 1 mole of CaCO₃. Combustion of C and decomposition of CaCO₃ results in 1.41 moles CO₂ emitted to the atmosphere. For each mole CaO dissolved in the ocean, 1.79 moles of CO₂ are absorbed by the ocean resulting in a net removal of $1.79 - 1.41 = 0.38$ mole CO₂ per mole CaO produced.

CO₂ could be alternatively used for the capture of combustion CO₂. In the second approach (System 3), oxygen separated from air is used for combustion in the kiln. With this approach, both the combustion and decomposition gases will be primarily CO₂ and water, and so the entire effluent can be dried, compressed and stored. In this way, System 3 avoids the need to separate the combustion and decomposition processes as was required in System 2.

The requirements of the three conceptual systems, sketched in Figs. 1–3, are listed in Table 1. All assume that the source of energy is coal. The basis for the mole flow rates shown in Figs. 1–3 is one mole of lime produced. Alternatively, the basis for the comparison in Table 1 is net removal of one mole of CO₂ from the atmosphere, i.e. the enhanced amount of CO₂ absorbed by the oceans as a result

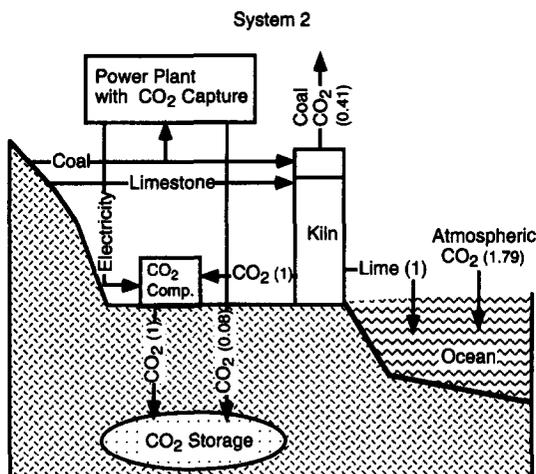


Fig. 2. Schematic of a conceptual process with limestone converted to lime by a high-efficiency kiln with the capture of the CO₂ from limestone decomposition. Combustion gases are emitted to the atmosphere. Decomposition CO₂ is captured and stored. Dissolution products of lime are put into the ocean, resulting in enhancement of the oceanic uptake of atmospheric CO₂. The estimated mole flow rates relative to that of produced lime are based on Table 1 and are shown in parentheses. For each mole of CaO produced, one mole of CO₂ from CaCO₃ decomposition must be compressed and stored, and electricity is required for this task. Electricity requires an additional 0.08 mole of carbon in coal to be burned for power generation, and requires the additional storage of 0.08 mole of CO₂. An efficiency of 0.3 (see Table 1) is used for power generation, and this efficiency accounts for the energy required for capture and storage of CO₂ generated by the power plant.

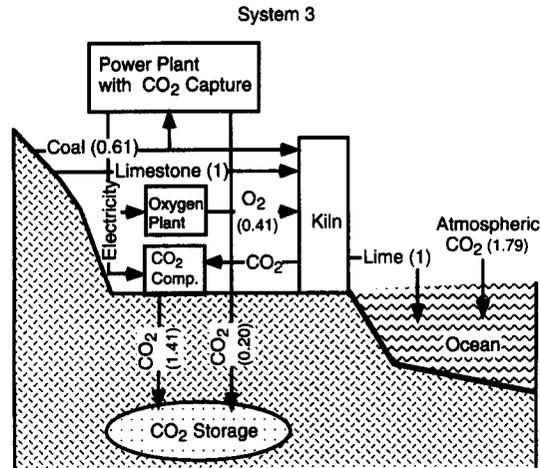


Fig. 3. Schematic of a conceptual process with limestone converted to lime by a high-efficiency kiln with oxygen feed. Combustion and decomposition CO₂ is captured and stored. Dissolution products of lime are put into the ocean, resulting in enhancement of the oceanic uptake of atmospheric CO₂. The estimated mole flow rates relative to that of produced lime are based on Table 1 and are shown in parentheses. For each mole of CaO produced, 0.41 mole of carbon in coal is burned in the kiln, 1 mole of CO₂ results from CaCO₃ decomposition, and 0.41 mole of oxygen must be separated from air. Electricity is required to separate the oxygen and compress the 1.41 moles CO₂ for storage. Electricity requires an additional 0.20 mole of carbon in coal to be burned for power generation, and requires the additional storage of 0.20 mole of CO₂. The capture and storage of power plant CO₂ is assumed to be accomplished through a technology, not necessarily dependent on oxygen separation.

Table 1. Analysis of systems to sequester CO₂ from the atmosphere by converting limestone to lime and then dissolving the lime in the mixed layer of the oceans to increase ocean alkalinity.

System	Requirements for Net Removal (Absorption - Emissions) per mol CO ₂ from the Atmosphere		
	Coal Consumed (mol C in Coal)	Lime Dissolved (mol CaO)	CO ₂ Stored (mol CO ₂)
1. Combustion & Decomposition Gases Emitted	1.10	2.66	0
2. Combustion Gases Emitted, Decomposition Gases Stored	0.36	0.73	0.79
3. Combustion & Decomposition Gases Stored	0.34	0.56	0.90

Total Heat Required for Kiln Operation ¹⁰	200 kJ/mol CaO
CO ₂ Sequestered per Mole CaO Dissolved (1)	1.79 mol CO ₂ /mol CaO
Coal Heat Value ^{18, 19}	29,000 kJ/kg Coal
Coal Carbon Content ¹⁸	60 mol C/kg Coal
Electricity Required for CO ₂ Compression from 1 to 60 atm ²⁰	12 kJ Elec./ mol CO ₂
Electricity Required for O ₂ Separation from the Atmosphere ²¹	28 kJ Elec./ mol O ₂
Efficiency of Coal Fueled Power Plant with CO ₂ Capture ²²	0.3 kJ Elec./kJ Coal

of increased ocean alkalinity minus any CO_2 emitted to the atmosphere by system operations. Three requirements are listed in Table 1 that could limit the effectiveness of a system: the quantity of coal required, the quantity of limestone required which is equal to the quantity of lime dissolved in the oceans, and the quantity of CO_2 that must be stored.

In System 1, lime is produced and both the combustion and the thermal decomposition gases are emitted to the atmosphere; this is the conventional method for lime production. This case requires, as seen in Fig. 1, the least quantity (of the three systems) of coal to produce lime and no storage site for CO_2 . However, the net amount of CO_2 removed from the atmosphere per amount of coal consumed, listed in Table 1, is the lowest of the three systems. This is because the emissions of decomposition CO_2 (1 mole per mole lime produced) plus combustion CO_2 (0.41 mole per mole lime produced) offsets most of the enhanced ocean uptake of CO_2 (1.79 moles per mole lime dissolved).

In System 2, combustion gases are kept separate from thermal decomposition gases, which are compressed and stored as shown in Fig. 2. The electricity used for CO_2 compression is assumed to be produced by coal and the emissions captured and stored. Note that there are many alternatives for power plant CO_2 capture;¹⁶ the efficiency listed in Table 1 represents a high efficiency technology with coal as the energy source. In this system, the coal (including the coal for electricity generation) and limestone requirements for sequestering atmospheric CO_2 are significantly less per unit of CO_2 sequestered in the ocean than in System 1, however, the quantity of CO_2 that must be stored is comparable to the net amount of CO_2 that is removed from the atmosphere.

In System 3, oxygen is separated from air and then fed into the kiln resulting in a concentrated CO_2 stream of both the combustion and decomposition gases which are compressed and stored as shown in Fig. 3. In this system electricity is used for both oxygen separation and CO_2 compression. This system requires slightly less coal and limestone and slightly more CO_2 storage per unit of CO_2 sequestered in the ocean than System 2. Both Systems 2 and 3 require the storage of large quantities of CO_2 , and sites^{13,17} for storage may be limited. And both require kilns which produce concentrated CO_2 , an unproven technology, at efficiencies equal to kilns (without CO_2 capture) of the best current technology.

The requirements and limitations of the capture and storage of power plant CO_2 (to avoid CO_2 emissions) is now compared to the requirements and limitations of Systems 2 and 3 proposed here. Leading technologies for the capture (including compression) of power plant CO_2 require (compare to Table 1) no limestone, an additional 0.25 mole of carbon in coal to avoid one mole CO_2 emissions and, correspondingly, require the storage of 1.25 moles CO_2 .¹⁸ In addition, the capture of power plant emissions is limited to those power plants that are close to storage sites, a small fraction of the total anthropogenic emission of CO_2 .

Each of the industrial systems, including the capture and disposal of power plant CO_2 , has different requirements. Power-plant-generated CO_2 must be captured and stored at the time of power generation and near the place of power generation which is often near population centers. Systems for increasing ocean alkalinity, and thereby sequestering atmospheric CO_2 in the oceans, need not be carried out near the place nor at the time of anthropogenic CO_2 emissions. Of course, all the methods for increasing ocean alkalinity must be carried out in close proximity to the oceans. In addition, Systems 2 and 3 need to be carried out at a location near a CO_2 storage site. If other non-fossil-fuel sources of energy which may become available in the future could be substituted for coal-fueled lime production, methods for increasing ocean alkalinity become even more effective means for sequestering CO_2 from the atmosphere. However, non-fossil forms of energy would likely be better used to offset the CO_2 produced by fossil fuel energy production, unless the time or the place at which the non-fossil forms of energy became available did not correspond to the demand for energy.

The methods for increasing ocean alkalinity that have been discussed in this section were chosen to show the tradeoffs between requirements for methods for which we know enough about to make a preliminary assessment. The choice of methods is not meant to be inclusive; for example, there are many other combinations of technologies for lime production. Nor does this analysis consider the details of the science and technologies that would be required before inclusion of this concept as a strategic response option; methods for mineral dissolution and the distribution of dissolution products, along with the concomitant environmental impacts, require study. A further search for less energy-intensive means for increasing ocean alkalinity may also be prudent. A natural response of the Earth system to increases in atmospheric CO_2 is the enhanced rate of alkaline mineral dissolution, although this feedback is thought to be too slow to significantly affect atmospheric CO_2 over the time scale of 100 years or

less. Explored in this paper are just one class of methods to accelerate this feedback, and others may well exist.

4. CONCLUSIONS

A preliminary analysis of a geoengineering option based on the intentional increase of ocean alkalinity to enhance marine storage of atmospheric CO₂ has been made. Ocean alkalinity might be raised by introducing the dissolution products of alkaline minerals into the oceans. Naturally occurring soda ash (Na₂CO₃) is readily soluble and easily mined. However, the world's recoverable resources of soda ash are estimated to be sufficient to sequester only enough CO₂ from the atmosphere to offset 1 year of fossil-fuel emissions. Limestone (CaCO₃) is abundant in the Earth's crust but is not readily soluble. Limestone can be converted into soluble lime (CaO) by thermal decomposition, an energy-intensive process that also releases CO₂ from the limestone. The resulting CO₂ can be stored in underground or undersea reservoirs. Estimates of resource requirements are made for three conceptual systems for increasing ocean alkalinity. Increasing ocean alkalinity might have the environmental benefit of chemically buffering the oceans to the increased concentration of CO₂, although there might be impacts by contaminants or local pH effects which would have to be evaluated.

Increasing ocean alkalinity is a means by which CO₂ might be sequestered from the atmosphere, but not without significant consumption of energy resources and significant cost. Nevertheless, this approach appears to be limited only by the availability of energy. Thermal decomposition of limestone to raise ocean alkalinity is one route to sequestering CO₂ from the atmosphere and is comparable to methods for avoiding power-plant emissions by capture and storage of CO₂. Obviously, the unknown potential side effects of most geoengineering options to limit climate change, including this option to enhance ocean alkalinity, make them unsuitable candidates for implementation in the near term. However, they should be assessed to determine their potential to contribute to development of a set of long-term, strategic response options, as knowledge improves.

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